



# Transfer from soil to grass and statistical analysis of naturally occurring radionuclides in soil from phosphate mining and processing sites in Maritime Region of Togo

Eyakifama Hazou<sup>1,2</sup> · Benjamin Zorko<sup>4</sup> · Milohum Mikesokpo Dzagli<sup>1,2</sup> · Essolakina Manakrihèa Haliba<sup>1,2</sup> · Shouph Cebastien Joel Guembou<sup>5,6,7</sup> · Moyo Maurice Ndontchueng<sup>5,6</sup> · Paalamwé Komi Tchakpele<sup>1,2,3</sup>

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## Abstract

Natural radionuclides activity concentrations of <sup>226</sup>Ra, <sup>228</sup>Ra, and <sup>40</sup>K in soil and grass samples, statistical analysis of these radionuclides, and transfer factors (TFs) from soil to the grass were carried out from phosphate mining and processing sites in southern areas of Togo using gamma-ray spectrometry-based high-purity germanium (HPGe) detector. The average activity concentrations of <sup>226</sup>Ra, <sup>228</sup>Ra, and <sup>40</sup>K were found within the range usually measured worldwide. Statistical analysis was done by investigating the skewness, the kurtosis, and the density distribution of radionuclides in the study areas. Both skewness and kurtosis displayed that the activity concentrations of soil and grass samples in the discharge waste site (Kpémé) and mining site (Hahotoé) exhibited slightly positively skewed, negatively skewed, leptokurtic, and platykurtic distributions. The TFs for <sup>226</sup>Ra, <sup>228</sup>Ra, and <sup>40</sup>K from discharge waste soil to grass were evaluated. The average transfer factors of <sup>226</sup>Ra, <sup>228</sup>Ra, and <sup>40</sup>K in the phosphate processing area were found to be 0.16, 0.32, and 8.42, respectively. <sup>226</sup>Ra and <sup>228</sup>Ra TFs values were found to be within the limit of worldwide measure values, while that of <sup>40</sup>K was found to be larger than the values measured in other countries. The average TF value of 8.42 for <sup>40</sup>K was eight times higher than the average value of unity, showing its large intake by vegetables. The high TFs for <sup>40</sup>K could be explained by the extensive application of phosphate fertilizers to soils when growing vegetables.

**Keywords** Phosphate · Gamma spectrometry · HPGe detectors · Soil · Grass · Transfer factors

## Introduction

Humans and biota are constantly exposed to natural ionizing radiations made of terrestrial, cosmic, and cosmogenic radionuclides, the main sources of natural radiation. The environmental background radiation is made of natural radiation sources and may be affected by anthropogenic radionuclides resulting from atmospheric nuclear weapon testing conducted in the past century, nuclear accidents, and any other human activities (Tagami and Uchida 2009; UNSCEAR 2017). Mining is harmful to human health because it is a natural source of radioactivity. Phosphate extraction and treatment activities in southern Togo could contribute potentially to an increase in radiation exposure to humans and its environment due to the high concentration of radionuclides. The impact of these radionuclides on human health depends on the amount of radionuclide humans are exposed to, through ingestion, inhalation, or direct exposition. It is then important to assess the

✉ Eyakifama Hazou  
constantcr7@gmail.com

<sup>1</sup> Department of Physics, Faculty of Science, University of Lomé, 01 P.O. Box 1515, Lomé, Togo  
<sup>2</sup> Laboratoire de Physique des Matériaux et des Composants à Semi-conducteurs (LPMCS), University of Lomé, 01 P.O. Box: 1515, Lomé, Togo  
<sup>3</sup> Department of Physics, FaST (Faculty of Sciences and Technics), University of Kara, P.O. Box: 404, Kara, Togo  
<sup>4</sup> Jožef Stefan Institute, Jamova cesta 39, 1000 Ljubljana, Slovenia  
<sup>5</sup> National Radiation Protection Agency, P.O. Box 33732, Yaoundé, Cameroon  
<sup>6</sup> Department of Physics, Faculty of Science, University of Douala, PO Box 24157, Douala, Cameroon  
<sup>7</sup> Atomic and Nuclear Spectroscopy, Archeometry, University of Liège, Bat. B15 Sart Tilman, 4000 Liege 1, Belgium

pathway radionuclides move from soil or atmosphere to the human body.

An important step to estimate the intake of radionuclides in the human food chain in the form of ingestion in the human organism is known as the soil–plant transfer factor (TF), which is defined as the ratio between the activity concentration of the plant and the activity concentration of the associated soil (Tagami and Uchida 2009; Banzi et al. 2017). TF is a quantity used in investigations to estimate the impact of routine or accidental monitoring releases of radionuclides into the environment for the most important agricultural products such as cereals, tubers, and other plants grown in the same area (Tagami and Uchida 2009; Adjirackor et al. 2014). Plants are drawing their nutrient resources from their soil roots and this phenomenon is the main carrier of radioactive contamination leading to radionuclides migration in the food chains. One of the most important parameters in the assessment of the environmental safety around nuclear installations such as nuclear power plants, factories and industries, is the TF installation–soil and installation–air parameter (IAEA 1994).

The transfer factors are necessary to estimate the environmental transfer models from the soil to plants that draw their nutrients from the same soil and which is an important aspect in predicting the level of radioactivity in agricultural crops and foodstuffs for assessment of dose to humans (IAEA 1994; Adjirackor et al. 2014). The most important natural radionuclides are  $^{226}\text{Ra}$  (referred to as  $^{238}\text{U}$ ),  $^{228}\text{Ra}$  (referred to as  $^{232}\text{Th}$ ), and  $^{40}\text{K}$ , with different physical and chemical properties related to their environments (Petrinec et al. 2017). The amount of radioactive material in various food and plants according to their absorption capacity is different depending on the different geological formation and composition of the area they grew, the weathering conditions, and the chemical and physical properties of the investigated area.

The phosphate well core is a sample in the solid state of rock in the form of a cylinder and is taken inside the extraction drilled (IAEA 1994; Banzi et al. 2017). These extraction and treatment processes produce waste that must be handled with care. The uncontrolled disposal of these types of waste could lead to environmental pollution and an increase in radioactivity levels in the waste discharge site and surrounding areas (Righi et al. 2005; UNSCEAR 2008). It is therefore possible to increase the radiological exposure of the public. With the increased public concern and awareness about radioactivity in the environment, the present study aims to statistically analyze the amount of each natural radionuclide transfer from soil to grass in the phosphate processing area in southern Togo. The increasing use of phosphate in industries worldwide (milling, processing, and fertilizer manufacturing), especially in Togo, justifies

the importance of investigating the potential radioactivity exposure of phosphate ores to the public and workers.

Particular areas of Kpémé (phosphate processing site: 6°12' 53" N, 01° 30' 16" E) and Hahotoé (phosphate mining site: 6°21' 45" N, 01°23' 48" E) have been identified as contaminated sites earlier by heavy metals and toxic chemicals due to the constant disposal of industrial effluent (Bouka et al. 2013; Aduayi-Akue and Gnandi 2015). As vegetables are an essential part of diets and are ingested both in cooked and raw forms by humans, it is worthy to investigate the transfer of radionuclides from soil to grass used as food. Soil to vegetable transfer of radionuclides is the major pathway of human exposure to potential radioactivity contamination (Righi et al. 2005). In this study, activity concentrations in soil and grass samples are assessed, then transfer factors are calculated based on the measured activity concentrations and statistical analysis of the radionuclide distribution in the investigated area. The estimation of transfer factors of these elements to grass is an important tool used to investigate the potential health effect of humans and animals that consume the products grown in the study region. All these parameters are accessed using some tools and techniques that are described in the following section.

The results obtained from this investigation would be useful for establishing a database in this area of phosphate ore processes and represent a basis to assess any future changes in the levels of radioactivity background due to modern agricultural technologies or any artificial influences around the phosphate ore mining and discharge waste areas.

## Materials and methods

The study area, equipment, procedures, and statistical analysis used in the study are presented in this section.

### Description of the study area

The soil and grass sampling areas were located at Hahotoé (phosphate extraction site) and Kpémé (discharge phosphate waste site), localities of south-east Togo. These places are part of the Maritime Region, in the prefecture of Vo and geographically located between 6°12' 53" N–6° 21' 60" N latitude and 1°23' 26" E–1°31' 41" E longitude. The Kpémé area is located on the coastal sedimentary basin of the Atlantic Ocean and extends along the Gulf of Guinea and is covered with grass, including foodstuffs (Aduayi-Akue and Gnandi 2015). The area of the Hahotoé mining site is affected by weathering and erosion processes which are created by torrential rains. The geological structure of the study areas consists of shale, phosphate shale, quartz, limestone, and phosphate limestone (Bouka et al. 2013; Hazou et al. 2019; Hazou and Patchali 2020). The Hahotoé

phosphate extraction site is the site where material resources are extracted and transported to the Kpémé treatment site. It is worth investigating both sites as there is a transfer of radionuclides from the first to the second and the crops grow on the second site.

### Sampling and sample preparation

Twenty soil samples and ten grass samples were collected from the Hahotoé phosphate extraction site (10 soil samples), from the Kpémé phosphate (10 soil samples) and from the Kpémé phosphate treatment site (10 grass samples). The grass samples were considered, respectively, as P1, P2, P3, P4, P5, P6, P7, P8, P9 and P10, as shown in Fig. 1. One soil sample was made off five sub-samples taken at the four corners and the center of a square of 25 × 25 m<sup>2</sup>. A global positioning system (GPS) was used to record the geographical coordinates of the sampling points to facilitate future investigations on radiological exposures in these mining and discharge areas.

Soil samples were taken from a depth of approximately 2–15 cm from the soil after removing debris from the top layer (first centimeter) to minimize contamination (Guembou Shouop et al. 2017; Penabei et al. 2018). Composite samples were then packaged in a polythene bag and transferred to the laboratory for preparation and measurement. Grass samples were taken around the Kpémé phosphate treatment area. The grass samples were washed in distilled water, dried at about 60 °C for 24 h, and ground to a fine powder in the laboratory (Forkapic et al. 2017; Glavič-Cindro et al. 2020).

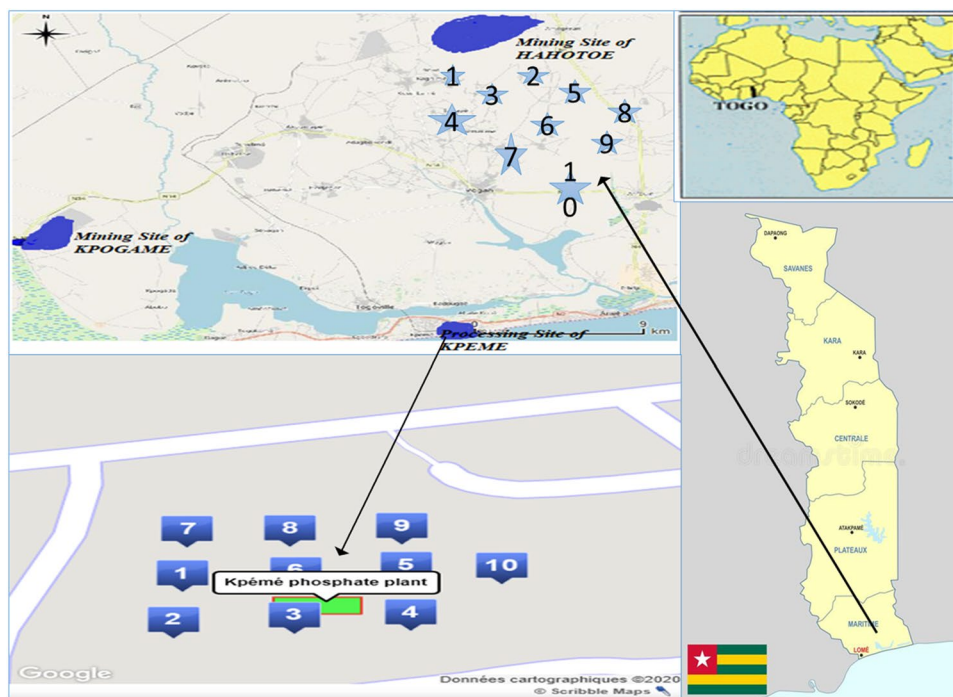
The activity concentrations’ measurement in samples was done using the formula suggested by Beretka (Beretka and Mathew 1985; Guembou et al. 2018). Each sample was prepared in beakers and closed with caps to ensure that the radon gas was confined in the volume and plastic tape was wrapped over the caps so that the daughter radionuclide would also remain in the sample before measurement. This is important to achieve secular equilibrium, as the activity of uranium and thorium series depends on their daughter’s radioactivity (Nguelem et al. 2013; Guembou Shouop et al. 2017; Hazou et al. 2019; Hazou and Patchali 2020).

### Measurement

The activity concentrations of the investigated radionuclides were determined on four HPGe detectors in the Laboratory for Radioactivity Measurements (LMR) of the Jožef Stefan Institute (JSI), Slovenia. These detectors have efficiency higher than 24% and cover the energy range from 4 to 2800 keV (Korun et al. 2012). All detectors have fit for purpose in-house built and constructed shields and each shield consists of an outer shell shield, inner shell shield, and graded layers. The shields are vented by aged (radon free) nitrogen air.

The results obtained from these measurements are traceable to the French national standard at LNE-LNHB (Glavič-Cindro et al. 2016). The geometry of the sample, its matrix, its density, the coincidence summing corrections, and the counting frequency known as systematic effects in gamma-ray spectrometry are taken into account in the estimation of

**Fig. 1** Sampling sites for paired soil and grass samples from Kpémé phosphate plant and soil samples from Hahotoé phosphate mining site



the activity concentration of the radionuclides (Vidmar et al. 2007; Glavič-Cindro et al. 2016). The measurement uncertainties are calculated following the Guide to the Expression of Uncertainty in Measurement rules (GUM 2008; Glavič-Cindro et al. 2020).

The activity concentrations of the long-lived radionuclides are evaluated by determining first the activity concentrations of daughter products which have short half-lives. Thus, the activity concentrations of the uranium series ( $^{238}\text{U}$ -series) were determined using the activity concentration of  $^{234}\text{Th}$  and  $^{234\text{m}}\text{Pa}$ , the activity concentration of  $^{228}\text{Ra}$  is calculated from the activity concentration of  $^{228}\text{Ac}$ , and the activity concentration of  $^{226}\text{Ra}$  is calculated from the activity concentration of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  by taking into account the radon exhalation from the sample matrix and the sample container (Korun and Kovačič 2011; Bučar et al. 2012; Hazou and Patchali 2020). The emission line at 1460.8 keV (10.7%) of  $^{40}\text{K}$  is used to determine directly its activity concentration, while the activity concentrations of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  were calculated based on the weighted average value of their decay products at equilibrium, respectively. The secular equilibrium of the daughter products of the U series ( $^{226}\text{Ra}$ ) and Th series ( $^{228}\text{Ra}$ ) is generally assumed for soil activity concentration; however, it is not true for vegetable samples as they are not direct earthly products with connection to emanation or exhalation. Plants are at least 10 cm from the ground level. All samples were packed in the same geometries, while the counting time for measurement process was 24 h. Therefore, the  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ , and  $^{40}\text{K}$  activity concentrations were used for both soil and vegetation to determine the transfer factors (TFs).

The activity concentrations of each radionuclide in the samples were evaluated using the following expression (Guembou Shouop et al. 2017; Joel et al. 2017a):

$$A(\text{Bq kg}^{-1}) = \frac{\frac{N_S}{t_S} - \frac{N_B}{t_B}}{M_s \times \varepsilon \times P_\gamma \times K_{SC} \times K_{SA} \times K_{DC}}, \quad (1)$$

where  $A(\text{Bq kg}^{-1})$  is the activity concentration of the measured radionuclide,  $N_S/t_S$  is the count rate of each radionuclide present in the sample,  $N_B/t_B$  is the count rate of each radionuclide present in the background,  $M_s$  is the dry weight of the sample,  $\varepsilon$  is the efficiency of the full energy peak,  $P_\gamma$  represents the emission probability,  $K_{SC}$  is the factor for cascade summing correction,  $K_{SA}$  is the correction factor for self-attenuation, and  $K_{DC}$  is the factor for decay correction for each radionuclide.

Parameters that influence the level of radioactivity in the soil and grass samples were evaluated by studying the density distribution of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Ra}$  (Xhixha 2012). The concepts of skewness and kurtosis were used to determine the distribution function of the naturally occurring radionuclides using R version 3.6.2 software (R Core Team 2019).

## Statistical analysis

### – Skewness

The skewness is about the average measure of the asymmetry of the distribution of the activity concentration of a given radionuclide. The skewness ( $S$ ) of a random variable  $x_i$  (the activity concentration in our case) with the data average value  $\bar{x}$  is the third standardized moment, defined by the following equation (R Core Team 2019):

$$S = \frac{\frac{1}{n} \sum_{i=1}^n (x_i - \bar{x})^3}{\left[ \frac{1}{n-1} \sum_{i=1}^n (x_i - \bar{x})^2 \right]^{\frac{3}{2}}}. \quad (2)$$

### – Kurtosis

The kurtosis indicates the degree of “flatness” or “peak-edness” in the same distribution of the activity concentration of a given radionuclide relative to the normal distribution or describes how the dataset is clustered or spread around the average. Kurtosis can either be mesokurtic, platykurtic, or leptokurtic. The kurtosis ( $k_u$ ) is the fourth standardized moment, defined as (R Core Team 2019):

$$k_u = \frac{\frac{1}{n} \sum_{i=1}^n (x_i - \bar{x})^4}{\left[ \frac{1}{n} \sum_{i=1}^n (x_i - \bar{x})^2 \right]^2} - 3 \quad (3)$$

## Transfer factors

The transfer factor (TF) defines the correlation between the activity concentration found in the soil sample matrix and the grass matrix for the same geographical location and the same radionuclide. It is expressed as a function described in Eq. (4) (Rosén et al. 1999). For natural radionuclides, instead of considering deposition on the ground, the activity concentration in plant matter is divided directly by the activity concentration in dry soil matter (Rosén et al. 1999):

$$TF = \frac{\text{Activity concentration plant dry matter}}{\text{Activity concentration in dry soil matter}} \left[ \frac{\text{Bq}_{\text{plant}} \text{ kg}_{\text{dw}}^{-1}}{\text{Bq}_{\text{soil}} \text{ kg}_{\text{dw}}^{-1}} \right] \quad (4)$$

In this study, dry weight of samples was preferred because the quantities of radioactivity per kilogram of dry weight are much less variable than the quantities per unit of fresh weight. This observation reduces uncertainties in

the measurements performed (IAEA 1994). This formula is appropriate as the radionuclides investigated are telluric radionuclides that use the specific activity in grass and in soil to evaluate the factor.

## Results and discussion

### Activity concentrations

#### – Soil samples

In the soil samples of phosphate discharge waste areas (Kpémé) from where the grass samples were collected, the activity concentration of <sup>226</sup>Ra, <sup>228</sup>Ra and <sup>40</sup>K ranged from 182 ± 16 to 919 ± 91 Bq.kg<sup>-1</sup>, 27 ± 1 to 67 ± 3 Bq.kg<sup>-1</sup>, and 37 ± 2 to 89 ± 5 Bq.kg<sup>-1</sup>, respectively, with an average of 442 ± 74 Bq.kg<sup>-1</sup>, 41 ± 4 Bq.kg<sup>-1</sup>, and 69 ± 6 Bq.kg<sup>-1</sup> for phosphate discharge waste (Kpémé) soil. For phosphate mining fields (Hahotoé), the activity concentration of these radionuclides in the soil samples were found to be within 63 ± 6–559 ± 51 Bq.kg<sup>-1</sup> for <sup>226</sup>Ra, 20 ± 1–61 ± 3 Bq.kg<sup>-1</sup>

for <sup>228</sup>Ra, and 14 ± 2–78 ± 4 Bq.kg<sup>-1</sup> for <sup>40</sup>K, with an average of 242 ± 42 Bq.kg<sup>-1</sup> for <sup>226</sup>Ra, 48 ± 5 Bq.kg<sup>-1</sup> for <sup>228</sup>Ra, and 38 ± 7 Bq.kg<sup>-1</sup> for <sup>40</sup>K.

When phosphate discharge waste and phosphate mining soils were compared taking into account their natural radionuclide activity concentrations, it was found that phosphate discharge waste soils contain slightly higher activity concentrations than phosphate mining soils, as shown in Table 1. This could be explained by the phosphate processing activities that increase the level of radionuclides by concentrating them in the areas where activities are undertaken. For comparison with the present work in Table 2, values of activity concentration for naturally occurring radionuclides in phosphate discharge wastes and phosphate mining soils in some studies in other countries around the world are shown (Table 2). Table 2 shows that the activity concentrations of natural radionuclides in phosphate discharge waste and phosphate mining soils of the investigated areas were higher than the activity concentrations of phosphate mining soils and phosphate discharge waste soil obtained by Bolca (Bolca et al. 2007) in Turkey, but lower than the activity concentrations of soil obtained by Uosif (Uosif et al. 2008) in Egypt (Abu-Tartur).

**Table 1** Measured average activity concentrations of <sup>226</sup>Ra, <sup>228</sup>Ra, and <sup>40</sup>K in phosphate ore from discharge waste (Kpémé) and mining (Hahotoé) sites in south-east Togo

Codes	Geographical coordinates	Soil type*	Activity concentrations in mining and discharge waste soil dry weight (Bq.kg <sup>-1</sup> )		
			<sup>226</sup> Ra	<sup>228</sup> Ra	<sup>40</sup> K
P1	(6°12' 51"N-1°30' 85"E)	DW	348 ± 33	28 ± 1	68 ± 4
	(6°20'11"N-1°23'13"E)	M	215 ± 19	54 ± 3	34 ± 2
P2	(6°12'50"N-1°30'71"E)	DW	398 ± 36	31 ± 2	87 ± 4
	(6°20'78"N-1°23'05"E)	M	246 ± 23	57 ± 3	26 ± 2
P3	(6°12'53"N-1°31'07"E)	DW	298 ± 27	30 ± 2	89 ± 6
	(6°19'82"N-1°23'40"E)	M	185 ± 17	51 ± 3	34 ± 2
P4	(6°12'53"N-1°31'52"E)	DW	919 ± 91	67 ± 3	55 ± 4
	(6°19'75"N-1°24'16"E)	M	559 ± 51	41 ± 2	78 ± 4
P5	(6°12'54"N-1°31'14"E)	DW	194 ± 18	56 ± 3	47 ± 3
	(6°19'87"N-1°24'52"E)	M	159 ± 14	61 ± 3	29 ± 2
P6	(6°12'53"N-1°30'47"E)	DW	501 ± 46	34 ± 2	89 ± 5
	(6°19'56"N-1°24'12"E)	M	261 ± 24	55 ± 3	29 ± 3
P7	(6°12'53"N-1°29'02"E)	DW	330 ± 30	27 ± 1	73 ± 4
	(6°19'49"N-1°24'80"E)	M	195 ± 18	59 ± 3	26 ± 2
P8	(6°12'54"N-1°29'27"E)	DW	536 ± 49	39 ± 2	81 ± 4
	(6°19'82"N-1°23'49"E)	M	201 ± 19	56 ± 3	32 ± 2
P9	(6°12'55"N-1°30'18"E)	DW	715 ± 66	47 ± 2	63 ± 5
	(6°19'32"N-1°24'63"E)	M	331 ± 30	26 ± 1	73 ± 4
P10	(6°12'55"N-1°31'88"E)	DW	182 ± 16	52 ± 3	37 ± 2
	(6°19'18"N-1°24'87"E)	M	63 ± 6	20 ± 1	14 ± 2
Averages ± SD		DW	442 ± 74	41 ± 4	69 ± 6
		M	242 ± 42	48 ± 5	38 ± 7

\*"DW" refers to discharge waste site (Kpémé), "M" to mining site (Hahotoé)



**Table 2** Comparison of the activity's concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ , and  $^{40}\text{K}$  in phosphate ore discharge waste and mining soil soils of Kpémé and Hahotoé with reported values from other countries

Countries	Soil samples	Activity concentrations in mining and discharge waste soil dry weight ( $\text{Bq.kg}^{-1}$ )			References
		$^{226}\text{Ra}$	$^{228}\text{Ra}$	$^{40}\text{K}$	
Egypt (Abu-Tartur)	DW	117.6	65	126	(Uosif et al. 2008)
	M	100	50	110	
Finland	DW	10	10	110	(Khan et al. 1998)
Morocco	DW	1600	20	10	
Sudan (Uro)	DW	4131	7.5	62.3	(Sam et al. 1999)
Turkey	DW	55	24.87	NF	(Bolca et al. 2007)
	M	36.77	22.08	NF	
USA (Florida)	DW	1600	20	NF	(Guimond 1990)
	M	1400	15	NF	
Togo	DW	442	41	69	Present work
	M	242	48	38	
Worldwide values	Phosphate ore	40	30	400	(UNSCEAR, 2000)

\*"DW" refers to discharge waste site (Kpémé) and "M" to mining site (Hahotoé)

**Table 3** Activity concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ , and  $^{40}\text{K}$  in grass samples from phosphate ore discharge waste Kpémé

Codes	Vegetables	Activity concentrations in Grass dry weight ( $\text{Bq.kg}^{-1}$ )			
		$^{226}\text{Ra}$	$^{228}\text{Ra}$	$^{40}\text{K}$	$^7\text{Be}$
P1	Grass	$56 \pm 9$	$6 \pm 1$	$421 \pm 21$	$91 \pm 11$
P2	Grass	$45 \pm 6$	$11 \pm 1$	$666 \pm 33$	$76 \pm 9$
P3	Grass	$132 \pm 20$	$26 \pm 2$	$615 \pm 31$	$167 \pm 27$
P4	Grass	$36 \pm 6$	$5 \pm 1$	$614 \pm 31$	$92 \pm 8.6$
P5	Grass	$70 \pm 12$	$6 \pm 1$	$467 \pm 23$	$68 \pm 14$
P6	Grass	$49 \pm 11$	$27 \pm 1$	$736 \pm 36$	$78 \pm 13$
P7	Grass	$26 \pm 7$	$7 \pm 1$	$395 \pm 19$	$67 \pm 15$
P8	Grass	$51 \pm 6$	$6 \pm 1$	$481 \pm 24$	$76 \pm 10$
P9	Grass	$48 \pm 9$	$12 \pm 1$	$586 \pm 29$	$31 \pm 6$
P10	Grass	$29 \pm 4$	$6 \pm 1$	$490 \pm 24$	$31 \pm 7$
Averages $\pm$ SD	Grass	$54 \pm 10$	$11 \pm 3$	$547 \pm 36$	$78 \pm 12$

#### – Mixed grass samples

The activity concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$ , and  $^7\text{Be}$  radionuclides in grass samples from ten different fields of phosphate discharge waste (Kpémé) are presented in Table 3. In the grass collected from agricultural fields, the activity concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$  and  $^7\text{Be}$  ranged  $26 \pm 7 \text{ Bq.kg}^{-1}$ – $132 \pm 20 \text{ Bq.kg}^{-1}$ ,  $5 \pm 1$ – $27 \pm 1 \text{ Bq.kg}^{-1}$ ,  $395 \pm 19$ – $736 \pm 36 \text{ Bq.kg}^{-1}$  and  $31 \pm 6$ – $167 \pm 27 \text{ Bq.kg}^{-1}$ , with an average of  $54 \pm 10 \text{ Bq.kg}^{-1}$ ,  $11 \pm 3 \text{ Bq.kg}^{-1}$ ,  $547 \pm 36 \text{ Bq.kg}^{-1}$  and  $78 \pm 12 \text{ Bq.kg}^{-1}$ , respectively. The  $^7\text{Be}$  was detected because the grass samples were measured directly without storage procedure like soil samples.

The highest activity concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$ , and  $^7\text{Be}$  were observed in grass sample P3, followed by grass sample P1 and P6, while the lowest activity concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$ , and  $^7\text{Be}$  were observed in grass sample P10, grass sample P7 and grass sample P2, respectively. The samples with high activity concentrations (P1, P3, and P6) were found to be closer to the processing plant than the samples with low activities (P2, P7, and P10) as can be seen in Fig. 1. Table 4 shows the average activity concentrations obtained in the present study [phosphate discharge waste (Kpémé)] compared to the results of other studies in other countries. It revealed that the activity concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$ , and  $^7\text{Be}$  in grass of the areas studied in phosphate discharge waste (Kpémé, Togo) was higher than the activity concentrations reported worldwide by the UNSCEAR (2000) and by Ahmed (Ahmed et al. 2005) in Egypt, but similar to those reported in Malaysia by Asaduzzaman (Asaduzzaman et al. 2014) and in Turkey by Bolca (Bolca et al. 2007).

## Statistical analysis

### – Skewness and kurtosis for $^{226}\text{Ra}$ in dry weight soil and grass samples

Statistics using R Core Team (2019) from Table 5 shows that the distribution of activity concentrations of  $^{226}\text{Ra}$  in both phosphate mining sites (Hahotoé) and discharge waste site (Kpémé) soil and grass samples is positively skewed (1.15, 0.73, and 1.55, respectively) at 5% level of significance; thus, the average activity concentration of  $^{226}\text{Ra}$  is greater than the modal activity concentration of measured soil samples (as shown in Fig. 2). The negative kurtosis

**Table 4** Comparison of the activity's concentrations of <sup>226</sup>Ra, <sup>228</sup>Ra, and <sup>40</sup>K in grass from phosphate ore discharge waste from different countries

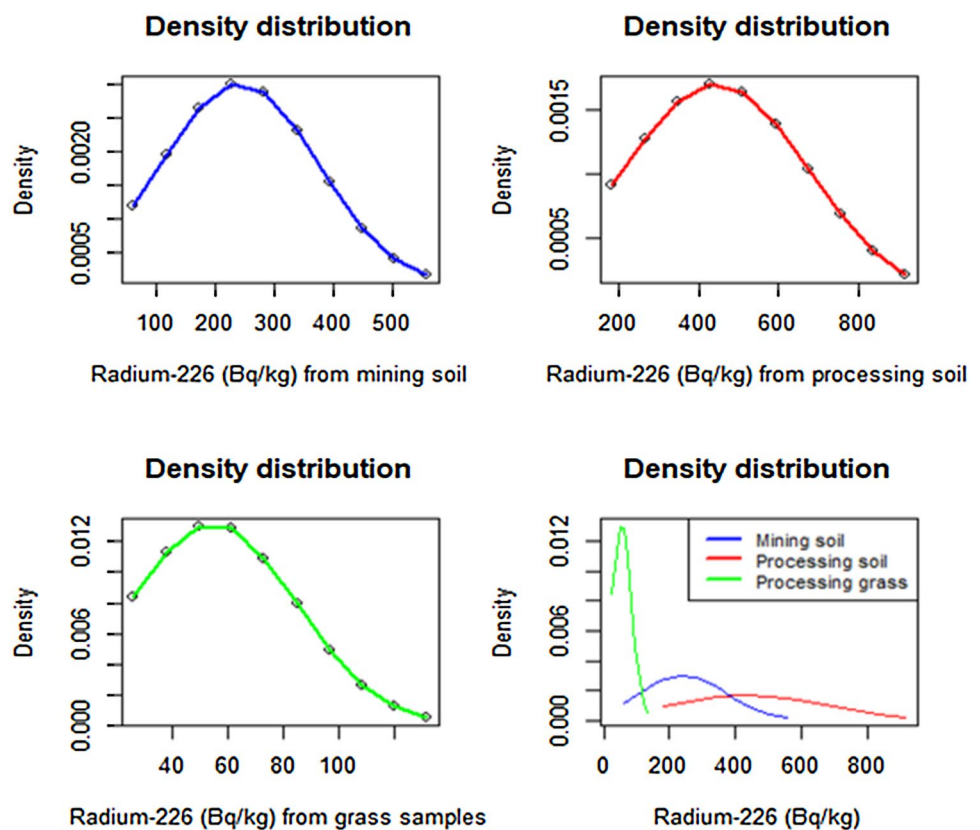
Countries	Samples	Activity concentrations in grass (grass) from phosphate discharge waste, dry weight (Bq.kg <sup>-1</sup> )			References
		<sup>226</sup> Ra	<sup>228</sup> Ra	<sup>40</sup> K	
Egypt	Grass	0.03	0.0005	–	(Ahmed et al. 2005)
Malaysia	Grass	10.14	17.38	4374.88	(Asaduzzaman et al. 2014)
Turkey	Grass	27.74	10.17	1497.88	(Bolca et al. 2007)
Togo	Grass	54	11	547	Present work
IAEA	Grass	3.3	0.45	432	(IAEA 1994)
Worldwide values	Grass	0.05	0.015	–	(UNSCEAR 2017)

**Table 5** Skewness and kurtosis for <sup>226</sup>Ra, <sup>228</sup>Ra, and <sup>40</sup>K in dry weight soil and grass samples from phosphate mining site (Hahotoé) and from discharge waste site (Kpémé)

Samples	<sup>226</sup> Ra		<sup>228</sup> Ra		<sup>40</sup> K	
	Skewness	Kurtosis	Skewness	Kurtosis	Skewness	Kurtosis
Soil (mining site)	1.15	0.79	−0.94	−0.82	1.02	−0.56
Soil (processing site)	0.73	−0.74	0.54	−1.32	−0.36	−1.43
Grass (processing site)	1.55	1.52	1.07	−0.66	0.18	−1.50

5% level of significance

**Fig. 2** Density distribution of activity concentrations of <sup>226</sup>Ra in soil and grass samples from mining and processing sites of phosphate



(−0.74) of discharge waste (Kpémé) soil shows that the distribution of activity concentrations of <sup>226</sup>Ra is platykurtic. The term platykurtic describes a statistical distribution with extremely dispersed activity concentrations of <sup>226</sup>Ra in

discharge waste soil, and the distribution will tend to have a flat top near the average rather than a sharp peak (as shown in Fig. 2). The positive kurtosis (0.79 and 1.52) of phosphate mining soil and grass from the processing area shows

that the distribution of activity concentrations of  $^{226}\text{Ra}$  is leptokurtic (as shown in Fig. 2). Leptokurtic describes a statistical distribution where the activity concentrations of  $^{226}\text{Ra}$  are clustered around the average activity concentration in the soil, and the distribution will tend to have a distinct peak near the average, which declines rapidly, and a heavy tail (as shown in Fig. 2).

- Skewness and kurtosis for  $^{228}\text{Ra}$  in dry weight soil and grass samples

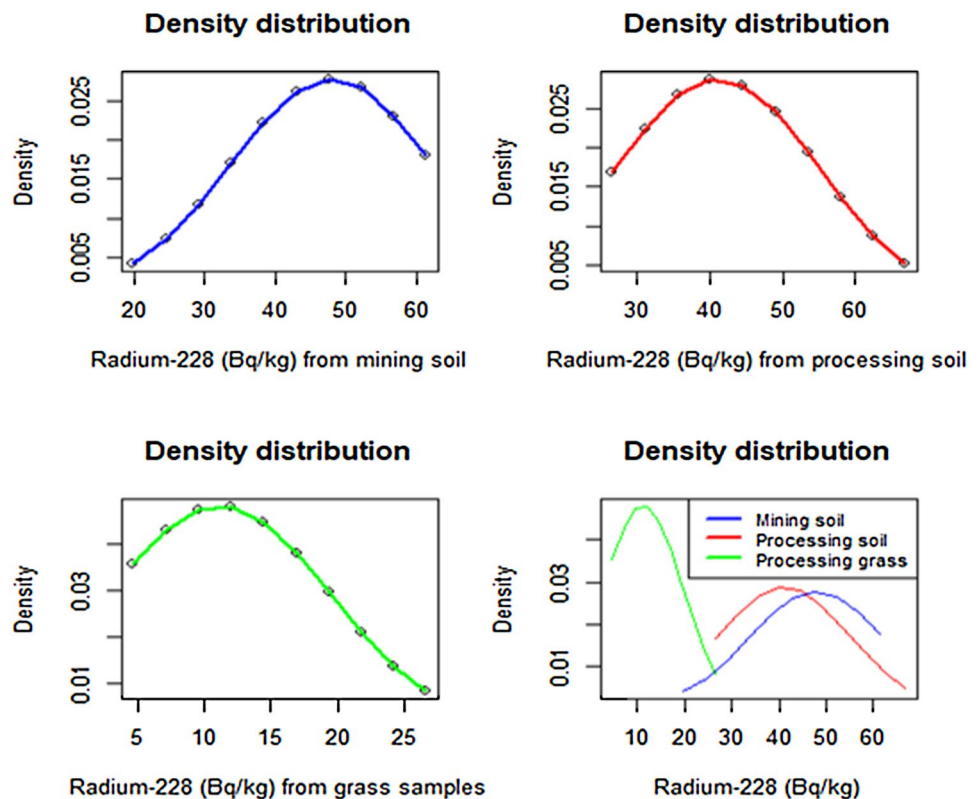
Statistics from Table 5 also shows that the distribution of activity concentrations of  $^{228}\text{Ra}$  in discharge waste site (Kpémé) soil and grass from the same area is positively skewed (0.54 and 1.07, respectively); thus, the average activity concentration of  $^{228}\text{Ra}$  in discharge waste soil and grass is greater than the modal activity concentration, or the soils and grass that measured relatively lower activity concentrations of  $^{228}\text{Ra}$  are more than those that measured higher activity concentrations (as shown in Fig. 3). The activity concentrations of  $^{228}\text{Ra}$  in phosphate mining soil is negatively skewed ( $-0.94$ ); thus, the average activity concentration of  $^{228}\text{Ra}$  in phosphate mining soil is less than the modal activity concentration, or soils that measured a relatively higher activity concentration of  $^{228}\text{Ra}$  are more than those that measured a lower activity concentration (as shown in Fig. 3). The negative kurtosis of both discharge waste

(soil and grass) and mining soil ( $-1.32$ ,  $-0.66$ , and  $-0.82$ , respectively) shows that the distribution of activity concentrations of  $^{228}\text{Ra}$  is platykurtic, describing a statistical distribution with extremely dispersed activity concentrations of  $^{228}\text{Ra}$  in phosphate soils and grass samples and the distribution will tend to have a flat top near the average rather than a sharp peak (as shown in Fig. 3).

- Skewness and kurtosis for  $^{40}\text{K}$  in dry weight soil and grass samples

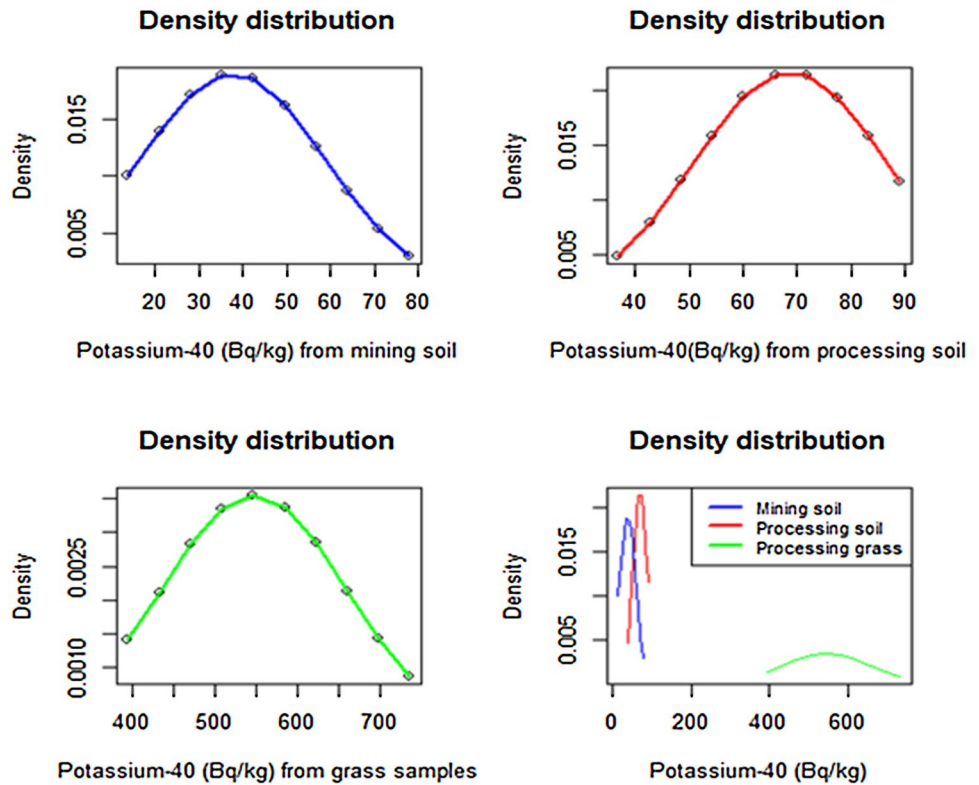
Statistics from Table 5 shows that the activity concentrations of  $^{40}\text{K}$  in the discharge waste site (Kpémé) soil is negatively skewed ( $-0.36$ ); thus, the average activity concentration of  $^{40}\text{K}$  in phosphate mining soil is less than the modal activity concentration, or soils that measured a relatively higher activity concentration of  $^{40}\text{K}$  are more than those that measured a lower activity concentration (as shown in Fig. 4). The distribution of activity concentrations of  $^{40}\text{K}$  in phosphate mining site (Hahotoé) soil and in grass samples from discharge waste site is positively skewed (1.02 and 0.18 respectively) (as shown in Fig. 4); thus, the average activity concentration of  $^{40}\text{K}$  in discharge waste grass samples and in phosphate mining soil is greater than the modal activity concentration, or the soils and grass samples that measured relatively lower activity concentrations of  $^{40}\text{K}$  are more than those that measured higher activity concentrations.

**Fig. 3** Density distribution of activity concentrations of  $^{228}\text{Ra}$  in soil and grass samples from mining and processing sites of phosphate





**Fig. 4** Density distribution of activity concentrations of <sup>40</sup>K in soil and grass samples from mining and processing site of phosphate



The negative kurtosis values (− 0.56, − 1.43, and − 1.50, respectively) of both phosphate mining site (Hahotoé) soil and discharge waste site (Kpémé) soil and grass samples show that the distribution of activity concentrations of <sup>40</sup>K is platykurtic. The term platykurtic describes a statistical distribution with extremely dispersed concentration of <sup>40</sup>K in these soils and grass samples and the distribution will tend to have a flat top near the average rather than a sharp peak (as shown in Fig. 4).

**Transfer factors of <sup>226</sup>Ra, <sup>228</sup>Ra, and <sup>40</sup>K from soil to grass in discharge waste (Kpémé)**

The TFs of <sup>226</sup>Ra, <sup>228</sup>Ra, and <sup>40</sup>K from the investigated phosphate discharge waste soil (Kpémé) to grass using Eq. (4) are presented in Table 6. The average values of TF were 0.16 ± 0.04, 0.32 ± 0.09, and 8.42 ± 0.80, respectively, for <sup>226</sup>Ra, <sup>228</sup>Ra, and <sup>40</sup>K. The highest TF of (0.44) for <sup>226</sup>Ra was found in the grass sample from P3, followed by grass sample from P5 with TF equal to (0.36). The average TF of (0.16) is higher than the result published by Hasan (Hasan et al. 2010). The highest TF values of (0.87) and (13.32) for <sup>228</sup>Ra and <sup>40</sup>K, respectively, were found in grass samples from P3 and P10, respectively. The average TFs for all grass samples for <sup>228</sup>Ra and <sup>40</sup>K were 0.32 and 8.42, respectively. These results are higher than the results published by Hasan (Hasan et al. 2010) in Pakistan.

**Table 6** Transfer factors of <sup>226</sup>Ra, <sup>228</sup>Ra, and <sup>40</sup>K in soil to grass samples from discharge waste (Kpémé)

Codes	TFs estimation (unit of Bq kg <sup>-1</sup> dry mass grass per Bq kg <sup>-1</sup> dry mass soil)		
	TFs ± SD		
	<sup>226</sup> Ra	<sup>228</sup> Ra	<sup>40</sup> K
P1	0.16 ± 0.03	0.23 ± 0.04	6.14 ± 0.31
P2	0.11 ± 0.02	0.36 ± 0.03	7.68 ± 0.38
P3	0.44 ± 0.07	0.87 ± 0.07	6.92 ± 0.35
P4	0.04 ± 0.01	0.07 ± 0.01	11.23 ± 0.56
P5	0.36 ± 0.06	0.11 ± 0.02	9.96 ± 0.49
P6	0.1 ± 0.02	0.79 ± 0.03	8.24 ± 0.40
P7	0.08 ± 0.02	0.25 ± 0.04	5.44 ± 0.26
P8	0.1 ± 0.01	0.16 ± 0.03	5.97 ± 0.30
P9	0.07 ± 0.01	0.24 ± 0.02	9.26 ± 0.46
P10	0.16 ± 0.02	0.12 ± 0.02	13.32 ± 0.65
Averages ± SD	0.16 ± 0.04	0.32 ± 0.09	8.42 ± 0.80

TF Transfer factor and SD standard deviation

The obtained results display the highest transfer factor for potassium. This is probably explained by the use of potassium as the crop’s fertilizer in the investigated area. However, high values of higher TF for potassium do not pose a risk to human health because <sup>40</sup>K has low contribution to internal dose, as <sup>40</sup>K content in the human body is homeostatically controlled (IAEA 1994; Banzi et al. 2017)).

TFs for  $^{226}\text{Ra}$  were found to be less than those of  $^{228}\text{Ra}$  and  $^{40}\text{K}$  in all grass samples in this study. This indicates that  $^{226}\text{Ra}$  is less effectively transported from soil to grass than  $^{228}\text{Ra}$  and  $^{40}\text{K}$ . The variation of soil-to-plant transfer factors depends on the soil's properties as the nutrient standard, the exchangeable K content, and the content of moisture in the soil (Frissel et al. 2002; Banzi et al. 2017). For the investigated radionuclides, the literature usually presents the same range of TFs values obtained in the present study except for the  $^{40}\text{K}$  that are slightly lower than those got in this work.

The observed variation of transfer factors for radium can be explained by its non-uniform distribution in various lands where crops grow, even for closest sampling points. This non-uniform distribution is also displayed in the results of statistical analysis as can be seen in Figs. 2, 3, 4. Investigations of grass samples showed that the most abundant radionuclide in grass is potassium 40, followed by radium 228 and radium 226, respectively. This explains the low absorption of radium isotopes by grass, with values of TFs less than the unity. The uptake of different radionuclides from the soil by grass depends on various properties, including soil texture and clay content, also taking into account the dominant clay minerals in the soil (IAEA 1994). The soil in these areas would be more inclined to the transport of potassium than other radionuclides.

The present study's results were compared to what has been done in similar regions in the world with mining and processing activity of phosphate. We can assume that the average values of the soil to grass transfer factors have been found to be in close agreement with those published by the IAEA (1994) except those for potassium which are higher than the IAEA's values as shown in Table 7. The TF of  $^{228}\text{Ra}$  in our study was found to be close to the result found by Harb (Harb et al. 2014) in Egypt, but less than the value found by Mohannad (Mohannad et al. 2014) in Palestine and much higher than that found by Hasan (Hasan et al. 2010) in Pakistan as shown in Table 7. This variation might be due to different soil's physical and chemical properties, the climate

in the area the study had been carried out, and some extra practices at operational processing sites.

This high transfer coefficient from soil to plant compared to other parts of the world can affect the animal grazing such as cows that are later used for milk and meat production. Therefore, the transport of radionuclides from soil to grass ( $^{40}\text{K} > ^{228}\text{Ra} > ^{226}\text{Ra}$  decrease in this order) affects animals that consume grass. Furthermore, by accumulating those radionuclides in the animal's body, the radiological risk of people who take milk or meat from those animals increases. The variations of TFs values from each country depend strongly on variability in contamination of soil due to non-homogeneous deposition of radionuclides into associated soil and can also be due to seasonal variations in diet and/or feeding behavior in dry or rainy season (IAEA 2010).

The obtained results demonstrate that each primordial radionuclide follows complex dynamics phenomena. These processes lead those radionuclides to be transported from soil to vegetables in different manner as described by Mohammed et al. (2014) and in IAEA (1994 and 2010). The portion of these radionuclides, which is in the soil matrix, can be drawn into vegetables through their roots. In some cases, this is facilitated by their chemical similarity with other elements that the vegetables normally use for their growth. This explains why there are differences between values of TFs observed between different vegetables in different locations around the world as shown in Tables 6, 7. Furthermore, radionuclides belonging to physiologically regulated elements, or their analogs, may be selectively adsorbed, whereas others may be excluded. In general, the highest activity concentration in vegetables was found in samples collected from areas with the highest radioactivity concentration in soil substrate (IAEA 2010; UNSCEAR 2017). This observation on the level of the activity concentration in vegetables is not linearly correlated to the activity in soils. Several authors reported that this nonlinear and non-uniform correlation in phosphate and mining sites is due to adsorption phenomena and the clayey nature of

**Table 7** Comparison of transfer factors of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ , and  $^{40}\text{K}$  in soil to grass samples from phosphate ore discharge waste with different countries

Countries	TFs estimation in the world (unit of Bq kg <sup>-1</sup> dry mass grass per Bq kg <sup>-1</sup> dry mass soil)						References
	$^{226}\text{Ra}$		$^{228}\text{Ra}$		$^{40}\text{K}$		
	Range	Averages	Range	Averages	Range	Averages	
Egypt	0.19–0.73	0.43	0.09–0.88	0.31	0.31–2.95	1.06	(Harb et al. 2014)
India	–	–	0.22–0.34	–	0.13–0.95	–	(Manigandan et al. 2008)
Pakistan	0.009–0.276	0.06	0.013–0.270	0.09	0.271–0.317	0.28	(Hasan et al. 2010)
Palestine	0.27–2.12	1.26	0.10–4.97	1.15	0.13–16.3	1.2	(Mohannad et al. 2014)
Saudi Arabia	0.06–0.19	0.12	–	–	0.08–0.23	0.16	(Alharbi et al. 2013)
Togo	<b>0.04–0.44</b>	<b>0.16</b>	<b>0.07–0.87</b>	<b>0.32</b>	<b>5.44–13.32</b>	<b>8.42</b>	<b>Present work</b>
IAEA	<b>0.01–1.00</b>	<b>0.11</b>	–	–	<b>0.49–5.60</b>	<b>1.4</b>	(IAEA 1994)

the phosphate soils (Frissel et al. 2002; Uosif et al. 2008; Alharbi et al. 2013; Asaduzzaman et al. 2014; Banzi et al. 2017). These two properties are the main reasons for the retention of radionuclides on the top layer of the soil. This situation decreases the mobility of radionuclides at depths. Therefore, the uptake of all radionuclides depends not only upon the mining and processing activities, but also on their chemical metabolism which defines the mobility of these radionuclides from soil to plant.

## Conclusion

The present study investigated the activity concentration of natural radionuclides in soils and grass samples from phosphate exploitations sites in Togo, discharge waste site of Kpémé and phosphate mining site of Hahotoé.  $^{226}\text{Ra}$  ( $^{238}\text{U}$ -series),  $^{228}\text{Ra}$  ( $^{232}\text{Th}$ -series), and  $^{40}\text{K}$  were found to be the main radiological constituents of soil and grass samples in the studied areas and  $^7\text{Be}$  was found in small concentration but in the range measured in other countries worldwide. Statistical analysis was performed by assessing the skewness, kurtosis, and the density distribution of the radionuclides in the investigated samples. The transfer factors of radionuclides from soil to grass obtained were within the range measured in other parts of the world except for that of  $^{40}\text{K}$ . The high TFs for  $^{40}\text{K}$  could be explained by the extensive application of phosphate fertilizers to soils when growing vegetables. Even though the fertilizer is not made of 100%  $^{40}\text{K}$ , it may slightly increase the activity concentrations of natural radionuclides. Its values are not a sign of significant radiological alarm as  $^{40}\text{K}$  is part of human/animal body and contribute to maintaining their life on Earth. The level of TFs for  $^{40}\text{K}$  highlighted the necessity of performing further investigation about its transfer from soil to grass and the characteristic of the discharge waste site.

Considerable variability was observed in the activity concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ , and  $^{40}\text{K}$  from all samples in both sites and all grass samples investigated. A high level of activity concentrations in discharge waste soil (Kpémé) in this study confirmed the previously reported values by Hazou et al. (Hazou et al. 2019). This leads us to conclude that phosphate treatment and waste storage increase the level of radioactivity in the processing area and therefore affect crops and pasture on the surrounding soils. TFs are statistically uncertain, thus for further investigations, there is a need to replicate TFs values for individual combinations of each radionuclide, soil type and crop, necessary to obtain reliable average values based on radiological assessment models.

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## Declarations

**Conflict of interest** The authors declare no conflicts of interest.

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