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Assessment of Natural Radionuclides Released into Different Environmental Compartments Caused by Usage of Phosphate Fertilizers

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ABSTRACT

The general goal of the current study is to measure radioactivity released into different environmental compartments (plant and soil) caused by use of phosphate fertilizers. The present study included 53 samples obtained from the Eastern side of the Nile Delta in the region including 20 soil samples before and after fertilization, 20 plant samples before and after fertilization, 10 samples of water wrapping the soil before fertilization and 3 Phosphate fertilizer samples. The mean concentration of ^{226}Ra in the soil and plant samples before and after fertilization is higher than the global average. After fertilization, mean value concentration of ^{40}K was higher than before fertilization and global average. The concentration levels of ^{226}Ra, ^{40}K and ^{228}Ra are greater than average background levels after fertilization. Regarding heavy metals concentrations in the plant samples, before and after fertilization, and also the concentration of Cd are higher than the maximum allowable limit. Pb element in plant samples after fertilization is higher than permissible limits. It could be concluded that after use of phosphate fertilizers, the total concentration of ^{226}Ra in soil and plant samples increases more than before fertilization. After fertilization, the average concentration of ^{40}K in the plant samples increases more than before fertilization and heavy metals levels of As, Cd and Pb are higher than guideline limits.

INTRODUCTION

A number of the public is regularly exposed to radiation from both natural and artificial sources, where 80% of our exposure comes from natural sources and only 20% comes from artificial sources, primarily from radiation applications used in medicine. Radioactive chemicals and atmospheric radiation can externally irradiate our bodies from the outside, or the chemicals in the air are inhaled, swallowed through the skin and wounds, and then, internally, they irradiate us from inside. Globally, the internal exposure doses are approximately the same the sources of radionuclides present in our atmosphere. Many elements such as ^{238}U and ^{40}K arise naturally, while others, such as ^{137}Cs are result from industrial or military activities. [1]

The acronyms NORM and TENORM indicate radioactive materials that occur naturally and radioactive materials that occurs naturally and that is technically enhanced. While radioactive material is not generated in TENORM-associated processes, the abundance or concentration of radionuclides that occur naturally is increased. This may be the result of radioactive material concentration or redistribution, for example, as a result of burning, extraction of uranium ore or construction materials. [2]

Naturally occurring radiation can be split into two classes firstly cosmic rays, which dominate the outer source of radiation as ^{7}Be. Secondary these strongly living terrestrial primordial radionuclides found in the soil, such as ^{40}K,^{238}U and ^{232}Th. [3] It is possible to classify primordial radionuclides into single radionuclides and members of the decay chain. There are currently three uranium series decay chains, the actinium series and thorium series, which end in a stable lead isotope. [4]

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The application of plant nutrients, including phosphate fertilizers, has greatly increased since the 1950s. About 30 million metric tons of phosphate fertilizers are used worldwide annually. Sedimentary and igneous phosphate fertilizers, such as Ra-226, can lead to high levels in some crops and other plants grown on the soil. Phosphate ores can be defined into two groups. This can cause increased exposure to radionuclides to humans and animals through the ingestion of these crops. [5]

The general goal of the present study is to make measurement of radioactivity released into different environmental compartments (plant and soil) and heavy metal such as As, Pb, Cu and Cd caused by use of phosphate fertilizers and assessment of the probable health risk.

MATERIALS AND METHODS

Study area

The study area is around the eastern side of the Nile delta of Egypt (Figure 1). 53 samples were collected from selected site in the area under investigation and they included 20 samples of soil before and after fertilization, 20 plant samples before and after fertilization, 10 water samples and 3 phosphate fertilizer samples.

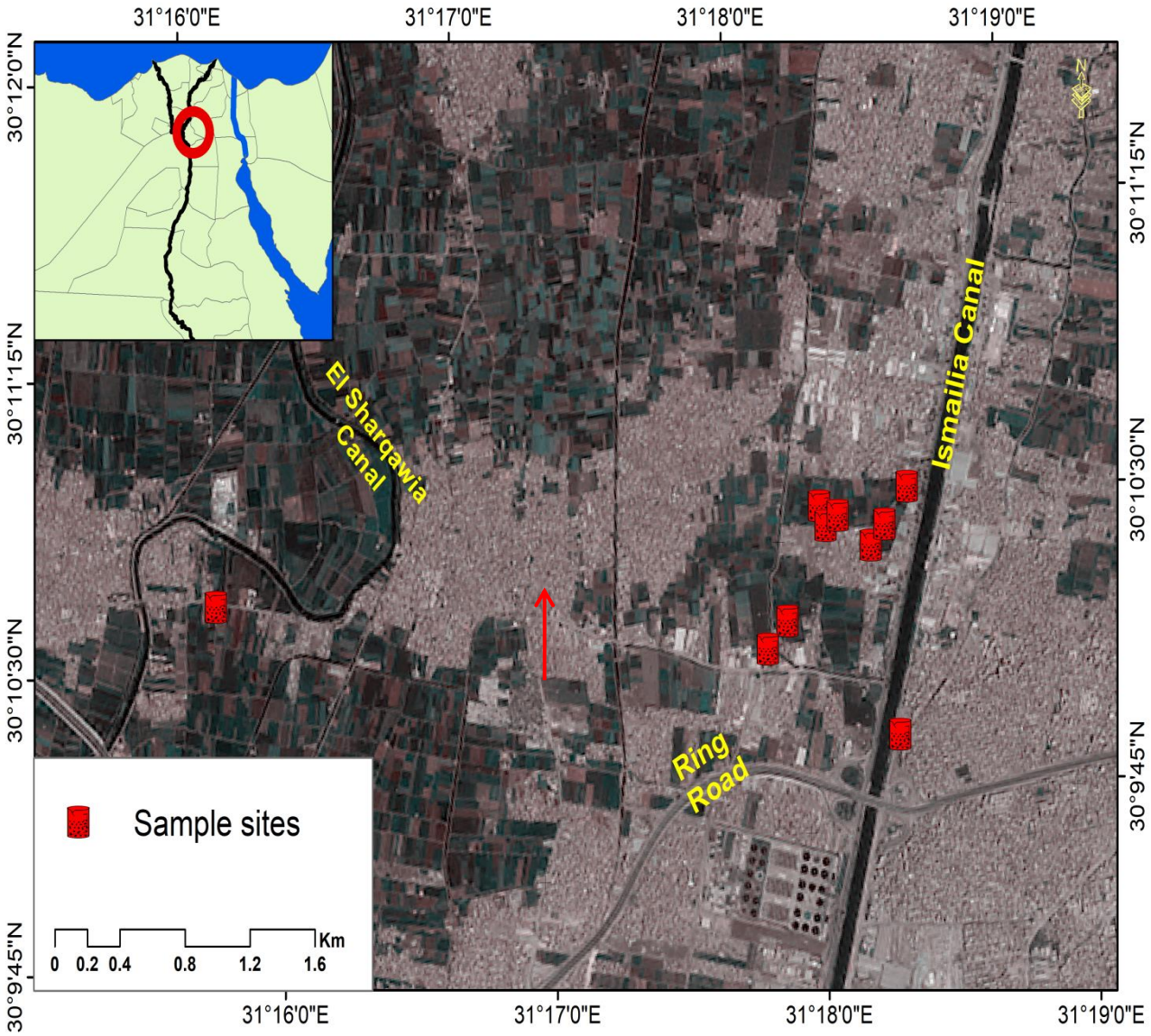

Fig. (1): Map around eastern side of Nile delta Egypt (Study area). Ten locations were studied. Soil, plant and water samples were obtained before fertilization at 1/6/2020 and after fertilization at 1/7/2020. The investigators took from each location, approximately 2 kilos from each sample (soil and plant) and 2 liter from water. The samples locations were detected by global position system (GPS) where plants samples are Cabbage, Lettuce, Rocca and Molokhia

Gamma rays spectrum analyses

The detected radionuclides were identified and quantified according to gamma energy lines in KeV, their emission probability Pɣ and gamma photopeak efficiencies calibration.

Minimum Detectable Activity (MDA)

The minimum detectable activity (MDA) for Radionuclides was calculated according to the following equation:

$$\mathbf{MDA} = \frac{LD}{T\ X\ Eff(E) X\ P\gamma(E) XM}$$

Where T is the counting time, Eff (E) is full-energy photopeak efficiency at photon energy E and Pγ (E) is emission probability, M is mass in Kg. LD is the detection limit calculated by the following equation:

$$\boldsymbol{L_D = L_C + K\,\sigma_D}$$

Where LC is the critical level below which no signal can be detected, D is the standard deviation and K σD is the error probability. [6]

Radiological Assessment of samples

Radium Equivalent (Ra_{eq})

The radium equivalent activity term was introduced as a common index by Brretka et al.[7], it was defined as a single quantity that represents the combined specific activities of ^{238}U, ^{232}Th and ^{40}K and develops numerical indicator of an external dose to public. 370 Bq/kg is limit for radium equivalent $^{226}Ra_{eq}$ was calculated from the following equation

$$Ra_{eq}=A_{Ra}+1.43A_{Th}+0.077A_K$$

Where A_{Ra} is the specific activity for ^{226}Ra (^{238}U) in Bq Kg-1, A_{Th} is the specific activity for ^{232}Th in BqKg-1, and A_K is specific activity for ^{40}K in BqKg-1. [7]

This equation depends on the fact that 370 BqKg-1 of ^{226}Ra or 259 BqKg-1 of ^{232}Th or 4810 BqKg-1 of ^{40}K produces the same gamma dose rate assuming radioactive equilibrium to be established in both ^{238}U and ^{232}Th series. [7,8]

Radiation Hazard Index

External radiation hazard (Hex)

Index Hex is the quantity that has provided a useful index for assessing the radiological health burden on the inhabitants. Exposure to radiation caused by 226Ra, 232Th and 40K. In terms of the external hazard index, it can be external and specified. [9]

$$H_{ex}= A_{Ra}/370+A_{Th}/259+A_K/4810$$

The activity concentration (Bq/Kg) of ^{226}Ra, ^{232}Th and ^{40}K, are denoted as A_{Ra}, A_{Th} and A_K respectively. To determine the interference or action limit of radiation exposure due to natural radioactivity in soil, this should be the appropriate criterion. To maintain the radiation risk to be low, the external hazard index should be less than unity. [10]

Internal hazard index (H_{in})

Radon and its short-lived products are also dangerous to the respiratory system. The inner exposure to radon and its daughter products are measured by the internal hazard index. The internal hazard index should be less than unity. [11]

$$H_{in}= A_{Ra}/185+A_{Th}/259+A_K/4810.$$

Absorbed Dose Rate

The rate of the absorbed dose in air expresses the received dose in the open air from the radiation emitted from radionuclides activity concentration in the environmental materials. This factor is important quantity to assess when considering radiation risk to a bio system. The absorbed dose rate, D (nGy/h) in air at 1m high, the ground level owing to the concentration of uranium-238, thorium-232 and potassium-40 [12] is given by:

$$D= 0.4299A_{Ra}+0.666A_{Th}+0.042A_K+0.112A_{Cs}+0.028A_{Be}$$

Where, D is the outdoor air absorbed dose rate. A_{Ra}, $A_{Th,}$ A_K, A_{Cs} and A_{Be} are the activity concentration levels (in Bq/Kg) of ^{226}Ra,^{232}Th, ^{40}K, ^{137}Cs and ^{7}Be respectively.

Studies indicate an average mean outdoor terrestrial gamma dose rate dose of 60 nGy/h. [13,14]

Annual effective Dose Equivalent (AEDE)

Annual effective Dose Equivalent (AEDE) was calculated from the absorbed dose by applying the dose conversion factor of 0.7 (Sv/Gy). [15]

$$AEDE\ (msv)=D(nGyh^{-1})\times 8760(h/y)\times 0.7(Sv/Gy)\ 10^{-6}\times 0.2$$

Where, D is the external absorbed gamma dose rate in nGy/h, 0.7 conversion factor with SV/Gy, 8760 hour/year, 0.2 external occupancy factors.

Excess Lifetime cancer Risk (ELCR)

Excess lifetime cancer risk (ELCR) could be defined as the excess likelihood of developing cancer at lifetime due to exposure level of humans to radiation. [16]

$(ELCR) = (AEDE) \times D_L \times R_F$

Where, ELCR is the Excess Lifetime Cancer Risk, (probability of cancer induction), AEDE is the annual effective dose equivalent, the D_L is the lifetime (70 years) and R_F is the risk factor (Sv-1), (Fatal chance of cancer per Sievert).

For low dose background radiations which are considered to produce stochastic effects, ICRP 60 utilizes values of 0.05 Sv-1 for the public exposure. [16]

Internal dose from ingested Vegetables

The annual intakes (Q) of ingested radionuclides special for ^{226}Ra, ^{232}Th and ^{40}K were determined in vegetables (Cabbage, Lettuce, Rocca and Molokhia) using the activity levels (C) in foods and the annual food consumption rates (F) [17]. The calculated annual intakes and the Internal Dose Conversion Factors (IDCF) were used to estimate the annual internal effective dose (H) [18],

$Q = C \times F$

$H = Q \times IDCF$

Transfer factors

TF=Plant specific activity/Soil specific activity

Measurement of heavy metals

Inductively coupled plasma optical emission spectroscopy (ICP-OES) was used to calculate heavy elements in the studied samples. [19]

Actual dermal dose and hazard quotient

Measured dermal dose due to skin contact with polluted atmosphere and calculated hazard quotient (HQ) where the ratio of exposure to hazardous material that affects associated target organs HQ where it is the non-cancer hazard quotient. And from the equation, we calculate the actual dermal dose:

(Dermal actual dose (mg/kg/day) = C x SA x SL x ABS x 10^{-9} x ED / (BW x AT)

Where (SA) is the surface area of the skin exposed to soil = 5800 cm2, (SL) is soil load = 1 mg/cm2/day, (ED) is the exposure duration =245 day/year, (BW) is the body mass= 70 kg and (AT) the days in lifetime = 25550 days/70-year, C is the concentration of metal in soil (mg/kg), ABS is the fraction absorbed across skin.

The hazard quotient Hazard index was calculated from equation (HQ = actual dose/ reference dose) [20]

Calculated hazard index

To calculate the hazard index (HI) for each organ, the hazard quotient (HQ) was obtained using this equation:

$(HI = \sum HQ)$

Where HQ is the hazard quotient and HI is the hazard index. [21]

Cancer risk

Cancer risk is used to describe the health effects of heavy metals and metalloid carcinogenic. This is defined by the amount of each route of exposure. The cancer slop factor (CSF) values for Cd, Cr, Pb and As are 6.3, 0.5, 0.0085 and 1.5 mg/kg/day, while the appropriate cancer risk value is less than 1.0×10^{-44}.

$CR = CDI \times CSF$

Where CR is the cancer risk, CDI is the dermal dose for non-carcinogenic metals and CSF is the cancer slop factor for arsenic. [21]

RESULTS

Average activity concentrations of detected radionuclide in phosphate fertilizers samples

Average concentrations of radionuclide in phosphate fertilizers samples in Bq Kg^{-1} of ^{238}U-series (^{238}U (^{234}Th), ^{226}Ra, ^{214}Pb, ^{214}Bi) and ^{232}Th-series (^{228}Ra(^{228}Ac), ^{212}Pb, ^{212}Bi, ^{208}Tl),^{40}K, ^{137}Cs and ^{7}Be are presented in Table (1).

Table (1): Average activity concentrations of detected radionuclide in phosphate fertilizers samples

Sample	^{238}U	^{226}Ra	^{214}Pb	^{214}Bi	^{210}Pb	^{228}Ra	^{212}Pb	^{212}Bi	^{208}Tl	^{40}K	^{7}B	^{137}Cs
PF_1	300.9± 0.9	566.6± 1.2	613.1± 1.23	592.6± 1.21	<15.8	<0.71	10.9 ± 0.2	<0.21	<0.14	26.2 ± 0.26	<0.33	10.4 ± 0.2
PF_2	787.1± 1.403	628.7 ± 1.25	558.9 ± 1.2	519.6± 1.14	<15.8	<0.71	19.5 ± 0.22	<0.21	<0.14	<1.21	<0.33	6.34 ± 0.13
PF_3	435.1± 1.04	1113.9± 1.7	569.4 ± 1.2	529.97± 1.2	<15.8	<0.71	24.1 ± 0.24	<0.21	4.84 ± 0.11	60.94± 0.4	<0.33	16.7 ± 0.205
average	507.7	769.73	580.5	547.4	<15.8	<0.71	18.2	<0.21	4.84	29.1	<0.33	11.14

Radionuclide in soil

Average concentrations of radionuclides in the soil samples in Bq Kg^{-1} of ^{238}-series (^{238}U (^{234}Th), ^{226}Ra, ^{214}Pb, ^{214}Bi) and ^{232}Th-series (^{228}Ra (^{228}Ac), ^{212}Pb, ^{212}Bi, ^{208}Tl),^{40}K, ^{137}Cs and ^{7}Be are presented in Table (2). Concentrations of ^{226}Ra, ^{228}Ra, ^{40}K, ^{7}Be, ^{137}Cs radionuclides in the soil samples before and after fertilization from different area, and the variation mainly depends on geological origins and mineralogical compositions then compared to the world average levels for ^{226}Ra, ^{228}Ra and ^{40}K (Figures 1-3).

Table (2): Average activity concentration of radionuclides in soil samples before and after fertilization

Sample type		Average activity concentration ($BqKg^{-1}$)										
		^{238}U	^{226}Ra	^{214}Pb	^{214}Bi	^{228}Ra	^{212}Pb	^{212}Bi	^{208}TI	^{40}K	^{7}Be	^{137}Cs
S_b	Min-Max	28.72-58.1	3.14-77.9	6.2-29.5	1.93-18.6	22.6-58.1	18-29.5	4.5-29.2	6.2-10.2	269.6-429.4	0 - 7.9	1.15-3.5
	average	35.23	37.7	25.8	13.9	35.23	25.8	8.81	7.6	397.9	0.79	2.28
S_a	Min-Max	1-57.7	23.64-80.8	12.24-20.4	15.6-19.8	22.5-36.1	19.2-29.9	10.5-23.4	6.9-12.7	350.02-429.4	-	0.86-2.9
	average	20.9	47.6	15.8	18.3	28.26	23.57	13.3	8.8	397.42	-	1.95

Sb: Soil before fertilization, Sa: Soil after fertilization

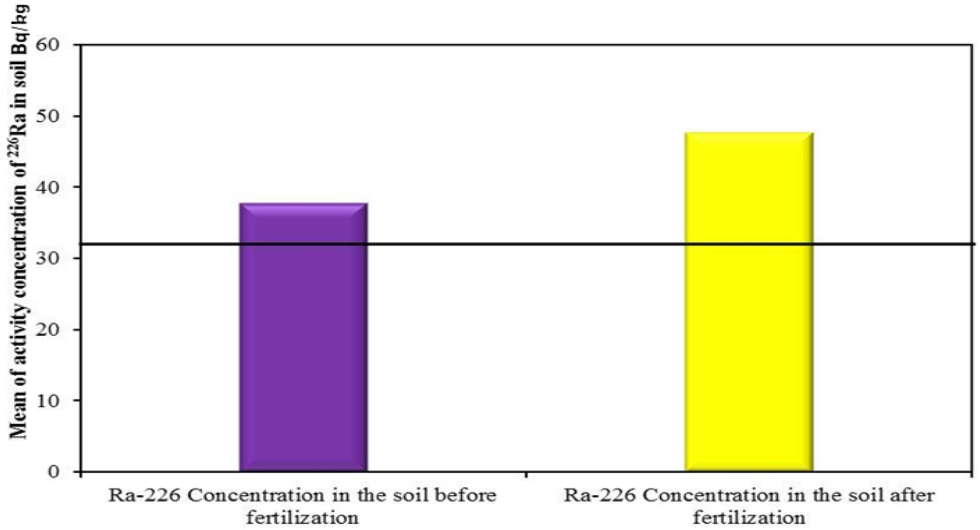

Fig. (1): Average activity concentration of ^{226}Ra before and after fertilization in soil samples

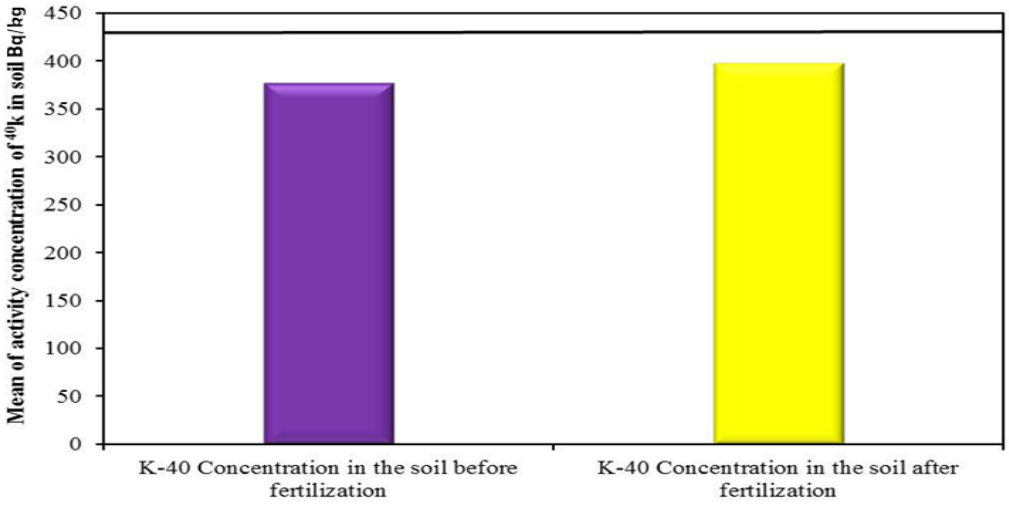

Fig. (2): Average activity concentration of ^{40}K before and after fertilization

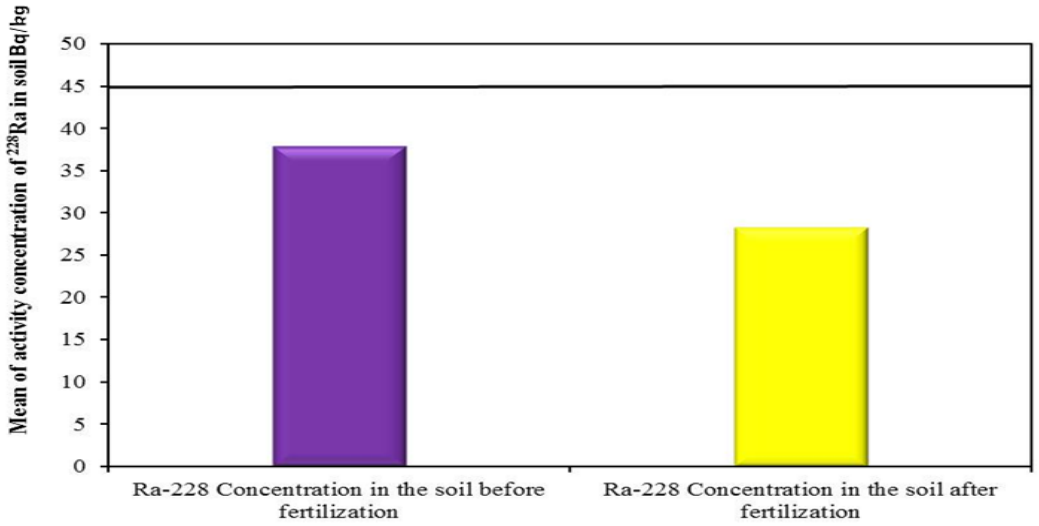

Fig. (3): Average activity concentration of ^{228}Ra before and after fertilization in soil samples

Calculated Ra_{eq} ($BqKg^{-1}$), H_{ex} and H_{in} indices for soil samples before fertilization

Radium equivalent activity (Ra_{eq}) results for all soil samples studied before fertilization are illustrated and the Internal (H_{in}) and external radiation hazard indices (H_{ex}) are revealed in Figures 4-5.

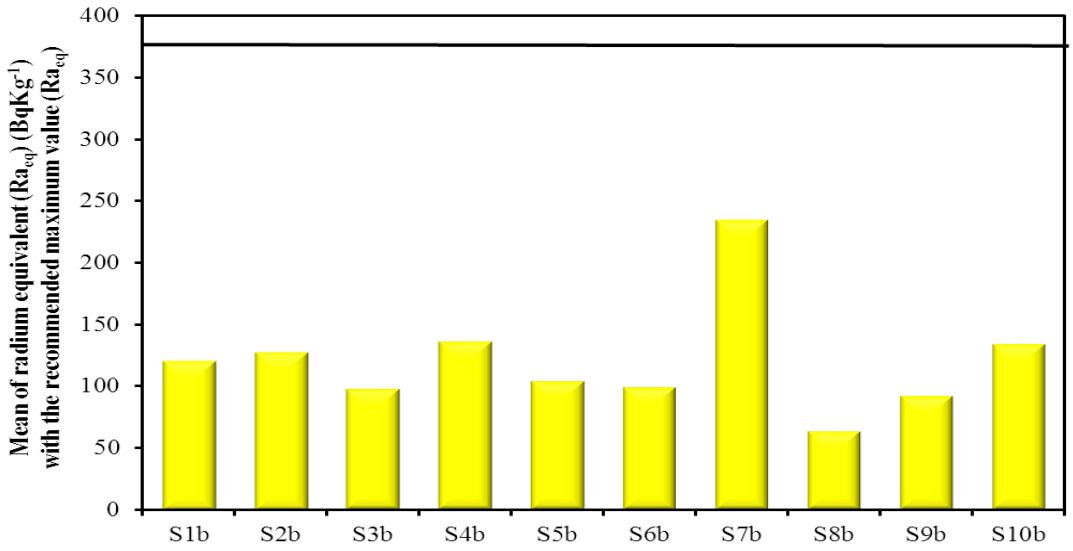

Fig. (4): Radium equivalent level for soil samples before samples fertilization

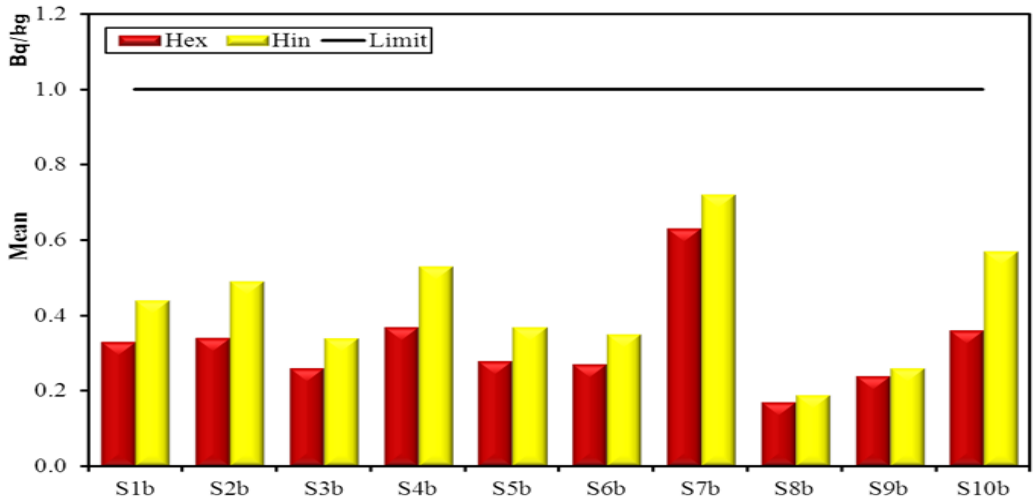

Fig. (5): External and internal radiation hazard index level of soil samples before fertilization

Calculated Ra_{eq} ($BqKg^{-1}$), H_{ex} and H_{in} indices for soil samples after fertilization

Radium equivalent activity (Ra_{eq}) results for all soil samples studied after fertilization are illustrated and the Internal (H_{ex}) and external radiation hazard indices ($H_{in}$are shown inFigures 6-7.

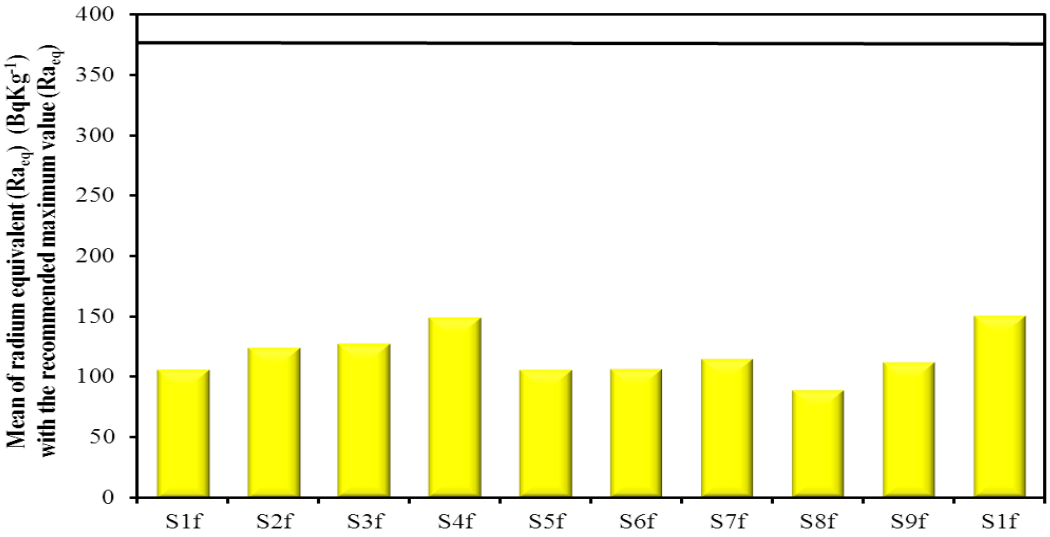

Fig. (6): Radium equivalent level of soil samples after fertilization

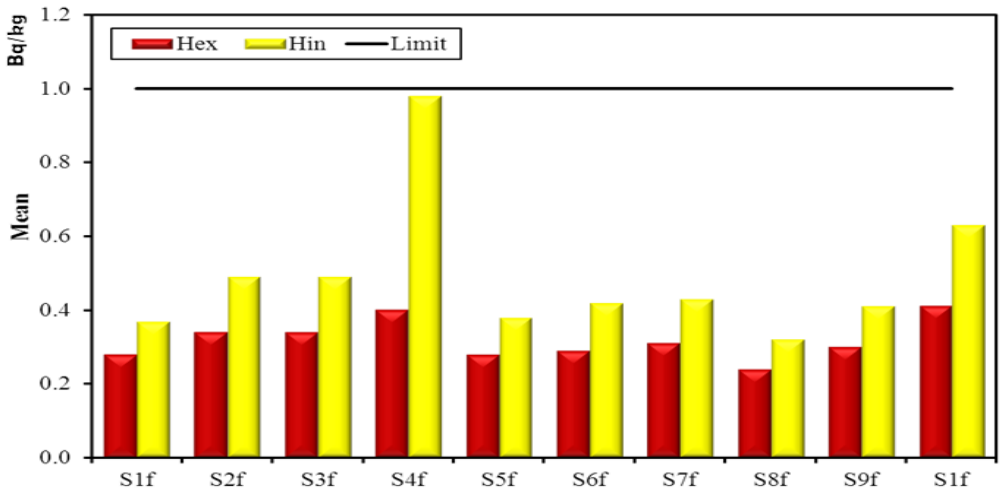

Fig. (7): External and internal radiation hazard index level of soil samples after fertilization

Absorbed dose rate to farmers, annual effective dose equivalent, and annual cancer risk of soil samples before fertilization

Table (3) shows the results of the absorbed dose rate in nGy/h, annual effective dose equivalent in mSv/y, annual cancer risk and excess lifetime risk of soil samples before and after fertilization respectively.

Table (3): A comparison between min, max and average absorbed dose, annual effective dose equivalent, and annual cancer risk of soil samples before and after fertilization

Sample type		Absorbed Dose rate nGy/h	Annual Effective dose Equivalent mSv/y	Annual Cancer Risk	Excess lifetime risk
Before fertilization	Min-Max	32.14-110.9	0.039-0.14	1.99×10^{-7}-7×10^{-7}	1.36×10^{-5}-4.9×10^{-5}
	Average	58.83	0.072	3.61×10^{-7}	2.5×10^{-5}
After fertilization	Min-Max	34.07-59.72	0.042-0.086	2.1×10^{-7}-4.3×10^{-7}	1.47×10^{-5}-3.01×10^{-5}
	Average	50.57	0.062	3.1×10^{-7}	2.17×10^{-5}

Radionuclides in plant

Activity concentrations of radionuclides in plant before and after fertilization

Average concentrations of radionuclides in plant samples in Bq Kg^{-1} of ^{238}U-series ^{238}U(^{234}Th),^{226}Ra, ^{214}Pb, ^{214}Bi) and ^{232}Th-series (^{228}Ra(^{228}Ac), ^{212}Pb, ^{212}Bi, ^{208}Tl),^{40}K, ^{137}Cs and ^{7}Be in granite samples are presented in Table (4).

Table (4): Activity concentrations of radionuclide in whole plant (Cabbage-Lettuce- Roca-Molokhia) before and after fertilization

Sample type		Average activity concentration ($BqKg^{-1}$)										
		^{238}U	^{226}Ra	^{214}Pb	^{214}Bi	^{228}Ra	^{212}Pb	^{212}Bi	^{208}TI	^{40}K	^{7}Be	^{137}Cs
plant before	Min-Max	5.1-64.7	5.2-636.6	16.8-66.6	28.2-32.6	-	11.06-76.6	-	0 -30.5	1791.4-7275.4	53.1-82.99	-
	average	9.9	121.6	16.97	12.03	-	20.02	-	30.5	3254.61	11.92	-
Plant after	Min-Max	2.3-40.2	8.7-464.2	2.44-28.2	5.2-59.4	6.3-609.7	3.4-78.6	2.44-28.2	3.4-72.7	932.2-3720.2	11.8-20.7	0.93-1.98
	average	11.9	67.98	12.7	6.37	102.7	13.7	12.7	9.06	2230.22	17.6	1.96

Minimum, maximum and average values of transfer factors (TF) for selected radionuclides

Table (5) shows ranges and average results of transfer factors (TF) for ^{226}Ra, ^{228}Ra, ^{40}K and ^{137}Cs radionuclides for before and after fertilization.

Table (5): Minimum, maximum and average values of transfer factors (TF) for selected radionuclides before and after fertilization

Samples		Transfer factors(TF)			
		^{226}Ra	^{228}Ra	^{40}K	^{137}Cs
TF from soil to plant before fertilization	Min-Max	3.96×10^{-3}-11.921	5.9×10^{-3}-0.031	4.31-26.98	0.08-0.24
	average	2.3	0.023	9.198	0.13
TF from soil to plant after fertilization	Min -Max	0.0038-13.9	0.02-2.1	0.094-23.5	0.06-1
	average	1.7	0.04	6.6	0.4

Calculated annual internal effective dose (mSv/year) of radionuclides for plant samples before and after fertilization

Tables (6) and (7) show the annual internal effective dose in mSv/y of radionuclide in plant samples before and after fertilization for different age groups for ^{226}Ra, ^{228}Ra, ^{40}K and ^{137}Cs.

Table (6): The annual internal effective dose (mSv/year) due to plant (Cabbage-Lettuce-Rocca-Molokhia) ingestion before fertilization

Element	Annual internal effective dose(mSv/y)					
	(0-1)year	(1-2)yrs	(2-7)yrs	(7-12)yrs	(12-17)yrs	>17yrs
^{226}Ra	37.98	7.76	5.01	6.5	12.12	2.3
^{40}K	1.34	0.91	0.45	0.28	0.16	0.13
^{228}Ra	-	-	-	-	-	-
^{137}Cs	-	-	-	-	-	-

Table (7): The annual internal effective dose (mSv/year) due to plant (Cabbage-Lettuce-Rocca-Molokhia) ingestion after fertilization

Element	Annual internal effective dose(mSv/y)					
	(0-1)year	(1-2)yrs	(2-7)yrs	(7-12)yrs	(12-17)yrs	>17yrs
^{226}Ra	21.24	4.34	2.80	3.61	6.8	1.3
^{40}K	9.2	6.23	3.11	1.92	1.13	0.92
^{228}Ra	12.7	2.41	1.44	1.65	2.24	0.042
^{137}Cs	0.04	0.023	0.18	0.019	0.025	0.025

Activity concentrations of radionuclides in liquid irrigation water

Table (8) shows the average concentration of radionuclides in liquid irrigation water in BqKg^{-1} of ^{238}U-series (^{238}U (^{234}Th), ^{226}Ra, ^{214}Pb, ^{214}Bi), ^{232}Th-series ^{228}Ra (^{228}Ac), ^{212}Pb, ^{212}Bi, ^{208}Tl),^{40}K, ^{137}Cs and ^{7}Be.

Table (8): Activity concentration (Bq/L) in liquid irrigation water

Sample ID	^{238}U					^{232}Th				^{235}U	^{40}K	^{7}Be	^{137}Cs
	^{238}U	^{226}Ra	^{214}Pb	^{214}Bi	^{210}Pb	^{228}Ra	^{212}Pb	^{212}Bi	^{208}TL				
W_1	1.2 ± 0.01	4.4 ± 0.02	<0.63	<0.56	2.7 ± 0.015	347.4 ± 0.2	3.3 ± 0.02	0.95 ± 0.1	<0.14	0.029 ± 0.01	5.6 ± 0.02	<0.3	<0.28
W_2	9.7 ± 0.03	4.7 ± 0.02	0.41 ± 0.01	<0.56	<15.8	<0.71	0.304 ± 0.01	<0.21	0.237 ± 0.05	0.450 ± 0.01	8.85 ± 0.3	1.21 ± 0.011	<0.28
W_3	3.4 ± 0.02	0.06 ± 0.008	<0.63	1.1 ± 0.01	<15.8	0.0000156 ± 3.9	0.57 ± 0.008	<0.21	<0.14	3.405 ± 0.004	3.52 ± 0.019	<0.3	<0.28
W_4	<0.2	4.02 ± 0.02	0.4 ± 0.006	0.12 ± 0.003	<15.8	0.00000328 ± 75 ± 17.44	<0.25	<0.21	0.07 ± 0.008	0.27 ± 0.005	4.2 ± 0.2	<0.3	<0.28
W_5	2.9 ± 0.02	2.2 ± 0.014	<0.63	0.45 ± 0.006	<15.8	<0.71	0.3 ± 0.005	<0.21	0.03 ± 0.002	0.2 ± 0.44	4.3 ± 0.02	<0.3	<0.28
W_6	7.3 ± 0.03	1.5 ± 0.01	0.13 ± 0.004	0.31 ± 0.005	<15.8	<0.71	<0.25	0.55 ± 0.007	<0.14	0.008 ± 0.0008	4.6 ± 0.02	<0.3	<0.28
W_7	2.1 ± 0.01	2.7 ± 0.02	0.6 ± 0.007	0.26 ± 0.005	<15.8	31.5 ± 0.05	<0.25	<0.21	<0.14	0.2 ± 0.004	3.5 ± 0.02	<0.3	<0.28
W_8	<0.2	0.54 ± 0.007	0.1 ± 0.003	<0.56	0.7 ± 0.008	<0.71	0.1 ± 0.003	<0.21	0.14 ± 0.011	0.03 ± 0.002	4.8 ± 0.02	<0.3	<0.28
W_9	3.73 ± 0.02	1.13 ± 0.01	0.1 ± 0.003	<0.56	<15.8	2.54 ± 0.02	<0.25	<0.21	1.7 ± 0.004	0.05 ± 0.002	4.2 ± 0.02	<0.3	<0.28
W_{10}	<0.2	2.8 ± 0.016	<0.63	2.8 ± 0.02	<15.8	<0.71	0.4 ± 0.005	<0.21	<0.14	0.22 ± 0.0045	5.6 ± 0.023	<0.3	<0.28

^{226}Ra, ^{238}U concentration and the U.S. Environmental Protection Agency standards for liquid discharges of naturally occurring radionuclides

^{226}Ra concentration was higher in all water samples (except sample W_3) than EPA standard limit. ^{23}U activity concentration was lower than EPA standard in all samples (Figure 8-9).

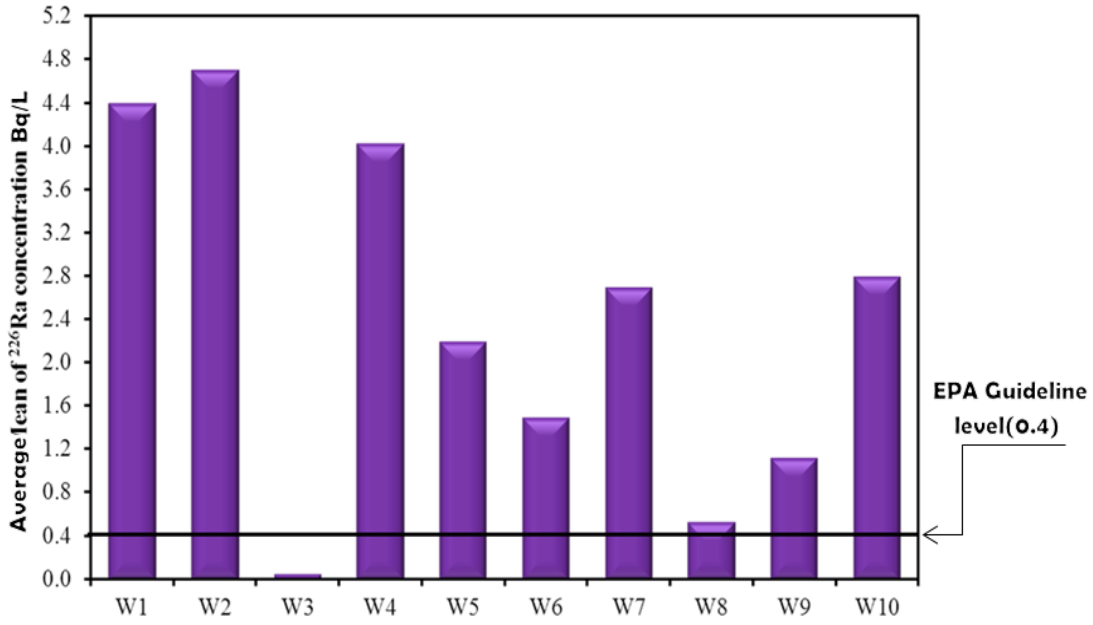

Fig. (8): The distribution of ^{226}Ra levels in waste water samples

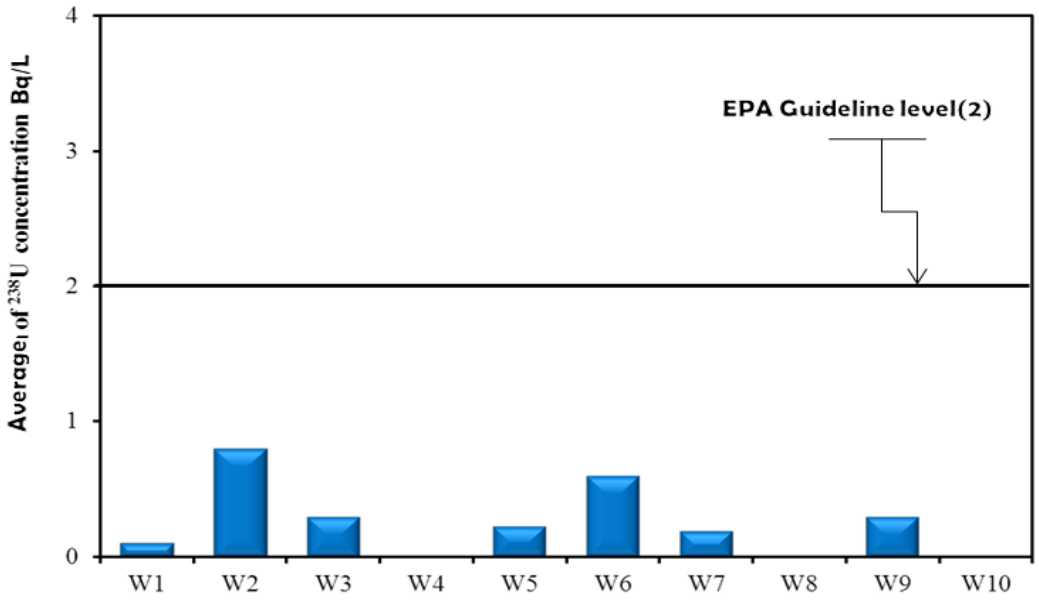

Fig. (9): The distribution of ^{238}U levels in waste water samples

Heavy metals concentrations in all studied samples

Average concentrations of heavy metals in all studied samples are presented in Table (9).

Table (9): Average concentrations of heavy metals in all studied samples

Number and Type of Sample	Average concentration (ɱg/ml)				
	As	Cu	Cd	Pb	Se
Sample fertilizer(1)	3.497	5.29	1.43	5.91	0.52
Sample fertilizer (2)	4.015	10.103	1.61	4.07	0.54
Sample fertilizer (3)	3.158	5.89	1.19	3.35	0.45
plant samples before fertilization	1.04	411.22	0.54	1.98	0.95
Plant samples after fertilization	1.111	9.802	0.58	4.07	1.01
Soil samples before fertilization	0.65	1.72	0.24	8.39	0.42
Soil samples after fertilization	1.49	1.6	0.24	5.43	0.42

Metals concentration, actual dermal dose, hazard quotient, and associated target organs for fertilizers samples

In Table (10), metals concentrations, actual dermal dose, hazard quotient, and associated target organs for fertilizers samples are given.

Table (10): Metals concentrations, actual dermal dose, hazard quotient, and associated target organs for fertilizers samples

Metal	Concentration (ɱg/ml)	Reference dose for non-carcinogenic mg/kg/day	Dermal actual Dose mg/kg/day	Hazard Quotient HQ	Target Affected Organs
Cd	**1.41**	**1×10^{-3}**	**1.12×10^{-12}**	**1.12×10^{-9}**	**Renal, and Respiratory**
Pb	**4.44**	**5.3×10^{-4}**	**2.5×10^{-10}**	**4.69×10^{-7}**	**Renal, Cardiovascular, and Nervous**
As	**3.56**	**2.8×10^{-4}**	**1.130×10^{-10}**	**3.98×10^{-7}**	**Cardiovascular, Developmental, and Nervous**
Se	**0.503**	**5×10^{-3}**	**$1.2x\ 10^{-11}$**	**3.98×10^{-9}**	**Nervous, Hematological, and Dermal**
Cu	**7.1**	**4×10^{-3}**	**3.96×10^{-12}**	**9.89×10^{-10}**	**Renal and Liver**

Metal hazard quotient (HQ) and hazard Index (HI) for affected organ for fertilizers samples

In Table (11) hazard quotient (HQ) was collected to calculate the hazard index (HI) for each organs from metals.

Table (11): Metal hazard quotient (HQ) and hazard Index (HI) for affected organ for fertilizers samples

metal	Renal	Respiratory	cardiovascular	Nervous	Developmental	Hematological	Liver
Cd	1.12×10^{-9}	1.12×10^{-9}					
Pb	4.69×10^{-7}		4.69×10^{-7}	4.69×10^{-7}			
As			3.98×10^{-7}	3.98×10^{-7}	3.98×10^{-7}		
Se				3.98×10^{-9}		3.98×10^{-9}	
Cu	9.89×10^{-10}						9.89×10^{-10}
Hazard Index HI	4.7×10^{-7}	1.12×10^{-9}	8.67×10^{-7}	8.71×10^{-7}	3.98×10^{-7}	3.98×10^{-9}	9.89×10^{-10}

Metals concentration, actual dermal dose, hazard quotient, and associated target organs for soil

In Table (12) measured metals concentrations, actual dermal dose, hazard quotient, and associated target organs for soil are presented.

Table (12): Metals concentrations, actual dermal dose, hazard quotient, and associated target organs for soil

Metal	Concentration (ɱg/ml)	Reference dose for non-carcinogenic mg/kg/day	Dermal actual Dose mg/kg/day	Hazard Quotient HQ	Target Affected Organs
Cd	1.41	1×10^{-3}	1.91×10^{-13}	1.91×10^{-16}	Renal, and Respiratory
Pb	4.44	5.3×10^{-4}	5.5×10^{-11}	1.04×10^{-7}	Renal, Cardiovascular, and Nervous
As	3.56	2.8×10^{-4}	3.4×10^{-11}	1.2×10^{-7}	Cardiovascular, Developmental, and Nervous
Se	0.503	$5\times^{-3}$	1×10^{-11}	2×10^{-9}	Nervous, Hematological, and Dermal
Cu	7.1	4×10^{-3}	3.96×10^{-12}	9.9×10^{-10}	Renal and Liver

Metal hazard quotient (HQ) and hazard Index (HI) for affected organ for soil

In Table (13) hazard quotient (HQ) was collected to calculate the hazard index (HI) by this equation (HI= ∑ HQ) for each organs from metals.

Table (13): Metal hazard quotient (HQ) and hazard Index (HI) for affected organ for soil

metal	Renal	Respiratory	cardiovascular	Nervous	Developmental	Hematological	Liver
Cd	1.91×10^{-16}	1.91×10^{-16}					
Pb	1.04×10^{-7}		1.04×10^{-7}	1.04×10^{-7}			
As			1.2×10^{-7}	1.2×10^{-7}	1.2×10^{-7}		
Se				2×10^{-9}		2×10^{-9}	
Cu	9.9×10^{-10}						9.9×10^{-10}
Hazard Index HI	1.5×10^{-7}	1.91×10^{-16}	2.24×10^{-7}	2.3×10^{-7}	1.2×10-7	2×10^{-9}	9.9×10^{-10}

Cancer risk due to exposure to arsenic and lead

Cancer risk is only due to the exposure to arsenic (1.95×10^{-14}) and lead(1.74×10^{-9}).

DISCUSSION

Results from the current work showed that the concentration of ^{226}Ra in the soil samples before fertilization are higher than the world average (32 Bqkg^{-1}) [15]. Moreover, in soil samples after fertilization, it is also higher than world average (32 Bqkg^{-1}) and higher than mean value of samples before fertilization. About 80% of the recorded values of ^{226}Ra were higher than the world average. This may be attributed to the successful fertilization of agricultural soil with fertilization and original radiological minerals in the soil.

For ^{228}Ra (^{228}Ac) levels assessed by ^{232}Th and ^{228}Ra concentrations before and after fertilization are lower than the world average value (45 Bqkg^{-1}). [3] This may be attributed to the low percentage of the original radiological minerals in the soil.

For ^{40}K, it is lower than the world average 420 Bqkg^{-1} [3] in soil samples before and after fertilization. About 20% of the record values were higher than the world average 420 Bqkg^{-1}.

In a study by saleh et al. [17] the radionuclides detected in soil were ^{226}Ra,^{232}Th,^{40}K and ^{137}Cs where the average concentration of ^{226}Ra was 16.43, ^{232}Th average was 18.31 Bqkg^{-1}. For ^{40}K the average was 268.8 Bqkg^{-1}. ^{137}Cs was detected only in 3 locations with a maximum level of 7.24 Bqkg^{-1}. It is clear that the levels of radionuclides in this study was much closer to those of the soil of middle and upper Egypt and lower than the world averages reported by UNSCER in 1988. [17]

In the current study, Ra_{eq} results for soil samples studied before and after fertilization all values are lower than 370 Bqkg^{-1} [22]. From results of Ra_{eq} before and after fertilization and after 104 times fertilization passed, the soil will become invalid either for agricultural purposes or as aggregates of building material.

In a study by Kolo et al. [23], the results obtained for Ra_{eq} showed an average value of 42.67 Bqkg^{-1}. The mean value was less than the healthy admissible value (370 Bqkg^{-1}) set by the Organization for Economic Cooperation and Development (OECD). Other additional criteria for assessing the radiological burden on a given

population are the hazard index of external radiation (H_{ex}) and the hazard index of internal radiation (H_{in}). [23]

In the current study, the values of external hazard index (H_{ex}) for soil samples before fertilization are below the guidance level of this index (1). The value of Hex is less than unity that keeps the risk of radiation insignificant for farmers. For the internal radiation hazard index (H_{in}) of soil samples before fertilization, they are below the guidance level of this index which is 1. Thus this index is less than unity that keeps the risk of radiation for general public due to exposure in the studied samples to natural radionuclides is insignificant.

For external hazard index (H_{ex}) of soil samples after fertilization, it was lower than the guidance level of this index which is 1. The value of this index is less than unity that keeps the risk of radiation insignificant for farmers. For the internal radiation hazard index (H_{in}) of soil samples after fertilization was below the guidance level of this index (1). The radiation risk to general public due to exposure from soil samples before and after fertilization to natural radionuclides is insignificant.

In the present study, the annual effective dose equivalent (HE) of outdoor received by the public is calculated using equation (HE=D×T×F). The values obtained for the soil samples before fertilization showed an average of 0.0717 $mSvy^{-1}$ for outdoor factor. The values obtained for the soil samples after fertilization showed an average 0.062 $mSvy^{-1}$. In a study by Anamika et al., the annual effective gamma dose rates indoors were less the average level (0.46 $mSvy^{-1}$) while the average annual outdoor dose was relatively larger than the advocated level of 0.07 $mSvy^{-1}$. These results are very useful from the radiation protection point of view. [24]

A comparison of mean concentrations of radium-226, thorium-232, and potassium-40 and cesium -137 in samples of whole plant before fertilization and (Cabbage-Lettuce-Rocca-Molokhia) after fertilization among the studied samples is given in present work. The mean concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in samples of vegetable plant before fertilizers are greater than activity concentrations of ^{226}Ra , ^{232}Th , ^{40}K , and ^{137}Cs in corn samples which were equal to 0.81 (0.08-3.93), 0.85 (0.33-1.14) , 101.52 (50.75-157.66) and 0.07 (0.03-0.34) (dry weight) respectively in terms of $Bqkg^{-1}$ .Moreover, for wheat samples, these values were equal to (0.05-1.4Bqkg-1) with average of 1.67 $Bqkg^{-1}$ for ^{226}Ra, (0.08-1.36 $Bqkg^{-1}$) with average of 0.5 $Bqkg^{-1}$ for ^{232}Th , (50.57-143.26 $Bqkg^{-1}$) with average 91.73 for ^{40}K and (0-0.18 $Bqkg^{-1}$) with average of 0.01 $Bqkg^{-1}$ for ^{137}Cs (dry weight) respectively. After fertilization it is clear that the average concentration levels 102.7 $Bqkg^{-1}$ for ^{228}Ra and 2230.22 $Bqkg^{-1}$ for ^{40}K are greater than the average concentration levels of ^{40}K and ^{232}Th for samples leaves of plant in the Western Ghats environment which were 163.32 and 15.69 respectively. [25]

It is clear that TF values for ^{226}Ra and ^{40}K are considerably larger than ^{228}Ra and ^{137}Cs. This indicates that some plant species concentrate higher ^{226}Ra and ^{40}K radionuclides than others, authors of a previous study [26] observed the same. Plants can take potassium from soil as an essential element of metabolism, and other radionuclides can be taken as a homologue of an essential element. [27] The effective dose is a valuable term that allows for the inclusion of radiation doses from various radionuclides and from different radioactivity levels and sources. Estimates of the health effects caused by radiation associated with the ingestion of radionuclides in the body are proportional to the overall dose of radionuclides in different organs. [28]

In plant samples, the annual effective internal dose due to public results from intake of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs before fertilization for various age groups. These doses are higher than the levels of normal background areas which are reported by UNSCEAR (1988)[22]. The annual effective dose equivalent of ^{40}K before fertilization is 1.34 $mSVy^{-1}$ from (0-1year), 0.91 $mSvy^{-1}$ from (1-2 yrs), 0.45 $mSvy^{-1}$ from (2-7 yrs), 0.28 $mSvy^{-1}$ from (7-12 yrs), these doses are higher than the levels of normal background areas reported by UNSCEAR (1988) [22]. The annual effective dose equivalent of ^{40}K before fertilization was 0.16 $mSvy^{-1}$ from (12-17) and 0.13 $mSvy^{-1}$ >17yrswhich are lower than the normal levels background reported by UNSCEAR.

The annual effective dose equivalent for individual exposure due to people ingestion after fertilization for ^{226}Ra are larger than the levels of the normal background areas which is reported by UNSCEAR, 1988. [22] The annual effective dose equivalent of ^{40}K after fertilization is higher than the levels of the normal background areas reported by UNSCEAR. The annual effective dose equivalent after fertilization for ^{228}Ra are higher than the levels of the normal background areas which is reported by UNSCEAR, 1988 [22].

These results can lead to Cancer risk which the risk incurred by a population and calculated by considering a linear dose-effect relationship with no threshold as per ICRP procedure. For fewer doses ICRP fatal cancer risk factor is 0.05 Sv^{-1} [29]. The risk factor states the probability of a person dying of cancer increases by 5% for a total dose of 1 mSv received during his lifetime.

Therefore, the probability of death from cancer due to 'natural incidence' increases from about 25% to 30% following a total lifetime exposure of 1 Sievert.

With respect to the activity concentration of radionuclides in irrigation samples, the present study results conducted a comparison with the limits on average concentrations in daily effluents for 30 consecutive days for total ^{226}Ra and for uranium ^{238}U. EPA standards for liquid discharges of naturally occurring radionuclides. It is noticed that the EPA standards limit is very low (0.4 BqL^{-1}) because the radium salts are solved in water and vital for all forms of life. [30] So, its mobility in the environment is high. Additionally Ra is a vital element for all forms of life. ^{238}U for all liquids of waste water is lower than that of Environmental Protection Agency standards for liquid discharges of naturally occurring radionuclides 2 mgL^{-1} [30].

Average concentration of As element in plant samples before and after fertilization were 1.04 and 1.111 mg/kg, respectively, which are greater than the acceptable limit of 1 mg/kg as recommended by the WHO in plants. As element average concentrations in soil samples before and after fertilization are 0.65 and 1.49 mg/kg respectively which are less than the acceptable limit of 20 mg/kg As recommended by the WHO [31]. In fertilizer samples, As element levels were 3.49, 4.015 and 3.16 mg/kg, respectively, which are less than the permissible limit for rock phosphate of North Africa 15 mg/kg [31,32].

Average Cd concentration shown in the plant samples before and after fertilization was 0.54 and 0.58 mg/kg, respectively, which are higher than the allowable limits where the Cd maximum limit in plants recommended by the WHO is greater than 0.02 mg/kg. Average concentrations of Cd element found in the soil samples before and after fertilization is 0.24 and 0.24 mg/kg, respectively, which are lower than the WHO suggested permissible limit of 0.8 mg/kg (Ogundele et al., 2015). The concentrations of Cd elements in fertilized samples 1, 2 and 3 were 1.43, 1.61 and 1.19 mg/kg, respectively which less than the permissible limit for rock phosphate of North Africa is 15mg/kg. [32&33]

The Pb levels in the plant samples before and after fertilization were 1.98 and 4.07 mg/kg, respectively, in which Pb increased after fertilization to be higher than the permissible limit in plants, recommended by the WHO (2mg/kg). The amount of Pb elements found in the soil samples before and after fertilization were 8.39 and 9.43 mg/kg, respectively, which is lower than the WHO suggested acceptable limit of 85 mg/kg [33]. The amount of Pb element in fertilized samples 1, 2 and 3 were 5.91, 4.07 and 3.35 mg/kg, respectively, less than permissible limit for rock phosphate of North Africa is 6 mg/kg [32].

Almost all heavy metals are serious toxicants as carcinogens. However due to their chemical and physiological properties, heavy metals are useful in industrial areas including alloy, smelting and products of commercial products. Such applications increase the opportunity for heavy metal exposure. Arsenic, cadmium, chromium, and nickel are classified as group 1 carcinogens by the International Agency for Research on Cancer resulting in increasing the risk of cancer and cancer related diseases [34, 35].

Cancer risks from ionizing radiation are better understood than those from other carcinogens largely because it is possible to quantify exposures and doses. Lifetime risk of developing or dying from cancer refers to the chance a person has, over the course of his or her lifetime (from birth to death). By comparing risk cancer from radiotoxicity (2.17×10^{-5}) and chemotoxicity (1.74×10^{-9}) from heavy metal exposure, the current study revealed that cancer risk from radiotoxicity is higher than chemotoxicity.

CONCLUSION

From the present study, it can be concluded that:

- After fertilization, the total concentration of ^{226}Ra in soil and plant samples increases than before fertilization, which is greater (……) than the world average (…….).
- After fertilization, the average ^{40}K concentration in plant samples increases than before fertilization.
- The soil would become unsuitable either for agricultural purposes or as aggregates of building material from Ra_{eq}'s results after 104 times fertilization has passed.
- The annual internal effective dose due to intake of ^{226}Ra, ^{40}K and ^{228}Ra from this outcome will contribute to cancer risk, which is determined by considering a linear dose-effect relationship with no threshold as per the ICRP method.
- Regarding heavy metals concentration in plant samples, before and after fertilization, the concentration of As as well as Cd are larger than the maximum allowable limit. While Pb in plant samples after fertilization is higher than the permissible limits.

RECOMMENDATIONS

The high levels of ^{226}Ra and ^{232}Th result from superphosphate fertilizer; the high frequency of soil fertilization can lead to soil accumulation of U and Th radionuclides

A similar research should be carried out annually to measure natural radiation in order to monitor the health of the residents of the area. This research provides a basis for determining the further risk of radiation and can be adopted in a future study on the mapping of natural radioactivity.

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CONFLICT OF INTEREST

No conflict of interest is declared.

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