Transfer from Soil to Grass and Statistical Analysis of Natural Occurring Radionuclides in Soil from Phosphate Mining and Processing Sites in Maritime Region of Togo.

Eyakifama Hazou (constantcr7@gmail.com)
University of Lome Faculty of Sciences: Universite de Lome Faculte des Sciences
https://orcid.org/0000-0003-2167-7945
Benjamin Zorko
Jozef Stefan Institute: Institut Jozef Stefan
Milohum Mikesokpo Dzagli
University of Lome Faculty of Sciences: Universite de Lome Faculte des Sciences
Essolakina Manakrihèa Haliba
University of Lome Faculty of Sciences: Universite de Lome Faculte des Sciences
Cebastien Joel Guembou Shouop
Universite de Douala Faculte des Sciences
Maurice Ndontchueng Moyo
Universite de Douala Faculte des Sciences
Paalamwé Komi Tchakpele
University of Lome Faculty of Sciences: Universite de Lome Faculte des Sciences

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Title:

Transfer from Soil to Grass and Statistical Analysis of Natural Occurring Radionuclides in Soil from Phosphate Mining and Processing Sites in Maritime Region of Togo.

HAZOU Eyakifama [a, b, *], ZORKO Benjamin [d], DZAGLI Milohum Mikesokpo [a,b, http://orcid.org/0000-0002-6021-3599], HALIBA Essolakina Manakrihèa [a, b], GUEMBOU SHOUOP Cebastien Joel [e, f, g], NDONTCHUENG MOYO Maurice [e, f], TCHAKPELE Paalamwé Komi [a, b, c].

[a] Department of Physics, Faculty of Science, University of Lomé, 01 P.O. Box 1515, Lomé, Togo.
[b] Laboratoire de Physique des Matériaux et des Composants à Semi-conducteurs (LPMCS), University of Lomé, 01 P.O. Box : 1515 Lomé –Togo.
[c] Department of Physics, FaST (Faculty of Sciences and Technics), University of Kara, P.O. Box: 404, Kara – Togo.
[d] Jožef Stefan Institute, Jamova cesta 39, SI-1000, Ljubljana, Slovenia.
[e] National Radiation Protection Agency, P.O. Box 33732, Yaoundé, Cameroon.
[f] Department of Physics, Faculty of Science, University of Douala, P.O. Box 24157, Douala, Cameroon.
[g] Atomic and Nuclear Spectroscopy, Archeometry, University of Liège, Bat. B15 Sart Tilman, 4000 Liege 1, Belgium.

Abstract

Natural radionuclides activity concentrations of ^{226}Ra, ^{228}Ra, and ^{40}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 ^{226}Ra, ^{228}Ra, and ^{40}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 dischargement waste site (Kpémé) and mining site (Hahotoé) exhibited slightly positively skewed, negatively skewed, leptokurtic, and platykurtic distributions. The TFs for ^{226}Ra, ^{228}Ra, and ^{40}K from dischargement waste soil to grass were evaluated. The average transfer factors of ^{226}Ra, ^{228}Ra, and ^{40}K in the phosphate processing area were found to be 0.16, 0.32, and 8.42, respectively. ^{226}Ra and ^{228}Ra TFs values were found to within the limit of worldwide measure values while that of ^{40}K was to be larger than the values measured in other countries. The average TF value of 8.42 for ^{40}K was 8 times higher than the average value of unity, showing its large intake by vegetables. The high TFs for ^{40}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.

1. 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 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; Mollah AS, 2014). 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 human (IAEA, 1994; Adjirackor et al., 2014). The most important natural radionuclides are ^{226}Ra (referred to as ^{238}U), ^{228}Ra (referred to as ^{232}Th), and ^{40}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 different the 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; Mollah AS, 2014). 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, justify the importance of investigating the potential radioactivity exposure of phosphate ores to the public and workers.

The 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, though 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 who consumes the products grown in the study region. All these parameters are accessed using some tools and technics 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 dischargement waste areas.

2. Materials and methods

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

2.1. Description of the study area

The soil and grass sampling areas were located at Hahotoé (phosphate extraction site) and Kpémé (dischargement 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 worthy to investigate both sites as there is a transfer of radionuclides from the first to the second and the crops grow on the second site.

2.2. Sampling and samples 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 Figure 1. One soil sample was made off five sub-samples taken at the four corners and the center of a square of 25x25 m^2. A Global Positioning System (GPS) was used to record the geographical coordinates of the sampling points in order to facilitate future investigations on radiological exposures in these mining and discharge areas.

Soil samples were taken from a depth of approximately 2 to 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, grounded 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; Joel, Penabei, Maurice, et al., 2017b). 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).

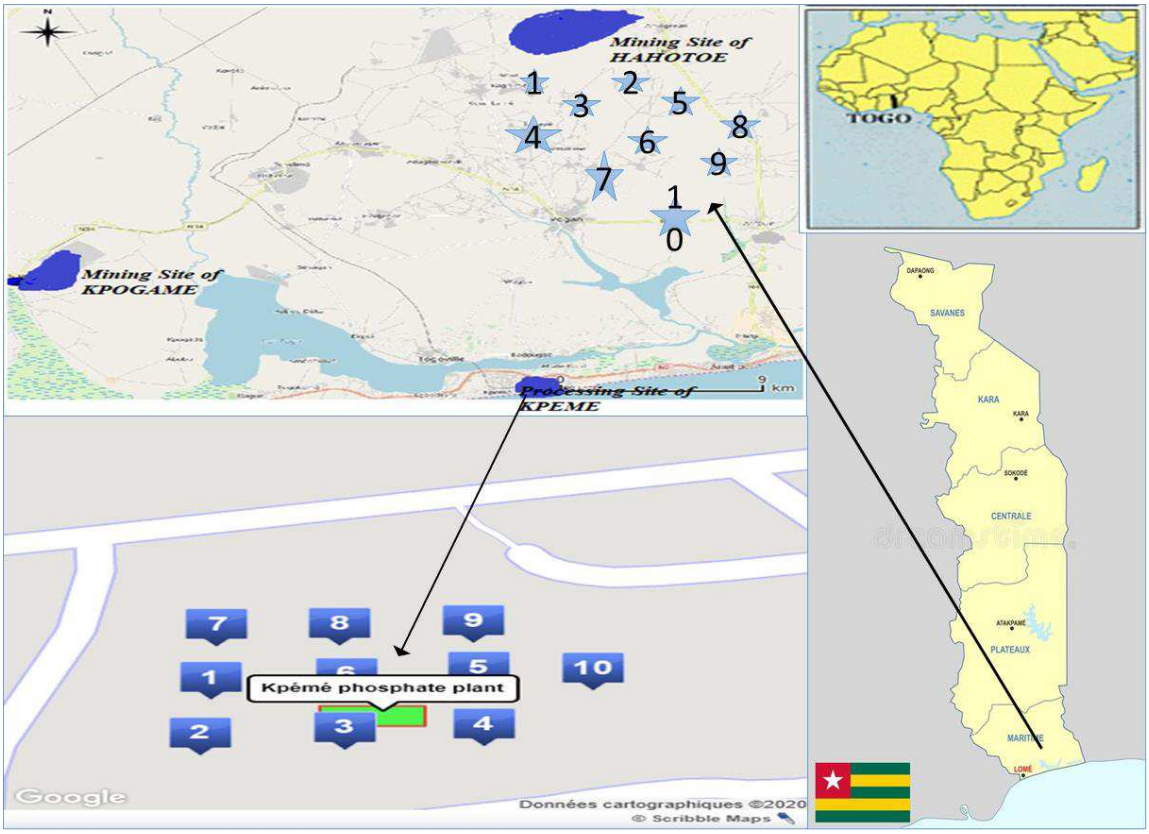

Figure 1: Sampling sites for paired soil and grass samples from Kpémé phosphate plant and soil samples from Hahotoé phosphate mining site.

2.3. 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 keV 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 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}U-series) were determined using the activity concentration of ^{234}Th and ^{234m}Pa, the activity concentration of ^{228}Ra is calculated from the activity concentration of ^{228}Ac, and the activity concentration of ^{226}Ra is calculated from the activity concentration of ^{214}Pb and ^{214}Bi by taking into account the radon exhalation from the sample matrix and the sample container (Korun and Kovačič, 2011; Bučar et al., 2012). The emission line at 1460.8 keV (10.7%) of ^{40}K is used to determine directly its activity concentration, while activity concentrations of ^{226}Ra and ^{228}Ra were calculated based on the weighted average value of their decay products at equilibrium respectively. The secular equilibrium of daughter products of U-series (^{226}Ra) and Th-series (^{228}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 upper the ground level. All samples were packed

in the same geometries, while the counting time for measurement process was 24 hours. Therefore, the ^{226}Ra, ^{228}Ra, and ^{40}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):

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

where $A(\mathrm{Bq\,kg^{-1}})$ is the activity concentration of the measured radionuclide, *Ns/ts* 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, ε is the efficiency of the full energy peak, P_γ represents the emission probability, K_{SC} is the factor for cascade summing correction, K_{SA} is the correction factor for self-attenuation, 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}K, ^{226}Ra, and ^{228}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).

2.4. Statistical analysis

- **Skewness**

The skewness is a measure of the asymmetry of the distribution of the activity concentration of a given radionuclide about its average. 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 “peakedness” 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):

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

2.5. 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 equation (**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):

$$TF = \frac{Activity\ concentration\ plant\ dry\ matter}{Activity\ concentration\ in\ dry\ soil\ matter}\left[\frac{Bq_{plant}\,kg_{dw}^{-1}}{Bq_{soil}\,kg_{dw}^{-1}}\right] \qquad (4)$$

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 tellurique radionuclides that use the specific activity in grass and in soil to evaluate the factor.

3. Results and discussion

3.1. Activity concentrations

- **Soil samples**

In the soil samples of phosphate dischargement waste areas (Kpémé) from where the grass samples were collected, the activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K, are ranged from 182 ± 16 to 919 ± 91 Bq.kg^{-1}, 27 ± 1 to 67 ± 3 Bq.kg^{-1}, and 37 ± 2 to 89 ± 5 Bq.kg^{-1}, respectively, with an average of 442 Bq.kg^{-1}, 41 Bq.kg^{-1}, and 69 Bq.kg^{-1} for phosphate dischargement 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^{-1} for ^{226}Ra, 20 ± 1 – 61 ± 3 Bq.kg^{-1} for ^{228}Ra, and 14 ± 2 – 78 ± 4 Bq.kg^{-1} for ^{40}K, with an average of 241 Bq.kg^{-1} for ^{226}Ra, 22 Bq.kg^{-1} for ^{228}Ra, and 37 Bq.kg^{-1} for ^{40}K.

When phosphate dischargement waste and phosphate mining soils were compared taking into account their natural radionuclide activity concentrations, it was found that phosphate dischargement 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 dischargement wastes and phosphate mining soils in some studies in other countries around the world were shown (Table 2). From Table 2, it comes out that activity concentrations of natural radionuclides in phosphate dischargement waste and phosphate mining soils of the investigated areas were higher than the activity concentrations of phosphate mining soils and phosphate dischargement 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 ^{226}Ra, ^{228}Ra, and ^{40}K in phosphate ore from dischargement waste (Kpémé) and mining (Hahotoé) sites in South-East Togo.

Codes	Geographical coordinates	Soil type*	Activity concentrations in mining and dischargement waste soil dry weight ($Bq.kg^{-1}$)		
			^{226}Ra	^{228}Ra	^{40}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		DW	442	41	69
		M	241	22	37

* "DW" refers to Dischargement Waste site (Kpémé) and "M" to Mining site (Hahotoé)

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

Countries	Soil samples	Activity concentrations in mining and dischargement waste soil dry weight (Bq.kg^{-1})			Reference
		^{226}Ra	^{228}Ra	^{40}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 M. 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	241	22	37	
Worldwide value	Phosphate ore	**40**	**30**	**400**	(UNSCEAR, 2000)

* "DW" refers to Dischargement Waste site (Kpémé) and "M" to Mining site (Hahotoé)

- **Mixed grass samples**

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

The highest activity concentrations of ^{226}Ra, ^{228}Ra, ^{40}K, and ^{7}Be were observed in grass sample P3 followed by grass sample P1 and P6, while the lowest activity concentrations of ^{226}Ra, ^{228}Ra, ^{40}K, and ^{7}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 Figure 1. Table 4 shows the average activity concentrations obtained in the present study (phosphate dischargement waste (Kpémé)) and previous studies for comparison. It revealed that the activity concentrations of ^{226}Ra, ^{228}Ra, ^{40}K, and ^{7}Be in grass of the areas studied in phosphate dischargement waste (Kpémé, Togo) was higher than the activity concentrations reported in worldwide by the UNSCEAR (2000) and by Ahmed (Ahmed NK et al., 2005) in Egypt but in the range of those reported in Turkey by Ekdal (Ekdal E et al., 2005) and Bolca (Bolca M et al., 2007).

Table 3: Activity concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K in grass samples from phosphate ore dischargement waste Kpémé

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

Table 4: Comparison of the activity's concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K in grass from phosphate ore dischargement waste from different countries

Countries	Samples	Activity concentrations in grass (grass) from phosphate dischargement waste, dry weight ($Bq.kg^{-1}$)			References
		^{226}Ra	^{228}Ra	^{40}K	
Egypt	Grass	0.03	0.0005	-	(Ahmed NK et al., 2005)
Turkey	Grass	10.14	17.38	4374.88	(Ekdal E et al., 2005)
Turkey	Grass	27.74	10.17	1497.88	(Bolca M et al., 2007)
Togo	**Grass**	**54**	**11**	**547**	**Present work**
IAEA	**Grass**	**3.3**	**0.45**	**432**	(IAEA, 1994)
Worldwide value	**Grass**	**0.05**	**0.015**	**-**	(UNSCEAR, 2017)

3.2. Statistical analysis

- **Skewness and kurtosis for ^{226}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}Ra in both phosphate mining sites (Hahotoé) and dischargement 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 concentrations of ^{226}Ra is greater than modal activity concentration of measured soil samples (as shown in Figure 2). The negative kurtosis (-0.74) of dischargement waste (Kpémé) soil shows that the distribution of activity concentrations of ^{226}Ra is platykurtic. The term platykurtic describes a statistical distribution with extremely dispersed activity concentrations of ^{226}Ra in dischargement waste soil, and the distribution will tend to have a flat top near the average rather than a sharp peak (as shown in Figure 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}Ra is leptokurtic (as shown in Figure 2). Leptokurtic describes a statistical distribution where the activity concentrations of ^{226}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 have a heavy tail (as shown in Figure 2).

Table 5: Skewness and kurtosis for ^{226}Ra, ^{228}Ra, and ^{40}K in dry weight soil and grass samples from phosphate mining site (Hahotoé) and from dischargement waste site (Kpémé)

Samples	^{226}Ra		^{228}Ra		^{40}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

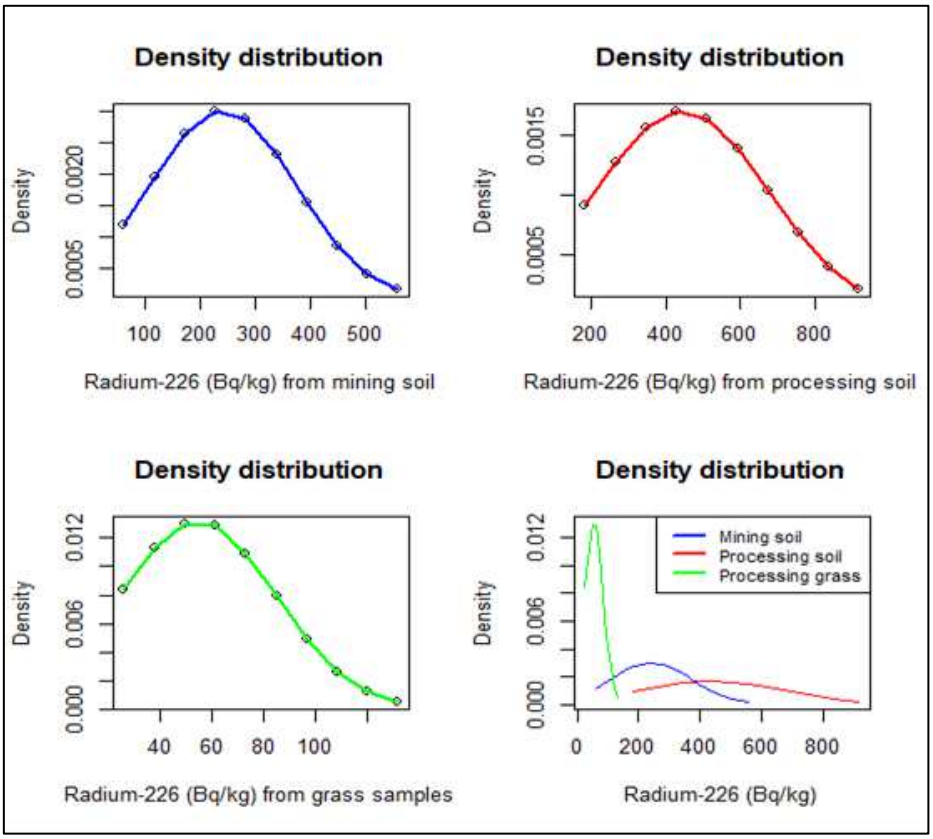

Figure 2: Density distribution of activity concentrations of ^{226}Ra in soil and grass samples from mining and processing sites of phosphate.

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

Statistics from Table 5 also shows that the distribution of activity concentrations of ^{228}Ra in dischargement 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}Ra in dischargement waste soil and grass is greater than the modal activity concentration, or the soils and grass that measured relatively lower activity concentrations of ^{228}Ra are more than those that measured higher activity concentrations (as shown in Figure 3). The activity concentrations of ^{228}Ra in phosphate mining soil is negatively skewed (-0.94); thus, the average activity concentration of ^{228}Ra in phosphate mining soil is less than the modal activity concentration, or soils that measured a relatively higher activity concentration of ^{228}Ra are more than those that measured a lower activity concentration (as shown in Figure 3). The negative kurtosis of both dischargement waste (soil and grass) and mining soil (-1.32, -0.66, and -0.82, respectively) shows that the distribution of activity concentrations of ^{228}Ra is platykurtic, describing a statistical

distribution with extremely dispersed activity concentrations of ^{228}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 Figure 3).

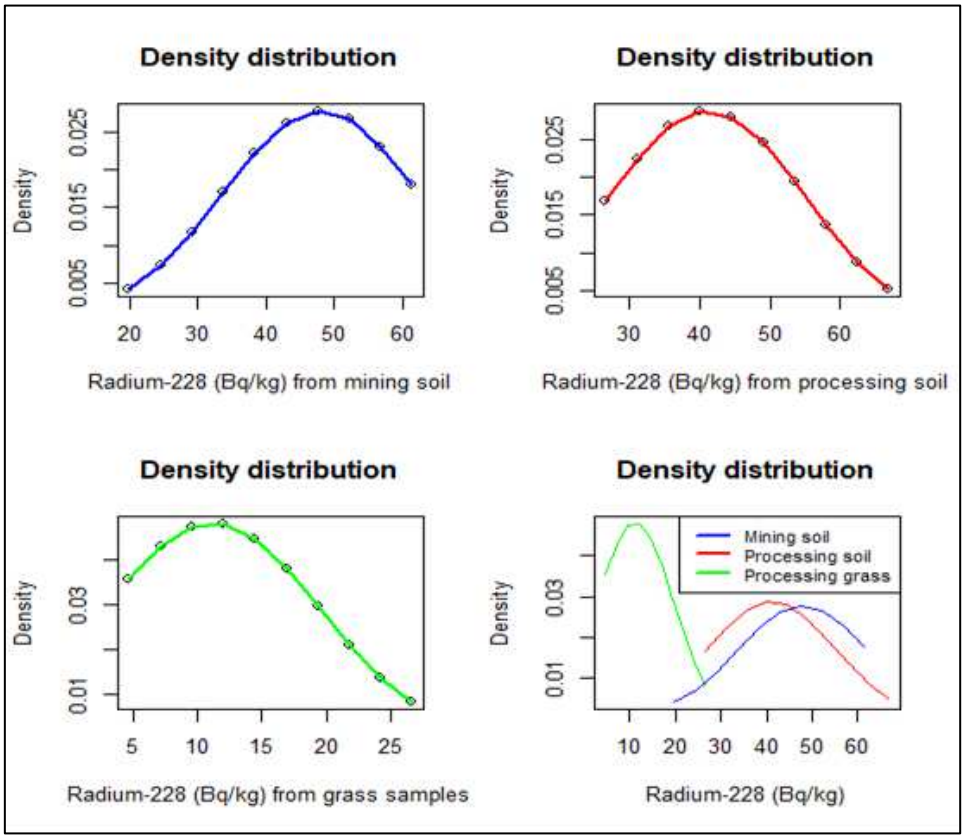

Figure 3: Density distribution of activity concentrations of ^{228}Ra in soil and grass samples from mining and processing sites of phosphate.

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

Statistics from Table 5 shows that the activity concentrations of ^{40}K in the dischargement waste site (Kpémé) soil is negatively skewed (-0.36); thus, the average activity concentration of ^{40}K in phosphate mining soil is less than the modal activity concentration, or soils that measured a relatively higher activity concentration of ^{40}K are more than those that measured a lower activity concentration (as shown in Figure 4). The distribution of activity concentrations of ^{40}K in phosphate mining site (Hahotoé) soil and in grass samples from dischargement waste site is positively skewed (1.02 and 0.18 respectively) (as shown in Figure 4); thus, the average activity concentration of ^{40}K in dischargement 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}K are more than those that measured higher activity concentrations. The negative kurtosis values (-0.56, -1.43, and -1.50 respectively) of both phosphate mining site (Hahotoé) soil and dischargement waste site (Kpémé) soil and grass samples show that the distribution of activity concentrations of ^{40}K is platykurtic. The term platykurtic describes a statistical distribution with extremely dispersed concentration of ^{40}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 Figure 4).

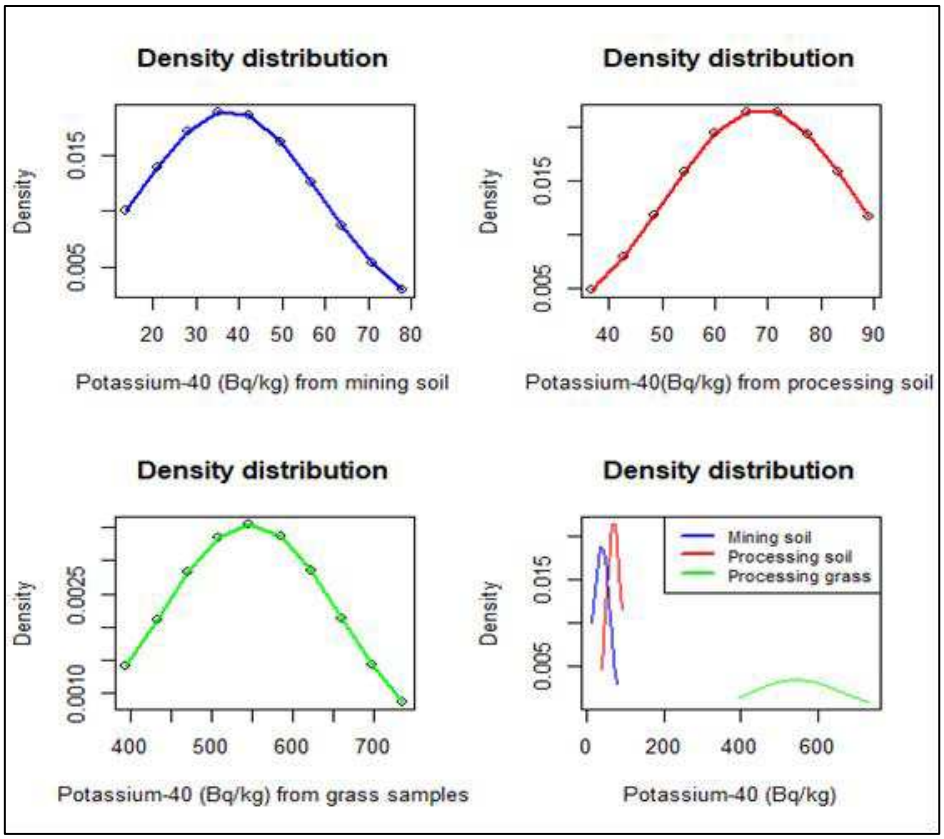

Figure 4: Density distribution of activity concentrations of ^{40}K in soil and grass samples from mining and processing site of phosphate.

3.3. Transfer factors of ^{226}Ra, ^{228}Ra, and ^{40}K from soil to grass in dischargement waste (Kpémé)

The TFs of ^{226}Ra, ^{228}Ra, and ^{40}K from the investigated phosphate dischargement waste soil (Kpémé) to grass using equation (**4**) are presented in Table 6. The average values of TF were 0.16, 0.32, and 8.42, respectively for ^{226}Ra, ^{228}Ra, and ^{40}K. The highest TF of (0.44) for ^{226}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 ^{228}Ra and ^{40}K, respectively, were found in grass samples from P3 and P10 respectively. The average TFs for all grass samples for ^{228}Ra and ^{40}K were 0.32 and 8.42, respectively. These results are higher than the results published by Hasan (Hasan et al., 2010) in Pakistan.

The obtained results display the highest transfer factor for potassium. This is probably explained by the use of potassium as a crop's fertilizer in the investigated area. However, high values of higher TF for potassium do not pose a risk to human health because ^{40}K has low contribution to internal dose as ^{40}K content in the human body is homeostatically controlled (IAEA, 1994; Mollah AS, 2014)). TFs for ^{226}Ra were found to be less than those of ^{228}Ra and ^{40}K in all grass samples in this study. This indicates that ^{226}Ra is less effectively transported from soil to grass than ^{228}Ra and ^{40}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; Mollah AS, 2014). For the investigated radionuclides, the literature usually presents the same range of TFs values obtained in the present study except for the ^{40}K, that are slightly lower than those get 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 Figure 2, Figure 3, and Figure 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, 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}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 finds by Mohannad (Mohannad et al., 2014) in Palestine and very 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 has been carried out, and some extra practices at operational processing sites.

This high transfer coefficient from soil to plan compared to other parts of the world can affect the animal grazing as cows that are later used for milk and meat production. Therefore, the transport of radionuclides from soil to grass (^{40}K > ^{228}Ra > ^{226}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 dried or raining season (IAEA, 2010).

Table 6: Transfer factors of ^{226}Ra, ^{228}Ra and ^{40}K in soil to grass samples from dischargement waste (Kpémé)

Codes	TFs estimation (unit of Bq kg^{-1} dry mass grass per Bq kg^{-1} dry mass soil)		
	FTs ± SD		
	^{226}Ra	^{228}Ra	^{40}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
AVR	0.16	0.32	8.42

TF: Transfer factor. AVR: Average

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

Countries	TFs estimation in the world (unit of Bq kg^{-1} dry mass grass per Bq kg^{-1} dry mass soil)						References
	^{226}Ra		^{228}Ra		^{40}K		
	Range	Average	Range	Average	Range	Average	
Pakistan	0.009 - 0.276	0.06	0.013 - 0.270	0.09	0.271 -0.317	0.28	(Hasan et al., 2010)
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.,2009)
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	**0.04 - 0.44**	**0.16**	**0.07 - 0.87**	**0.32**	**5.44 - 13.32**	**8.42**	**Present work**
IAEA	**0.01 - 1.00**	**0.11**	**-**	**-**	**0.49 - 5.60**	**1.4**	(IAEA, 1994)

Conclusion

The present study investigated the activity concentration of natural radionuclides in soils and grass samples from phosphate exploitations sites in Togo, dischargement waste site of Kpémé and phosphate mining site of Hahotoé. ^{226}Ra (^{238}U-series), ^{228}Ra (^{232}Th-series), and ^{40}K were found to be the main radiological constituents of soil and grass samples in the studied areas and ^{7}Be was found in small concentration but in the range measured in other countries worldwide. Statistical analysis was done 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 were investigated. The obtained data of TFs were within the range measured in other parts of the world except for that of ^{40}K. The high TFs for ^{40}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}K, it may slightly increase the activity concentrations of natural radionuclides. Its values are not sign of significant radiological alarm as ^{40}K is part of human/ animal body and contribute to maintaining their life on earth. The level of TFs for ^{40}K highlighted the necessity of performing further investigation about its transfer from soil to grass and the characteristic of the dischargement waste site.

Considerable variability was observed in activity concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K from all samples in both sites and all grass samples investigated. High level of activity concentrations in dischargement waste soil (Kpémé) in this study confirmed the previously reported values by Hazou (Hazou et al., 2019) let us 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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Conflicts of interest

Authors declare no conflicts of interest.

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Figures

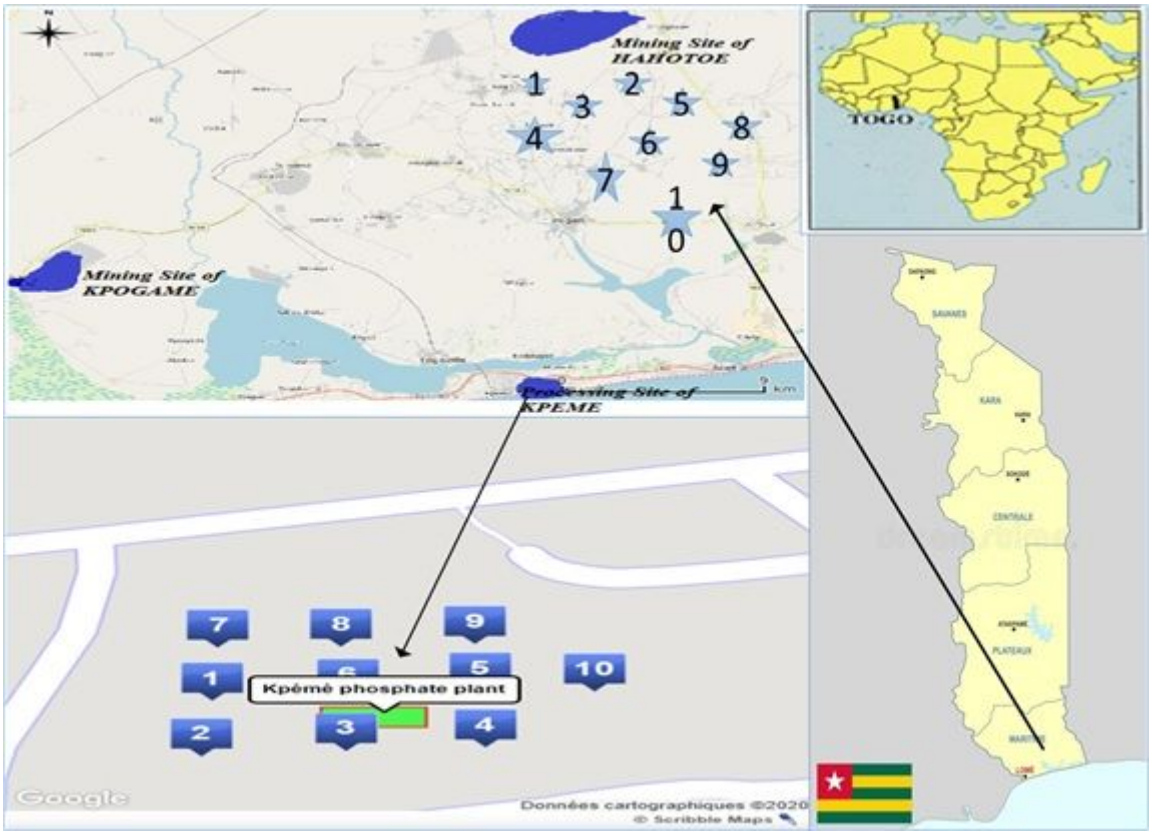

Figure 1

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

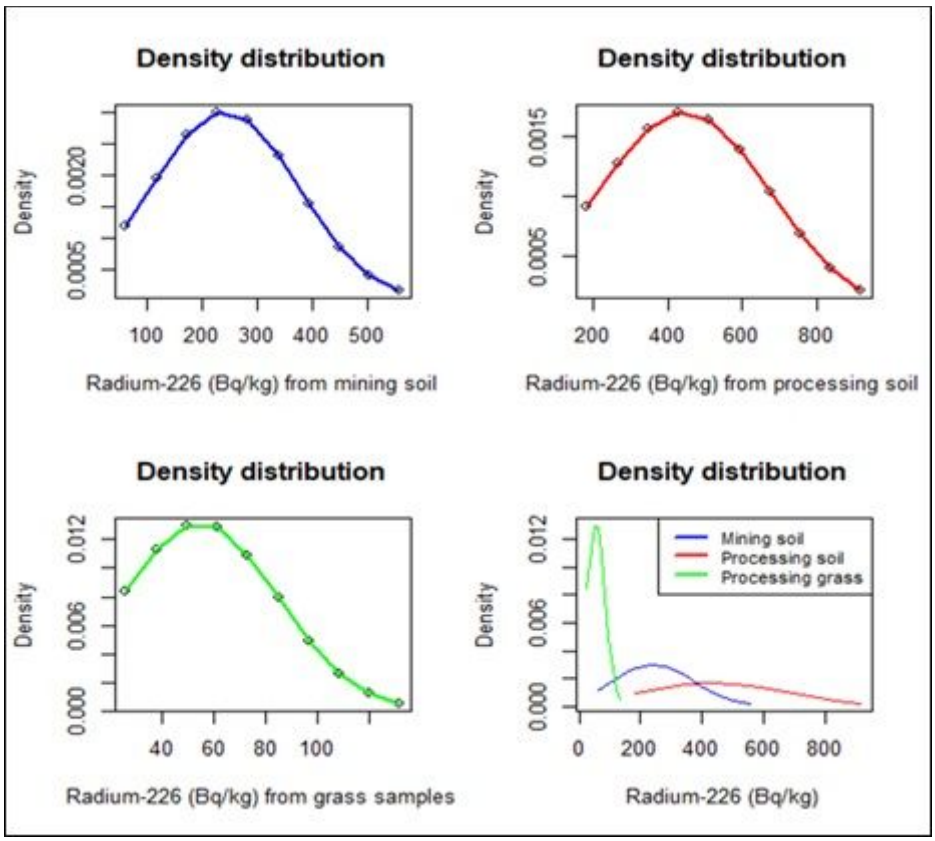

Figure 2

Density distribution of activity concentrations of 226Ra in soil and grass samples from mining and processing sites of phosphate.

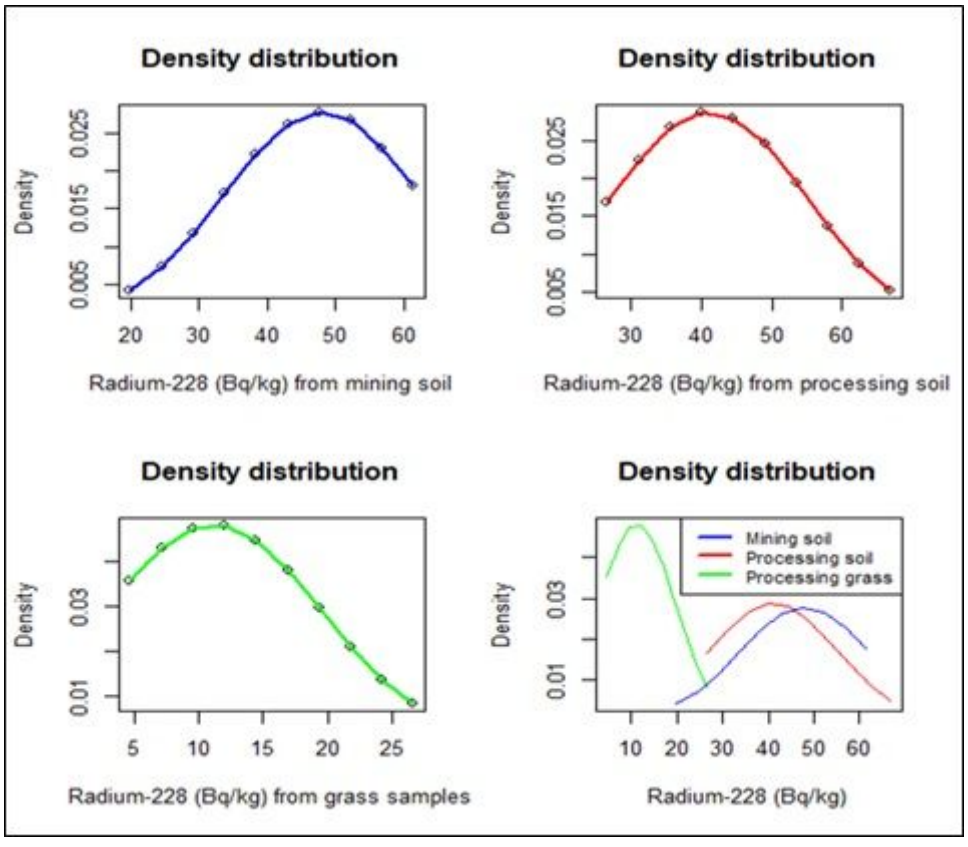

Figure 3

Density distribution of activity concentrations of 228Ra in soil and grass samples from mining and processing sites of phosphate.

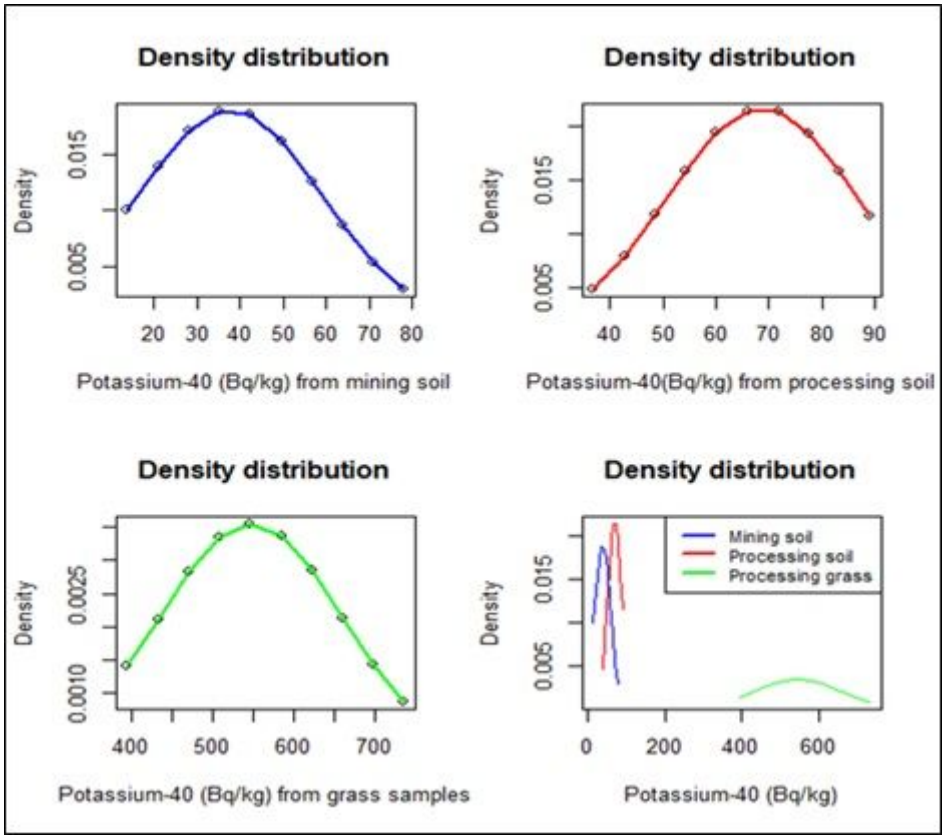

Figure 4

Density distribution of activity concentrations of 40K in soil and grass samples from mining and processing site of phosphate.