Radiological Risk Assessment of Soil using RESRAD-OFFSITE Code in Communities around Indorama Fertilizer Company Eleme, Port-Harcourt, Rivers State, Nigeria

Abstract:
Anthropogenic activities around a fertilizer company can cause elevation in the concentration of naturally-occurring radioactive materials (NORMs) within surrounding soils, as a result of effluent discharge or heaping of rocks used as raw materials. The aim of this study was to evaluate the human risk associated with exposure to NORMs in soils from communities around Indorama Fertilizer Company Eleme, Port-Harcourt, Rivers State, Nigeria. A sodium-iodide doped with thallium [NaI(TI)] detector was used to measure activity concentrations of these NORMs in 22 soil samples within the study area. The RESidual RADioactivity (RESRAD) OFFSITE modeling program (version 4.0) was then used to estimate the radiation doses and the cancer morbidity risk of uranium-238 (^{238}U), thorium-232 (^{232}Th) and potassium-40 (^{40}K) for hypothetical resident farmer scenario. According to the RESRAD prediction, the maximum total effective dose equivalent (TEDE) during 100 years was found to be 2.82 x 10^{-5} $mSvy^{-1}$ at year 12, while the maximum total excess cancer morbidity risk for all the pathways was 3.52E-08 at year 6. Therefore, results obtained from the RESRAD-OFFSITE code in this study has shown that the health risk from effluent discharge within the study area is within acceptable levels according to international standards.

Keywords: RESRAD-OFFSITE code; radionuclide; radiation dose; cancer morbidity risk; activity concentration

1. **Introduction**

"During the last decades agricultural activities had expanded widely, resulting in an increase in the application of different chemical fertilizers. More than 30 million tons of phosphate fertilizers are annually consumed worldwide" [1]. "However, fertilizers are compounds that provide necessary chemical elements and nutrients to the plants. As a matter of fact, phosphorus, potassium and nitrogen are essential elements for plants growth. Therefore, fertilizers have become essential to the agricultural field all over the world and they tend to increase crop production and also improve the nutrient-deficient properties of lands. However, phosphate rocks together with potassium ores and nitrogenous compounds which are found to contain naturally occurring radioactive materials (NORMs) are the main raw materials used in the industrial production of fertilizers" [1].

"By processing phosphate rock to fertilizers, the radioactivity of the ore is transferred to the products and likewise to the waste products" [2]. "The possible negative effects are the radiological contamination of cultivated lands or other lands where the waste products are discharged" [3]. "Relatively, large concentrations of natural radionuclides present in phosphate fertilizers contaminate the environment and agricultural lands during cultivation. The natural radionuclide of fertilizers consists mainly of uranium and thorium series radioisotopes and natural ^{40}K" [4]. "The level of activity concentration of radionuclides in phosphate fertilizers provides useful information in the monitoring of environmental contamination" [2].

"Generally, NORMs are available in the environment at levels that are not potentially harmful to human health. However, anthropogenic activities such as fertilizer application and discharge of waste from fertilizer producing plants onto soil may elevate the concentration of NORMs to a harmful level, a situation that had been of major concern for radiation protection" [5]. "NORMs

such as ^{238}U, ^{232}Th, and ^{40}K account for up to 85% of the annual dose exposure received by the world population" [6]. "They are known to provide significant sources of human exposure to ionizing radiation within the area of primary contamination" [7]. Hence, the radiological hazard due to the contaminated soil may be minimized by restricting the public access to this area, along with some cleanup measurement. Even so, radionuclide migration from contaminated site through soil via different physical processes to the clean offsite location also results in health hazard.

The mechanisms of transport that could result in cross contamination of offsite area are wind erosion, leaching, erosion by runoff, etc. Therefore, people in an uncontaminated area, i.e., outside the primary contaminated zone, will experience some radiological hazards by means of some exposure pathways such as direct exposure from contamination in soil, inhalation of particulates, ingestion of plant food (e.g., vegetables, grain, and fruits), ingestion of meat, ingestion of milk, ingestion of aquatic foods, drinking of water and incidental ingestion of soil. The extent of the possible risk of NORMs to the health of the population in the study area has not been fully documented. As a result, radiological measurements were conducted in this study, in order to perform off-site hazard assessment of contaminated sites using RESRAD-OFFSITE Code, especially around the Indorama Fertilizer Company located in Eleme, Port-Harcourt, Rivers State, Nigeria.

"The RESRAD (offsite) code is an extension of the RESRAD (onsite) code widely used for evaluating the radiological consequences to a receptor located onsite or outside the area of primary contamination. It evaluates the hazards by calculating radiological dose and excess lifetime cancer risk, predicting activity concentrations in the soil and deriving soil cleanup guidelines analogous to a particular dose limit" [8]. "It also uses the Gaussian Plume Model (GPM) owing to its less input parameters, simple and convenient calculation method as well as small computational costs" [9]. The hypothetical resident-farmer scenario which is adopted in this work includes all environmental pathways for on-site or near-site exposure that could results in the highest predicted lifetime dose.

2. **Materials and Methods**

2.1 **The Study Area**

"Eleme is among the twenty-three Local Government Areas that make up the present Rivers State of Nigeria. It is a coastal area located between Longitude 706'10" E and Latitude 4047'57" N" [10]. "The area is a famous town in Rivers State and is bordered on the north by Obio/Akpor and Oyigbo Local Government Areas, on the east by Tai Local Government Area, on the south by Ogu/Bolo and Okrika Local Government Areas as shown in Figure 1.Eleme is characterized with many industries including Indorama Fertilizer Manufacturing Company, Hamilton Technologies Nig. LTD, Eleme Petroleum Refinery, Dangote Cement, a sea port, Federal Lighter Terminal, Eleme Petrochemicals Limited among others" [11, 10]. "The popular East-West Road traverses through the length and width of Eleme, and as a matter of fact, serious vehicular traffic is frequently experience within the area. Eleme covers an area of 138 km^2 and according to 2006 census, its population was 190,884" [12]. "The occupation of most Eleme people is subsistence Agriculture and the following crops are grown; yam, oil palm fruit, bitter leaf, banana and plantain, sugar cane, cassava, vegetables among others" [11].

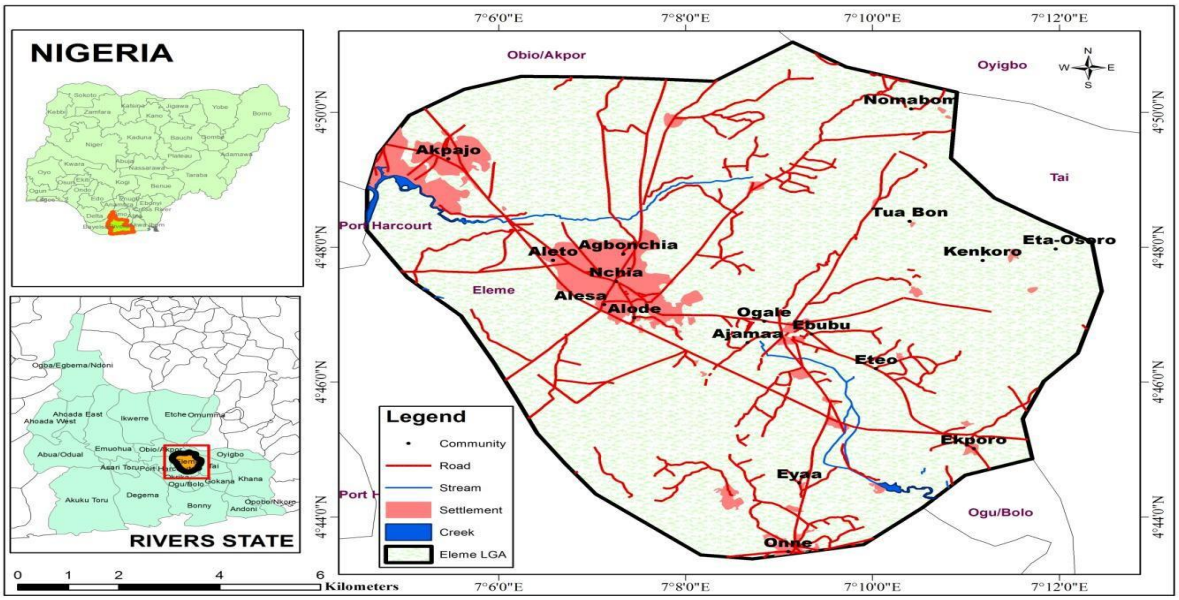

Fig. 1: Map of the Study Area [13]

2.2 **Samples Collection**

A total of 22 soil samples were collected in the research area from a depth of 0.5 m to 1.0 m with the aid of hand auger [14].Global Positioning System (GPS) receiver was also used to locate the sampling points. To avoid sample confusion, the samples were placed in clear polythene bags and labeled precisely using identification marks according to the IAEA [15]. Following proper marking, they were transported for processing and analysis at the environmental laboratory of the Nigerian Institute of Radiation Protection and Research (NIRPR), a division of the Nigerian Nuclear Regulatory Authority (NNRA), located at the University of Ibadan, Oyo State, Nigeria.

2.3 **Sample Preparation**

"At the laboratory, the soil samples were first spread out on a plastic sheet and allowed to air dry under the laboratory condition until constant weights were achieved. The dried samples were pulverized and homogenized using a motorized grinder and allowed to pass through a sieve of 200μm mesh size. The homogenized soil samples were then dried in a temperature-controlled oven at 105^0C for about 24 hours in order to eliminate organic matter content of the soil samples. They were then placed in Marinelli beakers (size 500ml each) and sealed accordingly to maintain their in-situ characteristics. The weights of the sealed samples were recorded using electronic weighing balance and then kept for twenty-eight (28) days in order to achieve radioactive secular equilibrium between parent radionuclides and their respective daughters" [14].

2.4 **Gamma Spectrometry**

The detector used was a Lead shield Canberra 76 mm x 76 mm NaI(TI) crystal, model number 802 series. It is a compatible sealed assembly that includes a photomultiplier tube, a high-resolution NaI(Tl) crystal, and a preamplifier base that feeds amplified electrical pulses into analyzer systems. The photomultiplier tube detects the tiny visible light photons produced in the crystal. The detector system was calibrated before carrying out actual measurement of the soil samples. In order to commence counting, three gamma standard sources ^{137}Cs, ^{241}Am and ^{60}Co were placed into 6cm lead shield of the detector chamber. This set up is aimed to minimize the effects of background and scattered radiation. By determining the correlation between the peak point in the spectrum and the associated gamma ray, the energy calibration was completed. Each pulse produced by a photomultiplier tube, as seen on the display output and the associated channel, has a height that is directly proportional to the original gamma energy that caused the pulse. The calibration was done using gamma emitter sources of known energies, these are ^{137}Cs and ^{60}Co source that emits with energies of 662 keV, 1332 keV and 1173 keV, and ^{241}Am which is an alpha emitter but also emits some gamma rays with energies 26.3 keV and 59.6keV. The gamma emitter sources were exposed to the NaI(TI) detector and gamma spectrum was acquired. These were done with the amplifier gain that gives 72% energy resolution for the 662 keV of ^{137}Cs and counted for 30 minutes. The net area corresponding to the photopeak's in the energy spectrum was computed by subtracting count from the background source from the total area of the photopeak's. The identification of individual radionuclides was performed using their gamma ray energies and the quantitative analyses of radionuclides were performed using gamma ray spectrum analysis software, Genie 2000.

2.5 **RESRAD-OFFSITE Computer Code**

The RESidual RADioactivity (RESRAD) offsite computer code was developed by the Argonne National Laboratory under the U.S. Department of Energy and the U.S. Nuclear Regulatory Commission as multi-functional software to assist in developing cleanup criteria and assessing the radiological dose or risk associated with residual radioactive materials. RESRAD-OFFSITE is an extension of the RESRAD-ONSITE computer code that was developed to estimate the radiological consequences to individual located onsite or outside the area of primary contamination. The code uses the Gaussian plume model, in which the radionuclide concentration and plume remain constant over time [16], IAEA safety reports series No.19 recommended the Gaussian model as a general model for the effects of radioactive emissions on the atmosphere. "In the evaluation procedure adopted in this study, the RESRAD code uses the U.S. Federal Guidance Report (FGR) 11 & 12 for internal and external dose conversion libraries, as well as the FGR 13 library for health risk. These served as the foundation for the radionuclide transition based on the International Commission on Radiological Protection-38 library. The radionuclide database also accounts for ingrowth of daughters from initially present parent radionuclides. However, RESRAD offsite code allows users to specify the features of their site and to predict the dose received by an individual at any time up to 1, 000 years. It uses more than 150 variables alongside ICRP-38 radionuclide database" [17].

2.6 **Input Parameters and Scenario Description**

"For the study area, a number of parameters were considered as inputs to the RESRAD-OFFSITE code as shown in Table 1. The resident farmer scenario was chosen as the critical receptor in the risk assessment. The parameter values were carefully selected to achieve a more realistic estimation of the dose or risk. Where necessary, site-specific parameters replaced default parameters" [18].

"Input parameters to the model include the measured activity concentrations of ^{238}U, ^{232}Th, ^{40}K, and meteorological data (annual wind speed and direction). Hydrogeological parameters for the various zones were also taken into consideration. These include the hydraulic conductivity and density of the soil. Some hydrological parameters, such as total porosity, field capacity, irrigation rate, erosion rate, precipitation rate, runoff coefficient and soil-specific exponential parameters were also estimated based on the condition of the study area , while the area and thickness of the different zones were taken into account" [18].

The major exposures in the resident farmer scenario include direct exposure to the radionuclides in soil, inhalation of dust, ingestion of contaminated vegetables, water, and soil. Dietary information on vegetables, water, and soil were also considered through regulatory guidelines as set by the South African National Nuclear Regulator [19], since the information is not readily available with the Nigerian Nuclear Regulatory Authority.

Table 1: Principal Input Parameters for the RESRAD-OFFSITE Code

Parameters	Value	Reference
Area of contaminated zone (m^2)	10, 000	Default
Density of contaminated zone (g/m^3)	1.44 g	[17]
Total porosity for contaminated zone	0.43	[17]
Hydraulic conductivity of contaminated zone (m/yr)	1090	[17]
[a]Soil-specific exponential b parameter	5.3	Default
Average annual wind speed (m/s)	4.1	[16]
Precipitation rate (m/yr)	1.0	[19]
Irrigation rate (m/yr)	0.2	Default
Runoff coefficient	0.65	[18]
Inhalation rate (m^3/yr)	8059.2	[18]
Exposure duration (yr)	30	Default
Soil ingestion rate (g/yr)	37	[18]

[a]The soil-specific b parameter = empirical and dimensionless parameter that is used to evaluate the saturation ratio (Rs) (or the volumetric water saturation of the soil, according to a soil characteristic function called the conductivity function).

2.7 **Transport Pathways Associated with NORMs in the Study Area**

The major mechanisms of radionuclide transport from potential primary contamination sources within the study area are the atmosphere, ground water sources, and surface water bodies. Air pollutants such as particulate matter from phosphate-rock dumps, water run-off from areas such as ore piles and waste water, constitute a major source of NORM's pollution to the environment within the vicinity of the fertilizer industry. Radionuclide(s) from these primary sources can also be transported to humans via the atmosphere as wind blow that could settle on farmlands or even residential buildings. They could also be leached into the underlying ground water aquifers or dissolved and drained through runoff into surface water bodies, thereby contaminating both soil

and water sources. The summary of scenario description for Offsite Dose Assessment within the study area is given in Figure 2.

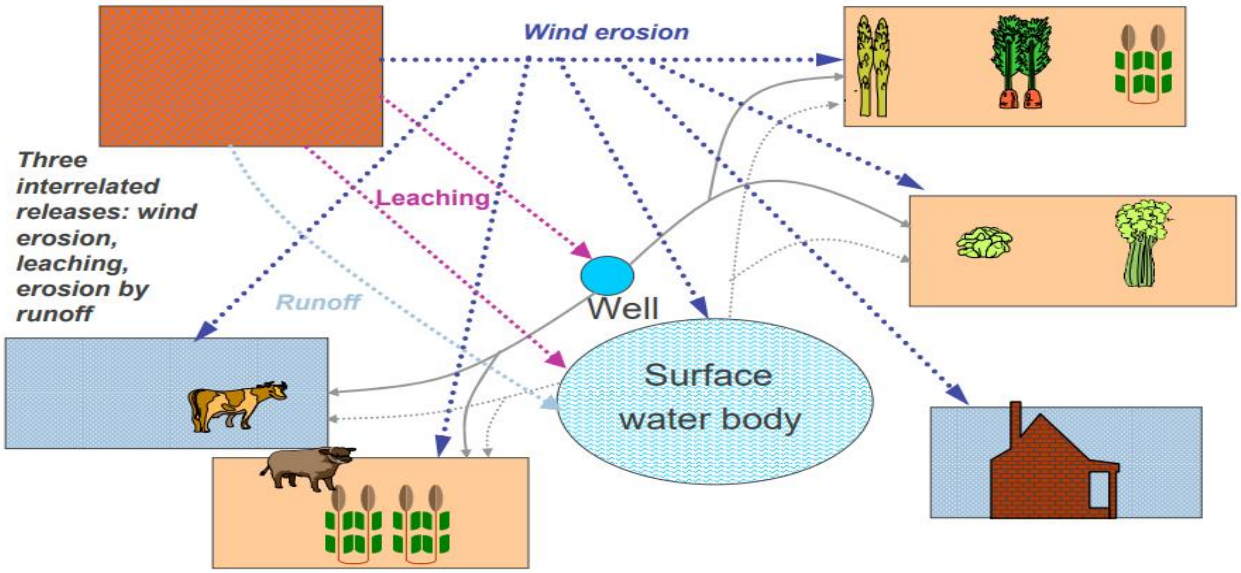

Fig. 2: Summary of Scenario Description for Offsite Dose Assessment [20].

3. **Results and Discussion**

3.1 **NORMs Activities Concentration in Soil**

The mean activity concentrations of radionuclide ^{238}U, ^{232}Th, and ^{40}K in soil samples within the study area were 3.91 $Bqkg^{-1}$, 1.53 $Bqkg^{-1}$, and 27.61 $Bqkg^{-1}$ respectively. These values were all below the UNSCEAR world standard of 35 $Bqkg^{-1}$, 30 $Bqkg^{-1}$ and 400 $Bqkg^{-1}$ for ^{238}U, ^{232}Th, and ^{40}K respectively [21], and were used as input parameters in the RESRAD-OFFSITE code to calculate the radiation dose, as well as the excess cancer risk. However, Figure 3 shows comparison of the mean activity concentration of naturally occurring radioactive materials (^{238}U, ^{232}Th and ^{40}K) in the sampling locations.

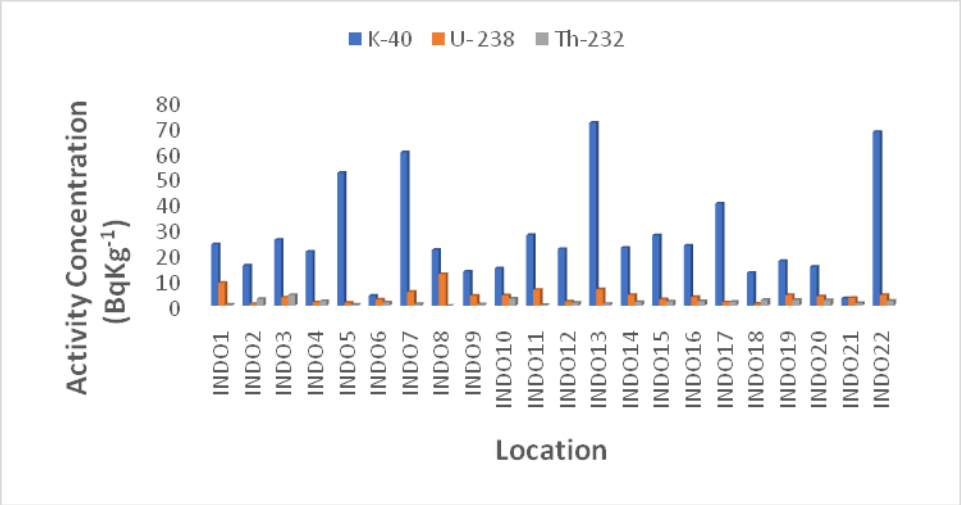

Fig. 3: Mean activity concentration of ^{238}U, ^{232}Th and ^{40}K in soil.

3.2 **Offsite Dose Assessments**

The total effective dose equivalent (TEDE) in $mSvy^{-1}$ predicted for an off-site public exposure using the RESRAD-Offsite computer code version 4.0 for a period of 100 years for all pathways summed is presented in Table 3. Meanwhile, Figure 4 shows the variation of TEDE with sample locations, while Figure 5 represents the variation of effective dose equivalent from the different radionuclide (^{238}U, ^{232}Th and ^{40}K) in conjunction with the variation of TEDE (all nuclides summed, all pathways summed) with time. From Figure 5, the contribution of each radionuclide to the TEDE was indicated, it can be seen that ^{232}Th gave the highest contribution and is followed by ^{40}K, while ^{238}U contributed minimally.

According to the RESRAD, the maximum TEDE value from the study area during 100 years period was 2.82×10^{-5} $mSvy^{-1}$ (at year 12), while the minimum TEDE value was 2.23×10^{-5} $mSvy^{-1}$ (at year 100). Figure 4 also showed that the TEDE value was lower at Indo 2, while it was higher at Indo 8 when the various sampling locations were considered. The highest dose in Indo 8 could be due to high level discharge of effluent from the fertilizer plant. This might result to long-term stochastic effects to the public. Nonetheless, the dose from Indo 8 is within the recommended ICRP public dosage boundary of 1 $mSvy^{-1}$. Also, all doses recorded from other study sites were below the recommended public dose boundary respectively.

However, the overall yearly radiation dose from the study area includes contributions from external exposure pathway via direct gamma irradiation, and internal radiation through inhalation and ingestion pathways. The inhalation dosage is ascribed to gaseous decay radon and thoron from ^{238}U and ^{232}Th while the external dose is attributed to direct gamma irradiation from ^{40}K which decays by releasing 89% beta and 11% gamma radiation which has low external dose contribution [22]. However, Faanu *et al.*, reported the annual dose due to mining in central region of Ghana as 0.918 $mSvy^{-1}$[23].This is by far higher than the value obtained in this study. Similarly, Nwankwo *et al.*, reported the annual dose from findings of public dose assessment around mining sites in Komu, Oyo State, Nigeria as 0.12 $mSvy^{-1}$ [24].The reported value is also higher than the value obtained in this study.

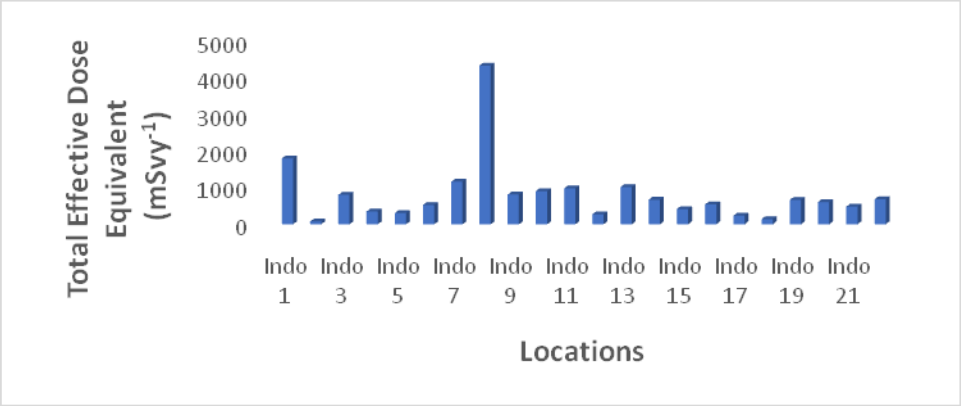

Fig. 4: Variation of TEDE with sample locations

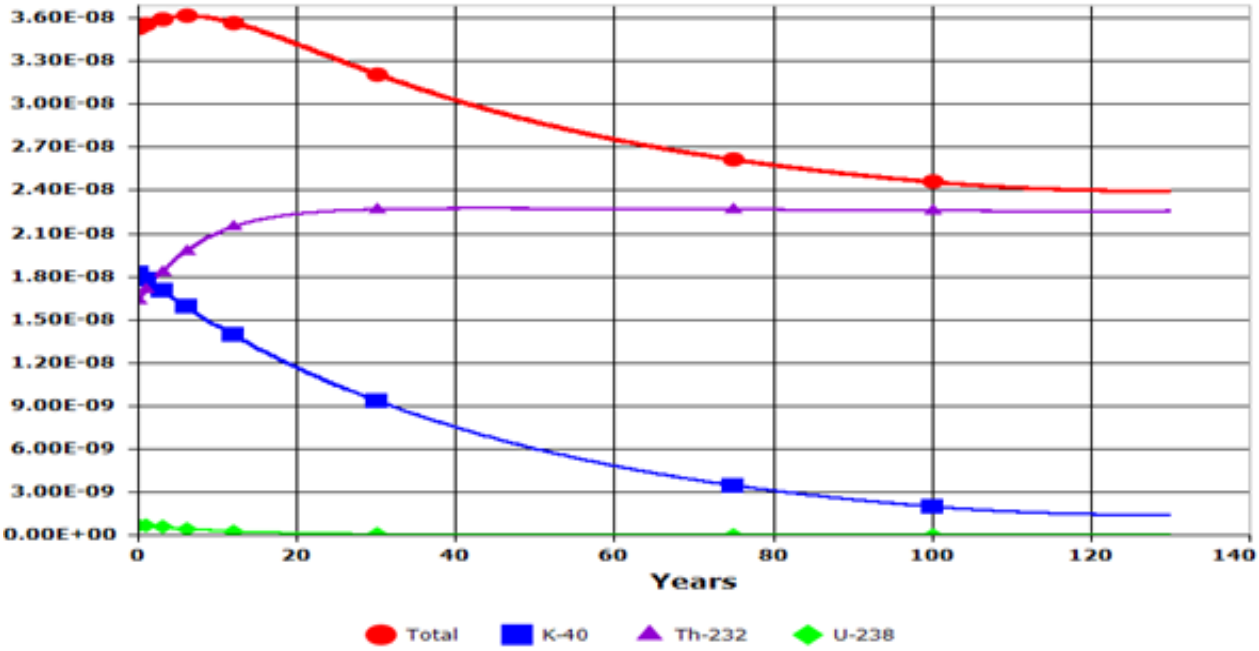

Fig. 5: Variation of TEDE with time (in Years)

3.3 **Assessments of Excess Cancer Morbidity Risk (ECMR)**

The results of the radiation doses were used to calculate the excess cancer morbidity risks for all of the pathways summed over duration of 100 years. Due to the tendency of low dose accumulation over a long period of time, which might result to stochastic effects, the ICRP reaffirms that there is no safe level of radiation exposure. A comprehensive review of available biological and biophysical data supports a "Linear-No-Threshold" (LNT) risk model; that the risk of cancer proceed in a linear fashion at lower doses without a threshold and that the smallest dose has the potential to cause a small increase in cancer risk to humans. The maximum total excess cancer morbidity risk from the study area was found to be 3.52E-08 at year 6, while the minimum value was 2.37E-08, obtained at year 100.

Figure 6 clearly shows the variation of ECMR with time (in Years). The maximum total ECMR progressively decreased to 2.37E-08 at year 100. ^{232}Th was the major driver of the risk effect, followed by ^{238}U and then ^{40}K. The US Environmental Protection Agency (USEPA) considers acceptable, for regulatory purposes, a cancer risk in the range of E-06 to E-04 [25]. Therefore, results obtained from the RESRAD-OFFSITE code had shown that the health risk from the study area is within acceptable limit based on international standards.

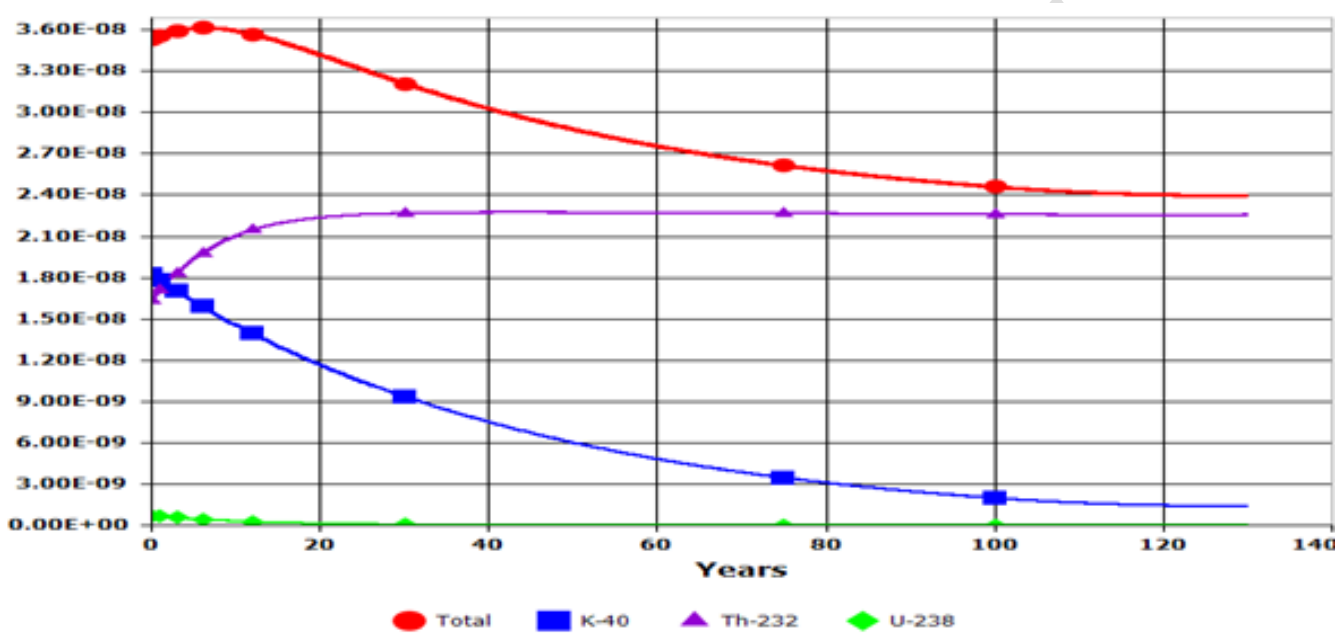

Figure 6: Variation of ECMR with time (in Years)

4. **Conclusions**

This study had evaluated the radiological risk to human associated with exposure to NORMs in soils due to the discharge of effluents around a fertilizer factory located in Eleme, Rivers State, Nigeria, using the RESRAD-OFFSITE code. The total effective dose equivalent (TEDE) from all nuclides was found to range from 2.23E-05 to 2.85E-05. The results of dose estimation from all the sample locations within the study area showed that all the dosages were lower than the recommended ICRP set limit of 1 $mSvy^{-1}$. However, in adherence to the principle of "As Low As Reasonably Achievable" (ALARA), the Competent Authorities need to conduct periodic assessment of radiation exposure from the study area in order to ensure that such exposure are kept below the dose limit of $1mSvy^{-1}$ or dose constraint of 0.25 $mSvy^{-1}$ set by the US Nuclear Regulatory commission (NRC), thereby protecting the public and the environment from the harmful effects of ionizing radiation.

The total excess cancer morbidity risk estimated from the model ranged from 2.37E-08 to 3.52E-08. However, the US Environmental Protection Agency considers acceptable for regulatory purposes a cancer risk in the range of E-06 to E-04. It has also been noted that ^{232}Th has a significant contribution to the radiation dose, as well as the excess cancer morbidity risk, compared to ^{238}U and ^{40}K. This may be attributed to higher dose conversion factors associated

with ^{232}Th compared to ^{238}U and ^{40}K. From the findings of this study, it can be deduced that the presence of Fertilizer Company within the study area and its activity there-off had a negligible risk based on international guidelines on radiation safety.

However, the modified Gaussian Plume Model has multiple input parameters and several corrections that make the model complex. Many input parameters are difficult to measure accurately; therefore their values are usually uncertain, if the model results are used directly to make decisions such as post-accident emergency response, there will be risks of making mistakes in decision-making which could be considered a major limitation. Therefore, other software program such as Environmental Risks from Ionizing Contaminants: Assessment and Management (ERICA) could be employed in the study area while their result compare to that obtained in this study.

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