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Evaluation of Radionuclide Concentration from Soil and Water Samples Collected from Around a Solid Waste Dumpsite

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ABSTRACT

This paper is focused on the re-evaluation of the radiological status of Eliozu dumpsite located near Port Harcourt, for which there exist a report for a previous study in 2011. Therefore, soil and groundwater samples were collected from short distances away from the dumpsite, and were subjected to radiological measurements using method of gamma-ray spectroscopy. From the results obtained, Potassium(K-40), Uranium(U-238) and Thorium (Th-232) were identified as the primary radionuclides. The mean activity concentrations of these radionuclides (K-40, U-238 and Th-232) in the soil samples were 619.77±5.74, 35.64±2.98 and 71.47±14.68 Bq/kg respectively, while that for the groundwater samples also showed: 27.50±1.93, 9.02±2.54 and 6.51±1.23 Bq/l respectively. The absorbed and equivalent dose rates were also estimated for human habitat around the dumpsite. The measured activity concentrations of radionuclides in the soil and groundwater samples for this present study was compared with the result of a previous study for the same dumpsite. It was revealed that there was significant increase in the radiological parameters. Although, the rate of radionuclide migration from soils around the dumpsite to the groundwater aquifer is slow, but there are strong indications that the dumpsite still receives radiologically contaminated solid wastes. Equivalent dose rates for radiation exposure in the community were within the WHO permissible limit.

1. INTRODUCTION

Waste can be defined as an unwanted material generated from human activities for which if not properly manage will cause harm to the inhabitants and the environment (Rao and Shantaram, 2003, Hoornweg and Bhada-Tata, 2012). These wastes may be in solid, liquid or gaseous forms, and most importantly they may contain pollutants that can be toxic or radioactive in nature. Radioactive pollutants come mainly from radioactive wastes materials which can be released from the manufacturing sector, health, mining and energy

industries. Just like other wastes, radioactive waste may also vary in origin, composition and physical state (Forsberg, 2000, Baisden and Choppin, 2009).

Generally, radioactive wastes sourced from industrial activities are called technologically enhanced naturally occurring radioactive materials (TE-NORMs). But specifically, they can be sourced from activities in nuclear fuel cycle and released as depleted uranium and spent fuel. The agriculture, mines and petroleum sectors can also release radioactive wastes in the form of metal scraps, ion exchange resins, mining wastes, scale in oil/gas pipes, oil/gas sludge and produce water, wastewater treatment and phosphate process solids (Ehirim and Itota, 2013). Others are diagnostic and therapeutic radionuclides (e.g. Co-60, I-131, Cs-137), granite, wood and coal ash, radon gas, food waste (e.g. K-40) etc. (Sutherland, 2018).

The pollution of an environment by radionuclides may arise, if radioactive wastes (or materials contaminated with radioactive wastes) are disposed in open dumpsites (or pits) without authorization, or without being subjected to proper treatment and monitoring prior to the disposal (Aderemi et al, 2011). The implication is that the radioactive wastes will degrade in the dumpsite, and release radionuclides which can be transferred into liquid medium to form solutions or leachates (Laconi et al, 2006).

The hydro-geological properties of the soils in the dumpsite environment can influence radionuclides migration within large extent area of dumpsite soil formation by several mechanisms: capillary action, eutrophication and gravity force. The radionuclides can also interact with surface and ground waters within and around the dumpsite. Consequently, soil and water environments near the dumpsite may become contaminated radiologically (Schueler and Mahlere, 2007; Abdus-Salam, 2009; De et al., 2016).

In Nigeria, there are reported studies to support these concerns. For instance, Kolo et al. (2016) found that the concentrations of Radium-226, Thorium-232 and Potassium-40 in coal samples collected in Nigeria are comparable with those of similar studies reported in literature, while Avwiri et al. (2011), Jibiri et al. (2014), Ademola et al. (2014), and Avwiri and Olatubosun (2014) have all reported independently on the assessment of radiological levels and heavy metal presence in selected dumpsites at different locations in Nigeria. The results generally indicated that the activity concentrations of Potassium-40, Radium-226 and Thorium-232 in dumpsites showed no significant radiological health hazards to the population around the dumpsites. Also, Ehirim and Itota (2013) have studied the radiological impact of solid waste dumpsite on soil and groundwater in Port Harcourt municipality using 2-D resistivity imaging and gamma-ray spectroscopy methods. The results obtained showed the presence of K-40, Ra-226 and Ra-228. The distribution of these contaminants in the formation was uneven and spotty. The activity concentrations and dose rates of the evident radionuclides in soil and water samples were permissible and were of no adverse effect. In the same vein, Oladapo et al. (2012) measured the levels of natural radionuclides in soil samples obtained around dumpsites in Lagos, Nigeria, using gamma-ray spectrometer. The results showed the presence of radionuclides: U-238, Th-232 and K-40 in soil samples from the dumpsites. The mean activity concentrations for these radionuclides were respectively for active dumpsite: 9.69±19.10, 14.49±3.22 and 409.45±86.08 Bqkg $^{-1}$, and for dormant dumpsite: 61.25 ± 21.82, 12.08 ±1.74 and 345.98 ±56.92 Bqkg $^{-1}$. The mean annual effective doses for both dumpsites were also respectively 0.28 and 0.25. Thus, by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) standards, both dumpsites were observed not to pose any immediate radiological hazards to human health.

Therefore, this study is aimed at re-evaluating the radiological status of Eliozu dumpsite in Port Harcourt, Nigeria, which was previously studied in Avwiri et al. (2011). The reason for this re-evaluation is based on the premonition of increased radiological exposure around the dumpsite environment within the time from previous study. Radionuclide migration and increased dumping of radiologically contaminated solid wastes evident from the increased urbanization and human population around the Eliozu dumpsite environment, including the rise in industrial activities in Port Harcourt were the factors considered. These factors were not considered in Avwiri et al. (2011).

2. MATERIALS AND METHODS

2.1. Materials

The main materials used for the study were soil and groundwater samples collected from around Eliozu dumpsite, Port Harcourt, Rivers State, Nigeria. Other materials used for this study were sampling device, sample container, paper tape and ice chest. A gamma-ray spectrometer which uses thallium-activated sodium iodide (NaI(TI)) as scintillator (or detector) was used for detecting and measuring the radiation emitting from the samples.

2.2. Methods

2.2.1. Collection of samples

Groundwater samples were collected from around the dumpsite at different distances (10, 18, 53, 156 and 164 m) measured from the base of the dumpsite. Plastic bottles were used for groundwater sample collection and were each labeled W1 – W5. Soil samples were also collected (at near surface depth) using hand auger from different distances (15.0, 20.0, 50, 80 and 100.0 m) measured from dumpsite edge. The soil samples were collected in black polyethylene bags, and were labeled S1 –S5 using paper tape in other to avoid contamination. The soil samples were preserved in an ice chest and transported to the laboratory for radiological analysis. At the laboratory, loss of radon was avoided by applying precautionary measures. Secular equilibrium was also established prior to gamma-ray spectrometric analysis (Zhang et al., 2014).

2.2.2. Radiation detection and measurement

A gamma-ray spectroscopy system which uses NaI(TI) as scintillator or detector was used for detecting and measurement of radiation activity. This scintillator glows or produces light when in contact with radiation emitting from the samples. The equipment was set vertically above ground, and connected to a multichannel analyzer (MCA) (see Figure 1). Before use, the detector was first enclosed in a 5 cm × 5 cm lead shield (in all side) to protect the environment from exposure and also to reduce radiation at the background (Viega et al., 2006). During measurement, soil and groundwater samples were each placed at 2, 5 and 10 cm distances from the face of the detector. The presence of radiation and quantity of radionuclides in the samples were detected and measured using gamma-ray spectrometer (with multichannel analyzer) which was calibrated before use, in other to attain the energy range of the radiation sources: 0.06 to 2.8 MeV. To calibrate the equipment, the reference materials employed were Uranium ore (RG U-1), Thorium ore (RGTh-1) and KCl powder of known activity concentrations and in line with the IAEA standards (IAEA, 2018).

In order to reduce errors in the measurement, as well as increase the efficiency of detection of the radionuclides, self-absorption of radionuclides (in the soil sample) was eliminated by ensuring that the soil samples were of similar geometries with standard solid sources. Also, adsorption of radionuclides on the walls of containers (in the groundwater sample) was eliminated by acidifying the water sample with 0.1 M HCl at the rate of 0.8 ml per liter of sample. Each sample was counted (or tested) in isolation from the three set distances from the detector face for about 30 mins while the average count per channel was recorded. The lowest level of measured activity concentration was determined from the background radiation spectrum. The absorbed dose and equivalent dose rates in respect of the radiation was also measured.

Among the list of radionuclides of natural origin, K-40, U-238 and Th-232 are the most significant for the purpose of environmental radiation protection and regulation. It also includes Radium-226 and Radon-222 which are decay products from U-238 and Th-232 respectively (Kuye and Uyigue, 2019). Thus, the specific activity concentrations (AC_K, AC_U and AC_{Th}) and absorbed dose rate (D), for K- 40, U-238 and Th-232 in

the samples were estimated using the equations proposed by Beck et al (1972) as shown in Equations 1 and 2.

$$AC = \frac{AC(s) \times m(s)}{A(s) \times M} \quad (1)$$

Where, AC = activity concentration (or radioactivity level) of sample, A = peak area of samples, AC(s) = radioactivity concentration level of standard sample, m(s) = mass of standard sample, A(s) = peak area of the standard sample, M = mass of sample.

Absorbed dose rate, D (nGy/h) for the samples were calculated relative to background concentrations of U-238, Th-232 and K-40 at all sampling points using Equation 2.

$$D = 0.042\,AC_k + 0.429\,AC_u + 0.666\,AC_{th} \quad (2)$$

Where, AC_K, AC_u and AC_{Th} are activity concentrations for K-40, U-238, Th-232 respectively while 0.042, 0.429 and 0.666 are the respective dose constants.

The conversion equivalent of dose measurement units from Gray to Sievert is expressed as in Equation 3.

$$S = Gy \times QF \quad (3)$$

Where, S = Sievert, Gy = Gray and QF = quality factor. The QF measures the relative hazard of an energy transfer by an emitted radiation of interest. QF for gamma radiation equals unity (Veiga et al., 2006).

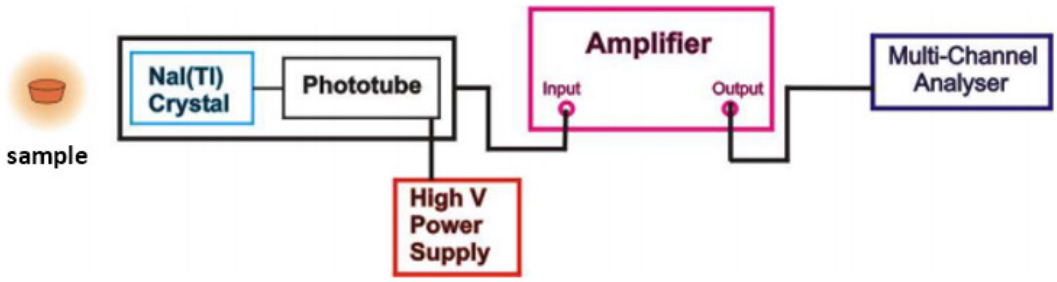

Figure 1: Schematic diagram for gamma-ray spectroscopy system using thallium-activated sodium iodide (NaI(TI)) detector

3. RESULTS AND DISCUSSION

The results obtained from this work are presented in Tables 1, 2 and 3, which represents the activity concentrations of radionuclides identified in the soil and groundwater samples collected from around Eliozu dumpsite. It also includes absorbed and equivalent dose rates for human habitat around the dumpsite. The result for the individual sample measurement is a mean of triplicate including the standard deviation.

Table 1: Activity concentration and half-life of identified radionuclide in soil and groundwater samples around dumpsite

Sample type (Soil and groundwater)	Identified radionuclide	Half-life, $t_{1/2}$ X 10^{15} (s)	Activity concentration in soil (Bq/kg)	Activity concentration in groundwater (Bq/l)
Sample 1	Potassium (K-40)	40.3	713.45 ± 2.94	33.12 ± 2.73
Sample 2			547.32 ± 5.63	19.42 ± 1.48
Sample 3			475.90 ± 8.75	31.97 ± 1.11
Sample 4			640.42 ± 6.48	28.35 ± 3.25
Sample 5			721.29 ± 5.21	24.58 ± 1.10
Mean value			619.77 ± 5.74	27.50 ± 1.93
Avwiri et al. (2011)			326 ± 66.7	24.77 ± 8.3
Sample 1	Thorium (Th-232)	443.6	81.53 ± 19.11	8.54 ± 1.24
Sample 2			60.54 ± 16.08	3.61 ± 1.03
Sample 3			79.29 ± 7.81	8.92 ± 1.09
Sample 4			111.12 ± 25.47	6.14 ± 1.38
Sample 5			24.69 ± 8.53	5.32 ± 1.12
Mean value			71.47 ± 14.68	6.51 ± 1.23
Avwiri et al. (2011)			19.27 ± 8.1	6.96 ± 2.4
Sample 1	Uranium (U-238)	141.0	61.53 ± 3.08	9.24 ± 2.10
Sample 2			28.35 ± 2.49	7.31 ± 1.82
Sample 3			31.86 ± 3.13	12.41 ± 3.11
Sample 4			30.13 ± 4.25	9.94 ± 3.14
Sample 5			26.35 ± 8.53	6.17 ± 2.50
Mean value			35.64 ± 2.98	9.02 ± 2.54
Avwiri et al. (2011)			27.41 ± 10.0	7.92 ± 2.7

3.1. Activity Concentration and Absorbed Dose from Around Dumpsite Soil Radionuclides

K-40, U-238 and Th-232 were radionuclides identified in the soil samples. K-40 activity concentration in the soil samples was significantly higher than those of U-238 and Th-232 radionuclides. The values ranged from 475.90±6.48 to 721.29±5.21 Bq/kg having a mean value of 619.77±5.74 Bq/kg (Table 1). The presence of a higher activity concentration of K-40 in the soil samples could be adduced to the dominance of certain wastes in the dumpsite such as banana, coal and fertilizer which are rich in isotopic potassium (Ademola et al., 2014). The lesser value of activity concentrations for Th-232 and U-238 in the dumpsite as indicated in Table 1 (Th-232(mean): 71.47±14.68 Bq/kg and U-238(mean): 35.64±2.98Bq/kg) are clear indications of low concentrations of wastes materials in the dumpsite that carries TE-NORM sources (e.g. spent fuel, nuclear tracer and nuclear diagnostic wastes). Comparing the measured activity concentrations of radionuclides in the soil samples around the Eliozu dumpsite in this present study with that of a previous study in Avwiri et al. (2011) for the same dumpsite showed higher concentrations of radionuclides. The absorbed dose rates from the dumpsite soils ranged from 32.76±7.10 to 90.36±18.63 nGy/h with a mean value of 65.23±11.77 nGy/h, while the equivalent dose ranged between 0.287±0.21 to 0.705±0.21 mSv/yr, having a mean value of 0.572±0.21 mSv/yr. Higher values of absorbed and equivalent dose were also evident when compared with Avwiri et al (2011) for the same dumpsite (see Table 2). These variations between the present study and that of Avwiri et al (2011) are indications that the radiological properties of Eliozu dumpsite are on the increase, which by implication may mean that the dumpsite still receives radiologically contaminated solid wastes.

Table 2: Activity concentration and dose rate analysis of radionuclide identified in soil sample around dumpsite

Soil sample	Absorbed dose rate (nGy/h)	Equivalent dose rate (mSv/yr)	Permissible dose limit (mSv/yr) (IAEA, 2018)
S1	80.44 ±14.17	0.705±0.21	1.0 ± 0.21
S2	54.99 ± 12.01	0.482±0.21	1.0 ± 0.21
S3	67.62 ± 6.95	0.593±0.21	1.0 ± 0.21
S4	90.36 ± 18.63	0.792±0.21	1.0 ± 0.21
S5	32.76 ± 7.10	0.287±0.21	1.0 ± 0.21
Mean Value	65.23 ± 11.77	0.572±0.21	1.0 ± 0.21
Avwiri et al. (2011)	38.17 ± 12.5	0.334 ±0.1	1.0 ± 0.21

3.2. Activity Concentration and Absorbed Dose of Radionuclides in Groundwater Sample Taken Around the Dumpsite

Although the concentration of radioactivity was low for all radionuclides identified in the groundwater samples, but it had a similar trend like the soil samples wherein higher activity concentrations were evident in K-40 than in U-238 and Th-232 (see Table 1). Therefore, K-40 had activity concentration values of range 19.42±1.48 to 33.12±2.73 Bq/l with a mean of 27.50±1.93 Bq/l. It is also observed that U-238 activity concentration is higher than that of Th-232 as indicated in Table 1, wherein U-238 had range 6.17±2.5 to 12.41±3.11 Bq/l and mean 9.02±2.54 Bq/l while Th-232 had range 3.61±1.03 to 8.92±1.09 Bq/l and mean 6.51±1.23 Bq/l. In the same vein, higher activity concentrations for radionuclides in groundwater samples taken around the Eliozu dumpsite were observed in the present study than that in a previous study in Avwiri et al (2011), except for Th-232 wherein the value in Avwiri et al (2011) was slightly higher by 6.9 %. Absorbed doses for groundwater samples were ranged from 4.73 ± 1.71 to 9.87 ± 2.11 nGy/h which had a mean activity concentration of 7.24±1.99 nGy/h. The equivalent doses also ranged from 0.042±0.04 to 0.087±0.04 mSv/yr with a mean value of 0.064±0.04 mSv/yr (see Table 3). Absorbed and equivalent dose in Avwiri et al. (2011) were observed to be respectively slightly higher than the result of this present study by 25 and 23 %. The overall observations for activity concentration and absorbed dose rate for both soil and groundwater samples collected from near the dumpsite for which higher activity concentrations were observed in the soil than in the groundwater samples, indicate of a slow migration rate for radionuclides from the soil to the water aquifer around the Eliozu dumpsite. However, the evaluated equivalent absorbed doses of radiation in the human habitat near Eliozu dumpsite are within the WHO standard for human permissible limit for radiation dose, which is not more than 1 mSv/yr. Hence, Eliozu dumpsite community is still radiologically safe for humans.

Table 3: Activity concentration and dose rate analysis of radionuclide in groundwater sample around dumpsite

Groundwater sample	Absorbed dose rate (nGy/h)	Equivalent dose rate (mSv/yr)	Permissible dose limit (mSv/yr) (WHO, 2011)
W1	8.73 ± 1.84	0.077 ± 0.04	1.0 ± 0.21
W2	4.73 ± 1.71	0.042 ± 0.04	1.0 ± 0.21
W3	9.87 ± 2.11	0.087 ± 0.04	1.0 ± 0.21
W4	7.27 ± 2.40	0.064 ± 0.04	1.0 ± 0.21
W5	5.60 ± 1.87	0.049 ± 0.04	1.0 ± 0.21
Mean value	7.24 ± 1.99	0.064 ± 0.04	1.0 ± 0.21
Avwiri et al. (2011)	9.03 ± 3.1	0.079 ± 0.03	1.0 ± 0.21

4. CONCLUSION

This study has re-evaluated the radiological status of the Eliozu dumpsite near Port Harcourt. Using gamma-ray spectroscopy, Potassium(K-40), Uranium(U-238) and Thorium (Th-232) were identified as primary radionuclides in the soil and groundwater samples collected around the dumpsite. Evidence of radiological parameters increase was significant for the soil and groundwater samples, on comparing the results of the present study with that of a previous one. Although, the rate of radionuclide migration from soils around the dumpsite to the groundwater aquifer is slow, there are strong indications that the dumpsite still receives radiologically contaminated solid wastes. Since the equivalent absorbed dose of radiation measured in this present study for soil and groundwater samples taken from around the human habitat near Eliozu dumpsite falls within IAEA and WHO permissible limit for human radiation dose, hence, the level of radiation exposure around Eliozu dumpsite is still permissible. However, agencies of Government responsible for checking radiation exposure should take urgent step in controlling the dumping at Eliozu dumpsite.

5. CONFLICT OF INTEREST

There is no conflict of interest associated with this work.

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