

A Study on Transfer Factors of Radionuclides from Soil to plant

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Abstract: Knowledge of various radionuclides in soil plays an important role in health physics and geo-scientific research and monitoring of any release of radioactivity to the environment is important for environmental protection. The activity concentrations and distribution of natural radionuclides in soil and plants in addition to transfer factor from soil to plant from Qassim area, Saudi Arabia were determined. The measurements were carried out through gamma-ray spectrometry using NaI (Tl) detector. The mean and range of the concentrations of ²²⁶Ra and ²³²Th were 12.96 ± 3.4 (9.6–19.1) and 16.6 ± 7.1 (9.2– 28.3) Bq kg⁻¹. The range of the concentrations of ⁴⁰K in soil samples was (542–773) Bq kg⁻¹ with a mean value of 618 ± 82 Bq kg⁻¹. These results were compared with reported ranges in the literature from other location in the world. The radium-equivalent, and total absorbed dose rate were evaluated and compared with internationally recommended values. The transfer factor for ²²⁶Ra and ⁴⁰K to Alfalfa and wheat and Palm dates were measured. ²²⁶Ra TF values from soil to Alfalfa were found to be higher than wheat grains and Palm dates. ⁴⁰K TF were lower than those values reported in other studies. 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

Natural radionuclides are present in every human environment; earth material, water, air, foods and even our own body contain naturally occurring radioactive materials (NORM). The radionuclides present in the environment are normally in very low activity concentrations. The sources of these radionuclides are natural as well as man-made. The natural radioactive background originates from uranium and thorium series, from potassium - 40 and from the interaction of cosmic radiation with matter, while man-made sources include various applications of radionuclides in medicine, industries, consumer products and nuclear weapon tests (1-3). Weathering of the earth's crust is the ultimate mechanism for the release of primordial radionuclides into the soil, which constitutes the principal source of natural background radiation. Plants acquire these radionuclides via their roots or leaves, and animals acquire them through consumption of plants, phosphate-based mineral food supplements and soils.

These radionuclides are ultimately transferred to man by eating animal meat or milk or directly from plants by using them as food. Radionuclides ingested in food and to a lesser extent, water, account for a substantial part of the average radiation doses received by various organs of the human body, especially the skeleton (4). Analysis of these radionuclides in foodstuff is an important part of the

environmental monitoring program. These natural radioactive sources are the largest contributor of the radiation doses received by mankind. In natural sources, the most important are ⁴⁰K and the member of ²³⁸U and ²³²Th decay series. Naturally occurring potassium (⁴⁰K) is present virtually in all foodstuff as an essential constituent of cellular material. The general public receives about 180 μ Sv/year from ⁴⁰K, an essential cellular constituent. The total potassium content in an average human being is about 0.14 kg of natural potassium, so ⁴⁰K is a predominant natural radioactive substance in our own body. Such natural background is considered as a constant source of radioactivity to man and, therefore, its adverse consequences cannot be ignored (5-6).

There are two mechanisms for the contamination of vegetables and vegetation, i.e., by root uptake or directly by aerial deposition of fallout radionuclides on plants. It is necessary to carry out accurate assessment of these radionuclides in the daily used food materials in order to ascertain the degree of risk and deleterious effects to the public health (7-9). Various radionuclides are always present in food samples from natural sources or as a result of discharges of radioactivity from industries, hospitals, research laboratories or from nuclear weapon tests fallout. Most of the internal radiation doses received by human beings are due to the consumption of food contaminated with different

radionuclides. Therefore, it is necessary to determine and estimate the activity of various radionuclides present in soil and their transfer factor to different plant samples, in order to assess the radiation doses to human beings.

2. Radiation Sources in the Environment

Since the bombing of Hiroshima and Nagasaki (1945) and the Chernobyl Catastrophe (1986), atomic energy became a symbol of power and evil. Radiation also, became a part of our physical world fact of every day life. The natural sources of radiation in environment are responsible for some background radiation (10). The major sources of external gamma radiation are K-40, uranium-238, thorium-232 and their decays products. The artificial radionuclides are those which are produced as a result of nuclear weapon tests and as by products from nuclear fuel cycle and other from mining, milling, fuel enrichment, fuel preparation for reactor use, and plant operation disposal (11).

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 (12). 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.

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 (13).

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 (14). 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 (15). Moreover, the behavior of radio nuclides in soil is affected by different biochemical processes (16-18).

Samples collection and preparation

A total of 30 soil, wheat grains, palm dates and alfalfa plant samples from Qassim province, Saudi Arabia were collected for investigation. Each sample, about 1 kg in weight. The Weighted samples were ground, homogenized and sieved to about 100 mesh by a crushing machine. The samples were then placed for drying at 70°C to ensure that moisture is completely removed. Weighted samples were placed in polyethylene beakers, of 350 cm³ volume. The beakers were completely sealed for more than one month to allow radioactive equilibrium to be reached. So that leakage of ²²²Rn produced from ²²⁶Ra decay was negligible. This step was necessary to ensure that radon gas is confined within the volume and that the daughters will also remain in the sample (19-20). The samples were marked, catalogued and brought to radiation physics lab. After ensuring secular equilibrium of the progeny of ²²⁶Ra and ²³²Th series by storage for 4 weeks. The sealed samples were ready for analysis (21)..

Samples counting

The naturally occurring radionuclides considered in the present analysis of the measured γ -ray spectra are: ²¹²Pb (with a main gamma energy at ~239 keV and a gamma yield of ~ 43.1%), ²¹⁴Pb (~ 352 keV, ~ 37.1%), ²¹⁴Bi (~ 609, 1120 and 1765 keV,

~ 46.1, 15 and 15.9% respectively), ^{228}Ac (~911 keV, ~ 29%), ^{208}Tl (~2615 keV, ~ 35.9%) and ^{40}K (~1461 keV, ~10.7%). Under the assumption that secular equilibrium was reached between ^{232}Th and ^{238}U and their decay products, the concentration of ^{232}Th was determined from the average concentrations of ^{212}Pb , ^{208}Tl and ^{228}Ac in the samples, and that of ^{226}Ra was determined from the average concentrations of the ^{214}Pb and ^{214}Bi decay products (22-25). Gamma-spectrometric measurements were performed with NaI (Tl) detector. The measuring time for gamma-ray spectra ranged was 12 h. In order to determine the background distribution due to naturally occurring radionuclides in the environment around the detector, an empty polystyrene container was counted in the same manner as the samples. After measurement and subtraction of the background, the activity concentration was calculated (22-25).

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

$$\text{TF} = \frac{\text{Bq.kg}^{-1} \text{ dry crops}}{\text{Bq.kg}^{-1} \text{ dry soil}} \dots\dots\dots (1)$$

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 (26).

3. Results and Discussion

Radioactivity content in soil samples

Naturally occurring radioactive materials (NORM) are widespread in the Earth's crust, On the other hand, the release of artificial radionuclides into the environment, from the testing of nuclear weapons and nuclear facility accidents, can result in the contamination of soil. Table 1 gives the concentrations of ^{226}Ra , ^{232}Th and ^{40}K in soil samples from Qassim area. The mean and range of the concentrations of ^{226}Ra and ^{232}Th were 12.96 ± 3.4 (9.6–19.1) and 16.6 ± 7.1 (9.2– 28.3) Bq kg^{-1} . The range of the concentrations of ^{40}K in soil samples was (542–773) Bq kg^{-1} with a mean value of 618 ± 82 Bq kg^{-1} . The results also showed that the concentrations of ^{226}Ra , ^{232}Th and ^{40}K fell within the world average and the contents of ^{226}Ra and ^{232}Th belonged to the lowest concentration ranges (1, 27).

The radium equivalent (Raeq) is a radiation hazard index used for the evaluation of the radiation hazards of the gamma rays due to the NORM radionuclides (28). It is given by the relation

$$\text{Raeq} = C_{\text{Ra}} + 1.43C_{\text{Th}} + 0.077C_{\text{K}} \dots\dots\dots (2)$$

Where C_{Ra} , C_{Th} and C_{K} are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , respectively. The calculated Raeq is shown in Table 1. The mean value and range of the radium equivalent are far below the maximum of 370 Bq kg^{-1} .

The absorbed dose rate in outdoor air (n Gy h^{-1}) at a height of 1m above the ground surface was calculated using the formula

$$D = 0.472 C_{\text{U}} + 0.662 C_{\text{Th}} + 0.0432 C_{\text{K}} \dots\dots (3)$$

where C_{U} is the activity concentration of ^{238}U . The mean value and range of the absorbed dose rate were 44 (35.4-54.8) nGy h^{-1} , which are below the internationally recommended value of 59 (18–93 nGy h^{-1}) (1). The contents of NORM are found to be dependent on the plant species. Also, the radioactivity contents in soil samples varied over narrow ranges except in some cases. The values of radium equivalent and absorbed dose rate are below the international recommend values.

The activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in soil samples from the studied areas was compared with those from similar investigations in other countries and summary results were given in Table 3. The comparison shows that the values of soils under consideration are extremely low in accordance with others.

Table 1: The activities of ^{226}Ra , ^{232}Th and ^{40}K in soil samples from Qassim with radium equivalent activity and radiation dose.

Sample No.	^{226}Ra Bq Kg ⁻¹	^{232}Th Bq Kg ⁻¹	^{40}K Bq Kg ⁻¹	Ra_{eq} Bq Kg ⁻¹	D n Gy h ⁻¹
1	9.8 ± 0.3	9.2 ± 0.3	571 ± 11	67.0	35.38
2	14.1 ± 0.3	17.2 ± 0.4	611 ± 11	85.7	44.43
3	15.6 ± 0.2	19.6 ± 0.3	651 ± 10	93.7	48.46
4	14.8 ± 0.3	16.7 ± 0.4	542 ± 10	80.4	41.45
5	12.1 ± 0.3	11.7 ± 0.3	679 ± 12	81.0	42.78
6	9.6 ± 0.1	15.1 ± 0.2	584 ± 8	76.1	39.75
7	10.4 ± 0.2	13.4 ± 0.3	702 ± 12	83.7	44.10
8	12.8 ± 0.2	12.5 ± 0.3	773 ± 13	90.2	47.71
9	10.3 ± 0.1	13.7 ± 0.2	739 ± 11	86.8	45.85
10	15.4 ± 0.3	28.3 ± 0.5	666 ± 11	107.2	54.77
11	14.9 ± 0.3	18.9 ± 0.4	634 ± 11	90.8	46.93
12	11.5 ± 0.3	15.5 ± 0.4	624 ± 11	81.8	42.64
13	14.4 ± 0.3	14.2 ± 0.4	543 ± 10	76.4	39.85
14	19.1 ± 0.3	19.4 ± 0.4	573 ± 10	91.0	46.61
15	9.7 ± 0.2	16.5 ± 0.3	553 ± 9	75.8	39.39
Mean ± SD	12.96 ± 3.4	16.6 ± 7.1	618 ± 82	84.5	44
Range	9.6–19.1	9.2– 28.3	542–773	76.1–107.2	35.4–54.8

Distribution of radionuclides in Alfalfa and wheat and Palm dates.

The distribution of the radionuclides ^{226}Ra , ^{232}Th and ^{40}K for leaves samples from alfalfa plant as food for calves, milking cattle and sheep are presented in Table 2. It shows that Ra-226 activity concentrations values were ranged from 1.39 to 2.20 Bq/kg, the corresponding values of Th-232 were ranged from 1.27 to 2.15 Bq/kg. The ^{40}K values ranged from 43.6 to 71.8 Bq/kg.

A major part of the human diet all over the world consists of cereals and legumes. According to World Health Organization estimates, 70% of human food comprises cereals and legumes. Grain is a small dry, one seeded fruit of a cereal grass, having the

fruit and the seed walls united. Table 2 shows that that Ra-226 activity concentrations values in wheat grains were ranged from 1.00 to 1.55 Bq/kg, the corresponding values of Th-232 were ranged from 1.05 to 1.20 Bq/kg. The K-40 values ranged from 103 to 120.3 Bq/kg.

The data presented here for the natural radioactivity levels in palm dates are a start to establishing a database for the Qassim palm dates. Table 2 shows that that Ra-226 activity concentrations values in palm dates were ranged from 1.10 to 1.50 Bq/kg, the corresponding values of Th-232 were ranged from 1.15 to 1.30 Bq/kg. The K-40 values ranged from 122.9 to 130.9 Bq/kg.

Table 2: The activities of ^{226}Ra , ^{232}Th and ^{40}K in (Bq/kg) for Leaves of Alfalfa, wheat grains and dates palm samples from Qassim.

Sample No	Type	Ra-226	Th-232	K-40
1	Alfalfa	1.85	1.40	43.6
2	Alfalfa	2.00	1.27	57.4
3	Alfalfa	1.39	2.15	58.2
4	Alfalfa	2.20	2.00	55.7
5	Alfalfa	1.59	1.45	71.8
6	wheat grains	1.53	1.16	103.0
7	wheat grains	1.55	1.17	106.5
8	wheat grains	1.21	1.20	115.0
9	wheat grains	1.00	1.05	115.1
10	wheat grains	1.47	1.13	120.3
11	Palm dates	1.40	1.15	130.9
12	Palm dates	1.47	1.22	122.9
13	Palm dates	1.39	1.22	126.9
14	Palm dates	1.10	1.20	130.6
15	Palm dates	1.50	1.30	125.9

Table3: Comparison between natural radioactivity levels in our soil with other studies worldwide.

Country	Activity concentration (Bq/kg)			Reference
	²²⁶ Ra	²³² Th	⁴⁰ K	
Qassim, Saudi Arabia	12.9	16.6	618	Present work
Canada (Saskatchewan)	19	8	480	(29)
Upper Egypt	15.7	16.5	227.5	(30)
Spain	39	41	578	(31)
Brazil (Rio Grande do Norte)	29.2	47.8	704	(32)
Turkey (Istanbul)	21	37	342	(33)
Denmark	17	19	460	(1)
Syrian	20	20	270	(1)
South India	35	29.8	117.5	(34)
Cyprus	7.1	5	104.6	(35)
Nigeria	16.2	24.4	34.8	(36)
Bangladesh (Southern districts)	42	81	833	(37)
Pakistan (Lahore)	25.8	49.2	561.6	(38)
Vietnam (South- east)	19.6	31	34.6	(39)
Egypt (Farm soil)	13.7	12.3	1233	(40)
Nile island's soil	11.9	10.5	1636	

Transfer factors from soil to Alfalfa and wheat and Palm dates

Many studies have been carried out to determine TFs for most important agricultural products (26). Several projects were run by the International Atomic Energy Agency (IAEA) to determine TF mainly for ⁹⁰Sr and ¹³⁷Cs (41). These data have been used extensively in radiological assessment models. Natural environmental radioactivity arises mainly from primordial radionuclides, such as ⁴⁰K, and the radionuclides from the ²³²Th and ²³⁸U series, and their decay products are considered to be the main contributor to internal radiation dose. Several studies on transfer of natural radionuclides from soil to plant have been carried out in different regions in the world (25, 42). However, there seem to be few data on transfer of natural radionuclides from soil to plant in semiarid environments. Therefore, the present study aimed to determine TF for natural radionuclides in semiarid settings to some agricultural crops under natural field conditions.

Transfer factor depends upon many factors such as: electrical conductivity (EC), PH, and bicarbonate contents of soil, etc, Soil of all samples under study was geologically same and climatic conditions are also similar. The TF values for ²²⁶Ra and ⁴⁰K to various agricultural products are shown in Table 3. These values were calculated as $TF = P/S$, where P is the radionuclide concentration in the plant (Bq kg⁻¹ dry wt.) and S is the concentration in corresponding soil (Bq kg⁻¹ dry wt.) Differences between ²²⁶Ra TF values for various plant species are due to the different characteristics of the plants. ²²⁶Ra TF values from soil to Alfalfa were found to be higher than wheat grains and Palm dates. Potassium is an

essential macronutrient for plants maintained in homeostatic equilibrium and plant regulate the uptake of essential elements from soil. In general, the values of ⁴⁰K TF were lower than those values reported in other studies (42-45) and the default values suggested by IAEA (26). The ²²⁶Ra and ⁴⁰K TFs to various plants can be arranged as in Table 4.

Table 4: Transfer factors from soil to Leaves of Alfalfa, wheat grains and dates palm samples

Sample No	TF		
	Type	⁴⁰ K	²²⁶ Ra
1	Alfalfa	0.08	0.19
2	Alfalfa	0.09	0.14
3	Alfalfa	0.09	0.09
4	Alfalfa	0.10	0.15
5	Alfalfa	0.11	0.13
6	wheat grains	0.18	0.16
7	wheat grains	0.15	0.15
8	wheat grains	0.15	0.09
9	wheat grains	0.16	0.10
10	wheat grains	0.18	0.10
11	Palm dates	0.21	0.09
12	Palm dates	0.20	0.13
13	Palm dates	0.23	0.10
14	Palm dates	0.23	0.06
15	Palm dates	0.23	0.15
mean		0.16	0.12

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 (14).

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 (17). 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 (46).

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 (17).

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 (14). 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 (47).

Organic fertilizer affects the ion exchange capacity, pH, stable element content of soil, as well as soil retention properties (48-49).

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 (49). In some situations of plants nutrient deficiencies they are useful because they decrease soil retention, therefore increasing plant uptake (46, 50). Their effectiveness depends upon soil properties (particularly soil pH), chemical form of the radionuclide and the nature and concentration of chelating agent.

Conclusion

The level of naturally occurring radioactivity in soil samples collected Qassim area was evaluated using NaI(Tl) gamma-ray spectrometry. The mean and range of the concentrations of ^{226}Ra and ^{232}Th were 12.96 ± 3.4 (9.6–19.1) and 16.6 ± 7.1 (9.2– 28.3) Bq kg^{-1} . The range of the concentrations of ^{40}K in soil samples was (542–773) Bq kg^{-1} with a mean value of 618 ± 82 Bq kg^{-1} . The radium equivalent and absorbed dose rate were evaluated. The results obtained from this study indicated that our soil have background radioactivity levels within natural limits for ^{226}Ra , ^{232}Th and ^{40}K . The transfer factor for ^{226}Ra and ^{40}K to Alfalfa and wheat and Palm dates were measured. ^{226}Ra TF values from soil to Alfalfa were found to be higher than wheat grains and Palm dates. ^{40}K TF were lower than those values reported in other studies. 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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