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Preliminary assessment of natural radioactivity and associated radiation hazards in a phosphate mining site in southern area of Togo

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Abstract

Introduction Because of the increasing use of phosphate in industries worldwide, especially in Togo, it is interesting to investigate the potential radioactivity exposure of phosphate ores, especially in the one being exploring in Togo nowadays. **Material and methods** The contents of natural radionuclides $(^{40}K, ^{226}Ra, ^{232}Th, ^{235}U$ and ^{238}U) were assessed in phosphate soil samples from Kpogamé, Dagbati and Kpémé in the maritime region of Togo by using gamma spectrometry-based Broad Energy Germanium detector (BEGe6530). Since no study was made prior to the exploitation, the samples from the control area of Anfoin-Kpota far away from the three others were considered as reference.

Results and discussion The results are discussed and compared with the data from other countries. The activity concentration of 40 K, 226 Ra, 232 Th, 235 U and 238 U are between (59.45 and 129.99), (20.19 and 779.93), (16.81 and 121.42), (2.26 and 52.03) and (16.66 and 841.14) Bq kg⁻¹, respectively. The values obtained shows that the exploitation sites (Dagbati and Kpogamé) and treatment site (Kpémé) have a very high level of radioactivity than the control area (Anfoin-Kpota). The Kpogamé and Dagbati exploitation and Kpémé waste discharging phosphate deposit sites were found to have higher activity concentration than many others exploited phosphate sedimentary deposits around the world. The average annual effective dose of the above studied sites is 0.36, 0.24 and 0.48 mSv year⁻¹, respectively. The value related to the discharge waste site is about 2% of the 1.0 mSv year−¹ recommended by the International Commission on Radiological Protection as the maximum annual dose to the public.

Conclusions The obtained result of both radioactivity and radiological level in the studied areas will be considered as a pre-operational baseline to estimate the possible radiological impacts due to mining and processing phosphate industrial activities.

Keywords Phosphate · Gamma spectrometry · BEGe6530 detector · Soil · Radiation hazard

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Introduction

Evaluation of radioactivity level in geological material is of great interest not only to control radiological exposure to naturally occurring radioactive material (NORM) for workers and public but lost for geochemist to characterize the type of rocks present in the study site [\[1](#page-7-0)[–4\]](#page-7-1). There are two main contributors to natural radiation exposures: high-energy incident cosmic ray particles on the earth's atmosphere and radioactive nuclides that originate from the earth's crust and are present throughout the environment, including the human body [\[4](#page-7-1)[–6\]](#page-7-2). In addition, industrial processes involving NORM can also carry an associated radiological risk and therefore identifying, quantifying, evaluating and managing any such risks are important nowadays [\[3,](#page-7-3) [7](#page-7-4)[–10\]](#page-8-0).

The natural radioactivity level in geological environment varies from one point to another. The natural radioactivity of geological sample such as soil, rock and sediment depends on their formation and transportation processes that were involved since soil formation; chemical and biochemical interactions influence the distribution patterns of uranium and thorium and their progenies $[4, 11-16]$ $[4, 11-16]$ $[4, 11-16]$. It results that thorium and uranium may be redistributed during igneous, sedimentary and metamorphic cycles of geological evolution, which might have resulted in small concentrations of deposits under favorable geological processes [\[17\]](#page-8-3). Study on the radioactivity levels and radionuclides distribution in soil provides vital radiological baseline information and understanding of the radiological implication of these elements due to the gamma-ray external exposure. Available information indicates that the deposits of monazite on the coastal areas (Kerala and Tamil Nadu) are formed due to the weathering of rocks (Western Ghats) [\[17\]](#page-8-3).

According to the estimation of IFG (International Fertilizer Group), the treatment of phosphate at Kpémé (southern Togo) emits about 3.5 million tons of phosphate mining waste (containing Cd and Pb) in the coastal waters of Togo [\[18\]](#page-8-4). During mining and processing of phosphate rock and soil, the waste containing radionuclides (uranium and thorium series) are discharged into the environment. The primary potential environmental radiation problem associated with phosphate mining and processing is related to processing waste products. While these materials do not present a direct radiation hazard, their use might create exposure problem: occupational exposure mainly occurs during mining, processing and transportation of phosphate-resulted product [\[4\]](#page-7-1).

Because of the increasing use of phosphate in industry worldwide, especially in Togo, it is interesting to investigate the potential radioactivity exposure of phosphate ores. The scope of the present study is to analyze the natural radionuclides in the Dagbati, Kpogamé and Kpémé phosphate exploitation and deposit sites in Togo and to assess the environmental implications resulting from mining and processing of the rock and soil phosphate. In addition, ^{226}Ra , 235 U, 238 U, 232 Th and 40 K activity concentrations were used to evaluate the potential radiation exposure to the occupational public and workers.

Materials and methods

Overview of the study area

of shale, phosphatic shale, quartz, limestone and phosphatic limestone. The relief is characterized by a vast plateau of bar ground. The zone is located on the coastal sedimentary basin of the Atlantic. Altitude of the study area varies between 51 and 75 m according to the position of landscape [\[19,](#page-8-5) [20\]](#page-8-6). This area of exploitation is discontinuous due to erosion and subdivided into three so-called spots, especially in the south the Aveta-Kpogamé spot; in the center the spot Hahotoé-Akoumapé; and in the north spot Dagbati (Table [1\)](#page-3-0) [\[18\]](#page-8-4).

Sample collection and preparation

Composites of eleven samples were randomly chosen from the four localities to cover the area and observe a significant local spatial variation in terrestrial radioactivity. Each composite sample was a mixture of five samples collected within an area of 5 m^2 . Each sampling point was marked using a Global Positioning System (GPS). Four samples were collected at the edges and one at the center. These five samples collected at a depth of approximately 20 cm from the top surface layer were mixed thoroughly to form a composite sample and packed into a polyethylene bag. The number of sample per site was dependent of the surface of the area and activity intensity. For that reason, 04 points were marked at Kpogame (20 samples), 03 at Kpeme (15 samples) and 02 at Dagbati (10 samples) and 02 at Afoin Kpota (10 samples). The vertical or near vertical surface was dressed to remove smeared soil. This was necessary to minimize the effects of contaminant migration interferences due to smearing of material from other levels.

Samples were labeled accordingly and were transferred to the laboratory. At the laboratory, the samples were prepared and stored in 120-ml cylindrical beakers for a period of thirty (30) days to reach secular equilibrium, following the rules describe by authors such as Guembou et al. [\[21\]](#page-8-7) and Dabayneh [\[22\]](#page-8-8). Sample mass measurements were made with the OHAUS brand scale which has an uncertainty of 0.1 g with 8.2 kg maximum scale.

Instrumentation and calibration of the detector

The activity concentration of ²²⁶Ra, ²³²Th, ⁴⁰K, ²³⁵U and 238U in soil samples was measured using a high-resolution gamma-ray spectrometric system. The system is consisted of a characterized broad energy germanium (BEGe) detector, BE6530 model, shielded with lead to reduce background. The active volume of the detector is about 6500 mm^3 with a relative efficiency of 60% at 1.33 MeV of 60 Co line [\[23\]](#page-8-9). The detector is placed in a low-level Canberra Model 747 lead shield with thickness of 10 cm. The resolution of this detector is 0.5 keV at 5.9 keV for 55Fe , 0.75 keV at 122 keV for 57 Co and 2.2 keV at 1332 keV for 60 Co as described by Guembou et al. [\[24\]](#page-8-10).

Fig. 1 Different sample sites: site of Kpogamé, site of Dagbati, site of Kpémé and site of Anfoin-Kpota

The energy calibration of the system was done using different point sources of 231 Am, 57 Co, 60 Co, 137 Cs, 55 Fe and ¹⁵⁵Eu. The absolute efficiency of the system was performed using ISOCS/LabSOCS mathematical calibration software inbuilt with Monte Carlo as described by Ndontchueng et al. [\[5,](#page-7-5) [6\]](#page-7-2) and Venkataraman et al. [\[25\]](#page-8-11). To avoid the influence of the decay scheme of nuclides, the sample geometry and composition, and the detector features on the activity concentrations of primordial radionuclides in sample, the cascade summing and auto-absorption effects were automatically corrected using the peak to total (P/T) curve and auto-absorption correction factors, respectively [\[26\]](#page-8-12). After measurement and subtraction of the background, the activity concentrations of the real sample were calculated.

Measurements of activity concentration and assessment of radiological parameters

• Activity concentration

Each sample was counted for 24 h, and spectra were analyzed using Genie 2000 software from Canberra Version V.3.2 based on the equation presented by Guembou et al. [\[21,](#page-8-7) [23\]](#page-8-9), Aoun et al. [\[27\]](#page-8-13).

Assuming a state of secular equilibrium between ²³⁸U and 232Th and their respective decay daughter products, the following relatively intense gamma-ray transitions were used to measure the activity concentrations for the above-mentioned radionuclides [\[5,](#page-7-5) [14\]](#page-8-14).

- (a) 226 Ra concentration was calculated as a weighted mean of the activity concentrations of the gamma rays of ²¹⁴Pb (295.1 keV, 351.9 keV), ²¹⁴Bi (609.3 keV and 1120.29 keV) and its specific gamma ray at 186.2 keV. Interference correction due to the presence of 185.7 keV energy peak of 235U has been taken into account and subtracted accordingly.
- (b) The gamma-ray photopeaks used for the determination of the 232Th contents were 338.4 keV, 911.2 keV and 969.11 keV of ²²⁸Ac and 238.6 keV of ²¹²Pb.

Table 1 Geographical coordinates of sampling points

- (c) 40 K was directly determined by using 1460.8 (10.7%) gamma ray.
- The radium equivalent activity (Ra_{eq})

Radium equivalent (Ra_{eq}) index in Bq kg⁻¹ is a widely used radiological hazard index. It is a convenient index to compare the specific activities of samples containing different concentrations of 226 Ra, 232 Th and 40 K. It was defined on the assumption that 10 Bq kg⁻¹ of ²²⁶Ra, 7 Bq kg⁻¹ of ²³²Th and 130 Bq kg⁻¹ of ⁴⁰K produce the same gamma dose rate. It is calculated using Eq. (1) [\[28\]](#page-8-15):

$$
Ra_{eq}(Bq kg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.07A_k
$$
 (1)

where A_{Ra} , A_{Th} and A_k are the activity concentrations of 226 Ra, 232 Th and 40 K in Bq kg⁻¹.

• External gamma dose rate

In order to assess radiological risk, external exposure to radiation arising from naturally occurring radionuclides can be determined in terms of the absorbed dose rate in air at 1 m above the ground surface. The mean activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K (in Bq kg⁻¹) in soil samples were used, applying the conversion factor given by UNSCEAR [\[4\]](#page-7-1) and Beretka and Mathew [\[28\]](#page-8-15) as follows:

$$
D_{\gamma}\left(\text{nGy h}^{-1}\right) = 0.462A_{\text{Ra}} + 0.604A_{\text{Th}} + 0.0417A_{\text{k}} \qquad (2)
$$

where D_{γ} is the absorbed dose in nGy h⁻¹.

• Annual effective dose associated with the radioactivity of the mine waste.

To assess exposure to radiation due to naturally occurring radioactive material from the studied soil samples, external annual outdoor equivalent effective dose was estimated and expressed in mSv year^{-1}. The conversion coefficients from absorbed dose in air to effective dose received by adults considered was that of UNSCEAR [\[4\]](#page-7-1).

$$
(AEDR)_{out} = (nGy h^{-1}) \times 0.7 (Sv Gy^{-1})
$$

× 0.2 × 8760 (hy⁻¹) × 10⁻⁶ (3)

The values of those parameters used in the UNSCEAR report [\[4\]](#page-7-1) are 0.70 Sv Gy⁻¹ for the conversion coefficient from absorbed dose in air to effective dose received by adults, 0.20 for the outdoor occupancy factor and 8760 is the conversion factor for year to hours (number of hours per years).

Results and discussion

Specific activity concentration

Evaluation of primordial radionuclide of 232 Th, 40 K, 238 U, ²³⁵U and ²²⁶Ra (daughter radionuclide of ²³⁸U) was carried out in soil samples of the two phosphate exploitation sites in Togo (Kpogamé and Dagbati) and the discharge waste site during treatment (Kpémé) using a characterized gamma-ray spectrometry-based broad energy germanium (BEGe6530) detector. In addition, soil samples from the control area of Anfoin-Kpota were also analyzed and used as reference to evaluate contamination of the population living around the phosphate exploitation site. To determine specific activity of the primordial radionuclide in soil sample as seen in Table [2,](#page-4-0) efficiency calibration of the broad energy germanium detector was done using LabSocs mathematical software inbuilt site

Table 2 Mean values of activity concentrations of ²²⁶Ra, ²³²Th, ⁴⁰K, ²³⁸U and ²³⁵U of radionuclides in the different zones (1st table for individual sample per site and in the 2nd table, the range and average values per site)

Sampling Number of Specify activity (Bq kg−1)

sampling point

with Monte Carlo method. It can be seen that specific activity of the primordial radionuclide in soil from the studied sites varies from one site to another. This variation in specific activity concentration is associated to the non-uniform distribution of naturally occurring radioactive material in the environment.

The average specific activity of $226Ra$, $232Th$, $40K$ and 238U was 215.91, 121.42, 78.23 and 300.88 for Kpogamé phosphate site, 367.04, 54.39, 129.99 and 439.78 for Dagbati phosphate site and 779.93, 82.94, 101.23 and 841.14 for Kpémé discharge waste site, respectively. The average activity concentrations of ²²⁶Ra, ²³²Th, ⁴⁰K and ²³⁸U in the studied soil of Anfoin-Kpota site used as reference data were 20.19, 16.80, 59.45 and 16.66 Bq kg⁻¹, respectively. Following the observed average specific activity of the primordial radionuclide in the soil of both phosphate exploitation and the discharge waste site as presented in Table [2,](#page-4-0) the average specific activity of 238 U was observed to be higher followed

by 226Ra. This is obvious that the main radioactivity content in soil from phosphate mining zone is due to 238 U and its decay products [\[29\]](#page-8-16). In addition, variation in activities of 238 U and its decay product of 226 Ra in the investigated soil of phosphate mining exploitation sites and the treatment site could be associated to the high percentage of P_2O_5 in the phosphate ore and the uranium solubility under oxidizing conditions [\[29–](#page-8-16)[32\]](#page-8-17). This variation in specific activity of ²²⁶Ra and ²³⁸U as compared to that of ²³²Th and ⁴⁰K may be associated to the type of rocks in the studied sites. The geological structure of the investigated sites (both phosphate mining exploitation sites and the discharge waste) is consisting of shale, phosphatic shale, quartz, limestone and phosphoric limestone $[20]$. The ²³⁵U average specific activity observed in soil of both phosphate exploitation sites and the discharge waste site were lower than that of 238 U. These low activity concentrations of 235 U are connected with the abundance of

both isotopes in the entire earth, i.e., 235 U (0.72%) and 238 U (99.27%) [\[2\]](#page-7-6).

Radioactivity level of the primordial radionuclide in soil of both Dagbati and Kpogamé phosphate mining sites is slightly different even though the geological structure is the same. This difference is associated to the non-uniform distribution of radioactivity in the environment [\[4,](#page-7-1) [5\]](#page-7-5). The observed radioactivity level in soil of the discharge waste site is relatively higher compared to both phosphate mining sites (Fig. [2\)](#page-6-0). In the purification process, the changes in radioactivity contents could be due to mechanical separation of radionuclides between the wet rocks and the different rejects and/or solubility of radionuclides [\[29,](#page-8-16) [33\]](#page-8-18).

The low level of radioactivity in soil of Anfoin-Kpota is obvious because this study is located out of the phosphate mining and exploitation sites. As a result, no contamination of this site can be observed as far as the activity concentrations of 226 Ra, 232 Th, 40 K and 238 U obtained are comparably lower than the worldwide average values.

Table [3](#page-6-1) presents the comparison of radioactivity levels of primordial radionuclide of ²²⁶Ra, ²³²Th, ²³⁸U and ⁴⁰K observed in the investigated soil samples from both phosphate ores deposit and the discharge waste with the available data from other countries and the reported range of the International Atomic Energy Agency in its technical report 419 series [\[8\]](#page-8-19). The type of phosphate in the studied sites is sedimentary phosphate with relatively high level of naturally occurring radioactive materials. The observed average values of 226Ra and 238U were comparably varied with the reported range of IAEA [\[8\]](#page-8-19) for different phosphate types.

The observed average activity concentrations of $226Ra$ in soil of the phosphate mining exploitation sites (Kpogamé and Dagbati) are lower than those reported in the soil of some other countries, with the exception of Finland and Egypt, reported by Uosif and El-Taher [\[34\]](#page-8-20) and Khan et al. [\[35\]](#page-8-21) as shown in Table [3.](#page-6-1) The observed 232 Th-specific activity in soil of Kpogamé and Dagbati is relatively higher that those reported values in some other countries as seen in Table [3](#page-6-1) except that of Tanzania published by Makweba and Holm [\[38\]](#page-9-0). The reported values of 40 K in soil of the studied sites are within the reported average values published by researchers in some other countries. Similar comparison was done with the obtained average values of primordial radionuclide in soil of the discharge wastes site of Kpémé with the reported values in some other countries as seen in Table [3.](#page-6-1) It was noticed that the observed average values of ^{226}Ra , ^{232}Th , ^{238}U and 40 K vary within the reported values. It is widely believed that the radioactivity related with phosphate rocks of sedimentary origin is formed by adsorption and co-precipitation of uranium with calcium [\[29,](#page-8-16) [31\]](#page-8-22). This variation in terms of activity concentration of the primordial radionuclide as compared to other reported values is associated to the source of phosphate rock, the depth of sampling and the migration rate (erosion) of radionuclide in soil [\[39–](#page-9-1)[41\]](#page-9-2).

Radiological study

To assess radiological exposure to natural occurring radioactive materials (NORMs) in soil of the studies sites, radiological safety parameters such as radium equivalent activity, the radiation-absorbed dose rate in air and the external exposure outdoor equivalent effective dose are estimated based on some assumptions as defined by UNSCEAR [\[4\]](#page-7-1) and reported in Table [3.](#page-6-1) As seen in Table [3,](#page-6-1) the average radium equivalent activity in soil from the studied sites was 395.57 Bq kg⁻¹ for Kpogamé phosphate site, 454.83 Bq kg−¹ for Dagbati, 906.33 Bq kg−¹ around the plant of Kpémé site and 48.80 Bq kg−¹ for the reference site of Anfoin-Kpota. The observed values of radium equivalent activity in both phosphate mining deposit sites and the treatment plant site are comparably higher than the reported average worldwide values of 370 Bq kg−¹ while the observed values in soil from Anfoin-Kpota known as reference site is approximately eight times lower than the recommended safe values of UNSCEAR [\[4\]](#page-7-1). The Ra_{eq} values were much higher in the terrestrial environment, mainly because of the phosphates rocks, which is rich in potassium and contains significant concentrations of the uranium, thorium, radium and their decay products [\[29,](#page-8-16) [30\]](#page-8-23).

The estimated gamma-absorbed dose expressed in $nGy h^{-1}$ received by the workers ranged from 52.35 to 538.22 with an average of 175.95 in Kpogamé phosphate mining site, from 141.73 to 254.97 with an average of 198.35 in Dagbati phosphate exploitation site, between 357.96 and 417.64 with a mean of 392.31 in Kpémé treatment plant and discharge site and from 20.11 to 24.52 with a mean of 22.32 in Anfoin-Kpota known as a reference site. These values are comparably higher than the recommended safe value of 90 nGy h−1, except the reference site of Anfoin-Kpota.

According to UNSCEAR [\[4\]](#page-7-1), on technologically enhanced naturally occurring radioactive material, workers in phosphate mines and treatment site are being exposed to radiation via two pathways such as external exposure to gamma radiation and inhalation exposure due to radon daughters associated with dust particles. The focus in this study was on the external gamma radiation exposure, considering that workers are exposed to both outdoor and indoor for a working period of 1820 h per year. The estimated average external exposure outdoor annual equivalent effective dose expressed in mSv year⁻¹ received by workers in the phosphate mines and the treatment site was 0.36, 0.24 and 0.48 for Kpogamé, Dagbati and Kpémé site, respectively (Table [4\)](#page-6-2).

Fig. 2 Comparison of the mean specific gamma activities of Ra-226, Th-232, K-40, U-238, U-235 and Raeq in soil from different studied sites

Table 3 Activity concentration of 226Ra, 232Th, 40K, (Bq kg−1) and radium equivalent Raeq (Bq kg−1) in phosphate rocks from different countries

KPOGAME (exploitation site) DAGBATI (exploitation site) KPEME (dischargement waste) ANFOIN-KPOTA (contrôle site)

A(Ra-226) A(Th-232) A(K-40) A(U-238) A(U-235)Raeq (Bq/Kg)

NF denotes to data not found in this reference

Table 4 Mean values of radiation hazard parameters of phosphate from Kpogamé, Dagbati, Kpémé and Anfoin-Kpota, Togo

These observed average values are comparably lower than the reported safe limit of 1 mSv year−¹ for the public. As shown in Fig. [3,](#page-7-7) it can be seen that the estimated annual equivalent effective dose is comparably far below the recommended safe values for workers in which there is no threshold below which there would be no effect [\[42\]](#page-9-3). However, any additional exposure source to ionizing radiation may increase in the chance of the health effect. The observed total annual effective dose

presents no significant radiation dose to staff from external gamma radiation in soil.

The observed total annual equivalent effective in soil of the reference site of Anfoin-Kpota is comparably lower than the recommended value of 1 mSv year⁻¹ [\[4\]](#page-7-1). This means no contamination as a result of mining is observed in the Anfoin-Kpota zone used as reference site in this investigation.

Conclusion

The activity concentration of 238 U, 235 U, 226 Ra, 232 Th and 40 K has been measured in soil samples from phosphate mining areas in Togo using gamma spectrometry-based broad energy germanium (BE6530) detector. A total of two exploitation sites, one discharge waste site and one reference site, have been assessed. The recorded mean values of 226 Ra and 232 Th are both higher than the worldwide average reported by UNSCEAR in all the investigated sites except that of the reference site, but lower than the reported value of mineral phosphate ore deposit zone in many countries reported in the IAEA technical report [\[8\]](#page-8-19). Only activity concentration of 40 K is lower than the worldwide average value of 370 Bq kg^{-1} . Considering the non-uniform distribution of radioactivity in soil matrix, rocks and sediment, the radium equivalent activity was calculated and observed to be comparably higher than the recommended safe value of 370 Bq kg⁻¹ reported by UNSCEAR. Both activity concentration of investigated radionuclides and radium equivalent activity calculated in this work are in the range of the values obtained in other countries by studying phosphate mining exploitation site.

Absorbed gamma dose rate and the corresponding external annual effective dose (outdoor) were estimated to assess external exposure to gamma radiation from soil in the study area. The obtained values were comparably higher than the recommended safe limits by UNSCEAR except those values related to the control site. Nevertheless, the obtained average values of external annual effective dose (outdoor) are lower than the recommended safe professional exposure limit of 20 mSv year⁻¹ [\[4\]](#page-7-1), which outcomes the interpellation of the regulatory authority for public and professional exposure on the investigated area. This work is a preliminary investigation on these areas of phosphates mining in Togo. It is expected that the results obtained may be used as baseline data for future work in the area of Kpogamé and Dagbati, the exploitation sites, and Kpémé, the discharging waste site.

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