RESEARCH ARTICLE

Assessment of radionuclide transfer factors and transfer coefficients near phosphate industrial areas of South Tunisia

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Abstract

The activity concentrations of naturally occurring and anthropogenic radionuclides in agriculture soils as well as in several food products at four locations within the phosphate area of South Tunisia were investigated. Soil-to-plant transfer factors as well as feed-to-animal products transfer coefficients were determined for the first time for the region. Activity concentrations of ^{40}K, ^{210}Pb, ^{226}Ra, ^{228}Ra and ^{137}Cs in soils of agriculture fields were lower than worldwide average values. The soil-to-plant transfer factors (TFs) for ^{40}K in leafy vegetables were higher than those in fruit vegetables. Soil-to-grass transfer factor (F_v) values were in the following order: ^{40}K > ^{210}Pb > ^{226}Ra. The grass-to-milk transfer coefficient (F_m) values for ^{40}K and ^{210}Pb ranged between 2×10^{-3} and 4×10^{-3} (day L^{-1}). The concentration ratios for the animal products (CR$_{milk-feed}$, CR$_{meat-feed}$ and CR$_{egg-feed}$) varied in the ranges of 2×10^{-2}–4×10^{-2} L kg^{-1}, 1×10^{-2}–2×10^{-1} (L kg^{-1}) and 5×10^{-2}–1 (L kg^{-1}) for ^{40}K, ^{210}Pb and ^{226}Ra, respectively. Transfer parameters determined in the present study were compared with those reported in International Atomic Energy Agency reports and other published values. The absorbed gamma dose rate in air and the external hazard index associated with these natural radionuclides were computed to assess the radiation hazard of radioactivity in this region, and the results indicated that these areas are within set safety limits.

Keywords Radionuclide activity concentrations · Soil-to-plant transfer factors · Feed-to-animal product transfer coefficients · Phosphate area · South Tunisia

Introduction

Over the past two decades, awareness of environmental issues has increased, prompted by evidence of the harm caused in the environment by industrially derived pollutants (UNSCEAR 2000). This subject is now under consideration by all relevant international organisations in the field of radiation protection, including the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), which aims at evaluating the sources and effects of radiation as the scientific

basis for estimating radiation risk, establishing radiation protection and safety standards and regulating radiation sources. Within the United Nations system, such radiological risk estimates are used by the International Atomic Energy Agency (IAEA) in discharging its statutory functions of establishing standards for radiation protection.

The environmental impact from the phosphate industries is mainly due to large volumes of residues produced during ore processing. Phosphate rocks are known to contain higher natural radioactive contents mainly originated from ^{238}U and ^{232}Th series (Tayibi et al. 2009). Typical activity concentrations of ^{238}U in phosphate rock range between 372 and 3200 Bq kg^{-1} (Bolca et al. 2007; Ekdal 2003), while the activity concentrations of ^{232}Th and ^{40}K are much lower and comparable to those normally observed in soil (Abbady et al. 2005). The worldwide average values for ^{238}U, ^{232}Th and ^{40}K concentrations in soil of normal-background regions are 35 Bq kg^{-1}, 30 Bq kg^{-1} and 420 Bq kg^{-1}, respectively (UNSCEAR 2000).

Radionuclides in soils are usually transferred to different plant tissues and may enter the human food chain through

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direct deposition on leaves or root uptake and translocation to parts of plants that are edible to humans and animals (Al-Hamarneh et al. 2016). The radionuclide transfer between compartments is commonly described by transfer parameters. These transfer parameters represent the ratio of concentrations of a radionuclide in two compartments for equilibrium conditions (IAEA 2010). The radiological impact of naturally occurring radioactive material (NORM) industries on the environment and the calculation of radiation dose to general population can be done from the knowledge of concentration of different radionuclides in different environmental matrices and the established transfer parameters. The evaluation of transfer factors for radionuclides in various food chains is essential to the knowledge of the long-term radiological hazard to the population of the region (IAEA 2010).

Tunisia is known as one of the large phosphate producers in the world with a production capacity of more than 10 million tons per year since the early 1990s (Khelifi et al. 2016). Phosphate mining was started in four regions of Gafsa City since 1899 (in Metlaoui, Redeyef, Moularas and Mdhilla), whereas phosphate treatment activity is existing in Sfax City since 1952 (Thyna and Skhira) and in Ghannouch, Gabes City, since 1972 (Khlifi et al. 2013). Additionally, this region of Tunisia is considered as a relatively polluted area due to the relatively high content of toxic metals in the environment (Gargouri et al. 2010; Houda et al. 2011; Serbaji et al. 2012). Despite these facts, no study has been carried out to assess the transfer factors of natural and anthropogenic radionuclide to the food chain around the phosphate industries. Therefore, generating new regional transfer factors would improve scientific knowledge of the mechanisms and factors that affect transfer of long-lived natural radionuclides into the environment. New regional transfer factor data will form a baseline for conducting accurate radiological assessments in similar environments for future assessments and provide useful information to IAEA and UNSCEAR databases. Hence, this study reports soil-to-vegetable and soil-to-grass transfer factors as well as transfer coefficient values for milk, beef and chicken meat and egg within these areas.

Materials and methods

Study area

The study was conducted around phosphate mines and phosphate processing plants in the south of Tunisia (Fig. 1). Sampling was done at locations situated 10–20 km from phosphate industries at both Sfax and Moularas cities, whereas at the coastal region surrounding the chemical complex at Gabes City, the samples were collected at the 0–5km region. Additionally, samples were collected at Gafsa City, which

was considered as a control area since this region has no mining and processing activities, for a comparative study.

Sampling and sample preparation

Soil samples

Three representative samples from the 0–20cm soil layer were collected from agriculture lands within each study area based on the root zones of target samples (IAEA 2010), following procedures of EMBRAPA and IAEA (Claessen 1997; IAEA 2010). The three samples collected from each site were mixed, and a composite sample, which is the representative sample of the field, was taken for analysis. To determine soil-to-grass transfer factors, soil samples were collected from grass fields (in which cows graze normally), in the same way as explained above.

Soil samples were then removed from any extraneous materials and stones and weighed. The samples were dried at 105 °C, and the dry weight is noted. It was then filled in a polypropylene container, sealed and stored for a minimum of 30 days to allow the radioactive equilibrium between ^{226}Ra and its daughters, prior to gamma spectrometric analysis.

Grass samples

The grass samples were collected from three different grazing areas. Grass from a 1m^2 area from pasture was cut just above the soil surface. Several such samples were collected within a large grazing area, and they were mixed to obtain a composite sample. They were then dried at 105 °C and weighed. The samples were homogenised, charred under low flame in a silica crucible and then subjected for dry ashing at 450 °C until a uniform white ash was obtained.

Food material samples

Samples of vegetables and animal origin products (milk, fish, beef meat, chicken meat and egg) were processed following the procedure described by Machraoui et al. (2018).

Vegetables were categorised based on the part that serves as food: leafy vegetables (i.e. parsley, spinach, lettuce), root vegetables (i.e. carrot, onion, radish) and fruit vegetables (i.e. tomatoes and pepper). This was consistent with the categorisation reported in UNSCEAR (2000) and IAEA (2010) reports.

Samples of cow milk were collected from dairy farms. The diet of dairy farm cows comprised of two parts: (i) grass/forage and (ii) nutrition-rich supplementary feed. To estimate the transfer of radionuclides to cow milk and beef meat, the grass was considered as the main source of radionuclide intake and intake through supplementary feed is not considered in this study as it is produced by local industries, which source

Fig. 1 Map showing the sites of phosphate industries in South Tunisia
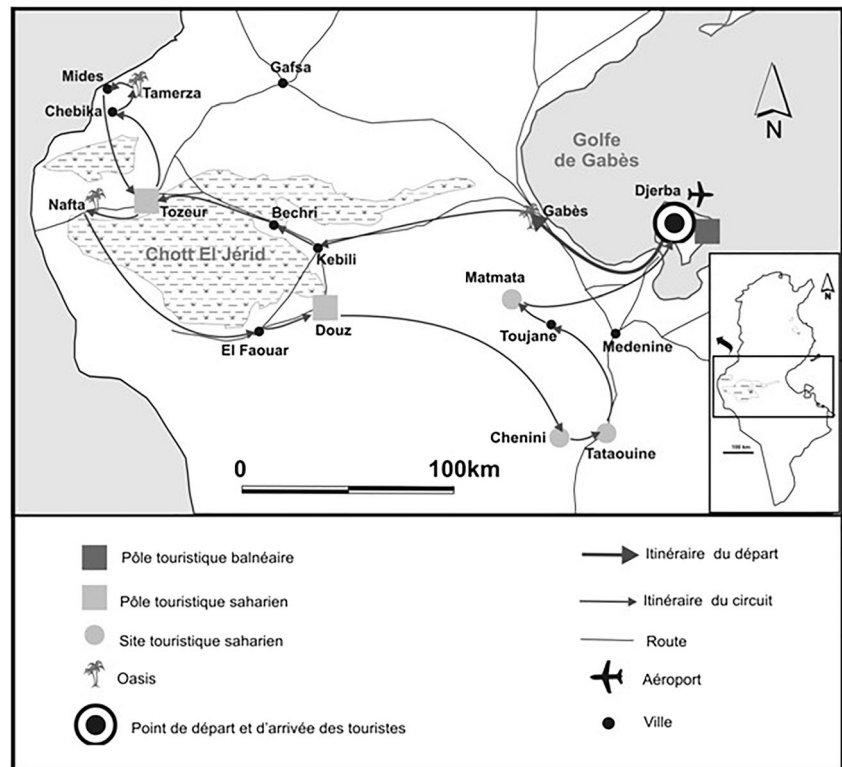
the raw material from northern parts of Tunisia where fodder and legume are produced. Samples of milk were collected from dairy farms. The sampling was done during December 2014 to July 2017.

Gamma spectrometry measurements

Gamma spectrometric analyses were performed at the Centre for Advanced Research in Environmental Radioactivity (CARER), Mangalore University, India, following the same procedure described by Machraoui et al. (2018).

Analysis of the data

Descriptive statistical analysis was performed on the data of the radionuclide activity concentration in the different matrices. The Shapiro-Wilk normality test was performed on the radionuclide activity concentration and transfer factor values for ^{40}K, which rejected the null hypothesis ($p = 1.5 \times 10^{-6}$ and $p = 8 \times 10^{-4}$, respectively) that the datasets come from a normal distribution with 95% confidence level and also indicates the non-existence of normal distribution. This was also confirmed by the skewness (1.38) and kurtosis (1.44) values. The log-normal fit for this suggests that the datasets are log-normally distributed with the geometric standard deviation (σ_g) value of 2.5 with the correlation coefficient (R) > 0.95. The datasets for ^{226}Ra and ^{210}Pb also exhibited the similar

trend of distribution ($p = 6 \times 10^{-8}$ and $p = 6.5 \times 10^{-10}$, respectively, for activity concentrations and $p = 2.3 \times 10^{-6}$ and $p = 4 \times 10^{-5}$, respectively, for transfer factor values). For log-normal distribution, geometric mean (GM) and geometric standard deviation (GSD) are better representative of the central value and associated deviation than the arithmetic mean.

Assuming normal distribution, an independent two-sample t test was performed to examine the mean difference of activity concentrations in soil samples between a potential site and the control site, using SPSS 22.0 (IBM Corp., Armonk, NY, USA).

One-sample t test was performed to determine the difference between the mean concentrations obtained from this study and the worldwide average values reported by UNSCEAR (2000) and IAEA (2010).

Determination of transfer factors and transfer coefficients

Soil-to-plant transfer factor

Transfer factors (TFs) (Eq. (1)) can be used as an index of the accumulation of either the trace elements by plants or of the soil-to-plant transfer of elements (Chen et al. 2005). Using the results of the activity concentration of a radionuclide in the soil and the plant, the transfer factor was calculated as follows:

$$TF = \frac{A_{plant}}{A_{soil}} \tag{1}$$

where A_{plant} is the activity concentration (Bq kg^{-1}, with respect to dry weight (dw)) of a particular radionuclide in plant and A_{soil} is the activity concentration (Bq kg^{-1}, dw) of that radionuclide in soil (IAEA 2010; IUR 1994; Madruga et al. 2001; Karunakara et al. 2013a).

The soil-to-grass transfer factor (F_v) for each radionuclide was expressed as:

$$F_v = \frac{\text{Activity concentration in grass } (Bq\,kg^{-1}\,dw)}{\text{Activity concentration in soil from pasture land} (Bq\,kg^{-1}\,dw)} \tag{2}$$

Transfer coefficient for milk (F_m)

The grass-to-milk transfer coefficient (F_m) values were estimated from the measured activity concentrations of different radionuclides as follows (IAEA 2010; Karunakara et al. 2013b):

$$F_m = \frac{A_m}{A_g \times Q_m} \tag{3}$$

where F_m is the transfer coefficient (day L^{-1}), A_m is the radionuclide activity concentration in milk (Bq L^{-1}, with respect to fresh weight (fw)), A_g is the radionuclide activity concentration in grass (Bq kg^{-1}, dw) and Q_m is the daily intake of grass by the cows (kg day^{-1}, dw).

Data on the daily intake of grass was obtained from several local farmers as well as from the local dairy farms from where milk samples were collected, and a mean value of 10 kg (dw) for the daily intake of dry weight was considered as the representative value for the F_m estimation.

Concentration ratio for animal product

In the present study, the concentration ratio (CR) which is the equilibrium ratio of the radionuclide activity concentration in animal product (fw) to that in feed (dw) is calculated as:

$$CR_{milk-feed} = \frac{\text{Activity concentration in milk}}{\text{Activity concentration in grass}} \tag{4}$$

$$CR_{beef-feed} = \frac{\text{Activity concentration in beef meat}}{\text{Activity concentration in grass}} \tag{5}$$

$$CR_{chicken-feed}$$
$$= \frac{\text{Activity concentration in chicken meat}}{\text{Activity concentration in grain for hen meat feed}} \tag{6}$$

$$CR_{egg-feed} = \frac{\text{Activity concentration in egg}}{\text{Activity concentration in grain for laying hen feed}} \tag{7}$$

Determination of the outdoor annual effective dose

The absorbed gamma doses, in nanograys per hour, were converted to annual effective dose in microsieverts per year, as proposed by UNSCEAR (2000). The annual effective dose rate (AEDR) was calculated using the following equation:

$$AEDR = D \left(nGy\ h^{-1}\right) \times 8760 \times 0.2 \times 0.7$$
$$\times 10^{-3} \left(\mu Sv\ year^{-1}\right) \tag{8}$$

where D is the absorbed dose rate in air (nGy h^{-1}), 0.7 is the dose conversion factor (S Gy^{-1}), 0.2 is the outdoor occupancy factor and 8760 is the time conversion factor (h year^{-1}).

Results and discussion

Radionuclide concentrations in soil

The activity concentrations (Bq kg^{-1}) of ^{40}K, ^{210}Pb, ^{226}Ra and, ^{228}Ra r ^{137}Cs were determined in the soil samples from the Tunisian phosphate area, and the results are given in Table 1.

The ^{228}Ra activity of soils presented here can be considered as that of ^{232}Th activity since one can assume that the secular equilibrium exists between these radionuclides in soil. A comparison of activity concentrations observed in the present study, with the worldwide average values of 35 Bq kg^{-1}, 30 Bq kg^{-1} and 400 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K (UNSCEAR 2000), respectively, suggests that soils studied in the present study had lower radionuclide activity concentrations ($p = 0.04$ for both ^{40}K and ^{226}Ra and $p = 1 \times 10^{-6}$ for ^{232}Th).

Figure 2 shows a comparison of the soil radioactivity in the control site with those collected around the phosphate industries. The results of statistical analyses indicated that the mean concentration of ^{40}K in soil samples collected from Sfax and Moularas cities was significantly higher ($p = 7 \times 10^{-10}$ and $p = 8 \times 10^{-9}$, respectively) than that in samples collected from the control site, while those from Gabes City were comparable ($p = 0.1$). Similar results were observed for ^{210}Pb ($p = 0.002$, $p = 2 \times 10^{-8}$ and $p = 0.06$, respectively, for soil samples from Sfax, Moularas and Gabes cities), whereas all the ^{226}Ra activity concentrations in soil samples collected from Sfax, Moularas and Gabes cities were significantly higher ($p = 4 \times 10^{-6}$, $p = 1 \times 10^{-5}$ and $p = 3 \times 10^{-4}$, respectively) than those observed for the control site. The mean ^{137}Cs concentration was significantly higher in soil samples collected from Moularas City ($p = 3 \times 10^{-7}$) when compared with those from the control site, whereas soils from Sfax and Gabes

Table 1 Geometric mean (GM) and ranges of radionuclide activity concentrations, dose rates (D) and hazard indexes (H_{ex}) for the soil samples collected near the phosphate industries (control site not included)

Sample (n = sample size)	Activity concentration (Bq kg^{-1}) with respect to dry weight					H_{ex} (nGy h^{-1})	D (nGy h^{-1})
	^{40}K	^{210}Pb	^{226}Ra	^{228}Ra (^{232}Th)	^{137}Cs		
Soils used for agriculture crops (n = 12)	250 (123–374)	41 (23–102)	26 (17–32)	13 (6–22)	3 (1–8)	2×10^{-1} (9×10^{-2}–3×10^{-1})	23 (13–32)
Soils from pasture land (n = 6)	230 (185–278)	27 (17–44)	22 (12–39)	4 (2–11)	4 (4–5)	1×10^{-1} (1×10^{-1}–9×10^{-1})	21 (17–26)
UNSCEAR (2000)	400 (140–850)	–	35 (17–60)	30 (11–64)	51	–	51 (32-107)

Values in italic are the activity concentration ranges

Values in parenthesis are the activity concentration range values

cities exhibited similar ^{137}Cs concentrations ($p = 0.1$ and $p = 0.5$, respectively).

The difference in the activity values of the naturally occurring ^{40}K radionuclide might be due to the use of fertilisers, which may lead to changes in their activity values. However, it must be noted that for a detailed impact assessment study, more samples, closer to the phosphate industries, need to be analysed.

External hazard index (H_{ex})

The external hazard index (H_{ex}) due to the emitted gamma rays for each sample was calculated according to the following formula (UNSCEAR 1988):

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \left(nGy\,h^{-1}\right) \qquad (9)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively (Beretka and Mathew

1985). The values of the indices should be < 1. Observed H_{ex} values in Table 1 are below 1 (set criterion value), indicating that the values obtained in the study are within the recommended safety limit.
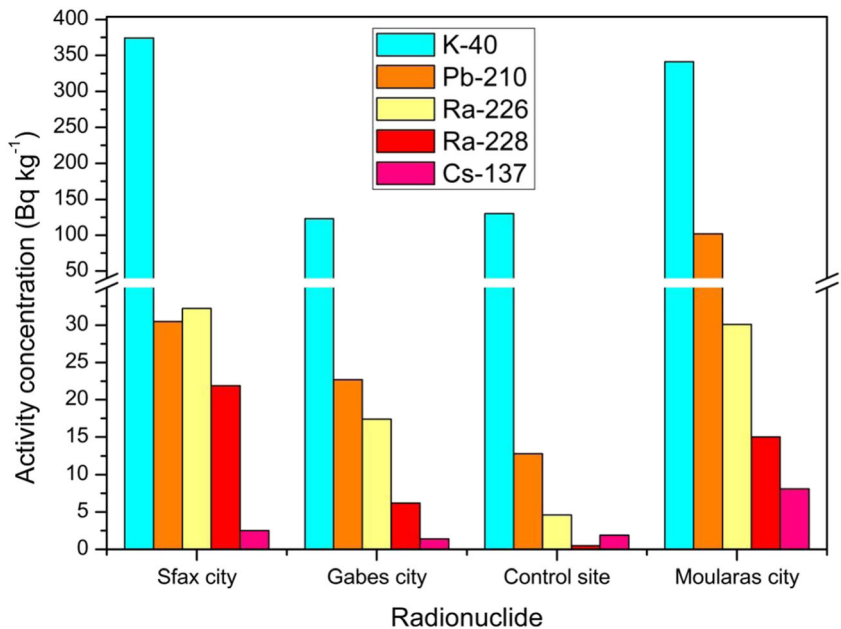
Absorbed gamma dose rates in air (D)

The absorbed dose rates in outdoor (D) due to gamma radiations in air at 1 m above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were calculated based on guidelines provided by UNSCEAR (2000). The conversion factors used to compute absorbed gamma dose rate (D) in air per unit activity concentration (Bq kg^{-1}, dw) were 0.427 nGy h^{-1} for ^{226}Ra, 0.662 nGy h^{-1} for ^{232}Th and 0.043 nGy h^{-1} for ^{40}K.

$$D = 0.427\,A_{Ra} + 0.662\,A_{Th} + 0.043\,A_K \left(nGy\,h^{-1}\right) \qquad (10)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra,

Fig. 2 Comparison of the activity concentrations of different radionuclides in soil samples collected from the control site and near the phosphate industries

^{232}Th and ^{40}K in Bq kg^{-1}, respectively. Table 1 also gives the results for the absorbed dose rate in air. According to UNSCEAR (2000), the mean value of outdoor gamma dose rate in air from terrestrial gamma rays for normal background radiation region is 51 nGy h^{-1} for a median value. The mean value of 21 nGy h^{-1} observed in the study area is thus lower than the worldwide average value. This shows that radiation in the study area was within the natural limits and did not significantly differ from that in other regions of the world.

Radionuclide concentration in vegetables and other food matrices

Table 2 shows the activity concentrations in vegetables, milk, meat, fish, egg, feed for chicken and grass samples on a dry weight basis.

The geometric mean values for different types of vegetables are plotted in Fig. 3 for a comparison. It is evident from the figure that the activity concentration of ^{40}K, ^{210}Pb and ^{226}Ra in leafy vegetables is higher when compared to that in fruit and root vegetables. While higher concentrations of ^{40}K and ^{226}Ra in parts of the plant above ground indicate a preferential translocation of these, the higher concentration of ^{210}Pb in above-ground organs may be because of the atmospheric deposition of this radionuclide on the plants. Similar findings were reported for Syria (Al-Masri et al. 2008) and for France (Jeambrun et al. 2012a). The reported ranges for ^{210}Pb in soil samples were 15–43 Bq kg^{-1} and 22–112 Bq kg^{-1} for Syria and France, respectively. The GM of ^{210}Pb (41 Bq kg^{-1}) in soil samples, observed in this study, is within the reported ranges. However, ^{210}Pb transfer to plant is not only via the roots of the plant but also via atmospheric deposition, and the transfer via the root system is considered rather small (Bunzl and Trautmannsheimer 1999). In a previous study on rice plants grown in the West Coast of India, Karunakara et al. (2013a) reported that the ^{40}K was higher in above-ground parts of the plants, whereas the ^{210}Pb was higher in roots. Their study have also indicated that significant difference in the activity concentrations of different radionuclides in different parts of the plant can occur within the same species of plant because of the difference in soil properties and variations in the type and extent of fertilisers used. The soils used for growing rice plant had organic matter content in the range of 9–20%, a very low clay content (varying in the range of 0–7%), high silt content (varying in the range of 46–76%) and low K content (Karunakara et al. 2013a). A previous study conducted in Sfax City reported that the soils over 2 km from the phosphate fertiliser plant were alkaline with low levels of organic matter and lime (Béjaoui et al. 2016). The soils are generally sandy or sandy-limestone in the study area (Choura 2007).

The activity concentrations of ^{228}Ra and ^{137}Cs in most of the samples analysed were below detection limits, only six vegetable samples showed a measurable activity concentration of ^{228}Ra and only one sample (leek) showed a measurable ^{137}Cs concentration. Hence, data on ^{137}Cs activity concentrations are not included in Fig. 3.

Table 2 also presents the activity concentrations of ^{40}K, ^{210}Pb and ^{226}Ra in milk, meat, fish, egg and animal feed such as grass, and chicken feed. For all types of foods, relatively large variability in radionuclide concentrations was observed, even within the same kind of food. ^{228}Ra could be detected in only two samples (fish and egg). The fish samples collected from the study area exhibited higher ^{210}Pb concentrations (p = 0.03) as that of observed in the UNSCEAR (2000) report, while those for ^{226}Ra (p = 0.06) were similar. It is well known that ^{210}Po and ^{210}Pb are absorbed by plankton in aquatic environments. The activities of these two elements may be increased by industrial waste and human activities (Manav et al. 2016). Štrok and Smodis (2011) measured ^{210}Pb activities in fish samples as 3 Bq kg^{-1}. The researchers stated that the higher ^{210}Pb activity concentrations for marine species in sea bass are due to the terrestrial feeding habit in mariculture.

To assess the impact of phosphate industries, the activity concentrations of radionuclides in different types of vegetables collected around different phosphate industries are compared along with those observed for the control site. The results of statistical analyses indicated that only the overall geometric mean concentration of ^{40}K (2402 Bq kg^{-1}) in vegetables collected from Moularas City was significantly different (p = 0.03) from that from the control site, while the ^{210}Pb and ^{226}Ra mean concentrations in vegetables collected from the phosphate industrial sites were comparable (p = 0.1 and p = 0.08, respectively) with those from the control site.

Soil-to-plant transfer factors

Soil-to-vegetable transfer factors

From the activity concentrations given in Table 2, the soil-to-plant transfer factors of different radionuclides were computed for leafy, root and fruit vegetable types. The corresponding geometric mean of transfer factor values is compared to each other in Fig. 4. The figure shows that the transfer factor values for ^{210}Pb and ^{226}Ra were in the following order: fruit vegetables > leafy vegetables > root vegetables. It is also noted that ^{40}K transfer factors were orders of magnitude higher when compared to the other radionuclides, and it was more than unity in all analysed vegetable samples. This may be because of the application of potassium-rich fertilisers to the soil during the process of cultivation, which will be taken

Table 2 Geometric mean values and ranges of the different radionuclide concentrations in food materials from the Tunisian phosphate area

Product category (sample size)	Site	Activity concentrations Bq kg^{-1} with respect to dry weight				
		^{40}K	^{210}Pb	^{226}Ra	^{228}Ra	^{137}Cs
Leafy vegetables (9)	Sfax	2212 (1965–2400)	6 (3–13)	2 (1–2)	1 ± 1	< 0.3
	Moularas	3753 (2425–5643)	64 (36–121)	9 (7–15)	< 1	< 0.3
	Gabes	424 (214–841)	11 (6–21)	2 (1–3)	1 (< 1–1)	< 0.3
	Gafsa (control)	1569 (1430–1722)	3 ± 0.2	1 ± 1	< 1	< 0.3
Root vegetables (10)	Moularas	1678 (1281–2149)	7 (5–11)	2 (2–3)	< 1	< 0.3
	Gabes	533 (317–813)	9 (5–15)	2 (1–10)	2 (1–3)	0.5 ± 0.2
	Gafsa (control)	529 (479–627)	< 2	1 (< 0.5–1)	1 ± 0.3	< 0.3
Fruit vegetables (7)	Sfax City	1182 ± 87	< 2	3 ± 0.5	< 1	< 0.3
	Moularas City	2104 (1259–3517)	< 2	< 0.5	< 1	< 0.3
	Gabes City	531 (186–1514)	9 ± 4	1 (1–2)	< 1	< 0.3
	Gafsa (control)	1381 (814–2344)	17 (9-69)	4 (3–5)	3 ± 1	< 0.3
Grass (4)	Sfax	1300 (855–1973)	63 (57–70)	2 (2–3)	< 1	< 0.5
	Moularas	208 ± 6	55 ± 2	8 ± 0.3	2 ± 0.4	< 0.5
	Gafsa (control)	1184 ± 30	7 ± 2	< 0.5	< 1	< 0.5
Milk* (5)	Sfax	58 (57–60)	1 ± 0.2 (< 2–1)	< 0.05	< 1	< 0.03
	Moularas	39 (30–48)	< 0.05	< 0.05	< 1	< 0.03
	Gafsa (control)	11 ± 0.3	0.2 ± 0.04	0.1 ± 0.01	< 1	< 0.03
Beef meat (3)	Sfax	797(362-1757)	< 2	< 0.5	< 1	< 0.3
	Gafsa (control)	301 ± 6	< 2	11 ± 2	< 1	< 0.3
Chicken meat (6)	Sfax	380 (282–601)	< 2	1 (1.1–1.2)	< 1	< 0.3
	Moularas	365 (321–449)	3 (< 2–3)	1 (< 0.5–1)	< 1	0.3 (< 0.3–0.3)
Egg (6)	Sfax	212 (176–281)	30 (< 2.0–30)	1 (0.8–1.5)	< 1	< 0.3
	Moularas	273 ± 8	< 2	1 ± 0.2	< 1	< 0.3
	Gabes	124 ± 3	5 ± 1	2 ± 0.1	1 ± 0.1	< 0.3
	Gafsa (control)	156 ± 5	< 2	1 ± 0.1	< 1	< 0.3
Fish (4)	Sfax	392 (219–703)	26.5 (3.5–30)	5 (1–24)	22 (< 1–22)	< 0.3
	Gabes	112 (26–485)	16	6 (4–10)	7 (2–25)	< 0.3
Laying hen feed (1)		35 ± 1	4 ± 1	5 ± 0.06	< 1	< 0.3
Meat hen feed (1)		236 ± 11	10 ± 0.5	11 ± 0.2	< 1	< 0.3

Values with the corresponding counting error are a single activity value

Values in parenthesis are the activity concentration range values

*For milk, the activity concentrations are reported with respect to the becquerels per litre

preferentially by the plant since it is an essential element for metabolism. ^{226}Ra has much lower transfer factor values, probably due to its high affinity with soil particles, resulting in reduced uptake of ^{226}Ra by plants (Vandenhove et al. 2009). Also, chemical properties such as the amount of exchangeable calcium in the soil will determine the rate at which radium will be absorbed by plants (James et al. 2011). The transfer factor values for ^{228}Ra and ^{137}Cs were not considered in this comparison since majority of the samples showed activity concentrations of these radionuclides below detection limit.

Vandenhove et al. (2009) have compiled a large number of soil-to-plant transfer factor data for natural radionuclides and have carried out critical reviews of the compiled data for their quality and usefulness. In their studies, the soil-to-plant transfer factors for the natural radionuclides U, Th, Ra, Pb and Po were reviewed and grouped according to crop group, soil group and organic matter content. The transfer factor values were generally about 10-fold lower for Th and Po than for U, Ra and Pb. The highest transfer factor values were obtained for fodder, pastures, grasses and leafy vegetables while the lowest transfer factor estimates were obtained for legumes and cereals. As evident from the results presented in Fig. 4, there is a large variability among the transfer factor data

Fig. 3 Comparison of the geometric mean values of radionuclide activity concentrations in the different types of vegetables
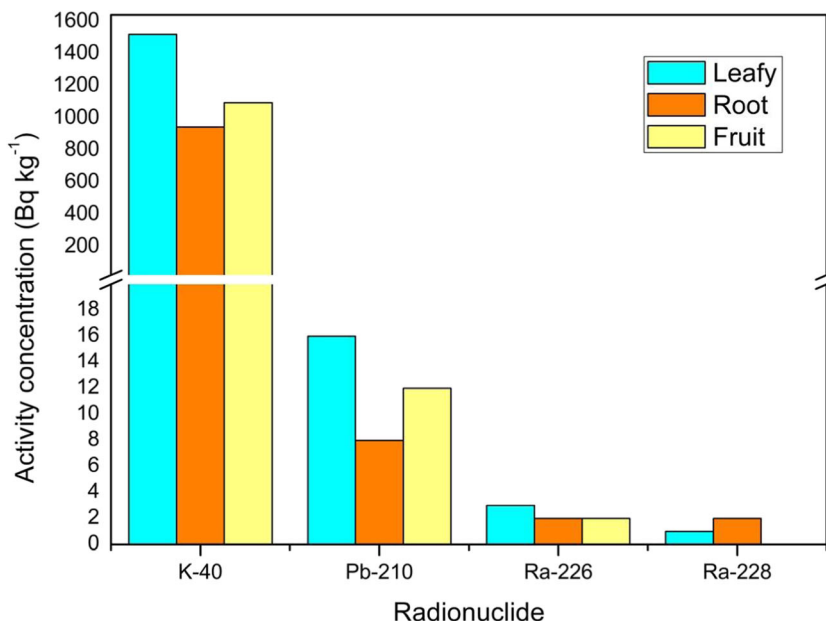
even at the crop group level. Al-Masri et al. (2008) have reported that differences between transfer factor values for various plant species are due to the different characteristics of the plants. Similar observations were reported by Djelic et al. (2016), Karunakara et al. (2013a) and Dreyer and Uozumi (2011).

The geometric mean values of transfer factors observed in the present study for different types of vegetables are compared with those reported by IAEA (2010). The geometric mean value for ^{40}K is significantly higher ($p = 0.04$), while that for ^{210}Pb and Ra isotopes is similar to most of the other reported values.

Soil-to-grass (F_v) transfer factors

Table 3 presents soil-to-grass F_v values observed in the present study which were in the following order: ^{40}K > ^{210}Pb > ^{226}Ra ∼ ^{228}Ra. The F_v values are also compared with those from the IAEA (2010) report for other countries. The IAEA-TRS-472 has compiled extensive data on F_v values published by different authors for different countries, and the range and mean of these data are also presented in Table 3. The F_v values summarised in the IAEA (2010) report has a wide range. The GM value of F_v for ^{210}Pb observed in the present study was higher ($p = 1 \times 10^{-4}$) when compared to that given in the

Fig. 4 Comparison of the geometric mean of the radionuclide transfer factors for the different types of vegetables
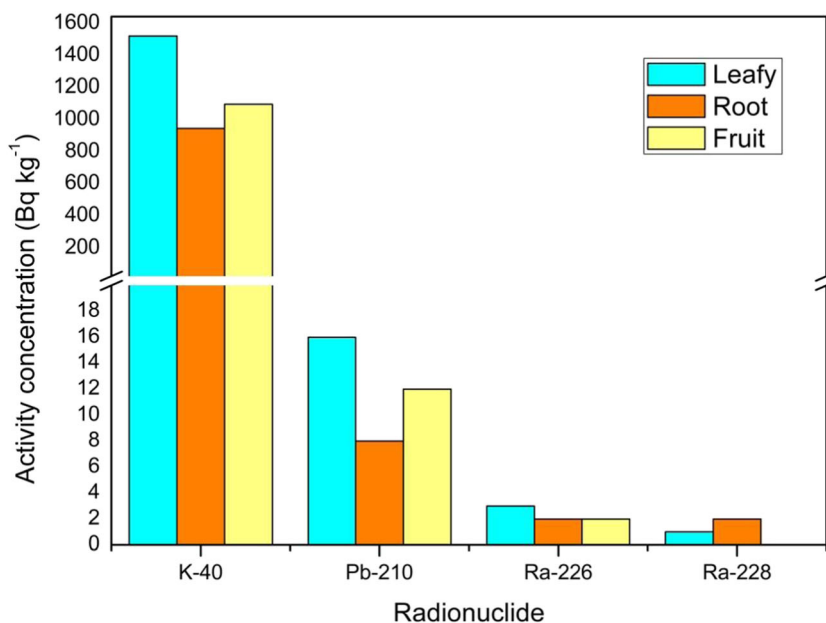
Table 3 Overall geometric mean (GM) values and ranges of soil-to-grass transfer factor (F_v) and comparison with the reported values (values in parenthesis are F_v ranges)

Product category (n = sample size)	Soil-to-grass transfer factor values (F_v)			
	^{40}K	^{210}Pb	^{226}Ra	^{228}Ra
Grass (n = 6)				
GM	3 (1–5)	8×10^{-1} (1×10^{-1}–4)	2×10^{-1}	2×10^{-1}
IAEA (2010)				
GM	–	3×10^{-1} (1×10^{-1}–1)	1×10^{-1} (4×10^{-3}–2)	
Chakraborty et al. (2013)				
GM	3×10^{-1} (2×10^{-1}–3×10^{-1})	–	5×10^{-2} (4×10^{-2}–8×10^{-2})	–
Jazzar and Thabayneh (2014)				
GM	1.2 (0.4–3)	–	1.3 (0.3–2)	–

IAEA (2010) report, while that for ^{226}Ra was similar (p = 0.08).

Transfer coefficients (F_m) and CRs for animal products

Beresford (2003) and Howard et al. (2001) have expressed that whether the transfer coefficient concept is more robust/generic or not, due to their wide variability, this is because of the fact that F_m values incorporate dry matter intake, which increases with animal size. An alternative method for quantifying the transfer of an element from feed to the animal product is the CR which is the equilibrium ratio of the radionuclide activity concentration in milk (fresh weight) to that in feed (dry weight).

F_m values can be derived by dividing the CR value by the daily dietary intake (kg day^{-1}), and CR values can be derived by multiplying the F_m value by the daily dietary intake. The CR has the advantage in the field studies that daily intake does not need to be calculated (IAEA 2010).

The results of transfer coefficient and concentration ratio values for ^{40}K, ^{210}Pb and ^{226}Ra are displayed in Table 4. The transfer coefficient values for milk (F_m) are by an order of magnitude less than the CR$_{milk-feed}$ values. The GM value of CR$_{beef -feed}$ for ^{40}K was also by an order of magnitude less than that observed for chicken meat. It was reported that the transfer coefficient values for meat varied over 3 orders of magnitude from 10 day kg^{-1} (chicken) to 8×10^{-3} day kg^{-1} (beef), and the range in estimated CR values was only two fold

Table 4 Overall geometric mean (GM) values and ranges of grass-to-milk transfer coefficients F_m (day L^{-1}) and feed-to-animal product concentration ratios (values in parenthesis are ranges)

Radionuclide	Grass-to-milk transfer coefficients (F_m)	Concentration ratios			
		CR$_{milk-feed}$ (L kg^{-1})	CR$_{beef\ meat-feed}$	CR$_{chicken\ meat-feed}$	CR$_{egg-feed}$
^{40}K					
GM	4×10^{-3} (9×10^{-4}–2 $\times 10^{-2}$)	4×10^{-2} (9×10^{-3}–2 $\times 10^{-1}$)	6×10^{-2} (3.5×10^{-2}–1 $\times 10^{-1}$)	2×10^{-1} (1.6–1.8 $\times 10^{-1}$)	1 (2–7×10^{-1})
MODARIA	3×10^{-3}–6×10^{-3}	–	–	–	–
^{210}Pb					
GM	2×10^{-3} (2×10^{-3}–3 $\times 10^{-3}$)	2×10^{-2} (2×10^{-2}–3 $\times 10^{-2}$)	–	–	6×10^{-1} (2.5×10^{-1}–1.5)
IAEA (2010)	2×10^{-4} (7×10^{-6}–1 $\times 10^{-3}$)	2×10^{-3} (10×10^{-4}–4 $\times 10^{-3}$)	8×10^{-2} (2×10^{-1}–1 $\times 10^{-3}$)	–	–
MODARIA	2×10^{-4} (1×10^{-5}–2 $\times 10^{-3}$)	5×10^{-3} (4×10^{-4}–4 $\times 10^{-2}$)	–	–	–
^{226}Ra					
GM	–	–	–	1×10^{-2} (9×10^{-3}–1 $\times 10^{-2}$)	5×10^{-2} (4×10^{-2}–8 $\times 10^{-2}$)
Jeambrun et al. (2012b)	–	–	–	–	7×10^{-2} (4.2×10^{-2}–1.3 $\times 10^{-1}$)

MODARIA Modelling and Data for Radiological Impact Assessments

(IAEA 2009). The CR values for egg, obtained in the present study, are also presented in Table 4.

The transfer parameters evaluated in the present study for animal products are compared with the literature values in the same table. The GM values of F_{m} and $\mathrm{CR_{milk\text{-}feed}}$ obtained in the present study for $^{210}\mathrm{Pb}$ were higher by an order of magnitude when compared to those reported in the IAEA (2010) document and Modelling and Data for Radiological Impact Assessments (MODARIA) 2016 cow-milk dataset (Howard et al. 2017) (Table 4). As mentioned earlier, in the present study, milk samples were collected from local dairy where milk samples from many cows are pooled and a representative sample is taken. Thus, CR values obtained in the present study do not give a cow-specific F_{m}; rather, it is a representative value. In contrast, studies included in the MODARIA dataset report an experimental work where cows were either adopted or identified for the purpose of grazing and the feeding pattern of cows was known accurately. Moreover, several reports on the determination of radionuclide transfer coefficients for food matrices are available for countries all over the world. Most of these values are compiled in the IAEA (2010) report, and an average weighted value is given.

Data on transfer coefficients for chicken meat and egg are reported for different radionuclides in the technical document No. 1616 of IAEA, but not on $\mathrm{CR_{chicken\text{-}feed}}$ and $\mathrm{CR_{egg\text{-}feed}}$. However, in a study carried in France (Jeambrun et al. 2012b), the $\mathrm{CR_{egg\text{-}feed}}$ mean value of 7×10^{-2} for $^{226}\mathrm{Ra}$ has been reported and this is similar when compared to the value obtained in the present study (Table 4). The study reported activity concentrations in chicken meat and eggs from five different regions of the French territory. One of these sites is located in the area influenced by the releases of the nuclear fuel fabrication facility to estimate the potential influence of the discharges.

Outdoor annual effective dose

The external dose due to the soil radioactivity in the study area was calculated (Eq. (8)), and the mean value was found to be 0.03 mSv year^{-1}. This is lower than the worldwide average of 0.07 mSv year^{-1} (UNSCEAR 2000). Radiation dose assessments from the ingestion of radionuclides through food samples collected form the same locations, studies in this work, were reported earlier (Machraoui et al. 2018). The doses estimated for the adult age group (>17 years) was computed to be 0.7 mSv year^{-1}, which suggests that the ingestion dose is an order of magnitude higher when compared to the external gamma dose due to the soil radioactivity.

Conclusion

This is the first systematic study aimed at evaluating radionuclide transfer parameters in a region of phosphate industries in Tunisia.

The activity concentrations of $^{40}\mathrm{K}$, $^{210}\mathrm{Pb}$, $^{226}\mathrm{Ra}$ and $^{228}\mathrm{Ra}$ in agriculture soils of phosphate industrial regions of Tunisia are lower when compared to the worldwide average values (UNSCEAR 2000). However, within the study region, a comparison of activity concentrations of radionuclides observed in agriculture soils of the control site with those collected from the 10–20km zone of the phosphate industries indicates enhanced radioactivity levels of $^{40}\mathrm{K}$, $^{210}\mathrm{Pb}$ and $^{226}\mathrm{Ra}$ in Sfax and Moularas cities. The H_{ex} and D values obtained in the study are within the recommended safety limit. This shows that radiation in the study area was within the natural limits and did not significantly differ from other regions of the world. The fish samples collected from Gabes and Sfax cities, where large quantities of phosphogypsum are released daily into the open sea without any treatment, exhibited higher $^{210}\mathrm{Pb}$ activity concentrations when compared to those reported by the UNSCEAR report. The transfer parameter values for $^{210}\mathrm{Pb}$ for food animal products were higher by an order of magnitude when compared to the weighted average values reported in IAEA (2010). A detailed radioecological impact assessment with a large number of samples within the 0–10km zone of phosphate industries is needed. These studies are now in progress.

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