Soil - to - Plant Transfer Factors of Naturally Occurring Radionuclides for Selected Plants growing in Qassim, Saudi Arabia

A. El-Taher [1,2] and Ahmad Al-Turki [1,3]

[1]Promising Research Center in Biological Control and Agricultural Information (BCARC),
[2]Physics Department, College of Science, Qassim University, P. O. Box 6622, Buraydah, 51452, Saudi Arabia
[3]College of Agriculture and Veterinary Medicine, Qassim University, P. O. Box 6622, Buraydah, 51452, Saudi Arabia.
atef_eletaher@hotmail.com

Abstract: Assessment of any release of radioactivity to the environment is important for the protection of public health, especially if the released radioactivity can enter the food chain. In the present work, a pot experiment carried out in greenhouse in Qassim University in order to examine the effect of contaminated irrigation water on migration and retention of radioactive elements in soil and plants. Natural radioactivity levels of ^{226}Ra, ^{228}Ra, ^{238}U and ^{40}K in nine samples (two soil samples and 7 dried plant samples) was evaluated using HPGe detector. Ra-226 showed results from below detection limit up to 22.9 Bq/kg^{-1} with an average 13.1 Bq/kg^{-1}. All U-238 showed results below detection limit except one soil sample with a result of 13.6 Bq/kg^{-1} (1.1 ppm). Ra-228 showed results from below detection limit up to 31.7 Bq/kg^{-1} with an average 17.5 Bq/kg^{-1}. K-40 showed results from 221 up to 1212 Bq/kg^{-1} with an average 151 Bq/kg$^{-1.}$ These results were compared with reported ranges in the literature from other location in the world. The correlation between the specific activities of the plant samples with of the associated irrigating water for ^{226}Ra and ^{228}Ra. and with total radium are calculated. The results would be useful for establishing of the database in the area under consideration and represent a basis to assess any future changes in the radioactivity background levels due to various geological processes or any artificial influences around the area.
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1. Introduction

Assessment of any release of radioactivity to the environment is important for the protection of public health, especially if the released radioactivity can enter the food chain. Contamination of the edible parts of a plant following soil contamination is the result of a series of closely coordinated steps from root uptake, transport across the root and transport to the aerial parts of the plant. The chemico-physical characteristics of the soil, root interception, ion uptake by roots, ion transport, ion translocation, remobilization of mineral nutrients, and root exudation are some of the factors affecting the transfer of radionuclides from soil to plants. The soil-to-plant Transfer Factor (TF). The TF relates the radionuclide concentration in edible products to that in the soil. TFs can be based on a quantity of soil and expressed as the ratio of Bq/kg dry weight of plant to Bq/kg dry weight soil or can be based on a surface area of soil and expressed as Bq/kg dry weight of plant to Bq/m^2 (IAEA, 1994). To facilitate comparisons, the contamination of crops is calculated on a dry weight basis to minimize differences due to environmental or varietal conditions. Some authors prefer to express TFs for fruit on a fresh mass basis because consumption data are given in fresh mass.

Radium is chemically similar to the essential element Ca and other alkaline earth elements, and is considered to participate actively in biogeochemical functions. In one study, the plants around uranium mine and mill complex were reported to contain elevated levels of uranium, thorium and radium; the associated soil-to-plant concentration ratios (CR) were also evaluated. However, some plants, such as Brazil nuts and ferns, are known to have high concentrations of ^{226}Ra and ^{228}Ra, or high soil-to-plant concentration ratios, even when grown under normal radiation conditions. Most plants selectively take up radium rather than its parent nuclides, uranium (^{238}U) and the thorium series (^{232}Th), from the soil. The long-lived ^{226}Ra ($t_{1/2}$ =1600 years) and ^{228}Ra ($t_{1/2}$ = 5.75 years) concentrations could be a consequence of long-term accumulation over months or years. Comparatively, ^{224}Ra would be taken up and then reside in plants with an equilibrium concentration of only a few days, because of its short half-life of 3.66 days. Therefore, the temporal change in level of ^{224}Ra could reflect the growth rate of plants and could be useful for understanding plant nutrition and mineral metabolism. To understand the metabolism of ^{224}Ra in plants, one must clarify the source of uptake of ^{224}Ra from soil and decay of its parent nuclides ^{228}Th and ^{228}Ra inside

the plant tissues. In the present work are, A pot experiment carried out in greenhouse in order to examine the effect of contaminated irrigation water on migration and retention of radioactive elements in soil and plants.

Radioactive Contamination of Soils

It is a well-known fact that a number of natural radioactive elements such as uranium, thorium, radium and potassium occurs in the soil. Thus the soils and also the crops harvested are somehow radioactive. A general review of the amounts of fallout which have reached the surface of the earth is given by (Garcia *et al.,* 1995). As a consequence, the radioactivity of the earth has increased and nowadays these artificial radioactive elements can be found in the soil, water and food items of man and animal. The increase in radioactivity of the soil is small compared to the radioactivity from naturally occurring radioactive nuclides, where the quantities of radioactive nuclides which have entered the soil are so small that no cases of solubility product of any compound reached (El-Taher and Makhluf., 2010).

Radioactive contamination of plant

1. Direct contamination of plant

When the so-called fallout or waste reaches the surface of the earth, it will pass the crop before it reaches the soil. i.e., direct contamination can take place where by radioactive material "sticks" to the surface of the crops.

2. Indirect contamination of the plants:-

With regard to the contamination of the crop via absorption of radioactive nuclides through the roots, three major factors may be distinguished besides the root pattern, as follows**:**

a- The availability for the plant of the nuclide after its reaction with the soil materials, this availability will be low for Cs-137, because of the strong fixation by the soil, it will be rather, higher for Sr-90, being only moderately strongly absorbed by the soil.

b- The capacity of the roots to take particular elements. This capacity is high for Sr-90 and Cs-137. Both have similar characteristics as calcium, and potassium respectively which are taken up by the plants in considerable quantities.

c- The possibility of transport of these elements from roots to the aboveground parts and to the tuber. The transport possibilities of strontium and cesium are large as the transport of calcium and potassium is also large (Jacobson and Overstreet., 1998).

Factors affect on the behavior of radionuclides in soil:

The concentration of naturally occurring radio nuclides in soil depends on the rock type from which the soil is formed. The soil can be contaminated by radio nuclides deposition either from what originally discharged into the atmosphere, or from direct discharge of waste to land or water ways. The amount of radio nuclides in the soil depends on its organic matter content, soil to water ratio, site characteristics, rate and amount of rainfall and soil drainage (NCRP., 1975). Moreover, the behavior of radio nuclides in soil is affected by different biochemical processes (Schulz,., 1965; John and Meyer., 1983; Abu – Khadra and Eissa.,.2008; El-Taher., 2011; El-Taher., 2012).

Transfer factor TF

Radionuclide uptake by plants from contaminated soil represents a key step of radionuclide input into human food chain; this phenomenon is described by soil-plant transfer factor that is defined as the ratio between plant specific activity and soil specific activity. Plants are the primary recipients of radioactive contamination to the food chain following atmospheric releases of radionuclides. The transfer factor (TF) is a value used in evaluation studies on impact of routine or accidental releases of radionuclide into the environment for most important agricultural products is known. For other areas and especially the developing countries TFs are less known. The soil –to-plant transfer factor is regarded as one of the most important parameter in environmental safety assessment needed for nuclear facilities. This parameter is necessary for environmental transfer models which are useful in prediction of the radionuclide concentrations in agriculture crops for estimating dose intake by man.

Transfer factor (TF) is defined as the ratio of radionuclide concentrations in vegetation and soil. The soil to plant transfer factors were determined according to the relation:

$$TF = \frac{Bq.kg{-}1\ dry\ crops}{Bq.kg{-}1\ dry\ soil} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

The dry weight was preferred because the amount of radioactivity per kilogram dry weight is much less variable than the amount per unit fresh weight. It reduces uncertainties (IAEA., 1994).

2.Experimental

A. Field Experiments:

Two types of soil (sandy and sandy loam) with very low background radioactivity content were used to grow two types of plants in pots. Three well-water varying in their radioactive contents used for irrigation. Each treatment has three replicates in a randomized complete design. Control treatment irrigated with free- water radioactive contents. Greenhouse condition is preferred in order to avoid contaminants and extreme weather changes and that may affect the experiment in addition to the unpredicted consequences of the materials being tested which may harm the environment. Usages of pots

provide more controlled compost and soil environments.

Samples preparation

Nine samples (two soil samples and 7 dried plant samples). Each sample was packed into 75 ml standard size beakers and tightly sealed to acquire secular equilibrium. Two reference materials were packed into the same standard size beakers in order to use them to perform efficiency calibration. A Hyper pure Germanium detector (HPGe), coaxial type, P-type with relative efficiency of 50% has been exploited. The detector is shielded with low-level background lead shield. The HPGe was calibrated for the efficiency using the reference material RGU-1 from IAEA. Efficiency calibration using RGU-1 has been tested several times in many proficiency tests and publications. It was assured that its radium-226 is in equilibrium with the uranium contents. The certified activity of uranium is 400 ppm which refers to 4960 Bq/kg^{-1}. Energy transition of the ^{226}Ra daughters (^{214}Pb and ^{214}Bi) were used to develop the efficiency calibration curve (El-Ther., 2010 a-f; El-Taher & Madkour., 2014). A fourth degree polynomial fitting was performed to reach the best R^2 value (≈ 0.999).

3. Results and Discussions

The results of natural radioactivity levels of ^{226}Ra, ^{228}Ra, ^{238}U and ^{40}K are shown in Table. 1. Ra-226 showed results from below detection limit up to 22.9 Bq/kg^{-1}. All U-238 showed results below detection limit except one soil sample with a result of 13.6 Bq/kg^{-1} (1.1 ppm). Ra-228 showed results from below detection limit up to 31.7 Bq/kg^{-1}. K-40 showed results from 221 up to 1212 Bq/kg^{-1}.

Ra-226

Radium-226 was measured using its progenies ^{214}Pb with energies 295.2 keV (19.3%), 351.93 keV (37.6%), and ^{214}Bi with energies 609.31 keV (46.1%), 1120.29 keV (15.1%), 1764.49 keV (15.4%). Radium was determined based on the above mentioned energy transitions after achieving secular equilibrium for 28 days after sample's packing. Average values were considered and occasionally outliers were excluded especially at low levels.

Ra-228

Radium-228 was assessed using the energy transitions of ^{208}Tl at 583.19 keV (84.5%), and ^{228}Ac at 911.2 keV (25.8%). Average values were calculated.

Uranium-238

Uranium-238 was determined based on its direct daughter ^{234}Th with the combined energies of 92.38 (2.81%) keV and 92.8 keV (2.77%). The energy transition of ^{228}Ac at 93.4 keV (5.6%) was not affecting the assessment due to the lack of thorium in most of the investigated samples with any contributing amounts. The choice of the above mentioned an energy transitions were based on their low minimum detectable activity (MDA).

K-40

Potassium-40 was assessed based on its energy transition of 1460.83 keV (11.0 %). Efficiency of each transition line was determined either by direct comparison for those energies existed in the reference material or derived from the above mentioned produced efficiency calibration curve.

Background

Background for each energy transition was routinely subtracted for the reference sample and the measured samples.

Minimum Detectable Activity

The detection limit was calculated based on the Currie detection limit method as follows;

$$MDA\ (counts) = 2.7+4.65\times\sqrt{BG}$$

Detection limit for each radionuclide was calculated separately for each sample based on the sample's weight as reported below (El-Taher & Madkour., 2011).

Quality Control Measures

The calibration was tested against IAEA proficiency and found no more than 5%.

Correlation of radium contents in the feeding water against the associated plants

The Transfer factors should be calculated based on the specific activities of soils against the associated plants. However the lack of enough data regarding the feeding soil to each plant limits the ability to perform this calculation. Alternatively, the correlation between the plant's specific activities and the associated irrigating water were reasonably presented. Figure 1 shows the correlation between the specific activities of the plant samples with of the associated irrigating water for ^{226}Ra and ^{228}Ra. Figure 2 shows the correlation between the specific activities of the plant samples with of the associated irrigating water total radium. Some initial discrepancies in the results were noticed due to the high uncertainty of the ^{226}Ra results. This is due to their relatively lower values compared to those of ^{228}Ra.

After excluding one outlier sample from the ^{226}Ra results, it is clear that there are considerably good correlations between radium contents the feeding water and those in the associated plants. However, these results are only initial results. Solid conclusions could not be achieved without comprehensive studies with large numbered representative samples.

Effect of Fertilizers applications

It is well known that phosphate fertilizers usually contain enhanced levels of uranium while phosphogypsum contains noticeable levels of radium. This is because of the partitioning processes during the beneficiation processes. Only two soil sample were

measured for their naturally occurring radionuclides inventories. The first was fertilized while the other was not. It is very hard to establish a solid and clear conclusion based on the measurement of those two soil samples. However, no preliminary evidence was found to indicate that there is a noticeable contribution due to the application of fertilizers. The lack of enough samples to compare against is a major factor in this uncertainty. Representative soil samples with enough fertilization history and their associated control samples would be essential to establish a good conclusion.

Table 1. Activity Concentrations of ^{226}Ra, ^{238}U, ^{228}Ra, and ^{40}K in soil and plant Samples in (Bq.kg^{-1}) Dry weight.

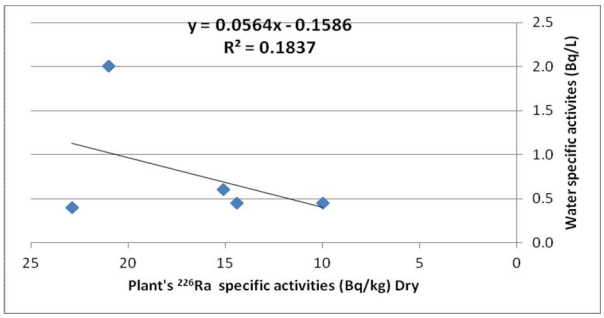
Sample Code	^{226}Ra	±SD	^{238}U	±SD	U (ppm)	^{228}Ra	±SD	^{40}K	±SD	^{40}K (%)
Soil (without Fertilizers)[463]	20.7	2.5	13.6	2	1.1	26.4	2.9	363	29	1.17
Soil (Fertilized)[464]	7.5	1.2	<7.5	—	—	7.8	1.3	221	18	0.71
A3 [465]	21	2.5	<29.6	—	—	31.7	3.2	1212	80	3.91
B4 [466]	15.1	1.6	12.1	—	—	22.7	2.5	1092	75	3.52
Co1 (1) [467]	22.9	2.6	<19	—	—	20.7	2.4	564	42	1.82
Co2 (2) [468]	14.4	1.5	<22	—	—	9.6	1.2	818	55	2.64
Date (1 -42-6) [469]	6.6	1	<15.4	—	—	<3.3	—	339	25	1.09
Dates (2-42-7) [470]	<2.1		<17.6	—	—	<3.9	—	418	30	1.35
5C [471]	10	1.2	<25	—	—	10.5	1	1047	81	3.37

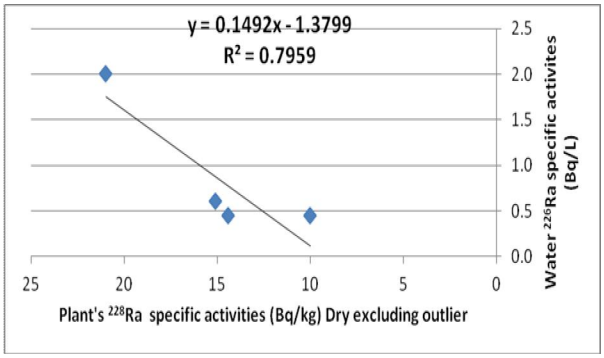
Table 2. Activity concentrations of ^{226}Ra and ^{228}Ra in groundwater (irrigation) samples in (Bq.L^{-1}).

Sample Code	^{226}Ra	±SD	^{228}Ra	±SD
A	2	0.11	6.81	0.31
B	0.62	0.06	3.44	0.12
C	0.45	0.04	1.24	0.05

Table 3. Results of the quality control samples compared to the reference (certified) values

QC Sample	IAEA Certified Value (Bq/g)	Lab Value	Bias (%)
IAEA-TEl-2011-03-sample #4	50.0	49.9	0.2
RGU-1	4960 (400 ppm)	4844 (390.6 ppm)	2.3





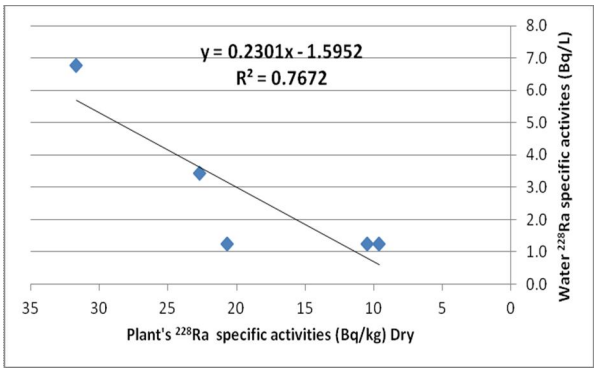
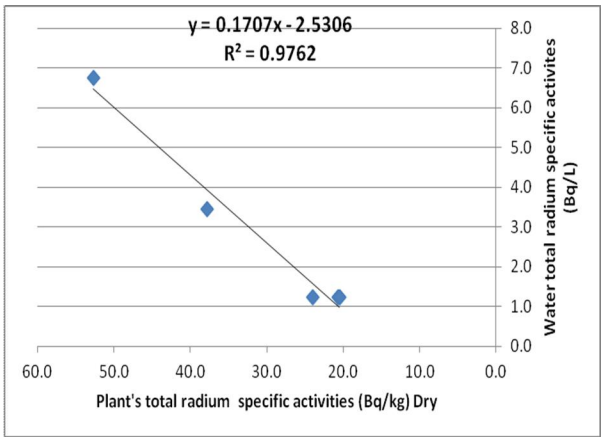


Figure 1: correlation between the specific activities of the plant samples with of the associated irrigating water for ^{226}Ra and ^{228}Ra



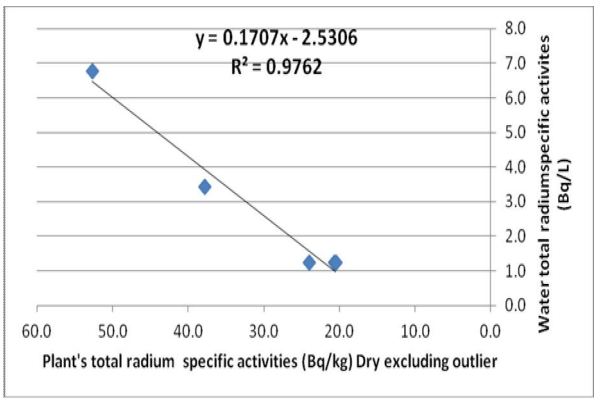


Figure 2: correlation between the specific activities of the plant samples with of the associated irrigating water total radium

Table 4: Comparison of natural radioactivity concentration (Bqkg^{-1}) in the soil Samples for from Qassim with previous study reported from different countries of the world.

Country	Mean activity concentration (Bqkg^{-1})			References
	^{226}Ra	^{232}Th	^{40}K	
Qassim, KSA	13.1(2.1 -22.9)	17.5 (3.3 – 31.7)	151(221- 1212)	Present study
Riyadh, KSA	14.5 (11 -30)	11(7 - 25)	225 (89- 320)	Alaamer, 2008
Albaha, KSA	37 (30 -45)	32(26 - 37)	343 (263- 435)	Alzahrani., 2012
Jeddah, KSA	9.3	7.4	369	Majid& Abulfaraj., 1992
Tourbh, KSA	4.35(1.95-13.07)	3.3(1.33-10.04)	71.74(39.92-193.71)	Alharbi., 2012
Makkah,KSA	13.3(10.1-18.1)	15.6(11.1-22.6)	514.3(454-592)	Hamidalddin. *et al.,* 2012
Oman	14.42(25.70-5.45)	9.95(20.18-0.88)	158.2 (282.86-10.49)	Saleh., 2012
Yemen Juban	44 (16.6 -84.4)	58(18-113)	823 (64 -1667)	El-mageed., *et al.,* 2011
Egypt Assiut	17(5-64)	18(2-96)	320(29-650)	UNSCEAR,2000
Iran	28(8-55)	22(5-42)	640(250-980)	UNSCEAR,2000
Aqaba Jordan	22-104	22-104	138-601	Al-Jundi *et al.,* 2003
Japan	33(6-98)	28(2-88)	400(15-990)	UNSCEAR,2000
Nigeria Minna	7.8 (1 -26.5)	29.4 (2.2 -70)	229 (43 – 468)	Kolo *et al.,* 2012
Nigeria Agbabu	9.38(4.98-18.6)	8.64(3.28-21.56)	65.75(8.5-165.77)	Isinkaye., 2008
Thailand	67.66 (25.73-156)	45 (6.77-91.28)	213 (62.4-949.44)	Kessaratikoon *et al.,* 2008
Turkey *Kirklareli*	37 (5 -111)	40(7- 151)	667 (87- 2084)	Taskin *et al.,* 2009
Algeria	50(5 - 180)	25 (2 - 140)	370 (66 - 1150)	UNSCEAR,2000
Sweden	42 (14 - 94)	42 (12 - 170)	680 (560 - 1150)	UNSCEAR,2000
China	32 (2 - 440)	41 (1 - 360)	440 (9 - 1800)	UNSCEAR,2000
India	41(14 – 160	29(7 – 81)	400(38- 760)	UNSCEAR,2000
United State	40(8 - 160)	35(4 - 130)	370(100 - 770)	UNSCEAR,2000
World average	32	45	420	UNSCEAR,2000

The results in parentheses correspond to the minimum and maximum values of the parameters.

Behavior of long-lived radionuclides in soil

The concentration of naturally occurring radionuclides in soil depends on the rock type from which the soil is formed. The soil is contaminated either by radionuclide deposition originally discharged into the atmosphere, or on the land surface by direct discharge of wastes. The concentration of radionuclides in soil increases by adsorption with soil particles and their precipitation on soil. The concentration decreases by a leaching process and also dilutes when the organic matter and soil water content increases behavior of radionuclides in soil on site characteristics, rate and amount of rain-fall and soil drainage (NCRP., 1975).

Losses of radionuclides from the plant root zone by infiltration into deeper soil layers are generally neglected in estimating radionuclide accumulation in soils. These losses are significant where soil permeability is high and the adsorption of radionuclides to soil particles is low. The low radionuclide adsorption capacity to soil particles leads to relatively high radionuclides uptake by plants. Moreover, the behavior of radionuclides in soil is affected by different biochemical processes, when organic matter decomposition changes soil property from an oxidizing to a reducing medium. This will affect the chemical form of the radionuclide present in soil. Radionuclide plant-soil ratio is affected by many factors that control plant uptake. These factors are:

1. Physico-chemical form of radionuclide.
2. Plant species and internal translocation mechanisms within the plant.
3. Soil characteristics.
4. Fertilizers and agricultural chemicals.
5. Chelating agents.
6. Distribution of radionuclides in soil

The physico-chemical form of the radionuclide strongly affects its retention by the soil particles and its availability for uptake by plants.

The soil type affects strongly the behavior of radionuclides in soil, and soil retention characteristics (Schulz., 1965). Sandy soils do not have the retention capacity of clay soils. Clay soils are composed of smaller particle sizes with larger surface area and negative charge surfaces (Nelson *et al.,* 1966).

The soil's pH value, affects the plants uptake. In alkaline soils (high pH) insoluble precipitates may be formed with carbonate, hydroxyl, phosphate or sulfide ions. These insoluble precipitates reduce the availability of radionuclides for plants. In acid soils (low pH), hydrogen replace the adsorbed cations which become more available to plants. In highly acidic soils (pH <5.5) some trace elements (particularly iron and manganese) may become toxic to plant growth (Schulz., 1965).

Fertilizers are chemical compounds added to increase the soil fertility and enhance plant production. They strongly affect both the stable element concentration, and soil acidity ((NCRP., 1975). The effect of lime stone ($CaCO_3$) addition appears to raise soil pH, increasing exchangeable calcium concentration, and decreasing the uptake of strontium. This is possibly due to the decreased solubility of $SrCO_3$ in alkaline conditions. Fertilizer with nitrogen in the nitrate form (potassium or calcium nitrates) and phosphate fertilizers may decrease soil acidity (Foth., 1978).

Organic fertilizer affects the ion exchange capacity, pH, stable element content of soil, as well as soil retention properties (Champlin. and Eichholz., 1967; Routson and Cataldo., 1978).

Chelating agents: Chelating agents are organic compounds which increase the ion mobility and reduce soil retention. This increases the plant uptake. Moreover, these agents enhance the translocation ability within the plant itself (Champlin. and Eichholz., 1967). In some situations of plants nutrient deficiencies they are useful because they decrease soil retention, therefore increasing plant uptake (Nelson *et al.,* 1966; Pickering *et al.,* 1966). Their effectiveness depends upon soil properties (particularly soil pH), chemical form of the radionuclide and the nature and concentration of chelating agent.

Conclusion

Radioactivity levels of ^{226}Ra, ^{228}Ra, ^{238}U and ^{40}K in nine samples (two soil samples and 7 dried plant samples) collected from Qassim area was evaluated using HPGe gamma-ray spectrometry. Ra-226 showed results from below detection limit up to 22.9 Bq/kg^{-1}. All U-238 showed results below detection limit except one soil sample with a result of 13.6 Bq/kg^{-1} (1.1 ppm). Ra-228 showed results from below detection limit up to 31.7 Bq/kg^{-1}. K-40 showed results from 221 up to 1212 $Bq/kg^{-1.}$ The correlation between the specific activities of the plant samples with of the associated irrigating water for ^{226}Ra and ^{228}Ra. and with total radium are calculated. Some initial discrepancies in the results were noticed due to the high uncertainty of the ^{226}Ra results. This is due to their relatively lower values compared to those of ^{228}Ra. it is clear that there are considerably good correlations between radium contents the feeding water and those in the associated plants. However, these results are only initial results. Solid conclusions could not be achieved without comprehensive studies with large numbered representative samples. The results would be useful for establishing of the database in the area under consideration and represent a basis to assess any future changes in the radioactivity background levels due to various geological processes or any artificial influences around the area.

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