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Heavy Metals and Naturally Occurring Radionuclides Distribution Risk Assessment in Wetlands in Kampala, Uganda

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Abstract: This study determined the effect of heavy metal concentration on naturally occurring radioactive material levels within wetlands in Kampala, Uganda. Samples from five wetlands (yams and their accompanying soils) were collected for heavy metal and NORMs analysis using an Atomic Absorption Spectrometer (AAS) and a Na(Tl) gamma ray spectrometer respectively. The study indicated that heavy metals Pb and Hg showed a mean concentration in all the sampled wetlands that were above the maximum acceptable values. The average activities of the radionuclides ^{232}Th and ^{226}Ra obtained in this study were above their maximum world acceptable limits in almost all the wetlands however the average activity of ^{40}K in all the wetlands were below the world acceptable limits. In order to determine the effect of heavy metal loading due to industrial waste disposal to the NORM activity concentrations in these wetlands, Pearson correlation coefficients were determined. These levels of activities however had biological effects to the general population so the dose rates at one metre above the ground and other biological effective dose rates were monitored. The Radium Equivalent (Ra_{eq}) values obtained in this study ranged from 71.9 Bqkg $^{-1}$ to 215.6 Bqkg $^{-1}$, the external values were in the ranges 0.2 to 0.6 while the internal hazard index values ranged from 0.3 to 0.8. The absorbed dose rate in indoor air ranged from 33.9 nGyh^{-1} to 94.8 nGyh^{-1} in the sampled wetlands and the annual effective indoor and outdoor dose rates ranged between 0.166 mSv to 0.593 mSv and 0.044 mSv to 0.115 mSv respectively.

Keywords: Heavy metals, Natural radioactivity, Wetlands, Pearson correlation coefficient, Radiological hazard indices, Absorbed dose rate.

1 Introduction

Radioactivity on earth comes from naturally occurring radionuclides (NORMS), such as the radioactive decay products of primordial radioactive elements in the outer layer of the earth and cosmic radiation interactions which is the source of most natural radioactivity [1]. The naturally occurring radionuclides such as potassium ^{40}K and radionuclides of radium ^{226}Ra and thorium ^{232}Th through decay series are present in the earth's crust and makes our planet expose us to various levels of ionizing radiations by releasing gamma rays [2–6]. The primary source of natural radioactivity is rock weathering and thermal springs due to geological degradation [7, 8]. Radioactive decays in soils contribute greatly to background radiation from the earth in the human environment [9–11]. Cultural environment involving agroecology, fossil fuel exploration and combustion, rapid urbanization, disposal of radioactive wastes and industrial effluents, among others escalate natural radioactivity in the environment [12–15].

The naturally occurring radionuclides pose a radiation health risk to people by entering the food chain through absorption via plant roots from the polluted topsoil, entering the edible sections of plant tissues [16–18]. Environmental pollution from industrial waste with heavy metals which are toxic and dangerous is one of the causes of elevated concentrations of NORMS. The commonly observed carcinogenic heavy metals include Chromium, arsenic, cadmium, mercury and lead in industrial waste can damage the functioning of the brain, heart, lungs, kidneys, and other vital organs [19–21]. The bio availability and bio accumulation of these metals have raised questions about crop quality, food security, and human health because they enter the animal food chain predominantly via contaminated soils [22–24]. The Ugandan government has strongly

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supported industrialization over the past few years, which has resulted in the loss of wetlands and the natural vegetation that filters heavy metals. As a result, manufacturing industries have grown quickly in the city, with little consideration given to the environmental effects that come with them [25]. The purpose of this study was to assess the health risk of heavy metals and Norms (^{226} Ra, ^{232}Th, and ^{40}K) in Kampala.

2 Materials and Methods

2.1 Sample Collection

Ten fresh yam samples and surrounding soils were collected randomly from five wetlands located in Kampala. The wetlands included Bukasa, Kitintale, Portbell, Busega, and Lubigi. This made a total of 50 samples for the yams and the soils.

2.2 Heavy Metal Determination

In the laboratory, sample portions of yam weighing 10–20g were dried in an oven at 100°C for approximately 4 hours to a constant weight. In the laboratory, 10–20g of yam sample portions were oven dried at 100^{o}C to a constant weight for approximately 4 hours. The dried samples were cooled at room temperature and ground to fine powder and 5g of the powder sample in a crucible was later ashed at 450^{o} C in a muffle furnace for approximately 12 hours. The ash was then cooled to room temperature and digested with 5 ml of 12M HNO_3 to produce a liquid solution. To produce a clear solution, the mixture was heated and 5 ml of H_2O_2 was added. By aspirating the solution in an AAS after completing the necessary standardization procedures for Pb and Hg from their respective hollow cathode lamps, the total concentrations of the metals in the digested samples were determined. The Atomic absorption spectroscopy detection parameters are shown in Table 1.

Table 1 The Atomic absorption spectroscopy detection parameters.

Heavy Metal	Pb	Hg
Wavelength (nm)	217.0	253.7
Detection limit (μg/g)	0.004	0.160
Slit Width (nm)	1.0	0.5
Oxidant	Air	Argon
Fuel	acetylene	Noflame

2.3 Radioactivity Measurement

The wetland soil samples obtained from the wetlands were dried in an oven at 100^{o}C for approximately 24 hours to remove the moisture. After the samples were dried, they were ground using a ceramic motor into a fine powder and were sieved to a uniform particle size using a 1mm sieve. Approximately 250g of the powdered soil samples were stored in an air tight malinelli beaker in order to ensure accumulation and minimize escape of gaseous daughter products that get emitted during the decay process. The soil samples were stored in the beakers for approximately 28 days in order to obtain radioactive secular equilibrium between the NORMs and their daughter products. The activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K present in the soil samples were detected using a gamma ray spectrometer coupled with a NaI(Tl) scintillation detector. The samples were counted for approximately 6000 seconds to obtain a clear spectrum and the background spectrum was deducted from the gross spectrum of each sample to produce the net soil sample spectrum. The evaluation of ^{232}Th, ^{226}Ra and ^{40}K was done using their corresponding spectral lines from the net soil sample spectrum [26].

2.4 Risk assessment

The radiation hazard index was used to account for the combined effect of the radionuclide activity concentrations in a material. Hazard indices were calculated using activity concentration of ^{40}K, ^{226} Ra and ^{232}Th that was determined from the collected wetland soil samples.

Radium Equivalent Activity (Ra_{eq})

For the past 40 years, Ra_{eq} has been used to assess the radiological hazard of radioactivity in agricultural soils [27]. It is calculated using equation (1) [28, 29].

$$Ra_{eq} = A_{Ra} + \frac{10}{7}A_{Th} + \frac{10}{130}A_K \quad (1)$$

Equation (1) makes an assumption that the gamma dose rates from 1 Bq kg^{-1} of ^{226} Ra, 0.7 Bq kg^{-1} of ^{232}Th, and 13 Bq kg^{-1} of ^{40}K are the same. In this situation, the limit value may have an annual effect of 370 Bq kg^{-1}, which is equal to the dose rate limit of 1.0 mSv y^{-1} [30–32]. Since Ra_{eq} is frequently used to determine exposure, radiation hazards could arise from any radium equivalent activity concentration that exceeds 370 Bq kg^{-1} [33].

External Hazzard Index (H_{ex})

H_{ex} is an index used to determine the amount of radiation exposure that occurs indoors as a result of gamma radiation exposure outside. It is calculated using the equation (2).

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (2)$$

For negligible radiation, the value obtained must ≤ 1 . This can only be possible if Ra_{eq} is less than 370 Bq kg^{-1} [34].

Internal hazard Index (H_{in})

H_{in} was calculated following equation (3)

$$H = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (3)$$

H_{in} should be less than unity for radiation hazard to be considered negligible [35]. This is only possible if the Ra_{eq} is less than 185 Bqkg^{-1} [33].

Absorbed Dose rates(D)

The absorbed dose rate, D (nGy h^{-1}), present in the swamp soil samples was calculated to estimate the external radiation exposure [36] using equation (4).

$$D = 0.462A_{Ra} + 0.604A_{Th} + 0.042A_K \quad (4)$$

Annual effective dose rate (AEDR)

With an outdoor occupancy of 20% and 80% for indoor, the average effective dose equivalent received by a member is calculated using a factor of 0.7 Sv/Gy [33, 37]. The Indoor annual effective dose is calculated using equation (5) while the outdoor annual effective dose is determined by equation (6) in (mSvy^{-1}).

$$Indoor = D \times 8760 \times 0.8 \times 0.7SvGy^{-1} \times 10^{-6} \quad (5)$$

$$Outdoor = D \times 8760 \times 0.2 \times 0.7SvGy^{-1} \times 10^{-6}$$

3 Results and Discussion

3.1 Heavy Metal Concentration

Elevated concentrations of lead and mercury are shown in all the sampled wetlands; Bukasa (BUK), Kitintale (KIT), Portbell (FORT), Busega (BUS), and Lubigi (LUB). Figure 1 shows that the mean concentrations of Pb in all the samples were above the acceptable range of 10 to 50 mg kg^{-1} in edible plants and above the orders of 2 to 4 mg kg^{-1} of Hg which are the maximum permissible in contaminated regions. These indicate a negative impact on disposal area's environmental quality. The burning of lead contaminated or containing products and disposal of untreated lead and mercury contaminated waste into the wetlands could have highly contributed to the high heavy metal concentration values in all the sampled wetlands. This will most likely result into increased Pb and Hg toxicity exposure levels to the population that practice agriculture and depend on the wetlands for food production. The informal disposal of waste into Nakivubo Channel and other wetlands by surrounding industries and the city waste also has a great pollution risk to the environment and the wetland soils, hence calling for strict and regular monitoring of heavy metals in the wetlands especially before their release in order to protect both the environment and human health.

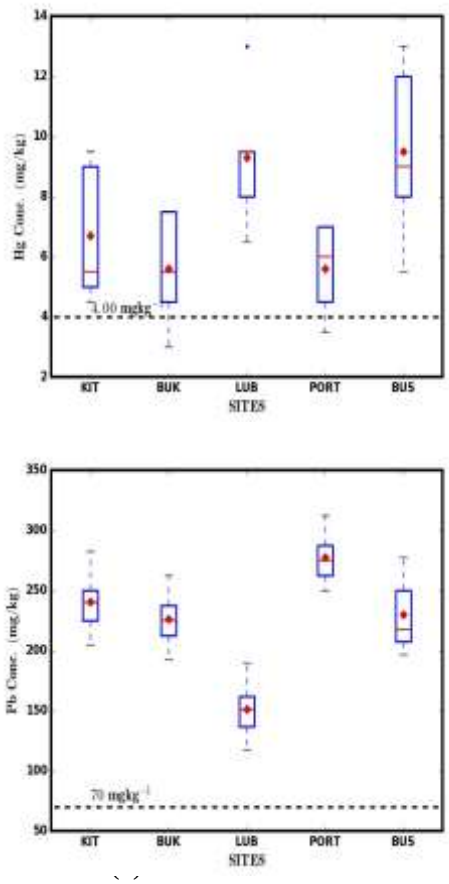

Fig.1: Box and whisker plots showing Hg and Pb concentrations.

3.2 Activity concentrations of NORMs

The activity concentrations of NORMs (^{232}Th, ^{226}Ra and ^{40}K) in the sampled wetlands were found to vary among the sampled areas as shown in Figure 2. These variations in the activities could be attributed to the surface geochemistry, physical soil properties and bioaccumulation from the disposed waste into the wetlands. The study showed high activity values of ^{232}Th and ^{226}Ra in the wetland soil samples and this may be due to thorium and uranium mineralization naturally in the soils and the content in the waste discharges. Potassium is chemically highly soluble in water and has increased concentration values in dry and rocky soils. The high moisture and organic matter content in the wetland soils attributed to the low levels of potassium in almost all the wetlands as shown in Figure 2. The study however indicates slightly elevated concentration values of ^{40}K in Portbell and this could be due to the run off of felsic rocks from the nearby mine.

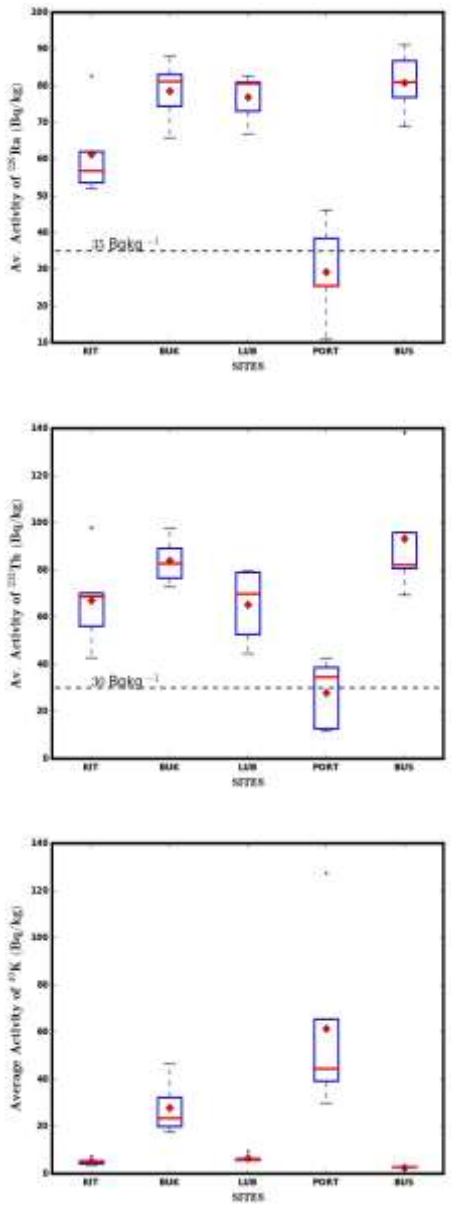

Fig.2: Box and whisker plots showing average activity for ^{226}Ra, ^{232}Th, and ^{40}K.

Table 2 shows the Pearson correlation coefficients between naturally occurring radionuclides in soil samples and heavy metals in yams. There was a moderate correlation in Pb and ^{226} Ra, according to the data in Table 2. Significant correlations were observed between ^{226}Ra and ^{232}Th, as well as with Pb and Hg.

Table 2 Correlations between heavy metals and naturally occurring radioactive materials

	^{226}Ra	^{232}Th	^{40}K	Hg	Pb
^{226}Ra	1.000				
^{232}Th	0.777	1.000			
^{40}K	0.477	0.499	1.000		
Hg	0.225	0.309	-0.447	1.000	
Pb	-0.541	-0.652	0.578	0.665	1.000

3.3 Radium Equivalent Activity (Ra_{eq})

It is observed in Figure 3 that Busega wetland shows a high Ra_{eq} mean concentration value of 215.6 Bq kg^{-1}. However, low values were observed in Portbell wetland with a mean of 71.9 Bq kg^{-1}. All the values obtained were less than the world threshold value of 370 Bq kg^{-1}.

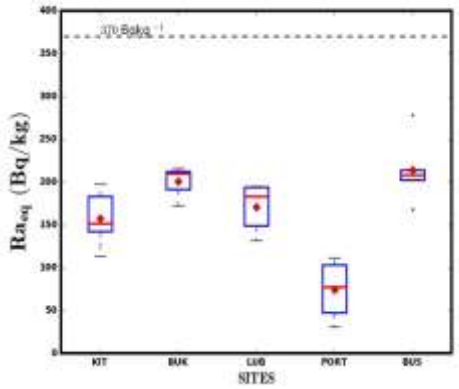

Fig. 3: A box and whisker plot of (Ra_{eq}) in all selected wetlands.

3.4 External and Internal Hazard Indices

The H_{ex} values obtained in Figure 4 (top) ranged from approximately 0.2 in Portbell to approximately 0.6 in Busega, while the H_{in} values obtained ranged from 0.3 Portbell to 0.8 in Busega wetland; Figure 4 (bottom). However, all the external and internal hazard indices calculated from all sampling sites are less than unity (1), and H_{in} values are greater than H_{ex} values obtained from all locations. The average hazard indices (both H_{in} and H_{ex}) for Portbell are relatively low in comparison to the indices of the other remaining four sampling sites.

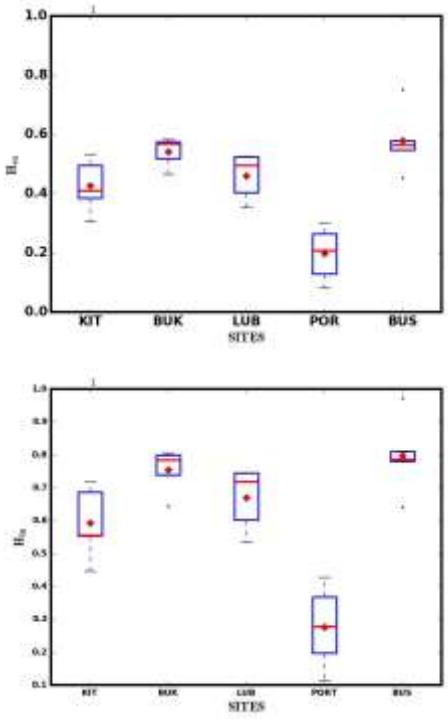

Fig. 4: Box and whisker plots of H_{ex} and H_{in} in all selected wetlands.

3.5 Absorbed Dose rates

Kampala Capital City has the highest population compared. There is an increased demand for employment and hence a high level of industrialization and agriculture for food safety [36]. This has led to some portions of different wetlands to become degraded and reclaimed for industrial and housing projects. The absorbed dose in indoor air were lowest for Portbell and highest in Busega wetland as shown in Figure 5. The values were in the ranged from 33.9 nGy h^{-1} to 94.8 nGy h^{-1} with an overall mean dose rate of 73.0 nGy h^{-1}. The absorbed dose rates were above the world acceptable value of 60 nGy h^{-1} in four of five sampled wetlands. This implies that these wetlands are not suitable for agriculture and housing projects due to the high radiation doses.

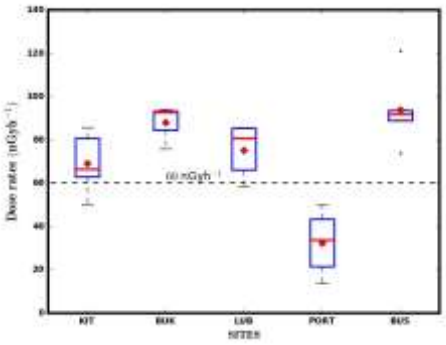

Fig. 5: Box and whisker plots of dose rates in all selected wetlands.

3.6 Indoor and Outdoor Annual Effective Dose

The Indoor annual effective dose ranged from 0.166 mSv to 0.593 mSv, while the Outdoor annual effective dose ranged from 0.044 mSv to 0.115 mSv in Portbell and Busega, respectively (Figure 6).

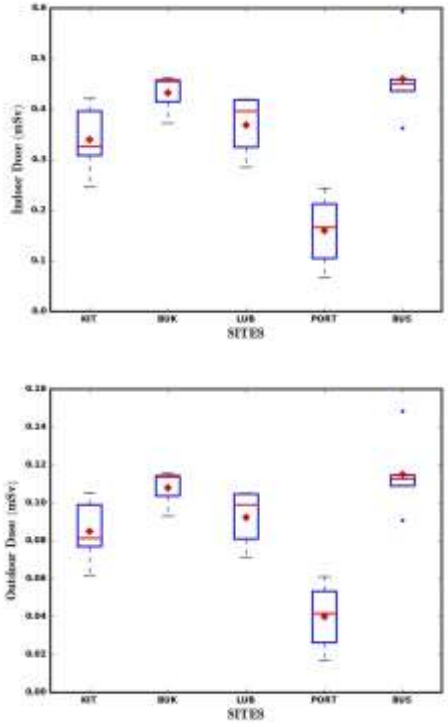

Fig.6: Box and whisker plots of Indoor and Outdoor Annual Effective Dose.

4 Conclusions

Distribution and effect of heavy metals and natural radionuclides in the Bukasa, Kitintale, Portbell, Busega, and Lubigi wetlands topsoil within Kampala has been investigated. The concentrations of Pb and Hg were above the world acceptable values in all the five sampled wetlands implying that there is less treatment of the sewage waters that are disposed of into the wetlands. The activity concentrations of ^{226}Ra and ^{232}Th in all the sampled wetlands below world mean values of these radionuclides for soil while the activity concentration of ^{40}K was generally lower than the world average value. According to statistical analysis using the Pearson correlation coefficient, there was a high correlation between ^{226}Ra , ^{232}Th and Pb while there were significantly low correlations between Hg and ^{232}Th. The Radium equivalent and hazard index values were all below the worldwide acceptable values. However, there were some sites with above the worldwide acceptable values in the absorbed dose rates including the Indoor and Outdoor annual effective dose rates. This implies that these

wetlands are not suitable for agriculture and housing projects due to high radiation doses.

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