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Activity of radionuclides in soils contaminated by scrap iron residues in the 1st District of Cotonou (Benin)

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Abstract

The scrap metal waste export activity has proliferated in Benin and in the West African sub-region in recent decades. It is a sector that employs a growing number of workers, particularly on the Ex SONICOG site in the first district of Cotonou in Benin. Unfortunately, this scrap metal waste can constitute chronic sources of radioactive contamination of the soil and of exposure of populations to ionizing radiation. The objective of this research work is to estimate the specific activities of the soil and the radionuclides involved on the site of the former SONICOG scrap metals, which has been in operation for more than two decades. The methodological approach consisted in taking soil samples using the diagonal method and analyzing them by gamma spectrometry using a Baltitic Scientific Instrument, Ltd (BSI) type device fitted with Gamma Pro analysis software. . The results show a stronger contamination of the superficial horizons by radionuclides such as lead 212Pb, thorium 232Th, cesium 137Cs in general and compared to the control soil. On the other hand, potassium (40K) is detected only in the surface horizon of the soil1 while actinium 228Ac was found in the first two surface horizons of the soil2; as for americium (241Am), it has been detected in the soil3 but it is not quantifiable (low concentrations). Moreover, all these radioelements are in low quantity in the control sample (sol0) chosen as the reference soil. This observation, coupled with an unequal distribution of radioactive sources in the soil horizons, confirms the hypothesis that the radioactive sources come from ferrous and non-ferrous metals from scrap metal waste on the various sites. Workers in the scrap metal sorting-recycling chain risk the effects of direct chronic exposure to low doses of ionizing radiation emitted by these sources and populations from indirect exposure.

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Introduction

Some radionuclides are produced by humans, such as cesium 137 (IRSN, 2005, Nifenecker, 2006, Saleh, 2017) and others are present in nature, such as potassium 40, lead 210, uranium 238 and thorium 232 (Knochel *et al.*, 1972; Desideri *et al.*, 2000; Desideri *et al.*, 2000; Shoeib and Thabayneh, 2014). Radionuclides are present everywhere in small quantities, the greatest exposure to radiation comes from radioelements that are naturally present in the environment (Planinsek *et al.*, 2018; Chakraborty *et al.*, 2013; Saba, 2019a).

These emanate from the earth's crust and cosmic radiation (from outside the earth's atmosphere). Since the atomic era, the migration of radioelements from anthropogenic sources into the food chain, particularly from contaminated sites and waste, has been a subject of concern for scientists because of the harmful effects of ionizing radiation on the human body (Saba, 2019b; Falandysz *et al.*, 2021). Indeed, radioelements are characterized by their nuclear instability which is manifested by the emission of particulate radiation and/or photons (Minty, 1997) of high energy which gives them this ionizing character (WHO, 1987; Mitchell *et al.*, 2013). For example, cesium 137 is found in the environment following accidental releases, aerial tests of nuclear weapons and accidents. In 1957 in Kyshtym (Chelyabinsk, Russia) and Windscale (United Kingdoms), Chernobyl in 1986 (Lestaevel *et al.*, 2010) and Fukushima Daiichi (IAEA, 2015, IRSN, 2015) on March 11, 2011. Since potassium is an essential element for life, potassium-40 is the main source of exposure to radioactivity of healthy organisms by ingestion (Trevisi *et al.*, 2012). Potassium K-40 is naturally β- and α $^{-}$ radioactive· It is naturally present in the earth's crust. Currently, about ten radionuclides are regularly measured in the French environment, including cesium 137 as well as americium 241 (Desideri, 2001). Actinium Ac-228 is naturally β– $^{\text{and}}$ α radioactive while americium Am-241 is naturally α radioactive. Cesium Cs-137, it is naturally radioactive β $^{-}$ and α. As for lead Pb-212, it is naturally radioactive β $^{-}$ and radiogenic belonging to the radioactive decay chain of Th-232.

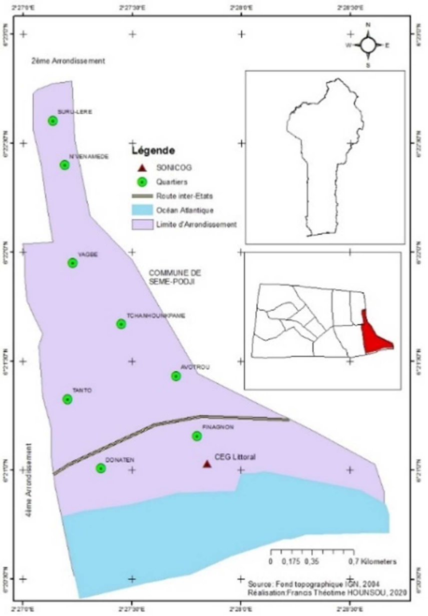
It is generally found in the impurities of metallic ores and especially in the nuclear fuel cycle (Ahamdach and Stammose, 2000; Chareyron Bruno, 2018). This radioactive radiation has the property of ionizing matter, unlike other electromagnetic radiation such as visible light, infrared rays or microwaves (Guénel *et al.*, 2003). Human beings are exposed daily to sources of ionizing radiation (Jeambrun, 2012) from radioactive materials of natural origin (NORM); but anthropogenic sources due to nuclear accidents are at the origin of the appearance of most pathologies, cancers, alteration of the immune system, genetic heritage and DNA over several generations (WHO, 1987). Different sectors of activity can be sources of exposure to radioelements: medicine, production and use of nuclear fuel, armies, industry, mining and extraction of raw materials, food processing, etc.

The doses incurred depend on the quantities ingested (Barthe *et al.*, 2998; Puncher *et al.*, 2012; Jibiri *et al.*, 2012). More than 60 radioelements are naturally present in soil, air and water (Jeambrun, 2012). Of all the heavy elements that descend from uranium or thorium, the one that contributes the most to internal exposure is lead-210, incorporated by inhalation and especially by ingestion (Souidi *et al.*, 2009). Uranium-238, with its two descendants, thorium-232, protactinium- 40 and then uranium-232 are essentially incorporated by ingestion and are mainly concentrated in the kidneys and bones. Thorium-230 and thorium-232 enter mainly through the lungs (inhalation). Their descendant's radium-226 and radium-228 are present in the diet (Kirby *et al.*, 1964; Basdevant, 2012). Metallurgical mining is also considered a potential source of contamination due to impurities; thus, the scrap metal waste resulting from the use of industrial products in the context of a consumption economy on a global scale arouses a dual scientific and ecological interest. In Benin and in particular in the city of Cotonou, scrap iron residues (metals) are found in all the waste scattered in nature. With the risk linked to exposure to anthropogenic radiation from scrap metal waste which pollutes the soil added to sources of terrestrial and cosmic origin, it is necessary to fear the contamination of agricultural products because the soil of the Beninese coast is recommended for various crops, planting forest species (Drechsel and Schmall, 1990), market gardening (Nagavallemma *et al.*, 2004) using compost household and urban waste. These agricultural amendment products can significantly influence the chemical properties of sandy soils (Mouton, 2008), because they are poor in minerals. Also, the former SONICOG site, receptacle for residue from refrigerators, batteries, of bacteria and any type of scrap metal, would it be a reservoir of radioelements (Hounsou *et al.*, 2020). Investigations on the presence of radioelements in this former SONICOG dump (Akpakpa, Cotonou) will make it possible to establish the degree of contamination of the soil by ferrous and non-ferrous waste by radioactive elements.

Material and methods

Choice of sampling sites

Four sites are chosen on the basis of different criteria: a control site (site0), a site in the area of the former SONICOG where scrap metal waste is deposited for sorting, recycling and recovery operations before export (site 1) and a secondary scrap metal waste disposal site located 1km south of site 1). Site 0 or even point P0 is the control site whose characteristics are those of a soil free of scrap iron or household or industrial waste.



Source: Fond topographique IGN, 2004
Réalisation: Francis Théotime HOUNSOU, 2020

Fig. 1. Location map of the study area.

Sampling

The material consists of soil samples taken from the site of the former SONICOG scrap metal dump at Akpakpa not far from the Cotonou lagoon (Fig. 1) and within a radius of 1 km from this site. Three of the four soil sampling points are landfills located on the SONICOG site (P1) and respectively on a radius of 1 km to the North (P2 or sol2) and to the south (P3 or sol3) of the SONICOG site. Points P2 and P3 are secondary burial sites located outside SONICOG. The fourth sampling point denoted by (P0) is free of scrap scrap; we had taken the control soil (Sol0) which will be used for the comparison of the specific activities. Soil samples are taken at three depth levels (at the surface or 0cm level, at 10cm and at 20cm depth) in horizontal areas of the identified sites, not subject to runoff, undisturbed and excluding any possibility of stagnation of rainwater. The three sampling points were randomly chosen. Once collected, the samples are weighed and then ground. The soil samples were taken using a steel shovel on a delimited surface of 1 m^2 side. The samples were taken at each corner as well as at the point of intersection of the two diagonals of this surface to constitute a composite sample, and then these composite samples are homogenized and treated by appropriate methods in order to ensure their stability.

Determination by gamma spectrometry

A conventional spectrum acquisition chain comprises, in addition to the detector and its preamplifier, an amplifier ensuring the amplification and shaping of the signal, a digital converter and a multi-channel analyzer. The principle of gamma spectrometry is based on the interaction of photons emitted by radioactive sources with matter. The interaction of photons with matter is done either by photoelectric effect, by compton effect or by creation of pairs (or materialization).

With a (point) reference source consisting of the deposit of a radionuclide, located at a distance d from the detection crystal, a spectrum is recorded for an acquisition period t. For each energy E emitted by the source, a peak is observed in the spectrum whose surface N(E) is proportional to the number of photons emitted by the source and to the efficiency of the detector for the energy considered.

The total absorption efficiency, RP (E) is determined according to: RP (E)= N(E)A×I(E)×t:

N (E): number of counts recorded in the total absorption peak

A: activity of source at time of measurement (Bq)

I (E): emission intensity of the energy line E; t: measurement duration (active time)

RP (E): efficiency for energy E and the source-detector geometric conditions of the measurement

Depending on the case, the number of counts in the total absorption peak must be corrected by different factors.

Sample analysis

The various analyzes of soil activity were carried out by gamma spectrometry at the Environmental Monitoring Laboratory (LES) in Benin. Gamma spectrometry is a fast, non-destructive and multi-element technique that does not require performing radiochemistry on the sample (Swiderski *et al.*, 2014). To do this, after mixing the samples on the squares of 1m sides, the composite sample obtained is dried in an oven (70°C) then ground and sieved. The fine fraction of the treated sample is passed to the spectrometer (Saba, 2019a; Annunziata, 2003) to determine the activity (in mBq/g) of the six radionuclides (^{212} Pb, ^{232} Th, ^{137} Cs, ^{40} K, ^{228} Ac, and ^{241} Am) detected. The ^{234} Th, ^{64} Cu and ^{22} ^{Na} radionuclides were searched for but were not detected.

The analysis of the samples was carried out using a gamma spectrometer of the Baltic Scientific Instrument (BSI) type fitted with Gamma Pro analysis software; this device has an analyzer which converts the energy spectra of radioactive sources into specific activity and applies corrections using the calibration curves in energy and efficiency. This spectrometer has three detection chains for the analysis of radioelements and the quantification of the activity of samples composed of a high-purity germanium-type semiconductor (hyper-pure Germanium HPGe) characterized by a high energy resolution of 1.8 Kev, excellent peak symmetry and a relative efficiency of 30%, housed in a vacuum chamber (cryostat) held by an electrically insulating support. A pre amplifier performs the charge-voltage conversion, and delivers a pulse proportional to the charge collected. An Amplifier allows the pulse received from the preamplifier to be converted into a Gaussian signal while the amplifier is being amplified. A multi-channel analyzer made up of a multi-channel analyzer board, inserted into the computer and comprising an ADC (Analog to Digital Converter) which collects the signals and builds the calibration curves in energy (Fig. 1) and in efficiency (Fig. 2) from the table of values.

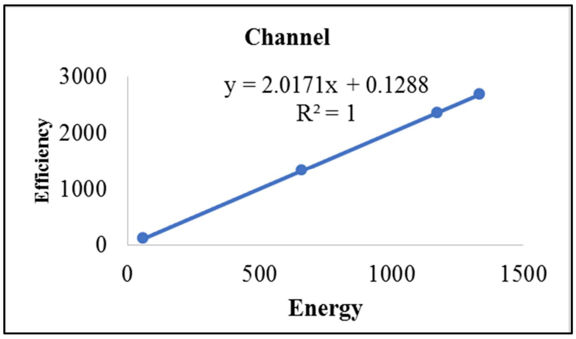


Fig. 2. Energy calibration curve.

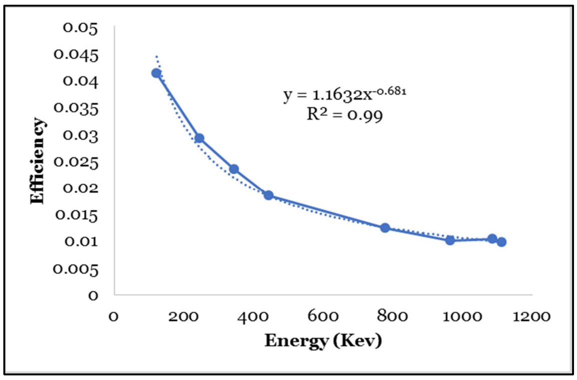


Fig. 3. Efficiency calibration curve of the HPGe detector.

Analysis and calculation of activity, uncertainty and detection limit of the radionuclides detected in the sample

Using the Spectrum of the soil sample, the specific activity and the limit of detection (LDD or AMD) of the radionuclides are calculated with the formulas below and a counting time **of** 161982s .

$$A = \frac{S_{réelle}}{\varepsilon * I * t} \text{ and } LDD\ ou\ AMD = \frac{4,66 * \sqrt{Nbf}}{\varepsilon * I * t * m}$$

N.bf: Surface of the background noise;

Ɛ: Efficiency calculated using the equation $y = 3.149x^{-0.793}$

The results of the calculations are presented in tables.

Separation of ^{137}Cs using a soil sample with ^{134}Cs as radioactive tracer

Radiochemical treatment

A mass of 200g of this sample was analyzed by gamma spectrometry, the result shows whether there is the presence of ^{137} Cs or other artificial radioelements in the sample. The radiochemical preparation consists of extracting the ^{137}Cs in a small mass of around 20g from the soil sample with hydrochloric acid for the destruction of the sample and its dissolution; the purpose of this is to prevent the fixation of the atoms of ^{137} Cs and of the ^{134} Cs added as a tracer. The correction of the activity of the tracer was carried out using the initial activity on the date of the analysis, the density of the extract and its half-life which is 2 year. The corrected activity on the injection date is denoted A_1.

The protocol consists of periodically adding about 50-100ml of hydrochloric acid to a beaker, stirring the mixture on a hot plate under a temperature between 130-150°C, then let the sample dry on the hot plate and continue attacking the sample with nitric acid HNO_3 still hot and finally allowing the sample to dry again on the hot plate.

Results and discussion

Presentation of the results

Recognition of radioelements by gamma spectrometry

Different radioelements have been sought on the four types of soil (Table 1) from the characteristic energies; the results compared with the quantification and detection thresholds made it possible to define the levels of soil contamination. Soil sample spectra were used to determine activities.

The values of the energies compared to the limits of detection (LDD) and of quantification made it possible to define different levels of evaluation: - not detected; Trace: < quantification threshold; Low: > detection threshold (or detected). The recognition of radioelements by gamma spectrometry was carried out from their energy in KeV at the surface, at 10cm and at 20cm deep (Table 1).

Table 1. Recognition of radioelements by gamma spectrometry from their energy in KeV at the surface (0cm), at 10cm and at 20cm.

Radionuclides	Ground	Floor 1	Floor 2	Floor 3
^{212}Pb (Lead-212)	238.6	238.6	238.6	238.6
^{232}Th (Thorium -232)	63.3	63.3	63.3	63.3
^{137}Cs (Cesium -137)	661.6	661.6	661.6	661.6
^{40}K (Potassium -40)	-	1525.3	-	-
^{228}Ac_ (Actinium-228)	-	-	911.1	-
^{241}Am (Americium-241)	-	-	-	59.5
^{234}Th (Thorium -234)	-	-	-	-
^{64}Cu (Copper-64)	-	-	-	-
^{22}Na (Sodium-22)	-	-	-	-

-: radioelements not found

The device has detected characteristic energies which signify the presence of the radioelements whose energies are presented in Table 1 above. This table shows that radioactive lead (^{212} Pb) , thorium (^{232} Th) and cesium (^{137} Cs) whose energies are presented were detected in the surface horizons in the four types of soil samples including samples control soils free of any contamination by burying scrap metal. On the other hand, potassium ^{40} K was detected only on point P1 of soil1 sampling whereas actinium (^{228} Ac) and thorium (^{241} Am) were detected respectively in soils 2 and 3 respectively corresponding to points P1 and P2. However, the distribution of radionuclides in soil horizons is not uniform. In the general case, the superficial horizon (0cm) presents specific activity values higher than the 0-10cm horizon and the latter higher than the 10-20cm horizon. The three radioelements are lower in the control sample compared to the soils sampled at points P1, P2 and P3 where scrap metal waste is exposed and buried. The main source of these radionuclides is exogenous, but the control samples show values characteristic of the geochemical background. On this control site, the soil horizons at depth are less contaminated or not contaminated. This confirms that the source of radioelements is anthropogenic. Except for Cs, the three elements found in all soil samples.

Distribution of average specific soil activities

The graph in Fig. 4 below shows the variations in the specific activities of the soils sampled for the three most represented radionuclides, namely Pb-212, Th-232 and Cs-137. The four soil sample collection points are designated soil 0 through soil3 in the graph. They present average specific activities of different intensity levels in the three depth levels. Solo or control soil exhibits the lowest specific activity intensities with average values ranging from 0.15 to 1.3 mBq /g regardless of the sampling depth. It is followed by sol3 (6.03 to 12.59 mBq /g), sol1 (6.57 to 14.91 mBq /g), and sol 2 (1.01 to 17.54 mBq /g).

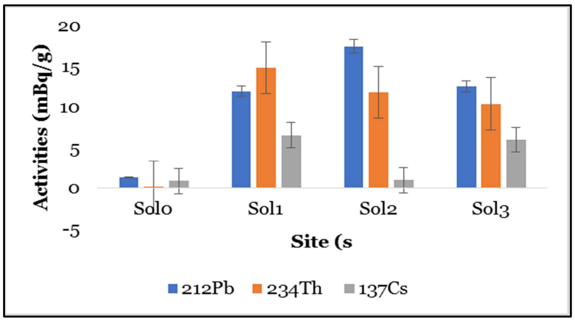


Fig. 4. Average specific soil activities in mBq /g.

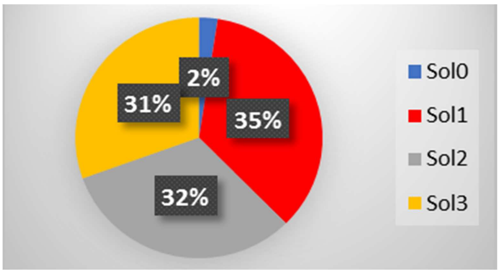


Fig. 5. Relative distribution (in%) of total specific soil activities of the three main radionuclides (Pb-212, Th-232 and Cs-137) in soil horizons.

The graph in Fig. 5 above shows the percentages of the specific activities of the soils sampled for the three most represented radionuclides, namely Pb-212, Th-232 and Cs-137. Considering the three main radionuclides, soil 1 confirms the highest radioactive contamination with 35% of the total radiation of the three sources. It is followed by the soils sol 2 with 32% and sol3 with 31% of the total activity of the sources.

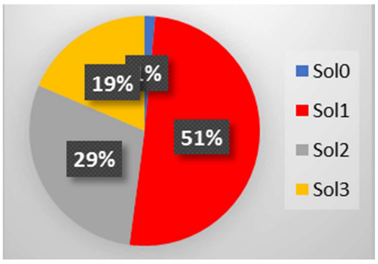


Fig. 6. Relative distribution (in%) of the total specific activities of the radionuclides studied (Pb-212, Th-232, Cs-137, K-40, Ac-228 and Am-241) in soil horizons.

In addition to the three main radionuclides detected in the control soil, new radionuclides are detected at the other sampling points. These are potassium 40 (^{40}K), actinium 228 (^{228}Ac) and americium 241 (^{241}Am). Considering all the radionuclides detected, soil 1 confirms the highest radioactive contamination with 51% of the total radiation from the sources, as does soil 2 with 29%. Sol3 comes in last position with 19% of the total activity of the sources. The control soil went from 1 to 2% despite no new radionuclides being detected. In fact, the soil1 revealed on the surface a source of potassium 40 with a high specific activity of 137.501 mBq /g. This potassium would certainly come from recent additions of sand or materials rich in this natural isotope, the most abundant of all the sources of natural human exposure. This is the most important specific activity recorded in this study. As for soil 2, it is characterized by the appearance of actinium on the first two depth levels at concentrations of 34.64 and 12.01 mBq /g. This decrease indicates an anthropogenic source unrelated to the parent rock.

Evolution of control soil activities (solo) as a function of depth

Table 2. Activities of radionuclides in the control soil sample (mBq/g) taken from the SONICOG compound.

Floor	Level	^{212}bp	^{232}Th_	^{137}Cs	^{40}K_	^{228}Ac_	^{241}Am
Solo	0-0	3.15	0.357	1,802	-	-	-
	0-1	0.75	0.09	0.802	-	-	-
	0-2	Trace	Trace	Trace	-	-	-
Mean		1.30	0.15	0.87	-	-	-

Legend: - not detected; Trace : < quantization threshold. Sampling levels: on the surface (Level 0-0), 0 to 10cm (Level 0-1) and 10 to 20cm (Level 0-2)

The device detected characteristic energies which indicate the presence of Pb-212, Th-232 and Cs-137 on the first two levels, namely on the surface and at a depth of 1cm. In general, the intensities are decreasing from surface to depth. It should be noted that there is a total absence of radioactive activity in the horizons of 2cm and more. We consider by hypothesis that the specific activity values obtained are characteristic of the geochemical soil background of the Akpakpa area where all the sampling sites are located.

Evolution of SONICOG burial soil activities as a function of depth

Table 3. Activities of radionuclides in soil sample 1 (mBq/g) taken from the SONICOG compound.

Floor	Level	^{212}bp	^{232}Th _	^{137}Cs	^{40}K _	^{228}Ac	^{241}Am
Floor 1	1 -0	34,488	47,728	14,802	137,501	-	-
	1 -1	12,075	11.75	10,602	Trace	-	-
	1 -2	0.126	Trace	-222	Trace	-	-
Mean		12.00	14.91	6.57	45.83	-	-

Legend: - not detected; Trace : < quantization threshold. Sampling levels: on the surface (Level 1-0), 0 to 10cm (Level 1-1) and 10 to 20cm (Level 1-2)

The device detected characteristic energies that confirm the presence of Pb-212, Th-232, Cs-137 and K-40. Soil samples 1 were taken in the main landfill area of the former SONICOG site in the part where the scrap metal waste was buried for about twenty years. This could explain the fact that samples from this site showed a high intensity of specific activity of radionuclides with a relatively higher specific activity with 34.488 mBq/g for the $^{21\ 2}$ Pb , 47.728 mBq/g for ^{232} The and 14.802 mBq/g for the ^{137} Cs. These values are significantly higher than those of the control site located in the same enclosure.

Evolution of secondary burial soil activities (soil2) as a function of depth

Table 4. Activities of radionuclides in soil samples 2 (mBq/g) taken 1 km from SONICOG to the north.

Floor	Level	^{212}bp	^{232}Th _	^{137}Cs	^{40}K _	^{228}Ac	^{241}Am
Floor 2	2 -0	37.37	24.35	2.31	-	34.64	-
	2 -1	15.24	11.29	0.72	-	12.01	-
	2 -2	Trace	Trace	-	-	Trace	-
Mean		17.54	11.88	1.01	-	15.55	-

Legend: - not detected; Trace: < quantization threshold. Sampling levels: on the surface (Level 2-0), 0 to 10cm (Level 2-1) and 10 to 20cm (Level 2-2).

In addition to soil radionuclides, the device detected characteristic energies that indicate the presence of actinium 228, a transuranic element.

Pb-212, Th-232 and Ac-228 have respectively a specific activity of 37.37 mBq/g, 24.35 mBq/g and 34.64 mBq/g in the surface samples on this site located 1km to the south of SONICOG . While Cs-137 exhibited a relatively lower specific activity of 2.31 mBq/g at the surface. Pb -212, Th-232, Cs-137 and Ac-228 have respectively a specific activity of 15.24 mBq/g, 11.29 mBq/g, 0.72 mBq/g and 12.01 mBq/g in samples taken at 10cm depth. We also detected the presence of these nuclides at a depth of 20cm in traces except for Cs-137 which was not detected at this depth.

Evolution of secondary burial soil activities (soil2) as a function of depth

Table 5. Activities of radionuclides in soil samples 3 (mBq/g) taken 1 km from SONICOG to the south (near the Atlantic Ocean).

Floor	Level	^{212}bp	^{232}Th _	^{137}Cs	^{40}K _	^{228}Ac _	^{241}Am
Floor 3	3 -0	24,514	21,185	12.83	-	-	Weak
	3 -1	13.25	10.12	5.25	-	-	Weak
	3 -2	Trace	Trace	Trace	-	-	Trace
Mean		12.59	10.44	6.03	-	-	-

Legend: - not detected; Trace: < quantification threshold; Low: > detection threshold Sampling levels: at the surface (Level 3-0), 0 to 10cm (Level 3-1) and 10 to 20cm (Level 3-2)

The device detected characteristic energies which make it possible to confirm the presence not only of Pb-212, of Th-232 and of Cs-137, radionuclides detected in the solo reference soil. In addition to soil radionuclides, the device has detected characteristic energies which indicate the presence of americium 241, a transuranic element.

Pb-212, Th-232 and Cs-137 have respectively a specific activity of 24.514 mBq/g, 21.185 mBq/g and 12,830 mBq/g in surface samples at this site located 1km north of SONICOG. While Am-241 exhibited relatively lower and unquantifiable specific activity at all levels. We also detected the presence of Pb-212 (13.25 mBq/g), Th-232 (10.12 mBq/g) and Cs-137 (5.25 mBq/g) of weak activity in the 10cm horizons and traces deeper at 20cm.

Discussion

I n this study on the four types of soil studied, certain radioelements are constantly found, namely lead (^{212} Pb), thorium (^{232}Th) and cesium (^{137}Cs). However, other types of radioactive elements such as potassium (^{40}K), actinium (^{228}Ac), and americanum (^{241}Am) appeared only in the scrap metal waste burial soils.

From the analysis of these results, we can propose two interpretations, namely, natural and artificial sources. Indeed, lead 212 is radiogenic and belongs to the family of radioactive decay of Th-232.

The presence of these two radionuclides in all the samples can be explained by their common origin, which is found in ferrous and non-ferrous metal ores. They could also originate from the natural geochemical backgrounds of the soils studied insofar as the control soil has characteristic concentration levels in the non-zero surface horizons. On the other hand, these metals disappear at the 10-20cm horizon on practically all the soil samples except at point P1 where the soil has a low specific activity of 0.126 mBq/g. This contradicts the first hypothesis based on the geochemical background. This is confirmed by the appearance only in the main burial area on the site of the former SONICOG of potassium 40 in a very high quantity (137.501 mBq/g), but this value is lower than that obtained by Chakraborty *et al.*, 2013 who reported a specific activity of 451.90 ±24.89 Bq /kg for this radionuclide in Chittagong city in Bangladesh. This high concentration could be explained by the fact that on this site, scrap metal waste was buried there for about twenty years. This is in agreement with a higher specific activity of the radionuclides present with 34.488 mBq/g for ^{212}Pb, 47.728 mBq/g for ^{232}Th and 14.802 mBq/g for ^{137}Cs. It should be noted that the Fig.s of 38.47±2.72 and 2.41±0.18 Bqkg^{-1} obtained by Chakraborty *et al.*, 2013 respectively for Th-232 and Cs-137 are lower than the values recorded for the soil1 which is the most contaminated by radioactive elements. On the other hand, Kall *et al.* (2015) reported a much higher level of specific activities for Th-212 (81±20 to 161± 23 Bq.kg^{-1}) and K-40 (112± 34 to 565± 50 Bq.kg^{-1}) soils on the shore of French Bay in the localities of Ankorikihely, betahitry and Lazaret-Nord. Phosphogypsum (PG) waste associated with phosphoric acid produced in industrial fertilizer and chemical production facilities in Egypt can contaminate soils (Elafifi, 2009). Indeed, Elafifi *et al.* (2009) found concentrations of 8.3 ±0.76 and 64.3±4.1 Bq/Kg of Th-232 and K-40 respectively in chemical fertilizer manufacturing residues in Egypt. The concentration obtained is much higher than our data for Th-232 and lower for K-40. The IAEA exemption limits are 10 Bq/kg for ^{40}K and ^{137}Cs. These limits are exceeded in most samples at the surface and at the second depth level (Planinsek *et al.*, 2000; Egidi *et al.*, 1997; Hounsou *et al.*, 2020).

They were actinium (^{228}Ac) and potassium (^{40}K) with a high specific activity (137.501 mBq/g) for potassium and 34.64 mBq/g for ^{228}Ac while the latter belongs to the $^{232\text{Th}}$ family (Kelly *et al.*, 2019). With the secondary soil samples located 1 km to the south (towards the sea) and 1 km to the north (on the mainland), we noted the presence mainly of ^{212}Pb, ^{234}Th and ^{137}Cs and the presence also potassium, actinium and americanum. Thorium has a specific activity of 21.185 Bq/g on the site 1 km towards the sea (soil 3) on the surface on the secondary site located to the south. On the site 1 km on the mainland to the north (soil 2) the presence of actinium was noted with a specific activity of 34.64 mBq/g. On the control site on which there was no burial of scrap metal waste, only a few rare radioelements such as Th, Pb and cesium, americium (^{241}Am) and ^{228}Ac with negligible specific activities . But, on the burial site, the presence of other types of heavy metals and radioelements was noted, the concentration of which decreases with depth. It appears that said soils are heavily contaminated by potentially toxic metals such as zinc, lead, chromium, americanium, acne. This contamination would come from sediments or sludge, phosphate residues, battery cells, vehicle batteries, iron and other non-degradable metals such as residues of copper rods, thorium from welders buried in the ground by scrap metal sorters. We also note an increase in the specific activity of the radionuclides which starts from double for lead and triple for thorium and is explained by an accumulation effect. If plants and/or vegetables were grown in this region, very high concentrations of radionuclides could be found in the leaves of these plants. However, high concentrations of lead have been found in leafy vegetables collected from areas of high concentration of household waste, in fish samples and in aquatic ecosystems in Benin (Drechsel and Schmall, 1990, Pazou *et al.*, 2010, Youssao *et al.*, 2011) with high risks that these lead residues contain radionuclides.

We have noticed that the different mass activities decrease progressively with depth to tend to disappear around 20cm for certain radionuclides such as ^{40} K and ^{228} Ac and according to the sampling site. It is deduced that the evolution of the concentration in the soils of these radionuclides is dependent on a large number of parameters, namely the physicochemical and biological nature (texture) of the soils, meteorological conditions (in particular precipitation) and runoff conditions and accumulation phenomena (Ferry, 2000). It is therefore customary to consider as a first approximation and in particular for the burials of recent radioactive scrap metal from the last 5 years an exponential decrease in the concentration of artificial radioelements according to the depth in the ground (Saba, 2019; Lennemann, 1978). There is surely a phenomenon which induces a decrease in the concentration of the radionuclide (Lebatard and Bourles, 2015).

This phenomenon evolves over time depending on the radioelement transfer conditions in the soil of sites containing high radioelement proportions such as the site of the former SONICOG company, the site located 1 km to the south and the site located 1 km to the north are sites polluted by radioactivity. This confirms that highly radioactive substances have been handled and stored in an uncontrolled manner, leading to the dispersion of these substances and presenting a potential risk depending on the use made of the sites: site for sorting and recycling waste from scrap.

In general, almost all soil and water rocks naturally contain small quantities of radioactive materials such as uranium, thorium, potassium and their descendants (Worden *et al.*, 2000; Larivière and Guérin, 2010; Vandenhove *et al.*, 2010). The work carried out by Cresswell *et al.* in 2013 also shows that urban areas have very complex radiative environments due to the diversity of sources of radioactive contamination and radioelements. These authors further demonstrated that the presence of Cs-137 identifies relatively undisturbed soil and a feature called "Naturally Occurring Radioactive Materials" (NORM). They can thus be found in building materials. These materials containing radionuclides naturally present in the earth's crust called NORM had been mentioned by Desideri *et al.*, 2006; these determined the geochemical distribution of natural (U, Th) and anthropogenic (Pu, Am, ^{137}Cs) radionuclides in three sediment samples of different origin. TE -NORM stands for technically enhanced natural radioactive materials for large volumes of waste with low specific activity emitted by several activities industries such as mineral extraction, beneficiation and chemical treatment of ores, the production of phosphate fertilizers, extraction and purification of trace elements, etc. The Pb-212 could have its origin in the rocks containing residues of Th-232 from which would come the buried metals. Indeed, Worden *et al.*, 2000 demonstrated that sandstones exceptionally enriched in uranium with concentrations of 0 to 70 ppm would be at the origin of the contamination of waters with Pb-210 from which it would come inside the reservoir (rock).

These concentrations, generally low, are observed on the control site, that is to say the site without burial of waste from scrap metal and vary between 3.21 mBq/g for thorium 2012 and 0 mBq/g for potassium. 40. The (TE-NORM) translates to Enhanced Technology Naturally Occurring Radioactive Materials and by technologically enhanced natural radioactivity is due to human activities operating in various industrial sectors (Mitchell *et al.*, 2013). Human activities can during the handling, storage, burial and storage of products and waste as well as production equipment concentrate materials on a site. Thus, the radioactivity is concentrated in the products and/or residues, including food products (Chakraborty *et al.*, 2013; Thabayneh *et al.*, 2012). And consequently the level of radioactivity becomes higher than that encountered in the natural state. This is verified on the main scrap metal waste landfill site and on all the secondary sites with respectively thorium which goes to 47.728 Bq/g, lead to 34.488 mBq/g and potassium to 137.501 mBq/g. It is important to note that the two secondary sites alone concentrate 63% of the total activity of the three main radionuclides found in all the sites, namely Pb-212, Th-232 and Cs-137. The natural radioactive elements which are the basis of the TE-NORM are present everywhere in the earth's crust, but the only difference in their distribution will be their affinity with respect to such or such other element and this will define their abundance from one environment to another, from one ore to another, hence their chemical speciation (Desideri *et al.*, 200). However, the physical observation of the waste on the various scrap processing sites reveals, before their burial, the presence, among other things, of aluminum residues, burnt copper, batteries and batteries of vehicles, electronic waste etc.

These areas typically contain fine sand particles, corrosion particles, paint chips, battery packs, and other fine particles breaking off from scrap metal deposits that can be the source of lead-212. Found in the TE-NORMs. These results agree with those of Egidi (1997) and Elafifi *et al.* (2006) on radionuclides emitted from NORM and TENORM.

Conclusion

This study was carried out to assess the radiological quality of scrap metal waste on the former SONICOG site in Cotonou. It revealed the presence of radioelements whose activity is generally low in samples taken at a depth of 20cm. This weak natural radioactivity would be reinforced by the waste which was on the sites and which produces a specific activity far superior to the exemption limits recommended by the IAEA. This observation confirms that the soils are contaminated by scrap metal waste buried underground. Under these conditions, environmental monitoring becomes vital and must above all be a major concern of environmental specialists for the prevention of the health of populations and especially of workers carrying out activities on the SONICOG site.

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