RADIOLOGICAL EXPOSURE RISK ASSESSMENT DUE TO 210PO FROM THE PROPOSED RADIOACTIVE WASTE STORAGE AREA OF PILIKWE, BOTSWANA.

Tshegofatso Solomon, Manny Mathuthu, Chamunorwa Oscar Kureba

Abstract— Determination of ^{210}Po activity concentration on the soil and water samples was given more emphasis for the radiological risk assessment as most studies focused on determining concentrations of ^{210}Pb by alpha spectrometry through its granddaughter ^{210}Po, assuming radioactive equilibrium between the two radionuclides. The average activity concentration was very low, for the soil sample being 3.10 ± 1.60 Bq.kg^{-1} and for water samples there was no activity concentrations recorded as it was below the detection limit of the Canberra Alpha analysis spectrometer instrument. The total excess cancer risk analysis by Residual Radioactivity (RESRAD) codes software values were less than 1, which indicated that there is no health risk to the residents of the study area.

Index Terms— Activity concentration, Excess cancer risk, Polonium-210, Radioactive waste storage

1 INTRODUCTION

Naturally occurring radionuclide ^{210}Po, arising from the uranium-radium decay series, is considered to be one of the contributing factors to human radiation exposure. Thus, it is investigated and analysed for different purposes, such as radiological impact assessment or as a tracer of environmental processes (Al-Masri, 2010). Poloniun-210 might be lost at temperatures above 100^{0}C, as it is volatile in conditions and therefore requires extra care during sample preparation and treatment (Sethy et al., 2015). There has been little development regarding analysis of polonium-210 in environmental samples since 1960 as radiochemical analysis of polonium-210 is quite straight forward due to ease of source preparation through auto deposition on to metal surfaces (Simion et al., 2017).

The Polonium (^{210}Po), ($T_{1/2}$ = 138.4 days) and lead (^{210}Pb), ($T_{1/2}$ = 22.3 years) radionuclide pair, continues to be broadly used in radiological risk assessment for the past several decades, for examining dissolved marine ecosystem particles (Taqi et al., 2016). Both radionuclides are members of the ^{238}U decay series, ^{210}Po being the decay daughter of ^{210}Pb via ^{210}Bi ($T_{1/2}$ = 5.0 days). In seawater, both ^{210}Pb and ^{210}Po are the reactive particles, nevertheless ^{210}Po also accumulates inside organic tissues (Casacuberta et al., 2012). These two radionuclides' specific activity difference is frequently employed in the water column quantitatively to assess export fluxes of material particulate sinking properties, such as dissolved organic carbon (DOC) with depth from the ocean surface to the bottom.

There is evident success in research conducted with these methods in the separation of Po and Pb, however during processing ^{210}Pb can be lost (Thabayneh, 2012). Due to this loss of ^{210}Pb, it is currently not clearly understood how to assess loss. This loss in most of the laboratories in the world is considered to be minor thus rendering it irrelevant and being ignored, whereas, where it is taken into consideration, more extensive procedures and process for its correction are being applied (Turhan et al., 2011).

When calculating radionuclide ingrowth, decay, and recovery during sample processing and error treatment, laboratories apply different methods, and this is based on a range of assumptions (Rigaud et al., 2013). Consequently, the accuracy and precision of ^{210}Po and ^{210}Pb measurements for radiological risk assessment result have always raised some questions. However, there is still a gap that needs to be closed regarding this method. An initial analysis of the precision and accuracy procedures of the assessment for ^{210}Po and ^{210}Pb measurement, was conducted during interlaboratory comparison exercise for calibrations for seawater particulate samples (Rigaud et al., 2013).

One of the most poisonous radionuclides known to human beings is ^{210}Po, yet it is abundantly around the earths crust. There are very low levels of ^{210}Po in the environment, and it is transmitted into the human body through the food chain, such as eating seafood (Singh et al., 2005). Due to its low levels in the environment, it does less harml to human health, except in smokers, who have higher levels due to smoking. However, if more amounts of it enters the human body is very lethal within days or weeks, therefore because of this toxicity, there is need for radiological risk assessment of polonium on the environment. The aim of the study was to assess the radiological risk due to exposure to ^{210}Po (from NORMS) in the proposed waste storage Facility of Pilikwe.

2 MATERIALS & METHODS

2.1 The study area

Detailed Kgosi Tshekedi Khama of Bangwato is believed to be the one who founded Pilikwe village which is a rural place in Central District of Botswana. The current chief of this village is Kgosi Gasebalwe Seretse and the commonly known native name for this village is Rametsana. Pilikwe village is located 10 km east from the Martin's Drift Tswapong highway and 32 km north-eastern from Radisele and is on its Northside is surrounded by a hill, its geographical coordinates are 22° 47' 0" South, 27° 14' 0" East as shown in figure 2.1.

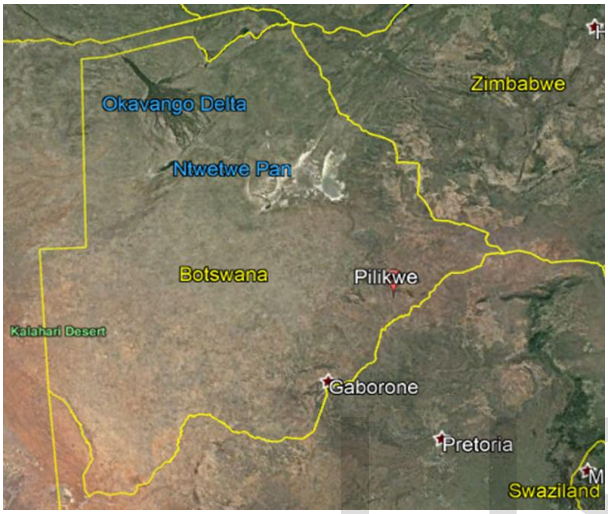

Figure 2.1 Google maps picture of Botswana showing the location of Pilikwe.

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2.2 Sampling

For Samples were collected at the site where the radioactive waste storage facility is proposed to be built in Pilikwe, and within the surrounding area of the village making a boundary. Sampling consisted of 60 soil and 2 borehole water. Figure 2.2 shows google maps picture of Pilikwe with the GPS locations of the samples collected from the study area.

2.2.1 Soil sample collection

Soil samples within the area of the storage facility were collected about 100 metres apart, while those in the village were collected 500 metres apart. Soil samples on the boundary were collected about 3 kilometres apart. All the soil samples were randomly collected systematically marking a boundar. The samples were collected by first removing the topsoil using a spade and then drilling the soil up to a depth of +/- 30 cm using a hand corer. The topsoil was removed in order to take away the organic matter. The location of soil samples is shown in figure 2.2 marked by a yellow Pin.

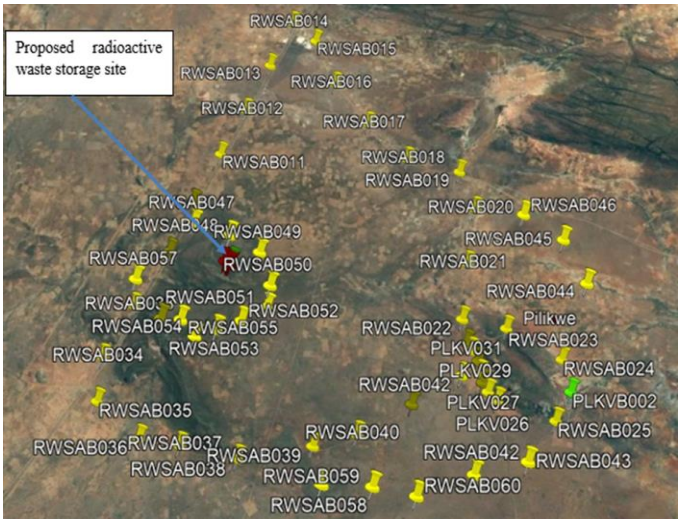

Figure 2.2: Google maps picture of Pilikwe with the GPS locations of the samples collection area**.**

2.2.2 Water Sample collection

Water samples were specifically collected from the only 2 borehole that are found in Pilikwe, as these boreholes are the source of water for dinking and farming by the residents of the study area. The boreholes depth were 260 m and 398 m; thus, water samples were manually pumped from the boreholes into 2 litre bottles for each sample. Borehole water samples colletcted are shown in figure 2.2 marked by a green symbol.

2.3 Instrumentation

Alpha spectroscopy was used for the identification and quantification of radionuclides that are alpha particle emitters during decay process. The energy spectra were acquired using the Canberra Alpha analysis spectrometer controlled by the Apex-Alpha; Alpha spectrometry software suite 1.1. The process of quantifying and identifying radionuclides that emit alpha particles is of very important use in environmental characterization and radiation protection. This makes alpha spectroscopy an important tool and technique for the analysis and providing guiding information in waste management, decommissioning and decontamination. A complete system of alpha spectroscopy will have:

- A high resolution, low background, and detection efficiency charged particle detector.
- Vacuum Chamber that enables alpha particles to reach the detector since they are attenuated easily by air.
- Recoil Contamination Avoidance Package (RCAP) to safeguard the detectors from contamination of daughter nuclides that may leave the sample material.
- Signal detector electronics for processing and storing

- Tshegofatso Solomon is a PhD alumin at North-West University (Mafikeng Campus), South Africa. Email: twsolomon1@gmail.com
- Manny Mathuthu is a Professor at North-West University (Mafikeng Campus), South Africa. Email: Manny.Miathuthu@nwu.ac.za
- Chamunorwa Oscar Kureba is a Senior Lecturer at Department of Physics and Astronomy, Botswana International University of Science and Technology. Email: kurebac@biust.ac.bw

the energy data.

- Analyzing software that controls the electronics, analyze spectra, establish quality control processes, and integrate data with laboratory data management system. This setup is shown in Figure 2.3.

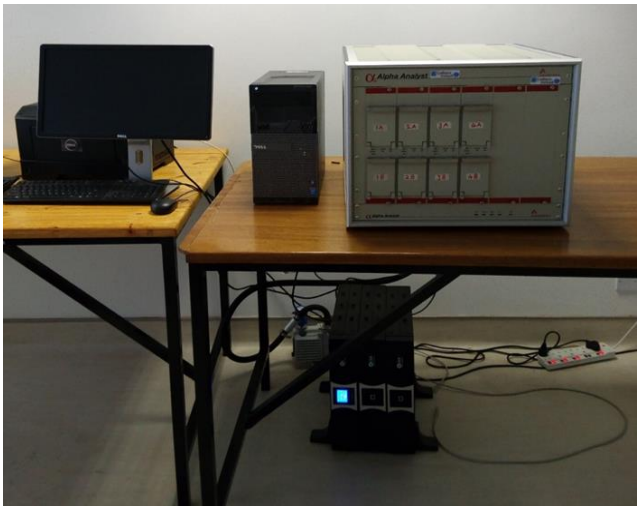

Figure 2.3: Alpha spectroscopy system used to identify and quantify radionuclides

2.4 Sample preparation

The soil samples were firstly prepared by digestion using the Milestone ethos easy microwave digestor. After completion of the digestion process, samples were loaded into the Canberra alpha analyst system spectrometer for sample run using Apex-Alpha; Alpha spectrometry software suite 1.1. These following steps were followed for sample digestion; Samples were weighed to approximately 0.200 g, added 8 ml of nitric acid (HNO_3) and 1.00 g of ^{209}Po tracer (≈ 0.188 Bq/g), after which the samples were then digested.

After digestion there was qualitative transfer from the digestor vessels to 210 ml beakers with addition of 5 ml HNO_3, then evaporation to dryness, and we added 3 ml hydrogen peroxide (H_2O_2) to oxidise organic matter followed by evaporation of the solution to dryness with the step repeated twice. Then 5 ml of hydrochloric acid (HCL) was added and evaporated the solution to dryness with the step repeated twice. Again 1 ml HCL was added and then diluted with ultrapure deionized water to a volume to 100 ml. Then ascorbic acid was added gradually until the yellow colour of Iron (Fe) ions disappeared. A magnetic stirring bar was placed into the reaction vessels, the temperature set to 85°C and speed set to 300 rpm in the stirring hotplate. The silver disc was then immersed into the reaction mixture and covered with parafilm. Deposition occurred during stirring and heating for 8 hours.
To measure the ^{210}Po (half-life = 138.376d), Polonium 209 Tracer, activity: 0.188 ± 0.004 Bq/g K=2 Ref date: 18 April 2018, was employed using the standard technique (the silver disc technique), whereby 2 g of soil samples were taken in duplicate samples having been spiked with 1 g of ^{209}Po tracer as a yield tracer during digestion of the samples.
Alpha counting of ^{209}Po and ^{210}Po were conducted using Canberra alpha analyst system spectrometry for sample run controlled via the Apex-Alpha; Alpha spectrometry software suite 1.1, with the background counts taken into consideration. The ^{210}Po activity was corrected for recovery by comparison with measured activity of the ^{209}Po yield tracer and for radioactive decay since the time of sampling (Taqi et al., 2016).
The ^{209}Po activity was determined using internal standard technique. The limit of lowest detection (MDA) of the method used was 0.4 Bq kg^{-1}. The determination of the MDA was done by analysing 10 replicates of the reference material: BOT7003 Sample 03 Soil with a known activity of ^{210}Pb (about 5 to 7 times of MDA). The standard deviation (STD) of the ten replicates was calculated using the equation below (Turhan et al., 2010).

$$MDA = 3STD \quad (1)$$

3 RESULTS AND DISCUSSION

The polonium determination method was applied on running both the soil and water samples respectively, the determination of polonium concentrations in the samples was crucial as ^{210}Po is a naturally occurring alpha emitter and exists in the environment because of the ^{210}Pb decay within the ^{238}U decay chain. Polonium has 25 known radioactive isotopes with mass numbers of 192 to 218, of which only the 208, 209 and 210 isotopes have half-lives longer than 1 day. Of these three, it is ^{210}Po which is of most interest from an environmental impact perspective (Casacuberta et al., 2012).

There is very little information known about the soil chemistry of polonium, but it is expected to be adsorbed onto soil particles, such as mineral colloids (Taqi et al., 2016). The ^{210}Po is considered to be in equilibrium with ^{210}Pb, suggesting that the ^{210}Pb in the soil is the main source of ^{210}Po irreversibly adsorbed on clay and organic colloids in the soil. The activity concentration levels of ^{210}Po in various soils that have been reported are in the range of 20 to 40 Bqkg^{-1} respectfully (Rigaud et al., 2013).

3.1 Determination of ^{210}Po in the soil samples

Table: 1. The average activity concentration of ^{210}Po in the soil samples

	Total Activity (Bq/kg)	**Total Activity on preparation date (Bq/kg)**
Min	0.498±0.053	0.498±0.053
Max	27.520± 1.840	27.520±1.840
Average	3.100 ± 1.600	3.100 ± 1.600

The average activity concentrations for soil samples from the study area in Table 1 above are way below the available reported and accepted activity concentration levels of ^{210}Po in various soils that have been reported ranging from 20 to 40

Bq/kg (Simion et al., 2017).

3.2 Calculation of ^{210}Po activity concentration

Figure The concentration of ^{210}Po in the samples were calculated backward to their date of collection, considering the decay of the original ^{210}Po and ingrowth from its progenitors ^{226}Ra, ^{210}Pb, and ^{210}Bi. This was done being mindful of Pb and Bi which are the major contributors to ingrowth over delayed period of time between sampling and separation. Therefore, to minimize the necessary correction, the delay time between sample collection and the separation for ^{210}Po measurement was kept as short as possible. This was very vital for these soil and water samples as their activity concentration for ^{210}Po was expected to be lower than those of its progenitors and the tracer since the ingrowth and decay calculations are dependent on the procedure followed (Turner, 2007).

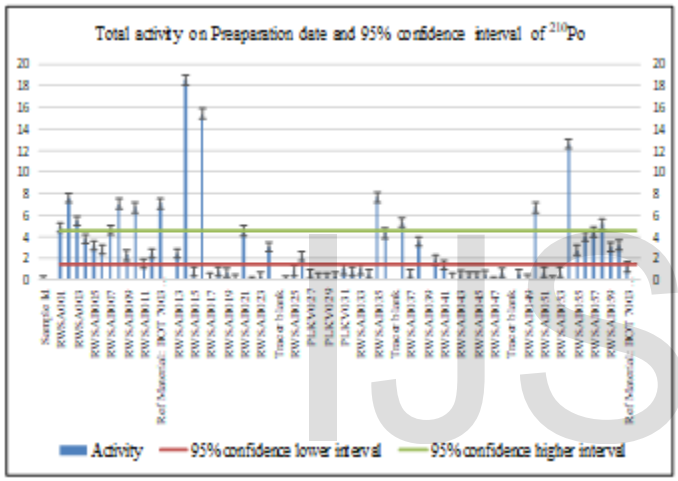

Figure 3.1: The soil samples total activity concentration of ^{210}Po at a confidence interval of 95%.

The average activity of ^{210}Po was 1.5 Bq/kg and 4.6 Bq/kg for the lower and higher interval of 95% confidence level respectively. Thus figure 3.1 shows the total activity concentration on preparation date of the soil samples with respect to the confidence level. The activity concentrations on preparation date are within acceptable range of 0 to 20 Bq/kg (Abdulrahman, 2011). The minimum, maximum and average recovery of ^{210}Po using ^{209}Po tracer was 65%, 100% and 86% respectfully.

3.3 RESRAD (Residual Radioactivity) radiological risk assessment

The radiological risk assessment for Pilikwe proposed waste storage facility using RESRAD software was determined by inputting the average activity concentrations of the soil sample results obtained by the use of the Canberra Alpha analysis spectrometer controlled by the Apex-Alpha; Alpha spectrometry software suite 1.1. The RESRAD was utilised to generate the amount of intake quantities QINT (i,p,t) for individual radionuclides (i) and pathways (p); Excess cancer risks CNRS (i,p,t) for individual radionuclides (i) and pathways (p) and fraction of total risk at time (t) in years and the total excess cancer risk for initially existent radionuclides and pathways as well as fraction of total risk at time (t).

The amount of radionuclides intake, excess cancer risk and the total cancer risk was estimated for a period of 0-year, 1 year, 3 years, 10 years, 30 years, 100 years, 300 years, and 1000 years respectfully. The results are shown in Figures 3.2 to 3.4.

3.3.1 Amount of radionuclides intake

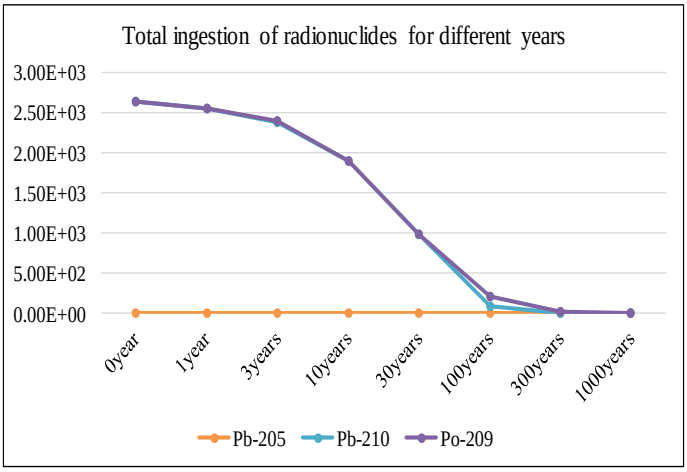

Figure 3.2: Estimated total intake quantities of individual radionuclides and pathways as pCi/yr. at time (t) in different years.

The projected amount of intake radionuclides quantities decreases with time as years increases from the initial amount of 2637 pCi/yr. for ^{210}Pb and 4.083 pCi/yr. for ^{209}Po. The total ingestion of radionuclides results was relatively very low for all the pathways thus considered to be cancer risk free.

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3.3.2 Total excess cancer risk

The total excess cancer risk for initially existent radionuclides and pathways and fraction of total risk at time (t) years for water dependent pathways and water independent pathways (inhalation excludes radon) show that the total cancer fraction and risk are very low and negligible to have the potential to cause any harm with respect from the initial (0 year) to the final (1000 years) year of interest from the study.

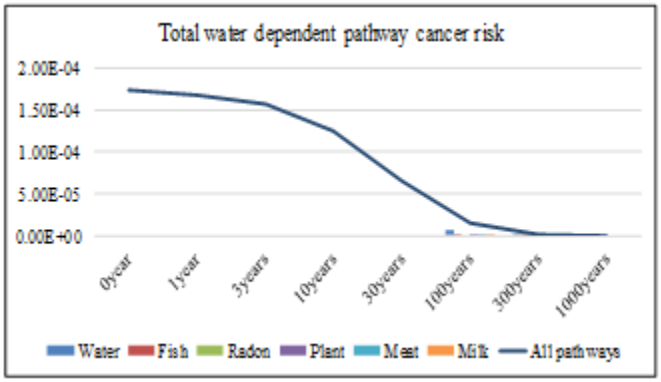

Figure 3.3: The total excess cancer risk for initially existent radionuclides and pathways at time (t) years for water dependent pathways (inhalation excludes radon).

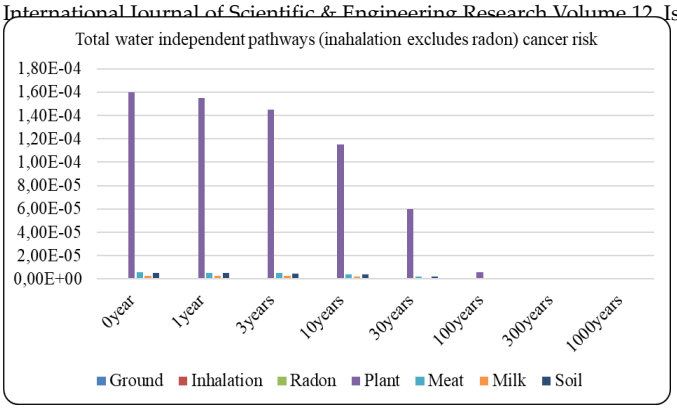

Figure 3.4: Total excess cancer risk for initially existent radionuclides and pathways at time (t) years for water independent pathways.

The cancer risk results from both the water dependent and independent pathways show that the risk decreases with time from the initial year (0 year) as shown in figure 3.3 and figure 3.4. The values are less than 1, which indicate that there is no health risk to the residents of the study area.

4 Conclusion

Extensive studies carried out over the years using alpha spectrometry provided alternative choices to this research for radiochemical analysis and determination of polonium in the water and soil samples. Polonium is considered to be poisonous thus if inhaled and deposited in the lung tissues, it will contribute to an increase in the internal radiation dose and in the number of lung cancer incidences observed among the resident of the area for the proposed radioactive waste storage. The concentration levels result for ^{210}Po obtained in this study indicated that the activity concentrations of ^{210}Po ranged from 0.498 ± 0.053 Bq/kg to 27.500 ± 1.840 Bq/kg and with an average of 3.100 ± 1.600 Bq/kg. The activity concentration levels of ^{210}Po in the proposed radioactive storage facility site and the village of Pilikwe was found to be below accepted activity concentration levels of ^{210}Po in various soils that have been reported as 20 to 40 Bq/kg (Sethy et al., 2015). The polonium was non-uniformly distributed within the samples thus the data obtained was indicating that it was generally from the dry or wet deposition of the radioactive fall-out into the soil.

The radiological risk assessment by the RESidual RADioactivity (RESRAD) model software for the analysis and modelling of cancer risk to the community residing in the study area was conducted by assessing the radionuclides activity concentrations results obtained by alpha spectrometry technique. The cancer risk for people living in the study area, because of Polonium in soil and water, was evaluated using the average concentrations of nuclides found in the samples to calculate the average intakes from 0-year,1 year, 3years, 10 years, 30 years, 100 years, 300 years and 1000 years respectfully for determination of cancer risk associated with the ingestion of the radionuclides.
The results obtained were very much lower than the acceptable worldwide value of 1x 10^{-4} (Thabayneh, 2012). These cancer risk results show that the Polonium concentrations in the Pilikwe area are safe for the community when ingested for a period from 0 year up to 1000 years without causing any health problems. Therefore, from the results of this baseline study, the proposed Pilikwe repository is currently free from radiological health risk and must be maintained thus after nuclear waste deposition has started.

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