

Determination of Soil-to-Plant Transfer Factors for ^{226}Ra and ^{228}Ra in Some Egyptian Crops

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Abstract— Soil to plant transfer factors (TF_s) of ^{226}Ra and ^{228}Ra have been determined for different crops, such as Cabbage, Cucumber, Kale, Aubergine, Pepper, Zucchini and Tomato. The crops were collected from some local farms in Al-Qalyubia governorate Egypt under outdoor ambient tropical conditions for two growing seasons 2014 & 2015. The crops samples and their corresponding soils were analysed for ^{226}Ra and ^{228}Ra using High Purity Germanium Detector (HPGe). Quality assurance objectives were followed through the efficiency calibration of the HPGe detector, background estimation, assessment of combined uncertainties and the analyses of IAEA certified soil and plant reference materials. The total activity concentrations of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K in soil were ranged from 6 ± 1.2 to 87.5 ± 4.5 , 3.8 ± 1.2 to 19.3 ± 2.0 and 91.3 ± 4.1 to 781.8 ± 37 Bq kg^{-1} respectively. The average activities of ^{226}Ra and ^{228}Ra in the crops under this investigation ranged from 0.1 to 2.8 and 0.2 to 13.9 Bq kg^{-1} dry weight respectively. Cabbage and Cucumber showed maximum concentration ratios of 0.07 for ^{226}Ra whereas for ^{228}Ra Cabbage and Aubergine had relatively higher values of 0.13. The results obtained are within the range of the corresponding values reported by IAEA in 1994 & 2010. The present work may provide a database of TF_s for these agricultural environments to be used in radiological safety assessment models.

Index Terms—Transfer Factor, Soil, Crops, Gamma Spectrometry, Natural Radioactivity, Egypt.

I. INTRODUCTION

Radium (Ra) isotopes are important from the point of radiation and environmental protection. Their high toxicity has stimulated the continuing interest in methodology research for determination of Ra isotopes in various media. Radium is a radioactive element for which no stable isotope is known. The mass numbers of the known isotopes range from 206 to 230. Among them, two radium isotopes, ^{226}Ra and ^{228}Ra are very significant from a radiological protection point of view due to their relatively long half-lives, presence in nature, and high dose conversion factors. Radium isotopes are important because they can be easily incorporated into bones due to having similar properties to other elements from Group II (i.e. Calcium) and produce short lived radionuclides of high specific activity [1]. The metabolic behavior of radium in the body is similar to that of calcium but radium poses an external as well as an internal health hazard since strong gamma radiation is associated with several short-lived decay products of ^{226}Ra . Inhalation risk is primarily associated with the alpha particles from radon and its short-lived daughters, which are emitted within the lung, where they can damage the cells

lining the airways potentially leading to lung cancer. Some studies indicate that chronic exposure to radium can also induce bone sarcomas [2].

The transfer of radionuclides from soil to plant is mostly expressed as plant/soil concentration ratios or transfer factors (TF_s). TF_s are used as parameters in radiological models for predicting the concentration of radionuclides in foodstuffs and for estimating dose impacts to man [3, 4].

Naturally occurring radionuclides like ^{226}Ra have not been studied as extensively as their artificial counterparts. The amount of relevant information about radium accumulation by plants is very limited as reported by Ewers et al., in 2003[5], Vasconcellos et al., 1987[6], Chen et al., 2005[7], Fernandes et al., 2006[8], Sam and Ericsson, 1995[9] and Markose et al., 1993[10] most of these studies have reported that radium concentrations in plants range from 0.5 to 410 Bq kg^{-1} dry weight (DW), depending on the type of soil and ^{226}Ra concentration.

The objective of the present study was to derive the radium transfer parameters from soil to plant in some Egyptian crops in different farms at Al-Qalyubia governorate. The work has been carried out for two seasons 2014 & 2015 to observe the modification of TF_s from soil to crops in the locations under this investigation.

II. SAMPLES COLLECTION AND PREPARATION

A. Soil Samples

Soil samples were collected from different farms in El-Obour city, Al-Qalyubia governorate that may be irrigated with ground water to determine the radium content Figure 1. The soil collected at depth about 5 to 10 cm from the surface, each sample about 1 kg was placed in plastic bags and transported to the laboratory. Physical and chemical properties were measured as illustrated in Table 1. The collected samples were prepared for counting by drying at 105°C , dry roots, grasses and other residual parts were discarded, crushed, homogenized and sieved through 200 mesh sieve shakers. Finally weighted and transferred the samples to 100 ml Marinelli beakers.

B. Crops Samples

The crops used in the present work were Cabbage, Cucumber, Aubergine, Pepper, Zucchini and Tomato. The experiments were performed outdoors, under ambient tropical and growth conditions. The moisture content of the soil used for these crops was maintained between 50% and 75% throughout the growing period. The deep tube well water may be used for irrigation. The amount and frequency of water application differed for the different crops. The experiment was conducted with four replications during the two growing seasons (2014 & 2015).

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The crops samples were collected after the cultivation. The crop samples were washed carefully with double distilled water twice to remove the dust. The samples were dried at 100°C until they acquired a constant dry weight and homogenized. The weigh 5 kg from crop samples, digested by acids according to the standard operating procedure (SOP) followed by NCNSRC in 2003 then packed into 100 cc Marinelli beakers [11].

C. Water Samples

Radium isotopes (^{226}Ra and ^{228}Ra) were determined in the water samples using the procedure described by A. El-Sharkawy et al., 2013 [12]. Four liters of each sample could pass through the purolite resin packed in columns, the resin was transferred to the counting containers and the containers were tightly sealed for about four weeks to allow secular equilibrium between ^{226}Ra , ^{228}Ra and their decay products. Efficiency calibration of the germanium detector for the radium determination was carried out using standard samples. Standard resin samples were prepared by spiking Double Distilled Water (DDW) with a known activity concentrations of the tracers ^{226}Ra and ^{228}Ra and pass it through the resin. The contaminated resin samples containing a known activity of the radionuclide of interest were used to provide an identical matrix and other parameters such as (flow rate, resin volume, counting time, geometry). All prepared samples sealed and stored for one month to insure the secular equilibrium between radium isotopes and their decay products before measured by gamma spectrometry.



Figure 1. Map of the sampling locations

III. RADIOACTIVITY AND TRANSFER FACTORS DETERMINATION

Activity measurements have been performed by gamma ray spectrometer at Nuclear and Radiological Regulatory Authority, Central Laboratory for Environmental

Radioactivity Measurements, Intercomparing and Training (CLERMIT) using vertical HPGe detector of relative efficiency 40% and Full Width at Half Maximum (FWHM) of 1.95 keV for ^{60}Co gamma energy line at 1.33 MeV. The detector was operated with Canberra Genie 2000 software for gamma acquisition and analysis. HPGe detector was contained in 5 cm thickness free standing lead castle providing a low background environment to shield the detector from lead fluorescent X-rays and bremsstrahlung, the lead is lined with 1.5 mm iron and 1.0 mm Cu metals, before performing the spectroscopic measurements, the spectrometer has been calibrated using multi nuclides standard sources distributed in a reference materials (soil) which have certified concentration of natural radioactivity which provided by the IAEA. The specific activity calculations of ^{238}U series and ^{232}Th series were obtained indirectly from the gamma rays emitted by their progenies which are in secular equilibrium with them while ^{40}K activities were determined from the gamma line at 1460.7 keV.

Transfer factors (TF_s), which are the ratios between the activity concentrations in crops and soil (Bq/kg Dry Weight, (DW) of crop divided by Bq/kg Dry Weight (DW) of soil). TF_s can be used as index for the accumulation of radium by crops or the transfer of radium from soil to crops as illustrated by Chen et al., 2005 [7,13].

$$\text{TF}_{\text{Crops}} = \frac{\text{Activity in dry crops (Bq/kg)}}{\text{Activity in dry soil upper 20 cm (Bq/kg)}}$$

IV. RESULTS AND DISCUSSIONS

The physical and chemical characteristics of the soil are illustrated in Table 1. Chemical characterization revealed that soils were possessed low fertility levels, this is related to low levels of nutrients (Ca^{2+} , Mg^{2+} , K^+ , and P^{3+}) as well as low Cation Exchange Capacity, (CEC) values 2.9. Low base saturation indexes indicated that small proportions of exchangeable Ca^{2+} , Mg^{2+} , and K^+ are occupying negative charged sites of the colloids. Conversely exchangeable Al^{3+} ions are dominant as indicated by relatively high Al saturation index ($m > 30\%$). Clay soil presented slightly higher fertility, with higher Ca and Organic Matter (OM) levels.

Table 1. Physical and chemical properties of the soil

$\text{CaCO}_3\%$	0.17
PH	8.5
Coarse Sand %	72.86
CEC mg /100g	2.91
Silt %	1.15
OM %	0.09
Fine Sand %	23.15
TSS %	0.05
Clay %	2.60
Structure	Sandy

TSS: Total Suspended Solids.

CEC: Cation Exchange Capacity.

OM: Organic Matter.

Table 2. Comparison between the activity concentrations of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K (Bq.kg⁻¹) in the soil samples from different countries and the present study.

Region/Country	^{226}Ra (^{238}U) Bq/kg	^{232}Th Bq/kg	^{40}K Bq/kg	Ref.
Cairo, Egypt	5.3-66.8	5-37.3	41.5-418	16
Spain	6-250	2-210	25-1650	17
Turkey	10-58	--	161-964	18
Jordan	16.3-57.3	7.6-16.2	121.8-244.8	19
Iran	8-55	5-42	250-980	20
Louisiana, USA	34-95	4-130	43-719	21
China	2-440	1-360	9-1800	20
India	7-81	14-160	38-760	21
World Average	35	30	400	20
El-Qalyubia, Egypt	(6.2±1.4) to (85.5±4.6)	(3.4±1.2) to (20.3±2.0)	(92.1±4.4) to (780.2 ±30.1)	Present study

Table 3. Specific activity (Bq kg⁻¹) or (Bq l⁻¹) for ^{226}Ra and ^{228}Ra in soil and water using γ -Spectroscopy

Sample Locations	Soil		Water	
	^{226}Ra (Bq/kg)	^{228}Ra (Bq/kg)	^{226}Ra (Bq/l)	^{228}Ra (Bq/l)
Site-1	37.8 ± 1.2	100.9 ± 3.2	4.70 ± 0.2	46.00 ± 3.7
Site-2	23.3 ± 0.8	94.7 ± 3.1	5.40 ± 0.2	31.70 ± 2.0
Site-3	26.3 ± 0.9	79.1 ± 2.6	7.20 ± 0.2	42.80 ± 1.2
Site-4	25.9 ± 0.9	89.2 ± 2.9	10.70 ± 0.3	75.40 ± 2.1
Site-5	19.4 ± 1.3	48.1 ± 1.6	9.00 ± 0.9	50.90 ± 1.4
Site-6	21.8 ± 1.6	40.5 ± 1.4	7.40 ± 1.0	24.10 ± 0.8
Site-7	11.1 ± 0.9	24.8 ± 0.9	LLD	10.90 ± 0.3
Site-8	19.1 ± 1.3	45.7 ± 1.5	LLD	7.90 ± 0.3
Site-9	21.0 ± 0.7	36.2 ± 1.2	LLD	LLD
Site-10	14.0 ± 0.5	36.7 ± 1.5	LLD	9.70 ± 0.5
Site-11	27.1 ± 1.3	38.0 ± 1.6	LLD	12.70 ± 0.5
Site-12	8.1 ± 0.7	10.3 ± 0.9	LLD	10.22 ± 1.0
Site-13	8.0 ± 0.7	13.8 ± 1.2	LLD	13.67 ± 1.1
Site-14	9.0 ± 0.8	16.9 ± 1.5	LLD	14.72 ± 1.5
Site-15	14.5 ± 1.3	28.6 ± 2.6	LLD	24.36 ± 2.6
Site-16	9.4 ± 0.8	12.9 ± 1.2	LLD	7.11 ± 0.7
Site-17	12.3 ± 1.1	21.2 ± 1.9	LLD	7.02 ± 0.6
Site-18	17.0 ± 1.5	31.6 ± 2.8	6.71 ± 0.7	53.72 ± 5.4

LLD: Low Limit of Detection.

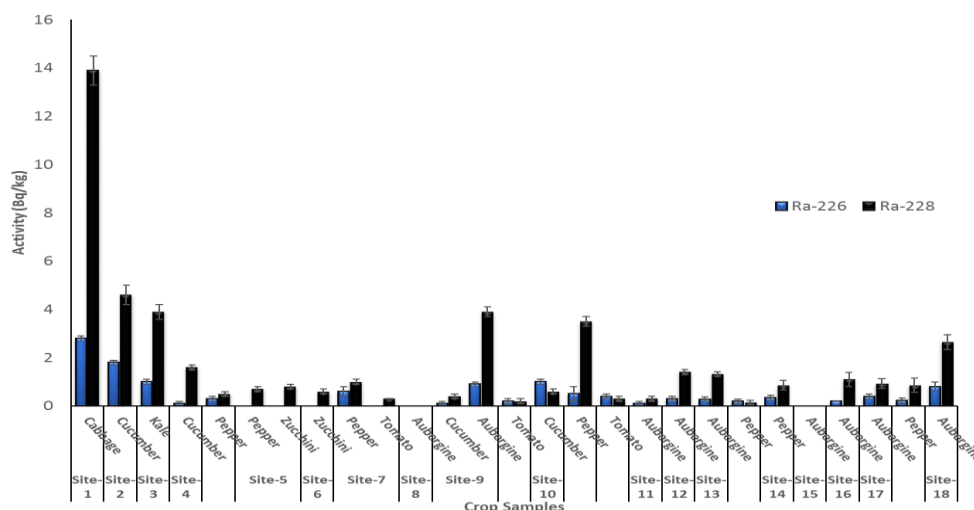


Figure 2. Activity concentrations of ^{226}Ra and ^{228}Ra isotopes in different crops at different sites.

The activity concentrations of ^{238}U (^{226}Ra), ^{232}Th and ^{40}K were determined depending on its corresponding locations for each soil samples. The ^{238}U (^{226}Ra) specific activities ranged from 6.2 ± 1.4 to 85.5 ± 4.6 Bq kg^{-1} with an average value of 31.1 ± 2.2 Bq kg^{-1} . ^{232}Th specific activities ranged from 3.4 ± 1.2 to 20.3 ± 2.0 Bq kg^{-1} with an average value of 11.01 ± 2.0 Bq kg^{-1} . ^{40}K specific activities ranged from 92.1 ± 4.4 to 780.2 ± 30.1 Bq kg^{-1} with an average value of 264.15 ± 12 Bq kg^{-1} . Table 2 compares the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in soil samples determined in the present study with those by other investigators in different countries in the world. In comparison, it was found that the obtained activity concentrations for ^{226}Ra , ^{232}Th and ^{40}K were almost matching those levels in other countries.

Table 3 presents the calculated specific activity (Bq kg^{-1}) for radium isotopes (^{226}Ra and ^{228}Ra) in soil and water by gamma spectrometry. As it can be verified that ^{226}Ra and ^{228}Ra concentrations in soil and water depending on the type of the fertilizers and quantity used in agriculture. In addition, the sandy soil usually has low ^{238}U concentration compared to other natural radionuclides.

The lowest specific activity concentrations in soil samples for ^{226}Ra and ^{228}Ra were (8 ± 0.7 Bq kg^{-1}) and (10.3 ± 0.9 Bq kg^{-1}) respectively for radium isotopes at sites 13&12. This could be attributed to either the nature of the soil itself or the fact that this sample location in the fallout pollutants from the industrial zone about 10 km. It could be also due to both of two reasons together. The highest specific activity values were (37.8 ± 1.2 Bq kg^{-1}) and (100.9 ± 3.2 Bq kg^{-1}) for ^{226}Ra and ^{228}Ra respectively for the soil collected from site 1 this was found in the site which used high content from fertilizers. In addition, the atmospheric releases from some pollutants due to the industrial zone that might contribute to the natural radionuclides concentrations of the soil samples.

The specific activity for radium in water samples ranged from Low Limit of Detection, (LLD) to 10.3 ± 0.9 Bq kg^{-1} and from LLD to 75.4 ± 2.1 Bq kg^{-1} for ^{226}Ra and ^{228}Ra

respectively. The activity concentration can be explained by the rock origin and the geological composition of ground well that used for the irrigation.

TF_s of ^{226}Ra and ^{228}Ra for the crops grown in two seasons are given in Table 4. The difference in the TF values for different crops seem to be due to soil parameters,

experimental conditions and plant species. The results show a significant influence of the plant species. These differences may be due to different soil properties and other factors because it is well known that the uptake of radionuclides by crops from the soil depends on the plant species as well as on the soil variables, e.g. clay content, pH, CEC, OM [14 & 15].

The results of the activity concentrations of ^{226}Ra , ^{228}Ra radionuclides in the crops (Cabbage, Cucumber, Kale, Aubergine, Pepper, Zucchini and Tomato) samples collected from the sites of the studied area as well as their TF_s are illustrated in Figures 2, 3 and Table 4. It was observed that, the activity concentration varies with the crops type and this agree with the results obtained from other investigators Mollah, A.S. et al., 1994[22] and Karunakar et al., 2003[23]. In the other publications it was clearly concluded that the TF_s of ^{226}Ra and ^{228}Ra from soil to plants are variable and depended on many factors that explained by IAEA, 1994 [14] and Ng YC, 1982 [15].

The averages activity concentrations of ^{226}Ra and ^{228}Ra were (2.8 ± 0.1 , 0.75 ± 0.1 , 1.0 ± 0.1 , 0.36 ± 0.08 , 1.26 ± 0.15 , LLD and 0.3 ± 0.1 Bq kg^{-1}) and (13.9 ± 0.6 , 1.8 ± 0.18 , 3.9 ± 0.3 , 1.5 ± 0.7 , 0.7 ± 0.1 and 0.27 ± 0.1 Bq kg^{-1}) respectively for the crops in the order Cabbage, Cucumber, Kale, Aubergine, Pepper, Zucchini and Tomato as illustrated in Table 4. The obtained results show differences in ^{226}Ra and ^{228}Ra uptake between crops species. The highest accumulation of the radionuclides ^{226}Ra and ^{228}Ra were detected in Cabbage-1 with activity concentration (2.8 ± 0.1 Bq kg^{-1}) and (13.9 ± 0.6 Bq kg^{-1}) respectively, which planted on sites 1.

The differences in accumulation of radium isotopes activities concentrations were notice between crops from the same type and between cultivars of the same plant species as in Figure 3. The calculated soil to crop transfer factors (TF_s) ranged from 0.004 to 0.077 for ^{226}Ra and ranged from 0.006 to 0.138 for ^{228}Ra . The highest TF_s for ^{226}Ra and ^{228}Ra were found in Cucumber-2 & Cabbage-1 respectively while the lower values observed in Cucumber-4 & Aubergine-11 for ^{226}Ra and Pepper-4 & Tomato-9 for ^{228}Ra as illustrated in Table 4.

In general, the obtained TF_s values are within the range that reported by Chen, S.B. in 2005[24], Markose, P.M et al 1993[25], Sam, A.K., Ericsson et al., in 1995[26] and Vasconcellos et al., 1987; [27]. Comparison of obtained TF_s

calculated with that obtained from other publications is very difficult because all the experiments were carried out in different histories and different substrate properties, including different nutrient and organic matter contents and this agree with the interpretation with Sam and Ericsson in 1995 [26]. The soil to crops TF_s in the present study for ^{226}Ra and ^{228}Ra are within the range that reported by IAEA, TECDOC NO. 472 [28]. Soil to plant TF_s of Ra isotopes for non-leafy vegetables, the mean value reported is 3.2×10^{-2} with a minimum of 5.2×10^{-4} and maximum of 7.0×10^{-2} . The leafy vegetables such as Cabbage and Kale the mean value reported is 2.7×10^{-2} with a minimum of 3.0×10^{-3} and maximum of 4.3×10^{-1} [28]. IAEA has compiled soil to plant TF of radium isotopes for fruits in tropical environment with an average

value 1.0×10^{-1} this value compatible with the results obtained in the present work. Finally, the concentration of radium in the crops depends on the radium isotopes content in soil, water and the metabolic characteristics of the crops under this investigation. The variations in radium levels in crops may be expected depending on the radium content of soil that varies widely. Chemical factors such as the amount of exchangeable cations in the soil that may be used for explaining the small portions and relatively high values of TF_s for ^{228}Ra in samples as Cabbage-1, Aubergine (9,12,13 and 16) and the results agree with published values obtained by James, J. P. et al. in 2011 [29].

Table 4. The average activity concentrations of ^{226}Ra and ^{228}Ra radionuclides in different crops and their transfer factors.

Locations	Crops type	Activity (Bq/kg) Dry Weight		TF_s	
		^{226}Ra (Bq/kg)	^{228}Ra (Bq/kg)	^{226}Ra	^{228}Ra
Site-1	Cabbage	2.8 ± 0.1	13.9 ± 0.6	0.074	0.138
Site-2	Cucumber	1.8 ± 0.1	4.6 ± 0.4	0.077	0.049
Site-3	Kale	1.0 ± 0.1	3.9 ± 0.3	0.038	0.049
Site-4	Cucumber	0.1 ± 0.1	1.6 ± 0.1	0.004	0.018
	Pepper	0.3 ± 0.1	0.5 ± 0.1	0.012	0.006
Site-5	Pepper	LLD	0.7 ± 0.1	--	0.015
	Zucchini	LLD	0.8 ± 0.1	--	0.017
Site-6	Zucchini	LLD	0.6 ± 0.1	--	0.015
Site-7	Tomato	LLD	0.3 ± 0.1	--	0.012
	Pepper	0.6 ± 0.2	1.0 ± 0.1	0.054	0.040
Site-8	Aubergine	LLD	LLD	--	--
Site-9	Aubergine	0.9 ± 0.1	3.9 ± 0.2	0.043	0.108
	Cucumber	0.1 ± 0.1	0.4 ± 0.1	0.005	0.011
	Tomato	0.2 ± 0.1	0.2 ± 0.1	0.010	0.006
Site-10	Cucumber	1.0 ± 0.1	0.6 ± 0.1	0.071	0.016
	Tomato	0.4 ± 0.1	0.3 ± 0.1	0.029	0.008
	Pepper	0.5 ± 0.3	3.5 ± 0.2	0.036	0.095
Site-11	Aubergine	0.1 ± 0.1	0.3 ± 0.1	0.004	0.008
Site-12	Aubergine	0.30 ± 0.1	1.41 ± 0.1	0.037	0.137
Site-13	Aubergine	0.28 ± 0.1	1.33 ± 0.1	0.035	0.096
	Pepper	0.19 ± 0.1	0.14 ± 0.1	0.024	0.010
Site-14	Pepper	0.34 ± 0.1	0.86 ± 0.2	0.038	0.051
Site-15	Aubergine	LLD	LLD	--	--
Site-16	Aubergine	0.20 ± 0.0	1.10 ± 0.3	0.021	0.085
Site-17	Aubergine	0.39 ± 0.1	0.93 ± 0.2	0.032	0.044
	Pepper	0.23 ± 0.1	0.86 ± 0.3	0.019	0.041
Site-18	Aubergine	0.80 ± 0.2	2.65 ± 0.3	0.047	0.084

LLD: Low Limit of Detection.

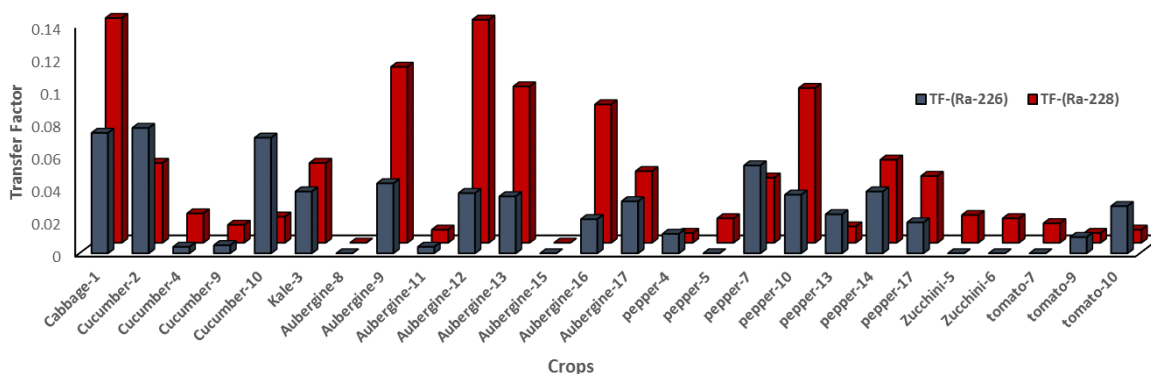


Figure 3. The transfer factors (TF_s) of ^{226}Ra and ^{228}Ra radionuclides in the crops at different locations

V. CONCLUSION

The natural radioactivity concentration in soil, water and transfer factor of soil to plant were determined in some locations at Al-Qalyubia governmental, Egypt. The activity concentrations of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K in soil samples were varied from (6.2 ± 1.4 to 85.5 ± 4.6), (3.4 ± 1.2 to 20.3 ± 2.0) and (92.1 ± 4.5 to 780.2 ± 30.1) Bq kg⁻¹ respectively. The average concentrations of ^{226}Ra and ^{228}Ra were (2.8 ± 0.1 , 0.75 ± 0.1 , 1.0 ± 0.1 , 0.36 ± 0.08 , 1.26 ± 0.15 , LLD and 0.3 ± 0.1 Bq kg⁻¹) and (13.9 ± 0.6 , 1.8 ± 0.18 , 3.9 ± 0.3 , 1.5 ± 0.7 , 0.7 ± 0.1 and 0.27 ± 0.1 Bq kg⁻¹) respectively for the crops in the order Cabbage, Cucumber, Kale, Aubergine, Pepper, Zucchini and Tomato. Soil to plants TF_s for ^{226}Ra and ^{228}Ra are within the range that reported by IAEA in 1994 [14] and 2010 [30]. However, the present study provides a database of TFs concerning some of the tropical environment to be used for radiological safety assessment, and it serves as a future study on the transfer of radionuclides by crops growing on different soils.

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