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Dose estimates to the public due to ^{210}Po ingestion via cocoa powder from Lolodorf high background radiation area, Cameroon

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Abstract – ^{210}Po activity concentrations have been measured in Lolodorf high background radiation area in cocoa beans which are hand-processed into cocoa powder for breakfast purposes to estimate radiological dose to human. ^{210}Po has been also measured in cocoa leaves and compared to the cocoa beans ^{210}Po content. The analysis has been carried out by CANBERRA alpha spectrometry using ion-implanted silicon detectors. ^{210}Po activity concentrations in cocoa beans varied from 2.31 ± 0.23 to $8.09 \pm 0.56 \text{ Bq.kg}^{-1}$, while these values varied from 21.7 ± 0.87 to $66.67 \pm 1.58 \text{ Bq.kg}^{-1}$ in cocoa leaves. The corresponding mean values are 4.96 ± 1.86 and $42.54 \pm 16 \text{ Bq.kg}^{-1}$ on a dry weight basis respectively. The obtained values confirm the fact that ^{210}Po activity concentrations in cocoa leaves are high compared to the cocoa beans due to the deposition of ^{222}Rn daughters in the atmosphere. The mean radiological doses to human were founded to be 0.227, 0.134, 0.083 and 0.062 mSv/year for children 2- to 7-year-olds, 7- to 12-year-olds, 12- to 17-year-olds and for adult respectively. Ingestion of cocoa powder by the most exposed group ages (children) might not exceed the recommended dose limit for members of the public, which is 1 mSv/year.

Keywords: ^{210}Po / activity concentrations / radiological dose / cocoa beans / cocoa powder

1 Introduction

There are seven polonium isotopes naturally present in the environment: ^{210}Po , ^{214}Po and ^{218}Po of the ^{238}U decay series; ^{212}Po and ^{216}Po of the thorium decay series; and ^{211}Po and ^{215}Po of the ^{235}U decay series. ^{210}Po has a half-life of approximately 138 days, which is long enough to play a significant role in many environmental processes. All of the other naturally occurring isotopes have half-lives of only 3 minutes or less.

^{210}Po is an alpha emitting radionuclide with no radioactive progeny and produces only very-low-intensity gamma rays at very low abundance; thus, the dose largely arises from internal exposure. The main reasons for its radiological importance are its relatively high activity concentrations in certain foods and its relatively high ingestion dose coefficient. Radiation doses from ^{210}Po arise owing to natural occurrences of the radionuclide as well as to human activities (IAEA, 2017). As ^{210}Po is part of the ^{238}U decay series, it is naturally occurring and is found in varying amounts worldwide. The analysis of the radionuclide contents of foods and water, along with bioassay data and knowledge of the metabolic behaviour of the radionuclides, provides an alternative basis for dose estimation (UNSCEAR, 2000). ^{210}Po contributes a substantial portion

of the radiation dose to human. According to Bulman et al., the dose due to ingestion of ^{210}Po was about 7% of the natural internal radiation dose (Bulman et al., 1995). The main source of ^{210}Po in the environment is ^{222}Rn gas which diffuses into the atmosphere from rocks and soil where it ultimately decays to ^{210}Pb , ^{210}Bi and then to ^{210}Po in the atmosphere. ^{210}Po attaches itself further electrostatically to aerosol particles and are transported back to earth's surface to soil, plant and aquatic environments by dry deposition and wash out (Santos et al., 1990; Pietrzak-Flis and Skowronska-Smolak, 1995; Karali et al., 1996). Consumption of food is usually the most important route by which natural radionuclides can enter the human body and assessment of their levels in different foods is therefore important to estimate the intake of these radionuclides by man.

Ngombas and Melondo, which are two localities of Lolodorf subdivision of the south region of Cameroon, were considered as study areas. According to Chapaud in its work performed in 1966 on Cameroon's cocoa economy, it has been established that south region of Cameroon is the main cocoa production area (Champaud, 1966). Since cocoa is a fragile tree, it supports neither too low temperatures nor too strong, intensive zone for cocoa activities in Cameroon is a relatively narrow band of 50 to 70 km wide which widens in the south in the departments of Ntem and Dja-et-Lobo, and towards Lolodorf. The optimum temperature seems to be around 27 °C;

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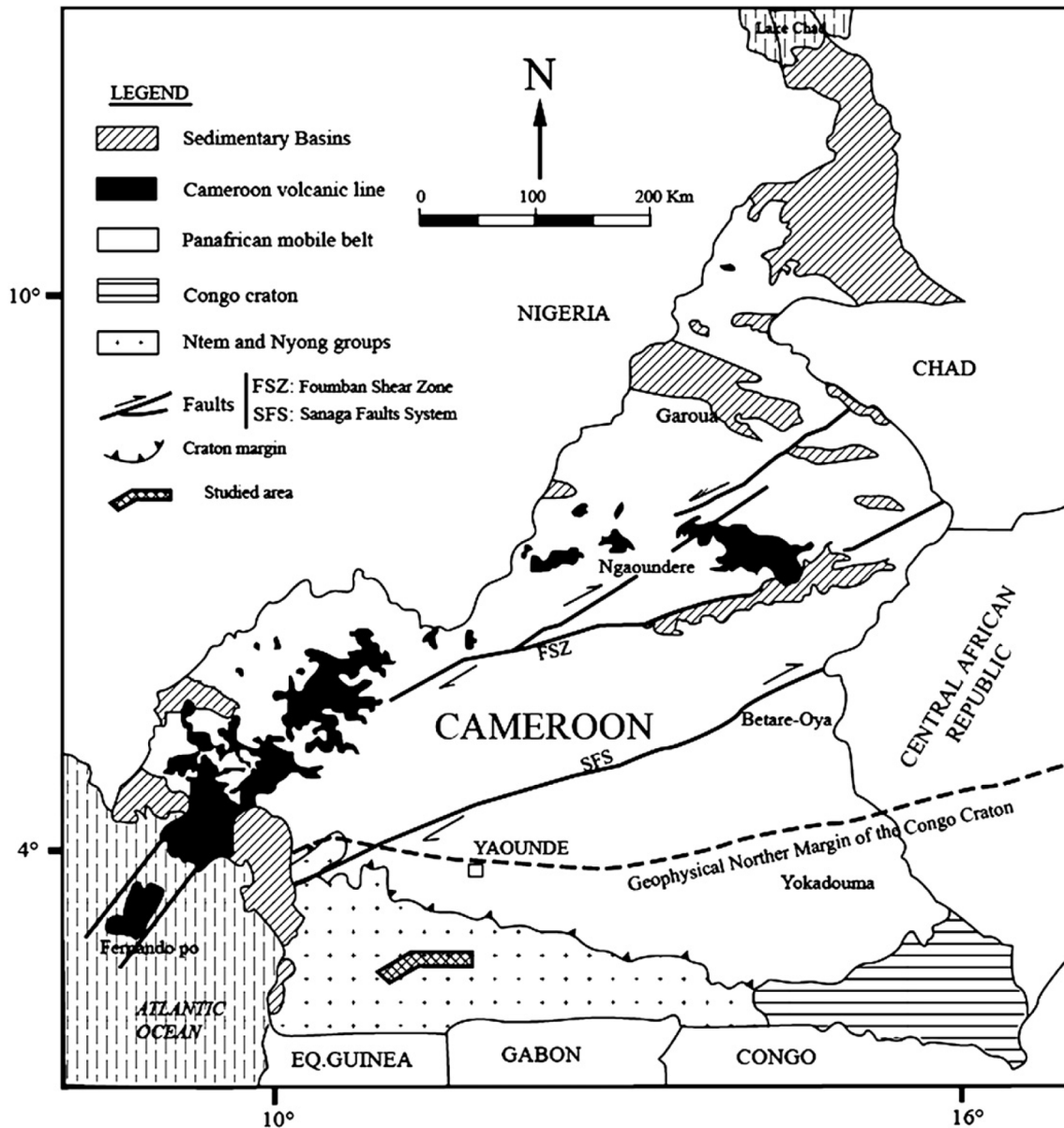


Figure 1. Geological map of Cameroon.

it cannot tolerate temperatures higher than 32°C , nor those lower than 15°C (Champaud, 1966). The entire population lives from cocoa production which is hand-processed into cocoa powder for local consumption and on the other hand involved in world trade for the manufacture of the chocolate. Between 1978 and 1985, this subdivision has been identified as uranium ores deposits by French Office of Geological and Mining Research (Maurizot *et al.*, 1986). For this previous investigation which consisted of helicopter-borne radiometric survey was to evaluate the mineral potential of the region. Recently, gamma spectrometry has been performed to determine natural radioactivity in soil and rocks samples from this area (Ele Abiama *et al.*, 2010; Beyala Ateba *et al.*, 2010, 2011, 2017; Ben-Bolie *et al.*, 2013; Saïdou Shinji *et al.*, 2015). These studies identified Lolodorf as High Background Radiation Area (HBRA). According to Beyala Ateba *et al.*, study made at Ngombas and Bikoe gives the mean activity

concentrations of $1482 \pm 280 \text{ Bq.kg}^{-1}$ for ^{40}K , $134 \pm 64 \text{ Bq.kg}^{-1}$ for ^{226}Ra and $177 \pm 102 \text{ Bq.kg}^{-1}$ for ^{232}Th in soil and the average outdoor absorbed dose rates in air, 1 m above the ground surfaces, were estimated to be 218 ± 61 and $250 \pm 97 \text{ nGy.h}^{-1}$ in the locations of Ngombas and Bikoue, respectively (Beyala Ateba *et al.*, 2010).

Mvondo *et al.* (2017) studied the soil-fern transfer of naturally occurring alpha emitting radionuclides in the southern region of Cameroon. This study concluded the fact that the study area is a high background radiation area. Figures 1 and 2 show the study area position in Cameroon map and radiometric map of both localities.

Since ^{210}Po is considered to be one of the most important environmental radionuclide due to its wide distribution and potential for human radiation exposure through ingestion and inhalation (Martin and Ryan, 2004), the main objective of the study is to measure ^{210}Po activity concentrations in cocoa

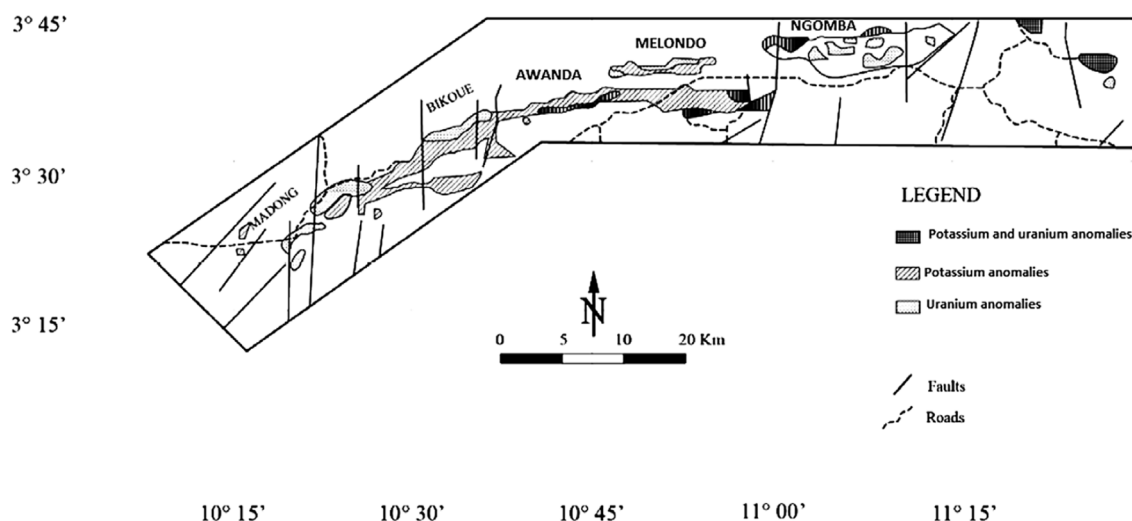


Figure 2. Geological and radiometric anomalies of Lolodorf locality.

beans and leaves and estimate radiological dose to human due to the consumption of the cocoa powder in the study area.

2 Materials and methods

2.1 Sampling collection and preparation

Samples have been collected by 10 points over an area of approximately 1 hectare for both localities. The present study was conducted on adult cocoa trees with an average height of 2 m. Most branches are close to the ground to collect the leaves without climbing. For a given tree, about a number of 10 wet leaves and 02 fruits were collected per leaves and beans sample respectively. Mature fruits were cut from the stems and split with a machete to extract the beans.

Cocoa beans and leaves sample were collected, washed, frozen and lyophilized for a week in a lyophilizer at -40°C . After lyophilization, the dried samples were ground into powder and homogenized.

2.2 Polonium analysis

The preparation procedure has been followed according to Mvondo *et al.* One hundred milligrams of dry weight of each powder sample was spiked in a beaker and 0.5 mL solution of ^{209}Po as yield tracer is added (Mvondo *et al.*, 2017). Each sample was digested for overnight using a mixture of 100 mL of 65% nitric acid and 50 mL of 37% hydrochloric acid (aqua regia) and adding 0.2 mL of octanol to facilitate the digestion of organic material. After digestion, the mixture is gradually evaporated on a hot plate at 200°C , occasionally adding concentrated nitric acid and a few drops of hydrogen peroxide (H_2O_2) to help oxidizing the organic compounds. After complete evaporation, the residue was dissolved and diluted with concentrated hydrochloric acid and evaporated again to completely remove nitric acid. The residue is dissolved in concentrated hydrochloric acid with 50 mL of distilled water mixed with 10 mL of ascorbic acid for iron reduction prior to overnight deposition of ^{210}Po on a silver disc under magnetic

stirring. Polonium is deposited onto silver foil 99.9% pure, cut in the laboratory with an extruder in 24 mm diameter discs. Discs are used once and discarded.

2.3 Alpha counting

Measurements were done using CANBERRA alpha spectrometer with ion-implanted silicon detectors. The acquisition of the spectrum in the computer was made using MAESTRO software. The detector efficiency was previously performed using a reference source which consists of a mixture of ^{237}Np ($T_{1/2} = 2.14 \times 10^6$ years), ^{241}Am ($T_{1/2} = 433.176$ years) and ^{244}Cm ($T_{1/2} = 18.1$ years) whose activities are known at the measurement date.

3 Results and discussion

3.1 Activity concentrations of ^{210}Po in cocoa beans and leaves

Analytical results were obtained with the efficiency detectors range between 20 and 30% and with chemical yield of 80% approximately. Table 1 presents activity concentrations of ^{210}Po in cocoa beans and leaves. ^{210}Po activity concentrations in cocoa beans varied from 2.31 ± 0.23 to $8.09 \pm 0.56 \text{ Bq.kg}^{-1}$, while these values varied from 21.7 ± 0.87 to $66.67 \pm 1.58 \text{ Bq.kg}^{-1}$ in cocoa leaves on a dry weight basis. The corresponding mean values are 4.96 ± 1.86 and $42.54 \pm 16.21 \text{ Bq.kg}^{-1}$ respectively. The obtained values confirm the fact that ^{210}Po activity concentrations in cocoa leaves were higher than in the cocoa beans due to the deposition of ^{222}Rn daughters in the atmosphere. The contamination of vegetation by the ^{210}Po is largely by deposition on the leaf, which depends on parameters such as growing season rainfall, and size and morphology of leaves (Francis *et al.*, 1968; Skwarzec *et al.*, 2001).

It also appears in Table 1 that ^{210}Po activity concentrations in the cocoa samples in Ngombas are relatively high compared to those measured in Melondo due to the fact that soil activity concentrations in Ngombas are high. The previous studies

Table 1. ^{210}Po activity concentrations in cocoa beans and cocoa leaves.

Sampling Locality	Location ID	^{210}Po activity concentration in the samples (Bq/kg dry weight)	
		Cocoa beans	Cocoa leaves
Melondo	M1	2.31 ± 0.23	21.74 ± 0.87
	M2	4.63 ± 0.42	31.79 ± 1.12
	M3	4.22 ± 0.4	41.54 ± 1.37
	M4	3.42 ± 0.36	23.74 ± 0.77
	M5	3.87 ± 0.33	33.92 ± 0.97
Ngombas	N1	4.88 ± 0.39	66.67 ± 1.58
	N2	8.09 ± 0.56	66.53 ± 1.56
	N3	6.41 ± 0.47	57.48 ± 1.46
	N4	4.12 ± 0.38	40.07 ± 1.02
	N5	7.69 ± 0.47	41.89 ± 1.22
Maximum	–	8.09 ± 0.56	66.67 ± 1.58
Minimum	–	2.31 ± 0.23	21.74 ± 0.87
Mean	–	4.96	42.54
Standard deviation	–	1.86	16.21

Table 2. ^{210}Po soil-cocoa beans and ^{210}Po soil-cocoa leaves transfer factors (in kg/kg on a dry weight basis) at each sampling point.

Sampling locality	Location ID	^{210}Po activity concentration in Soil	Soil-cocoa beans	Soil-cocoa leaves
			transfer factor	transfer factor
Melondo	M1	81 ± 7	0.0285	0.2684
	M2	152 ± 11	0.0305	0.2091
	M3	176 ± 15	0.0240	0.2360
	M4	112 ± 9	0.0305	0.2120
	M5	132 ± 8	0.0293	0.2570
Ngombas	N1	246 ± 24	0.0198	0.2710
	N2	231 ± 18	0.0350	0.2880
	N3	251 ± 23	0.0255	0.2290
	N4	189 ± 12	0.0218	0.2120
	N5	224 ± 16	0.0343	0.1870
Maximum	–	251 ± 23	0.0350	0.2880
Minimum	–	81 ± 7	0.0198	0.1870
Mean	–	179.4	0.0279	0.2370
Standard deviation	–	59.2	0.0051	0.0329

based on gamma ray spectrometry established high ^{238}U and ^{226}Ra in soil samples of Ngombas subdivision (Beyala Ateba *et al.*, 2010, 2011; Ele Abiama *et al.*, 2010). According to Coppin and Roussel-Debet (2004), the global range of activity concentrations of ^{210}Po in plants varied from 0.1 to 160 Bq/kg (Coppin and Roussel-Debet, 2004). The values obtained for cocoa beans and leaves in this study are in this range. In addition, the relative importance of exposure pathways depends on the concentration of the radionuclides in the soil, the soil-plant transfer factors (TF) and the rate of deposition onto plant parts above ground.

According to the study of Mvondo *et al.* (2017) on soil-fern transfer of naturally occurring alpha emitting radionuclides in the southern region on Cameroon, where activity concentrations in soil of different alpha emitting radionuclides are known, soil-cocoa transfer factors of ^{210}Po have been determined. Table 2 presents soil-cocoa beans and soil-cocoa leaves transfer factors. Soil-cocoa beans transfer factors varied from 0.0198 to 0.0350, while soil-cocoa leaves transfer factors varied from 0.187 to 0.2880. The corresponding mean values are 0.0279 and 0.237 respectively. Soil-cocoa leaves transfer factors are 10 times high compared to soil-cocoa beans transfer factors. Transfer factors (TFs) range across orders of magnitude, depending on the plant and soil type variations assessed (IAEA, 2010). It should be noted that the radioactivity in the plant is not only acquired through root transfer. The above ground biomass might also be contaminated because of resuspension or direct deposition of radionuclides from the atmosphere (Pietrzak-Flis and Skowronska-Smolak, 1995).

3.2 Estimation of radiological dose to human

Based on the collaboration of several families regularly consuming cocoa powder in their breakfast, a consumption rate of 1 kg/week of cocoa powder for a family of about 5 persons has been assumed. The ingestion doses were estimated using

the activity concentration of ^{210}Po determined in cocoa beans and the appropriate dose conversion factor recommended by ICRP (ICRP, 1996). Table 3 shows radiological dose to human due to the consumption of cocoa powder in Lolodorf HBRA. The mean radiological doses to human were founded to be 0.227, 0.134, 0.083 and 0.062 mSv/year for children 2- to 7-year-olds, 7- to 12-year-olds, 12- to 17-year-olds and for adult respectively. Ingestion of cocoa powder by the most exposed group ages (children) might not exceed the recommended dose limit for members of the public, which is 1 mSv/year. The values from the present study were relatively high compared to those obtained from study performed in Qena city, Egypt by Salahel Din in 2011 which revealed that the annual dose received by the general public via ingestion of ^{210}Po varied from 0.008 to 38.3 $\mu\text{Sv}/\text{year}$ (Salahel, 2011). Similar study focused on dose estimates to the public from ^{210}Po ingestion via dietary sources at Kalpakkam, India reported values from 0.08 to 128 $\mu\text{Sv}/\text{year}$ (Kannan *et al.*, 2001). These results are in the same range with those obtained in the present study. In 2014, Carvalho *et al.*, in their study on intake of radionuclides with the diet in uranium mining areas, confirmed the fact that the largest contribution to radiation dose from the diet comes from ^{210}Po in vegetables and from ^{226}Ra as well (Carvalho *et al.*, 2014).

4 Conclusion

Activity concentrations of ^{210}Po in cocoa beans and cocoa leaves collected from Lolodorf HBRA were determined using alpha spectrometry. Activity concentrations in cocoa leaves were higher than in the cocoa beans. It may conclude that the unsupported ^{210}Po in air is deposited onto leaves and thus result in a higher ^{210}Po concentration. Ingestion doses to the public resulting from the consumption of cocoa powder were estimated and the results revealed that children are more

Table 3. Effective radiation dose extrapolated to annual basis for members of public.

Locality	Location ID	Absorbed dose from ingestion (mSv/year)			
		More than 2 to 7 years	More than 7 to 12 years	More than 12 to 17	Adult
Melondo	M1	0.106	0.063	0.038	0.029
	M2	0.212	0.125	0.077	0.058
	M3	0.193	0.114	0.070	0.053
	M4	0.156	0.093	0.057	0.043
	M5	0.177	0.105	0.064	0.048
Ngombas	N1	0.223	0.132	0.081	0.061
	N2	0.370	0.219	0.135	0.101
	N3	0.293	0.173	0.107	0.080
	N4	0.189	0.111	0.069	0.051
	N5	0.352	0.208	0.128	0.096
Maximum	–	0.370	0.219	0.135	0.101
Minimum	–	0.106	0.063	0.038	0.029
Mean	–	0.227	0.134	0.083	0.062
Standard deviation	–	0.087	0.051	0.031	0.023

expose than adults. Values of ingestion dose due to ^{210}Po in the study area were relatively high compared to those obtained from study performed in Qena city of Egypt and in the same range with the study performed at Kalpakkam, India.

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