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Evaluation of Natural Radioactivity and Radiation Hazards of Soils around Petroleum Products Marketing Company using Gamma Ray Spectrometry

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Abstract

This study aimed at evaluating natural radioactivity and radiation hazards of soils around a petroleum product marketing company due to anthropogenic influences using gamma ray spectrometry. Top soils (5 cm deep) were collected from diverse locations inside the premises of the Company using standard analytical technique, while control samples were acquired at approximately 200 m from the study area. Mean activity concentrations of the radionuclides in the test soils were 89.36 Bq/kg for ^{40}K, 92.1 Bq/kg for ^{238}U and 10.95 Bq/kg for ^{232}Th, which are more than that of control samples with statistically significant differences. Assessment of the radiological levels of the oil-contaminated soils were done using internal hazard index, external hazard index, annual gonadal dose equivalent, annual effective dose rate, radium equivalent, total absorbed dose rate and representative level index ($I_{\dot{Y}}$). The results showed that mean annual equivalent dose rate of the oil-contaminated soil (0.46 mSv/yr) fell below the permissive annual dose limit (1.0 mSv/yr). Mean internal hazard index (0.55 Bq/kg) and external hazard index (0.30 Bq/kg) values were less than the minimum permissible limit (1.0). Mean annual gonadal dose equivalent value (358.46 μSvy^{-1}) of the radionuclides was close to the world average of 300 μSvy^{-1}, while the mean representative gamma index value (0.78) was also less than unity. Estimated mean value (114.65 Bq/kg) of the radium equivalent was also below the world average of 370 Bq/kg, the values are also more than that of the control samples. Though the soils exhibited low gamma radiation, however lengthy term exposure may pose an inherent cancerous ill-health. It is vital to repeat this investigation periodically to avert precarious effects of anthropogenic influences; also indiscriminate discarding of oils should be discouraged.

Keywords: Gamma ray spectrometry, radionuclide, refined petroleum products, risk assessment, soil.

Introduction

Radioactivity is a phenomenon associated with unstable atomic nuclei which spontaneously decompose emitting particles such as beta, alpha and neutron or electromagnetic radiation in the form of gamma rays (UNSCEAR 2010). Radioactivity of a particular radioisotope can be accounted for by decay constant and half-life ($t_{1/2}$). Half-life is the time required for the half of the original value of a radioisotope to decay, however ten half-lives are required for a radionuclide to be removed (Murray 1989). The time required for the total decay of radioisotopes ranges from seconds to billions of years, and this varies from one radioisotope to another (ATSDR 1999).

The existence of radionuclides in the environment can occur naturally either as cosmogenic or terrestrial radionuclides. Some of the cosmogenic radionuclides include ^{14}C, ^{3}H, ^{39}Ar, $^{7,10}Be$, and ^{26}Al. They are the major radionuclides generated as a result of the interaction between cosmic rays and atmospheric gases. Rocks, minerals, and soil, generally are composed of naturally occurring radioactive materials, and they are known to have half-lives longer than usual and are referred to as terrestrial radionuclides (ATSDR 1999, Botezatu and Iacob 2004). Among the major terrestrial radionuclides are ^{232}Th, ^{40}K and ^{238}U decay series. The world average soil activity values are 32 Bq/kg for ^{226}Ra, 420 Bq/kg for ^{40}K, and 45 Bq/kg for ^{232}Th (UNSCEAR 2000).

Anthropogenic activities that are linked to the advancement of energy production and usage from nuclear materials have been attributed to be a major source of environmental degradation. From the mid-19th century, contamination due to radioactive sources has been identified through discharges as a result of anthropogenic radionuclides, thus making energetic radiation an important factor in an ecosystem including some other degradations that are associated with soil such as biological, physical, and chemical degradations (Aleksakhin 2009, Carvalho et al. 2014). The radioactive contamination of the environment requires utmost priority attention due to the damaging effects of the ionizing radiations on tissues of living organisms, but their occurrence is very rare. The negative impacts are proportional to the magnitude of energy absorbed, exposure duration, penetrating power of the radiation, and the rate of reproduction of cells of certain tissues (Smičiklas and Šljivić-Ivanović 2016).

Petroleum depots are generally located in remote areas. Experience shows that with the passage of time, it gets surrounded by residential and/or industrial installations due to possible gains. However, as human dependency on crude-oil increases, the dangers that are accompanied with it also increase. Spillages and leakages as a result loading and offloading of refined petroleum products in the premises of depots and dewatering of the refined oil storage facilities have adverse effects on the receiving sinks.

The existence of petroleum products has significant environmental effects due to diverse petroleum development processes. It has been reported that crude oil usually accumulates radionuclides through its handling and refining processes, source rocks, sea salt intrusion, and migration to its reservoir. These contaminants can enter the ecosystems, affect flora, fauna and eventually enter the food chains. There is a need to assess the concentrations of these contaminants using soil in the premises of the study area as indicator of environmental contamination, hence this study. Thus, this study determined the human and ecological risks associated with oil depot practices with respect to the radioactivity levels of oil-impacted soils in the premises of Pipelines and Product Marketing Company (PPMC), Ibadan, Nigeria.

Experimental

Study area

The area studied is within Ibadan, Oyo State capital, Nigeria; it is a city in South-Western Nigeria located on latitude 7º15'2" N and longitude 5º12’36” E. It is situated within the tropical rainforest of Nigeria (Wikipedia 2021). A map showing the sampling points in the study area is given in Figure 1.

The oil depot of the NNPC in Nigeria that supplies products to retailing outlets and end users in southwestern part of the country, as well as some northern states is suited in Apata, Iddo local government area of Oyo State. The depot has the largest loading facilities in southwestern Nigeria. It was established in a remote area suitable for depot activities, however, as a result of population explosion, expanse of land close to the perimeter fence of the storage facilities are currently being exploited for residential purposes and the occupants engage in farming activities even within and around the perimeter fence of the depot. A shallow stream transverses the depot premises and extends to the residential area. There may be great health and environmental hazards to

humans, aquatic life and other forms of life in the area as a result of infiltration, plant uptake, bioaccumulation and biomagnifications in the food chains when contaminated with oil products.

Some of the refined petroleum products that are stored by the depot include Automotive Gas Oil (AGO or diesel), Dual Purpose Kerosene (DPK or kerosene), and Premium Motor Spirit (PMS or petrol or gasoline).

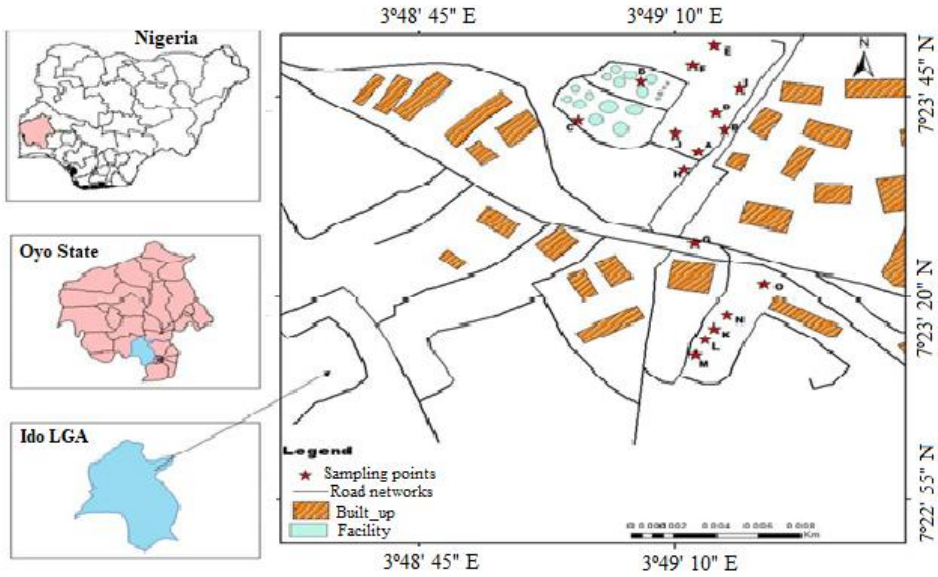

Figure 1: Map showing the sampling locations of the study area.

Sample collection and preparation

Surface soil samples from different points at the Pipelines and Product Marketing Company (PPMC), Apata, Ibadan, Nigeria were collected. This was done by taking the soils (10 samples) using hand trowel and placing them into labelled air tight containers, then conveyed to the laboratory for analysis. Five control samples were collected from outside the study area where there were little or no anthropogenic influences. Table 1 shows the sampling point coordinates.

Table 1: Sampling sites and their coordinates

S/N	Site	Coordinates
1.	A	07°23.592' N 003°49.187' E
2.	B	07°23.640' N 003°49.233' E
3.	C	07°33.009' N 003°24.519' E
4.	D	07°23.677' N 003°49.221' E
5.	E	07°23.658' N 003°49.080' E
6.	F	07°23.742' N 003°49.115' E
7.	G	07°23.406' N 003°49.176' E
8.	H	07°23.564' N 003°49.170' E
9.	I	07°23.743' N 003°49.264' E
10.	J	07°23.633' N 003°49.161' E
11.	CTR 1	07°23.260' N 003°49.195' E
12.	CTR 2	07°23.250' N 003°49.182' E
13.	CTR 3	07°23.231' N 003°49.173' E
14.	CTR 4	07°23.280' N 003°49.206' E
15.	CTR 5	07°23.337' N 003°49.265' E

The apparatus used in the study were washed using non-ionic detergent solution, rinsed thrice with distilled water, soaked for 48 hours in 2 M HNO_3 before usage, rinsed with the distilled water again, and subsequently dried at 70 ºC temperature overnight in Gallenkamp oven. Solvents and reagents used in this research work were of analytical grade.

Rocks, pebbles, and stones were removed from the soils prior to air–drying at room temperature and then ground with a pre-cleaned agate mortar and pestle. The pulverized soils were made to pass through a 2 mm mesh size sieve to accomplish homogeneity of the soil particle magnitude for the radioactivity level determination.

Radioactivity determination

The pulverized and homogenized dried soils were sealed for a month by means of cylindrical plastic vessels enfolded with a masking tape so as to achieve radiative secular equilibrium before being determined using the gamma-ray spectrometer at the Environmental Laboratory, Department of Physics and Engineering Physics, Obafemi Awolowo University, Ile-Ife, Nigeria. Gamma counting equipment employed for radionuclide content measurement composed of a calibrated NaI (Tl) detector.

Energy and efficiency calibrations were also accomplished by a well-calibrated mixed source soil standard (IAEA-375 reference soil) acquired from the International Atomic Energy Agency (IAEA). The energy and efficiency calibrations aided the identification and quantification of the radionuclides in the soils. Analysis time for each sample was 10 hours, while the convention standard and background were strictly adhered to. The detector of the spectrometer was sealed off in 100 mm thick lead shield in order to curtail the interfering consequences of natural radiation from the atmosphere where the measurement was being done (IAEA 2009). A SAMPO 90 computer programme was used to analyze resultant spectra. This programme is designed to be able to match the gamma energies at a number of levels detected in the samples to a library of potential isotopes obtainable in the standard. Since secular equilibrium was recognized, the specific activities of ^{232}Th (as ^{208}Ac) and ^{238}U (as ^{214}Bi) in the separate parent nuclides of the natural decay series of thorium and uranium were measured indirectly from the gamma energies released by their progenies. Therefore, the activity concentration of ^{214}Bi (determined from its 1760 keV or 1.760 MeV gamma-ray peak) was selected to offer an estimate of ^{226}Ra (^{238}U) in the samples, while that of the daughter radionuclide ^{208}Ac (determined from its 911.2 keV or 0.9112 MeV gamma-ray peak) was selected as an indicator of ^{232}Th, while the non-series of ^{40}K was directly acquired through its 1460.8 keV or 1.4608 MeV gamma line. The net area under the parallel peaks in the energy spectrum was computed by subtracting counts due to Compton scattering of higher peaks and other background sources from the total area of the peaks. From the net area, the activity concentrations in the samples were achieved using the procedures reported by Jibiri and Emelue (2008).

$$C\ (Bq/kg) = kC_n \qquad (1)$$

Where $K = \frac{1}{EP\gamma Ms}$, C = activity concentration of the radionuclide in the sample given in Bq/kg, Cn = count rate under the corresponding peak, E = detector efficiency at the specific gamma-ray energy, P_γ = absolute transition probability of the specific gamma-ray, and M_s = mass of the sample (kg). The detection limit of a measuring system defines its operating proficiency without the interference of the sample. The detection limit (DL) in Bq/kg essential to assess the minimum detectable activity in a sample was achieved:

$$DL\ (Bq/Kg) = 4.65\frac{\sqrt{Cb}}{tb}.K \qquad (2)$$

Where C_b is the net background count in the corresponding peak, t_b is the background counting time (s), k is the factor that converts cps (counts per second) to activity concentration (Bq/kg) (Metzger et al. 2002, Abey et al. 2017).

Data handling

The analysis of geochemical data collected in this research involved the use of statistical approaches which are descriptive statistics (range, mean, standard deviation), and t-test. Additionally, assessment of the radiological level of the soils were performed through external hazard index (H_{ex}), annual gonadal dose equivalent (AGDE), annual effective dose rate (AEDR) and radium equivalent (Ra_{eq}), total absorbed dose rate (ADR), internal hazard index (H_{in}), representative level index ($I_{\dot{Y}}$).

Radiological impacts

The absorbed dose rate (ADR) of soils under investigation was in conformity with (UNSCEAR 2000) using the equilibrium expression below when air from external radiation (gamma) at minimum 1 m above the ground level due to accessible of evenly distributed natural radionuclides

$$ADR\ (nGy/hr) = 0.0417A_K + 0.462A_U + 0.604A_{Th} \quad (3)$$

A_K, A_U and A_{Th} are the activity concentrations determined in Bq/kg for ^{40}K, ^{232}Th (as ^{208}Ac) and ^{238}U (as ^{214}Bi), respectively. Nonetheless, secular equilibrium was anticipated for calculation of dose.

The radiological hazard assessment for populations living around the PPMC was appraised in terms of the Annual Effective Dose Rate (AEDR), thus gives explanations for the dose rate that will be operative in causing biological damage to human tissues (Veiga et al. 2006). The annual effective dose rate was calculated by converting the absorbed dose rate in air to the effective dose and the outdoor occupancy by dose conversion coefficient due to naturally emanating radionuclides (UNSCEAR 2000). The AEDR was determined by embracing the formula stated by Jibiri and Adewuyi (2008)

$$E = TfQD\text{Ɛ} \quad (4)$$

Where: T = the time in seconds in a year (D, 8760) = absorbed dose rate in air (nGy/hr)E = the effective dose rate measured in mSv/yr, f = the occupancy factor that corrects the average time spent outdoors in the sites (0.2), Q quotient = the effective dose rate and absorbed dose rate in air (0.7 Sv/Gy), while Ɛ is the factor translating nano (10^{-9}) into micro (10^{-6}).

The internal exposure to ^{222}Rn and its hazardous progenies into the respiratory systems is ordered by the internal hazard index (H_{in}) and it is as specified below.

$$H_{in} = (A_{Ra}/185 + A_{Th}/259 + A_K/4810) \leq 1 \quad (UNSCEAR\ 2000) \quad (5)$$

A_U substituted the progeny A_{Ra} in the calculation, while decay product ^{226}Ra substituted the parent nuclide ^{238}U, however, there could be inequality between ^{238}U and ^{226}Ra. It is given as 1.03 by UNSCEAR (2000). The two factors which can explain rate of exposure for an individual are: the outdoor duration and the concentration of radionuclides in the soil (Avwiri and Ononugbo 2012). A widely established hazard index for external exposure can be denoted as external hazard index H_{ex} and the equation is as presented as follows:

$$H_{ex} = (A_{Ra}/370 + A_{Th}/259 + A_K/4810) \leq 1 \quad (UNSCEAR\ 2000) \quad (6)$$

Bone surface cells, gonads, and the active bone marrow are recognized as the organs of concern (UNSCEAR 2000). Therefore, Annual Gonadal Dose Equivalent (AGDE) for the residents of the area under this study as a result of the specific activities of ^{40}K, ^{226}Ra, and ^{232}Th was carried out using the equilibrium expression below:

$$AGDE\ (\mu Svy^{-1}) = 3.09\ A_{Ra} + 4.18\ A_{Th} + 0.314\ A_K\ (Arafa\ 2004) \quad (7)$$

Representative gamma index $I_{\gamma r}$ is additional radiological factor which can be calculated by the equation:

$$I_{\gamma r} = A_{Ra}/150\ Bqkg^{-1} + A_{Th}/100\ Bqkg^{-1} + A_K/1500\ Bqkg^{-1} \quad (8)$$

The activity level in terms of mass which can be credited to ^{226}Ra, ^{232}Th and ^{40}K is known not to be fairly distributed throughout in sample (Tufail et al. 2006). Radium equivalent activity (Ra_{eq}) offers information with respect to the non-uniformity in the

exposure of soil to radiation. It is thus an object that reflects the hazards linked to radiation with ^{226}Ra, ^{232}Th and ^{40}K. It can be determined as (Rahman et al. 2008):

$$Ra_{eq} = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_K \quad (9)$$

Nonetheless, A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively, but A_U generally substitutes A_{Ra} in calculations.

Results and Discussion

Activity concentrations of radionuclides in the oil-impacted soils

The radioactive daughters of ^{238}U are ^{214}Bi, ^{214}Pb and ^{226}Ra; likewise, the daughters of ^{232}Th are ^{208}Ti and ^{228}Ac. ^{40}K is also naturally occurring but a non-decay series radioactive isotope. Employing the gamma spectrometric analysis, three naturally occurring radionuclides namely ^{40}K, ^{232}Th (as ^{208}Ac) and ^{238}U (as ^{214}Bi) were measured in the oil-impacted and control soils. In the oil-impacted soils (n = 10) the mean activity concentrations ± standard deviation of ^{40}K, ^{232}Th and ^{238}U were 89.36 ± 2.48 Bq/kg, 10.95 ± 1.14 Bq/kg and 92.11 ± 3.37 Bq/kg, respectively. For the control soils (n = 5) the mean activity concentrations ± of ^{40}K, ^{232}Th and ^{238}U were 77.70 ± 2.85 Bq/kg, 8.82 ± 1.13 Bq/kg and 72.38 ± 3.80 Bq/kg, respectively (Figure 2).

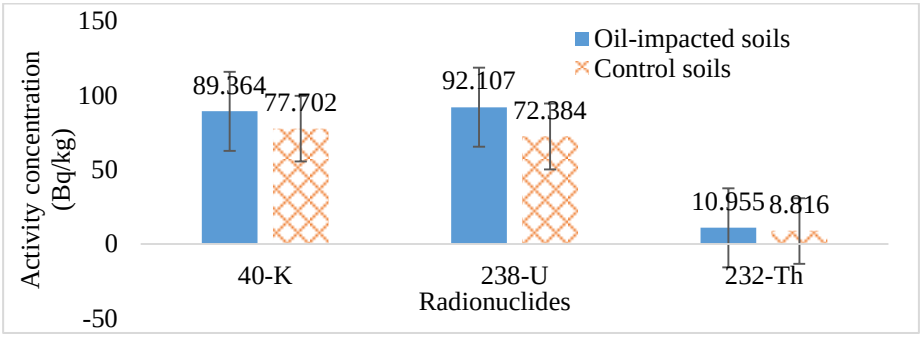

Figure 2: Comparison of the activity concentrations of the identified radionuclides in the oil-impacted and control soils.

Table 2 shows the comparison of the activity concentrations of the identified radionuclides in the oil-impacted and control soils. Of the three radionuclides, ^{238}U had the highest activity in the oil-impacted soils, while ^{40}K had the highest activity in the control soils. Generally, the mean activity concentrations of the radionuclides were higher in the oil-impacted soils than that of the control soils. This suggested that anthropogenic activities, most particularly indiscriminate discharges of oils within the oil depot must have contributed to the levels of radionuclides in the soils. Comparison with similar studies showed that these values were also less than those reported by Olalekan and Adebiyi (2020), as well as Adebiyi and Ore (2020) on researches carried out on petroleum product retailing stations and mechanic workshops in Ile-Ife, Nigeria, respectively, except for ^{232}Th in the report of Adebiyi and Ore (2020) (Table 2).

Table 2: Comparison of the activity concentrations of the identified radionuclides in the oil-impacted soils with similar studies (Mean ± STD) Bq/kg

Radionuclides	This study (n = 10)	Olalekan and Adebiyi 2020	Adebiyi and Ore 2020
^{40}K	89.36 ± 2.48	131.11 ± 3.76	51.52 ± 0.06
^{238}U	92.11 ± 3.37	106.03 ± 3.68	132.13 ± 0.08
^{232}Th	10.95 ± 1.14	17.17 ± 1.56	0.89 ± 0.16

STD = Standard deviation.

T-test results

A comparison of the activity concentrations of the radionuclides in the oil-impacted and control soils by t-test values at 95% confidence interval is obtainable in Table 3.

Table 3: T-test comparison of the activity concentrations of the radionuclides in the oil-impacted and control soils at 95% confidence interval

Radionuclides	$t_{experimental}$	p-value	Remark
^{40}K	1.810	$p > 0.05$	NSD
^{238}U	3.358	$p < 0.05$	SD
^{232}Th	3.292	$p < 0.05$	SD

SD = Significant difference, NSD = No significant difference, $t_{critical} = 2.13$.

The statistical analysis indicated significant difference if $t_{experimental}$ value is greater than 2.13 at 95% confidence limit and vice versa. The results of the t-test showed that there is a significant difference between the activity concentrations of ^{238}U and ^{232}Th in this study with the exception of ^{40}K.

Radiological assessment of the identified radionuclides in the oil-impacted soils

The sum-ups of the radiological assessments of the oil-impacted and control soils are presented in Tables 4 and 5, respectively.

Table 4: Absorbed dose rate, annual equivalent dose rate, internal hazard index, external hazard index, annual gonadal dose equivalent, representative level index and radium equivalent activity of the radionuclides in the oil-impacted soils

Value	ADR (nGy/hr)	AEDR (mSv/yr)	H_{in}(Bq/kg)	H_{ex}(Bq/kg)	AGDE (μsvy^{-1})	$I_{\gamma r}$	Ra_{eq}
Range	38.40–73.07	0.35–0.64	0.40–0.77	0.22–0.42	260.72–494.78	0.56–1.08	83.06–158.51
Mean	52.89	0.46	0.55	0.30	358.46	0.78	114.65

ADR = Absorbed dose rate, AEDR = Annual Equivalent Dose rate, H_{in} = Internal Hazard Index, H_{ex} = External Hazard Index. AGDE = Annual Gonadal Dose Equivalent. $I_{\gamma r}$= Representative Level Index and Ra_{eq} = Radium equivalent.

Table 5: Absorbed dose rate, annual equivalent dose rate, internal hazard index, external hazard index, annual gonadal dose equivalent, representative level index and radium equivalent activity of the radionuclides in the control soils

	ADR (nGy/hr)	AEDR (μSv/yr)	H_{in}(Bq/kg)	H_{ex}(Bq/kg)	AGDE ($\mu svyr^{-1}$)	$I_{\gamma r}$	Ra_{eq}
Mean	42.00	0.36	0.44	0.24	284.91	0.62	90.97
Range	40.09 – 43.49	0.35 – 0.38	0.42 – 0.45	0.23 – 0.25	271.82 – 295.03	0.59 – 0.64	86.88 – 94.23

ADR = Absorbed dose rate, AEDR = Annual Equivalent Dose rate, H_{in} = Internal Hazard Index, H_{ex} = External Hazard Index. AGDE = Annual Gonadal Dose Equivalent. $I_{\gamma r}$= Representative Level Index and Ra_{eq} = Radium equivalent.

The mean absorbed dose rate of the oil-impacted soil is 52.89 nGy/hr. The dose rate value is less than the population weighted average absorbed dose rate (60 nGy/hr) in outdoor air from terrestrial gamma radiation (UNSCEAR 2000) and that of the worldwide average of 55 nGy/hr (Faweya et al. 2014). The mean annual equivalent dose rate of the oil-impacted soil was 0.46 mSv/yr, which falls within the range for individual countries (0.3–0.6 mSv/yr range) (UNSCEAR 2000). The annual dose rate of the radionuclides in

the oil-impacted soils was below the world permissive annual dose limit (1.0 mSv/yr), but more than that of the corresponding control soils. This specifies that the dose rate may not lead to respiratory illness viz, asthma and cancer or external ailments like erythema, skin cancer and cataracts. Therefore, for the oil-impacted soils, there was no concern from a radiological point of view.

The internal hazard index of the oil-impacted soil is 0.55 Bq/kg, while the external hazard index is 0.30 Bq/kg on the average. The values of the indices (H_{ex}, H_{in}) must be less than 1.0 for the radiation hazard to be insignificant. The internal hazard and external hazard indices of the oil-impacted soil in this study are both less than 1 and so, the radiation hazard is insignificant.

The mean annual gonadal dose equivalent of the radionuclide in the oil-impacted soil is 358.46 μSvy^{-1}. This value is close to the world average of 300 μSvy^{-1}. The mean representative gamma index value of the oil-impacted soil is 0.78. This value is less than unity and this indicates that the oil-impacted soils exhibit low gamma radiation.

The estimated mean value of the Ra_{eq} in this current study is 114.65 Bq/kg. This value is below the world mean of 370 Bq/kg (UNSCEAR 2000), but more than that of the corresponding control soils. The radiological assessments carried out using the above parameters all suggest that the soils of the oil depot exhibited low gamma radiation.

Conclusion and recommendations

Oil-impacted soils in the premises of Pipelines and Product Marketing Company (PPMC), Apata, Ibadan, Nigeria were analyzed for radioactivity levels using gamma spectrophotometry. These were determined to assess the levels of radionuclides and the human and ecological risks of the contaminants in the area under study. The results of the radioactivity level of the soils were interpreted in terms of its absorbed dose rate, the annual equivalent dose rate, the internal hazard index, the external hazard index, the radium equivalent, annual gonadal dose equivalent and the representative gamma index. Of all the indices of measuring the radiological hazard, only the annual gonadal dose equivalent was slightly above the world average. The other indices of radiological hazard, particularly the representative gamma index was below their standard permissible limits. These risk assessment parameters confirmed that the oil-impacted soils of the oil depot exhibited low gamma radiation; nonetheless extensive term exposure may pose an inherent cancerous illness.

It is thus recommended that the levels of radioactivity for long-term effects of anthropogenic contributions should be monitored continuously, while indiscriminate disposal of oils should be prevented in its totality.

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