

# Natural Radionuclides in Cultivated and Virgin soil of the Same Origin Using NaI Gamma-ray Spectrometer and the Potential Phosphate Fertilizers Impacts

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**Abstract:** Soil samples were collected from an arid environment in the central region of Saudi Arabia, 28 samples from selected 14 locations within an agricultural farm (300 km<sup>2</sup> area). Two composite samples, cultivated (C) and virgin (V) soils, of the same origin were collected from each location. This work aims to evaluate the long term (25 years) impacts of various agricultural practices on the activity concentration of some naturally occurring radionuclides (NOR) and some of soil's chemo-physical parameters. The activity concentration, in Bq/kg, of <sup>226</sup>Ra (<sup>238</sup>U series), <sup>228</sup>Ra (<sup>232</sup>Th series) and <sup>40</sup>K were measured using well calibrated gamma-ray spectrometer based on NaI (TI) detector. The soil physical and chemical properties [e.g. pH, EC, particle size distribution (clay, silt and sand percentages), CaCO<sub>3</sub> %, soluble cations (Ca, Mg, Na and K) and soluble anions (CO<sub>3</sub>, HCO<sub>3</sub>, Cl and SO<sub>4</sub>)] were determined. The radium equivalent activity, in Bq/kg was calculated. Generally, there were no noticeable changes that could be related to agricultural practices and/or strong correlations between natural radionuclide and soil's chemo-physical parameters. That could be due to the sandy nature of the soil, the applied agricultural practices, and the effects of adsorption-filtration processes on the behavior and the distribution pattern of NOR in arid environment. Therefore, the environmental impacts of different man-made activities on underground resources should be carefully considered due to the possible filtration behavior of different pollutants in dry-land environment.

**Keywords:** Soil, gamma-ray spectrometry; Natural radionuclides, Phosphate fertilizer.

## 1 Introduction

Soil is a dynamic earth's top layer in which many chemical, physical and biological activities are going on constantly. In soils, metals and radionuclides can dissolve in solution, or undergo ions exchange processes. They can also form complexes with soil organics or inorganic solid constituents. Soluble constituents are subject to migration with soil water, uptake by plants or loss due to infiltration into deep soil's layers [1].

Naturally occurring radionuclides (NOR), <sup>238</sup>U series, <sup>232</sup>Th series and <sup>40</sup>K, have a wide range of concentrations in soils that depends on their origins, their physical and chemical properties, and on some man-made activities such as agricultural application of phosphate fertilizers and different ores mining processes [2]. The long-term soil

cultivation and application of organic and inorganic fertilizers could affect various soil's physico-chemical properties, nutrient content as well as the concentration of various kinds of potential contaminants. For example, long-term phosphate fertilization could elevate and redistribute some natural radionuclides especially uranium-238 series radionuclides within the soil environment and soil profiles, altering their availability within the soil-water- plant system and consequently leading to their potential enhanced transfer to the food chain, mainly in acidic soils. It can also raise-up the concentration of these elements in irrigation runoff/drainage waters [3, 4, 5]. This work aims at study of the alteration of some natural radionuclides (<sup>226</sup>Ra, <sup>228</sup>Ra and <sup>40</sup>K) activity concentration and soil's physicochemical properties of cultivated and virgin soil sample of the same origin after long time (25 year) of different agricultural practices in an arid environment in Saudi Arabia.

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## 2 Experimental Works

### 2.1 Study Area

Hail agricultural development company's (HADCO) farm is located in Hail province in the Northwest to center of Saudi Arabia, between latitudes 26.8–28.3°N and longitude 41.1–43.5°E. It is the oldest agricultural company in Saudi Arabia, established on 1982 and covers an area of about 300 km<sup>2</sup>. The Saq groundwater aquifer bore wells are the only water resource for the agricultural activities using a central pivot irrigation system [6]. Cultivated soils were composed of reclaimed circles and each occupies an area of about 80 hectare.

### 2.2 Sampling and Samples Preparation

Twenty-eight top soil (20 cm depth) samples were collected from 14 cultivated circles within HADCO agricultural farm using core method. Each cultivated (C) soil sample was a composite sample where 6 subsamples were collected and well mixed to represent the soil inside each circle. The corresponding virgin (V) soil sample was collected from the area outside the circle that was assumed to have the same origin. Samples were dried at about 80 °C, then pulverized, homogenized and sieved through a 2 mm mesh sieve [7].

### 2.3 Analytical Techniques

Gamma spectrometric analysis; the dried samples were transferred to polyethylene containers of 100 cm<sup>3</sup> capacity and sealed at least for 4 weeks to reach secular equilibrium between radium and thorium, and their progenies. The activity concentration in Bq/kg dry weight of <sup>226</sup>Ra (<sup>238</sup>U) series, <sup>232</sup>Th series and <sup>40</sup>K were measured using well calibrated gamma spectrometry based on 3''x3'' NaI detector. The detector has full width at half maximum (FWHM) of 6% for <sup>137</sup>Cs gamma energy line at 661.6 keV is coupled with 14 pin photomultiplier tube (PMT) and Canberra DSA 1000 that include a power supply, amplifier and multichannel analyzer (MCA) of 8 k channels. The gamma spectra were measured and analyzed using Genie 2000 software by Canberra. The detector was housed inside low background lead shield of 10 cm thick and 0.8 mm thick copper sheet. The efficiency calibration of the gamma-ray spectrometer was performed using three standard reference materials RGU-1 (uranium ore), RGTh-1 (Thorium ore) and RGK-1 (Potassium sulphate) that were obtained from International Atomic Energy Agency (IAEA) [8,9]. The gamma transmissions used for activity calculations and spectrometer calibration for <sup>40</sup>K, <sup>226</sup>Ra and <sup>228</sup>Ra were 1461, 1764 (<sup>214</sup>Bi) and 2614 (<sup>208</sup>Tl) keV, respectively [10, 11].

Theoretical calculation of radium equivalent value (Ra-eq) in Bq/kg was described in other reference [10]. Physical and chemical soil saturation percentage (SP), pH, electric conductivity (EC), clay %, Silt % and sand%] and chemical

properties, soluble cations and soluble anion in meq/L (CaCO<sub>3</sub>, Ca, Mg, Na, K, CO<sub>3</sub>, HCO<sub>3</sub>, Cl and SO<sub>4</sub>) were determined using standard methods that were described by [12].

## 3 Results and Discussion

The activity concentrations in Bq/kg dry weight of <sup>226</sup>Ra, <sup>228</sup>Ra and <sup>40</sup>K using NaI gamma-ray spectrometer, and calculated radium equivalents (Ra-Eq), and activity ratios of <sup>226</sup>Ra/<sup>228</sup>Ra and <sup>40</sup>K/<sup>226</sup>Ra in both cultivated (C) and virgin (V) soil of the same origin were given in Table 1. Some of their descriptive parameters (mean, standard error, standard deviation, minimum and maximum) were given in Table 2 and shown in figures 1 and 2. It is well known that the activity concentrations of natural radionuclides (such as <sup>238</sup>U series, <sup>232</sup>Th series and <sup>40</sup>K) in soil varied widely where the world population weighted average activity concentration (range) of <sup>238</sup>U, <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in soil were 33 (16-110), 32 (17-60), 45 (11-64) and 420 (140-850) Bq/kg, respectively [2].

In General the physico-chemical properties of soils depend on various factors such as the type of the source rock from which soils were formed and the land-use. Agricultural processes could change soils' physico-chemical and biological properties as well as their elemental and compounds constituents [13, 14, 15]. The average activity concentrations ±standard error (range) of <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>40</sup>K and calculated Ra-eq, Table 2, were 20.92±1.29 (12.22-28.10), 25.97±1.99 (15.57-41.07), 553.96±8.05 (502.98-597.53) and 98.83±3.31(81.72-125.72) Bq/kg dry weight for cultivated soil samples, respectively; 18.62±1.60 (7.95-26.61), 23.28±2.11 (11.50-35.50), 536.91±13.02 (460.64-609.13), and 93.19±3.58 (75.74-114.19) Bq/kg dry weight for virgin soil samples of the same origin, respectively. The comparison between the average activities of <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>40</sup>K and calculated Ra-Eq (Bq/kg dry weight) in all, cultivated and virgin soil samples of the same origin was shown in Figure 1.

Although their average values could show a slight elevation in cultivated soil over the virgin soil samples that will be vanished if the standard deviation values were considered. The individual data for <sup>226</sup>Ra indicate that 9 locations have not shown noticeable variation between their activity concentrations of the cultivated and virgin soils due to long term cultivation (about 25 years). However, <sup>226</sup>Ra activity concentrations have been increased and noticed in soil in 4 cultivated locations and only at one location in virgin soil. The activity concentrations of <sup>228</sup>Ra have not been changed at 5 locations, and enhanced at 5 cultivated soils and one uncultivated soil. The pattern of radium isotopes variation in the studied locations could be dependent on their original concentrations in soils at each location that affected by the soil origin (rock type from which soil were formed) and their addition through cultivation processes. Usually <sup>226</sup>Ra concentrations in phosphate deposits (and consequently

phosphate fertilizers) are higher than  $^{228}\text{Ra}$  concentrations [10]. Potassium-40 ( $^{40}\text{K}$ ) concentration have not been changed at 4 locations, and enhanced at 6 cultivated soils and 4 uncultivated soils. High solubility and competing chemical behavior of potassium, and application of fertilizers containing potassium regularly could be considerably appreciated in order to understand their concentration variation in relation to agricultural practice. Radium equivalent values have not been changed at 2 locations, and have been enhanced at 10 cultivated soils and 2 virgin soils.

The average activity concentration ratios  $\pm$  standard error (range) of  $^{226}\text{Ra}/^{228}\text{Ra}$  and  $^{40}\text{K}/^{226}\text{Ra}$  were  $0.77\pm 0.07$  (0.41-1.22) and  $28.17\pm 2.18$  (17.90-44.97) in cultivated soil samples, respectively;  $0.88\pm 0.10$  (0.38-1.37) and  $32.79\pm 3.76$  (19.02-65.24) in virgin soil of the same origin, respectively. These ratios clear up that the activity concentrations of  $^{228}\text{Ra}$  were higher than that of  $^{226}\text{Ra}$  in some of the soil samples. While the chemical behavior of both isotopes are identical but their activity concentrations depend on their concentration in soil's parent rock as well as the additional input due to land use as an example phosphate fertilizers could be a source of some natural radionuclides and other contaminants into soils' ecosystem [4,5].

Descriptive statistical parameters (mean, standard error, median, standard deviation, sample variance, kurtosis, skewness, minimum, maximum, sum and count) of soil physical [saturation percentage (SP), pH, electric conductivity (EC), clay %, Silt % and sand%] and chemical properties, soluble cations and soluble anion in meq/L ( $\text{CaCO}_3$ , Ca, Mg, Na, K,  $\text{CO}_3$ ,  $\text{HCO}_3$ , Cl and  $\text{SO}_4$ ) were given in table 3.

The correlation coefficients between the activity concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{40}\text{K}$ , and physical [saturation percentage (SP), pH, electric conductivity (EC), clay %, Silt % and sand%] and chemical properties, soluble cations and soluble anion in meq/L ( $\text{CaCO}_3$ , Ca, Mg, Na, K,  $\text{CO}_3$ ,  $\text{HCO}_3$ , Cl and  $\text{SO}_4$ ) in cultivated (C) and virgin (V) soil samples of the same origin were given in table 4. There is no noticeable strong correlation between natural radionuclides  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{40}\text{K}$ , and physical [saturation percentage (SP), pH, electric conductivity (EC), clay %, Silt % and sand%] and chemical properties, soluble cations and soluble anion in meq/L ( $\text{CaCO}_3$ , Ca, Mg, Na, K,  $\text{CO}_3$ ,  $\text{HCO}_3$ , Cl and  $\text{SO}_4$ ) Table 4: The correlation coefficient between the activity concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{40}\text{K}$ , and soil's physical and chemical properties, and soluble cations and soluble anion in cultivated (C) and virgin (V) soil samples of the same origin. For example, the correlation coefficient between soluble K and  $^{40}\text{K}$  in C and V soils were -0.03 and 0.26 respectively. Although, the K contain fertilizers increase its concentration in C soils but due to the relatively high solubility of K and plants intake, the soluble and geogenic origin K in soil could behave

differently.

The ratios (cultivated to virgin - C/V- soil samples of the same origin) of activity concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{40}\text{K}$  and calculated radium equivalent (Ra-Eq), and activity concentration ratios of  $^{226}\text{Ra}/^{228}\text{Ra}$  and  $^{40}\text{K}/^{226}\text{Ra}$ , and their descriptive statistical parameters (mean, standard error, median, standard deviation, sample variance, kurtosis, skewness, minimum, maximum, sum and count) were given in tables 5 and 6, and shown in figure 2. The average values (C/V), table 5, for  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and calculated radium equivalent (Ra-Eq) were higher than unity that could give an indication of changes in their activity concentration due to agricultural processes. While, ratio (C/V) values for  $^{40}\text{K}$ ,  $^{226}\text{Ra}/^{228}\text{Ra}$  and  $^{40}\text{K}/^{226}\text{Ra}$  were close to unity that gave an oppose indication. The range of these ratios (C/V) were varied widely; (0.54-2.99) for  $^{226}\text{Ra}$ , (0.48-2.62) for  $^{228}\text{Ra}$ , (0.9-1.30) for  $^{40}\text{K}$ , (0.76-1.54) for Ra-Eq, (0.30-2.82) for  $^{226}\text{Ra}/^{228}\text{Ra}$  and 0.38-1.73) for  $^{40}\text{K}/^{226}\text{Ra}$ . For more deep analysis of C/V ratios, less than unity ratio indicates decreasing of radionuclide due to agricultural processes, which was observed or encountered for in 2, 1 and 0 sites for  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{40}\text{K}$ , respectively. Ratios (C/V) less than unity were observed for in 6, 7 and 2 sites for  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{40}\text{K}$ , respectively. Ratios (C/V) equal  $1\pm 0.1$  were observed for in 4, 6 and 12 sites for  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{40}\text{K}$ , respectively. The variation in the elemental concentrations in soil could be due to the application of phosphate fertilizers and other agricultural chemical, dilution when organic matter and soil's water increases, migration of elements within the soil, leaching and absorption, uptake by plants and other various reasons [16]. The average specific activities of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$ , and Rad-Eq value have not shown any obvious changes due to agricultural processes especially due to continuous application of phosphate fertilizers. Khater 2008, report that the average concentrations (Bq/kg)  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$ , and Rad-Eq value in granule compound phosphate fertilizers used in Saudi Arabia were 75, 23, 2059 and 226 Bq/kg and annual application rate of about 600 kg/ha. Continuous fertilization will add annually more natural radionuclides and their added concentrations will depend on their average concentrations in fertilizers and application rate. While their variation in soil before and after cultivation will depend additionally on the geochemical behavior of each radioelement and soil physical and chemical properties [4]. The behavior of radionuclides depends on the elemental properties of radionuclides, on the mineral and organic inventory of the soil and the chemical reaction milieu. Many processes are ran simultaneously [17]. Descriptive statistical parameters of the ratios (cultivated to virgin - C/V- soil samples of the same origin) of Physical [saturation percentage (SP), pH, electric conductivity (EC), clay %, Silt % and sand%] and chemical properties, soluble cations and soluble anion in meq/L ( $\text{CaCO}_3$ , Ca, Mg, Na, K,  $\text{CO}_3$ ,  $\text{HCO}_3$ , Cl and  $\text{SO}_4$ ) were given in table 7. These data indicate that there were no obvious changes in physical properties e.g. SP, pH, EC, clay %, silt % and sand % and

some chemical properties e.g. concentration of Mg, K, CO<sub>3</sub>, and Cl.

The activity concentrations <sup>226</sup>Ra, <sup>228</sup>Ra and <sup>40</sup>K in Bq/kg in cultivated (C) and virgin (V) soils and their ratio (C/V) in different countries were given in table 9. It was noticeable

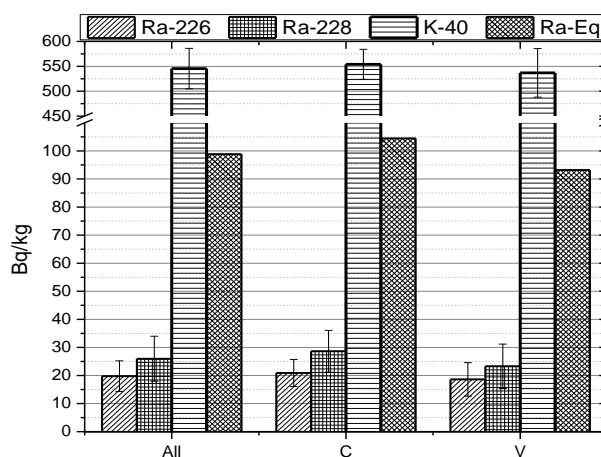
that the activity concentration of natural radionuclides in soil samples within the world range. The ratios C/V values of <sup>40</sup>K were obviously unchanged that could be explain due to the balance between soluble K input into soil, plants uptake and infiltration, and insoluble geogenic origin K inside the soil matrix.

**Table 1:** Activity concentration in Bq/kg dry weight of Ra-226, Ra-228 and K-40 using NaI gamma-ray spectrometer, and calculated radium equivalent (Ra-Eq), and activity ratios of Ra-226/Ra-228 and K-40/Ra-226 in cultivated (C) and virgin (V) soil of the same origin.

Ser	Code	Ra-226	Error	Ra-228	Error	K-40	Error	Ra-Eq	Ra226/ Ra228	K40/ Ra226
1	C2 – C	21.56	3.08	33.87	2.73	504.93	15.60	109	0.64	23.42
2	C2 – V	22.90	2.16	18.16	2.97	560.22	11.54	92	1.26	24.46
3	E10 – C	23.89	2.23	33.79	2.60	535.39	14.88	113	0.71	22.41
4	E10 – V	14.12	3.00	34.92	2.55	578.12	12.66	108	0.40	40.93
5	E14 – C	28.10	2.57	23.10	2.64	502.98	12.52	100	1.22	17.90
6	E14 – V	24.25	2.94	23.86	2.90	495.75	13.24	96	1.02	20.44
7	E17 – C	24.59	2.99	41.07	3.17	551.91	13.08	126	0.60	22.45
8	E17 – V	23.16	2.99	35.50	3.21	524.14	13.31	114	0.65	22.63
9	E20 – C	18.36	2.95	15.57	3.14	595.43	12.80	86	1.18	32.43
10	E20 – V	21.26	2.44	32.43	3.18	590.65	13.47	113	0.66	27.79
11	F2 – C	20.00	3.15	35.77	3.10	535.65	13.66	112	0.56	26.78
12	F2 – V	26.61	2.19	25.49	2.75	506.12	13.06	102	1.04	19.02
13	F5 – C	17.50	2.20	23.11	2.66	547.06	11.60	93	0.76	31.26
14	F5 – V	12.29	2.34	24.38	2.96	478.70	12.59	84	0.50	38.95
15	H1 – C	12.86	2.23	18.14	2.72	558.41	11.78	82	0.71	43.42
16	H1 – V	10.93	2.27	12.57	2.91	609.13	11.15	76	0.87	55.74
17	H11 – C	20.76	3.06	36.86	3.01	597.53	12.97	119	0.56	28.78
18	H11 – V	20.42	1.94	15.02	2.69	460.64	10.92	77	1.36	22.56
19	H15 – C	17.64	2.64	28.53	3.44	580.44	13.70	103	0.62	32.90
20	H15 – V	13.80	2.27	28.99	2.46	513.18	12.06	95	0.48	37.20
21	H18 – C	12.22	2.52	29.79	2.80	549.37	12.86	97	0.41	44.97
22	H18 – V	22.72	1.93	16.62	0.00	590.25	11.39	92	1.37	25.98
23	J20 – C	25.38	2.53	30.09	3.13	553.05	13.77	111	0.84	21.79
24	J20 – V	15.65	2.76	11.50	3.05	593.64	12.05	78	1.36	37.93
25	J6 – C	26.28	2.43	29.58	2.59	548.54	13.60	111	0.89	20.87
26	J6 – V	24.69	1.98	25.85	2.48	497.51	11.59	100	0.96	20.15
27	K3 – C	23.79	1.78	21.93	2.18	594.69	11.95	101	1.08	24.99
28	K3 – V	7.95	2.44	20.69	2.83	518.70	12.55	77	0.38	65.24

**Table 2:** Descriptive statistical parameters of activity concentration in Bq/kg dry weight of Ra-226, Ra-228 and K-40 using NaI gamma-ray spectrometer, and calculated radium equivalent (Ra-Eq), and activity ratios of Ra-226/Ra-228 and K-40/Ra-226 in (I) all, (II) cultivated (C) and (III) virgin (V) soil of the same origin

		Ra-226	Ra-228	K-40	Ra-Eq	Ra-226/Ra-228	K-40/Ra-226
Mean	I	19.77	25.97	545.4	98.83	0.82	30.48
	II	20.92	28.66	553.96	104.48	0.77	28.17
	III	18.62	23.28	536.91	93.19	0.88	32.79
Standard Error	I	1.03	1.51	7.69	2.63	0.06	2.18
	II	1.29	1.99	8.05	3.31	0.07	2.18
	III	1.60	2.11	13.02	3.58	0.10	3.76
Standard Deviation	I	5.45	8.01	40.68	13.90	0.31	11.54
	II	4.82	7.44	30.11	12.37	0.24	8.16
	III	5.97	7.91	48.73	13.41	0.37	14.09
Minimum	I	7.95	11.50	460.64	75.74	0.38	17.90
	II	12.22	15.57	502.98	81.72	0.41	17.90
	III	7.95	11.50	460.64	75.74	0.38	19.02
Maximum	I	28.10	41.07	609.13	125.72	1.37	65.24
	II	28.10	41.07	597.53	125.72	1.22	44.97
	III	26.61	35.50	609.13	114.19	1.37	65.24



**Fig.1:** Average activity concentration in Bq/kg dry weight of Ra-226, Ra-228 and K-40 using NaI gamma-ray spectrometer, and calculated radium equivalent (Ra-Eq) in cultivated (C) and virgin (V) soil samples of the same origin.

**Table 3:** Descriptive statistical parameters of physical [saturation percentage (SP), pH, electric conductivity (EC), clay %, Silt % and sand%] and chemical properties, soluble cations and soluble anion in meq/L (CaCO<sub>3</sub>, Ca, Mg, Na, K, CO<sub>3</sub>, HCO<sub>3</sub>, Cl and SO<sub>4</sub>) cultivated (C) and virgin (V) soil of the same origin.

	Mean	Standard Error	Median	Standard Deviation	Sample Variance	Kurtosis	Skewness	Range	Minimum	Maximum	Count
SP	34.39	0.56	33.5	2.91	8.46	0.36	0.41	12.73	28.81	41.54	27
PH	7.88	0.04	7.88	0.2	0.04	0.07	-0.2	0.85	7.39	8.24	27
EC	5.42	1.01	3.8	5.25	27.61	2.1	1.61	20.15	0.85	21	27
Ca	24.2	4.93	13.75	25.63	656.7	1.67	1.57	86.75	3.75	90.5	27
Mg	9.84	1.57	6.76	8.16	66.58	2.08	1.52	33.28	1.3	34.58	27
Na	34.14	7.56	20.09	39.3	1544.31	1.18	1.49	137.49	0.51	138	27
K	0.67	0.14	0.41	0.73	0.53	4.17	2.06	2.82	0.12	2.93	27
CO <sub>3</sub>	0.53	0.03	0.5	0.17	0.03	1.58	0.77	0.75	0.25	1	25

HCO <sub>3</sub>	3.27	0.17	3	0.86	0.74	0.66	0.87	3.5	2	5.5	27
CL	39.7	8.79	22.5	45.69	2087.72	3.47	1.94	180	2.5	182.5	27
SO <sub>4</sub>	10.77	1.87	6	9.74	94.85	-0.62	0.81	33.1	0	33.1	27
Clay%	20.05	1.06	21.88	5.52	30.45	-0.33	-0.72	19.6	8.88	28.48	27
Silt %	13.96	1.18	14	6.13	37.58	1.14	0.35	29	1	30	27
Sand%	65.99	1.18	65.52	6.11	37.31	1.42	0.57	28.4	55.12	83.52	27
CaCO <sub>3</sub> %	7.5	0.5	6.77	2.61	6.8	0.34	0.7	10.92	2.97	13.89	27

**Table 4:** The correlation coefficient between the activity concentrations of <sup>226</sup>Ra, <sup>228</sup>Ra and <sup>40</sup>K, and physical [saturation percentage (SP), pH, electric conductivity (EC), clay %, Silt % and sand%] and chemical properties, soluble cations and soluble anion in meq/L (CaCO<sub>3</sub>, Ca, Mg, Na, K, CO<sub>3</sub>, HCO<sub>3</sub>, Cl and SO<sub>4</sub>) in cultivated (C) and virgin (V) soil samples of the same origin.

	All			Cultivated			Virgin		
	Ra-226	Ra-228	K-40	Ra-226	Ra-228	K-40	Ra-226	Ra-228	K-40
Ra-228	0.28			0.29			0.18		
K-40	-0.16	-0.10		-0.28	-0.24		-0.18	-0.16	
SP	0.18	0.47	0.03	0.20	0.29	0.10	0.09	0.60	-0.11
PH	0.13	0.12	0.36	0.03	-0.35	-0.24	0.11	0.29	0.53
EC	0.26	0.22	-0.10	0.39	0.22	-0.03	0.20	0.30	-0.12
Ca	0.22	0.14	-0.18	0.43	0.18	-0.10	0.14	0.23	-0.16
Mg	0.33	0.20	-0.09	0.43	0.20	-0.01	0.28	0.28	-0.13
Na	0.30	0.31	-0.10	0.45	0.42	-0.17	0.24	0.33	-0.02
K	0.12	-0.33	0.06	0.31	-0.03	-0.03	0.08	-0.52	0.26
CO <sub>3</sub>	-0.14	0.20	0.15	0.01	-0.03	0.23	-0.26	0.34	0.10
HCO <sub>3</sub>	0.10	0.23	0.29	0.10	-0.07	0.24	0.00	0.40	0.25
CL	0.25	0.27	-0.09	0.36	0.23	-0.04	0.18	0.38	-0.10
SO <sub>4</sub>	0.24	-0.11	-0.15	0.48	0.10	0.00	0.18	-0.16	-0.16
Clay %	-0.13	0.20	-0.21	-0.29	0.27	0.35	-0.14	0.00	-0.61
Silt %	0.05	0.13	0.00	0.33	-0.03	-0.14	-0.06	0.30	0.09
Sand %	0.07	-0.31	0.19	-0.03	-0.23	-0.20	0.18	-0.33	0.42
CaCO <sub>3</sub> %	-0.05	-0.22	-0.32	0.16	-0.52	-0.38	-0.17	0.02	-0.29

**Table 5:** The ratios of activity concentrations <sup>226</sup>Ra, <sup>228</sup>Ra and <sup>40</sup>K and calculated radium equivalent (Ra-Eq), and activity concentration ratios of <sup>226</sup>Ra/<sup>228</sup>Ra and <sup>40</sup>K/<sup>226</sup>Ra of cultivated (C) to virgin (V) soil samples of the same origin.

	C/V	R-226	Ra-228	K-40	Ra-Eq	Ra226/Ra228	K40/Ra226
1	C2 WI	0.94	1.86	0.90	1.18	0.50	0.96
2	E10 EI	1.69	0.97	0.93	1.04	1.75	0.55
3	E14 EI	1.16	0.97	1.01	1.03	1.20	0.88
4	E17 EI	1.06	1.16	1.05	1.10	0.92	0.99
5	E20 EI	0.86	0.48	1.01	0.76	1.80	1.17
6	F2 EI	0.75	1.40	1.06	1.10	0.54	1.41
7	F5 WI	1.42	0.95	1.14	1.10	1.50	0.80
8	H1 WI	1.18	1.44	0.92	1.08	0.82	0.78
9	H11 EI	1.02	2.45	1.30	1.54	0.41	1.28
10	H15 EI	1.28	0.98	1.13	1.09	1.30	0.88
11	H18 EI	0.54	1.79	0.93	1.06	0.30	1.73

12	J20 NI	1.62	2.62	0.93	1.43	0.62	0.57
13	J6 WI	1.06	1.14	1.10	1.11	0.93	1.04
14	K3 WI	2.99	1.06	1.15	1.30	2.82	0.38
	Ave	1.28	1.34	1.05	1.14	1.15	0.96

**Table 6:** Descriptive statistical parameters of the ratio of activity concentration of Ra-226, Ra-228 and K-40 and calculated radium equivalent (Ra-Eq), and activity ratios of Ra-226/Ra-228 and K-40/Ra-226 of cultivated to virgin soil samples of the same origin.

C/V	R-226	Ra-228	K-40	Ra-Eq	Ra226/Ra228	K40/Ra226
Mean	1.26	1.38	1.04	1.14	1.10	0.96
Standard Error	0.16	0.16	0.03	0.05	0.19	0.10
Median	1.11	1.15	1.03	1.10	0.92	0.92
Standard Deviation	0.59	0.61	0.12	0.19	0.69	0.36
Sample Variance	0.35	0.37	0.01	0.03	0.48	0.13
Kurtosis	5.72	0.27	0.11	1.63	1.55	0.41
Skewness	2.07	0.92	0.68	0.55	1.19	0.52
Minimum	0.54	0.48	0.90	0.76	0.30	0.38
Maximum	2.99	2.62	1.30	1.54	2.82	1.73
Sum	17.58	19.28	14.56	15.94	15.41	13.41
Count	14	14	14	14	14	14

**Table 7:** Descriptive statistical parameters of the ratios (cultivated to virgin - C/V- soil samples of the same origin) of the physical [saturation percentage (SP), pH, electric conductivity (EC), clay %, Silt % and sand%] and chemical properties, soluble cations and soluble anion in meq/L (CaCO<sub>3</sub>, Ca, Mg, Na, K, CO<sub>3</sub>, HCO<sub>3</sub>, Cl and SO<sub>4</sub>) cultivated (C) and virgin (V) soil of the same origin

	Mean	Standard Error	Median	Standard Deviation	Sample Variance	Kurtosis	Skewness	Range	Minimum	Maximum	Count
SP	1.03	0.03	1	0.11	0.01	-0.97	0.13	0.35	0.86	1.2	13
PH	1.01	0.01	1	0.03	0	-0.67	0.35	0.09	0.97	1.07	13
EC	0.98	0.16	1.13	0.57	0.32	-0.32	0.29	2.01	0.09	2.1	13
Ca	0.87	0.17	0.75	0.62	0.39	0.3	0.9	2.13	0.07	2.2	13
Mg	0.91	0.16	0.88	0.59	0.35	0.88	0.98	2.1	0.15	2.25	13
Na	1.1	0.34	0.85	1.21	1.47	1.24	1.29	3.97	0.01	3.98	13
K	0.69	0.23	0.39	0.84	0.71	8.28	2.73	3.11	0.15	3.26	13
CO3	1.17	0.21	1.1	0.7	0.49	4.72	1.91	2.5	0.5	3	11
HCO3	1.18	0.11	1.14	0.41	0.17	1.15	1.11	1.44	0.7	2.14	13
Cl	1.06	0.23	0.66	0.82	0.68	-0.51	0.91	2.44	0.09	2.53	13
SO4	1.53	0.41	0.92	1.37	1.89	-1.13	0.81	3.61	0.14	3.75	11
Clay %	1.33	0.18	1.16	0.64	0.4	0.75	1.13	2.08	0.5	2.58	13
Silt %	1.22	0.3	0.89	1.08	1.17	3.33	1.89	3.93	0.07	4	13
Sand %	0.99	0.04	0.97	0.14	0.02	0.64	0.57	0.53	0.76	1.29	13
CaCO3%	1.06	0.14	0.96	0.5	0.25	4.24	1.85	1.86	0.56	2.42	13

**Table 9:** The activity concentrations  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{40}\text{K}$  in Bq/kg in cultivated (C) and virgin (V) soils and their ratio (C/V) in different countries.

	Ra-226			Ra-228			K-40			Reference
	C	V	C/V	C	V	C/V	C	V	C/V	
Present study	20.92	18.62	1.26	28.66	23.28	1.38	554	537	1.04	Present study
India (Srirangam)	8.4	6.8	1.24	98.4	87.5	1.12	436	419	1.04	[18]
Cameroon	34.5			16.6			187			[19]
Pakistan (Faisalabad)	32.6	24.2	1.35	62.3	53.3	1.17	584	558	1.05	[20, 21]
Algeria	53	47	1.13	50	33	1.52	311	329	0.95	[14]
Egypt (Qena)	13.7			12.3			1233			[22]
Barzil (Panama)	10.22	1.69	6.05	7.2	5.3	1.36	55	34	1.62	[23]
India (Mumbai)		21-674			11-44			51-295		[24]
India (Western Ghats)		36.3			107.8			232		[25]
India (Kashmir)		6.4-18.8								[26]
World Average	32 (17-60)			45 (11-64)			420 (140-850)			[2]

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