**Radiological Risk Assessment of SoilusingRESRAD-OFFSITE Code in Communities around Indorama Fertilizer Company, Eleme, Port-Harcourt, Rivers State.**

**Abstract:**
Anthropogenic activities around a fertilizer company can cause elevation in concentration of naturally-occurring radionuclide materials (NORMs) on 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 fromcommunities around Indorama Fertilizer Company, Eleme, Port-Harcourt, Rivers State. A sodium-iodide [ NaI (TI)]detector was used to measure activity concentrations of these NORMs in 22 soil samples within the study area. The RESidualRADioactivity (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 a hypothetical resident scenario. According to 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. The US Environmental Protection Agency considers acceptable for regulatory purposes a cancer risk in the range of E-06 to E-04. 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 have expanded widely, resulting in an increase in the applications of the different chemical fertilizers. More than 30 million tons of phosphate fertilizers are annually consumed worldwide [1]. However, fertilizers arecompounds 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 increasecrop 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 for fertilizers in industrial production.

By processing phosphate rock to fertilizers, the radioactivity of the ore is transferred to the products and to 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 anddischarge of waste from fertilizer producing plants onto soil may elevate the concentration of NORMs to a harmful level, a situation that has been of major concern for radiation protection [5]. NORMssuch 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, theradiologicalhazarddue to the contaminated soilmay be minimized by restricting the public accessto 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 would 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 waterandincidentalingestion of soil. The extent of the possible risk of NORMs to the health of the population in the study area has not been fullydocumented. As a result, radiological measurements were conducted in order to perform off-site hazard assessment of contaminated sites using RESRAD-OFFSITECode, especiallyaround the Indorama Fertilizer Company located in Eleme, Port-Harcourt, Rivers State.

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]. The hypothetical resident-farmer scenario which is adopted in this work includes all environmental pathways for on-site or near-site exposure that results in the highest predicted lifetime dose.

## 2.**Materials and Methods**

### 2.1 The Study Area

Eleme is among thetwenty-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 [9]. The area is a famous town in Rivers State and isbordered 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[10,9]. The popular East-West Roadtraversesthrough 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 138km$^2$and according to 2006 census, its population was 190,884[11]. 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[12].

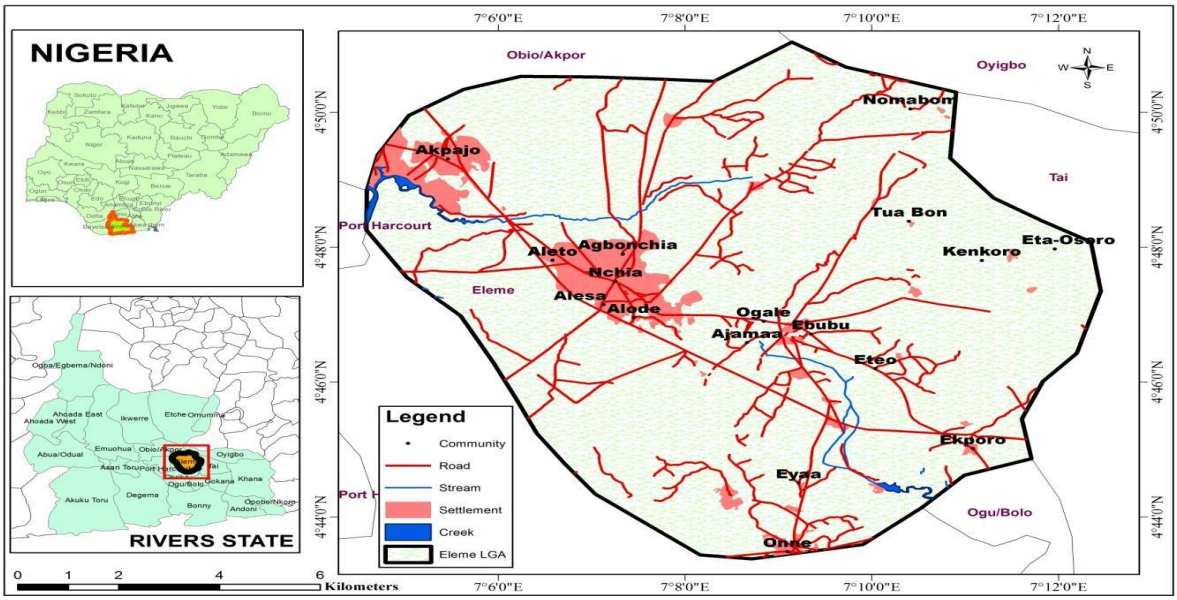


**Figure1:**MapoftheStudyArea(Moses *et al*., 2020)

## 2.2 **Samples Collection**

A total of 10 soil samples were collected in the research areafrom a depth of 0.5 m to 1.0 m with the aid of hand auger[13].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 preciselyusingidentification marks according to the IAEA [14].Following proper marking, they were transported for processing and analysisatthe environmental laboratory of theNigerian 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 andallowed to air dry under the laboratory conditionuntil 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 $105^0$C 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[13].

## 2.4 **Gamma Spectrometry**

The detector used was a Lead shield Canberra 76 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 Cs-137, Am-241 and Co-60 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 gammaray, 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 Cs137 and Co-60 source that emitswith energies of 662 keV, 1332 keV and 1173 keV, and Am-241 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 Cs-137 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 the residual radioactive material. RESRAD-OFFSITE is an extension of the RESRAD-ONSITE computer code that was developed to estimate the radiological consequences to individuals 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 [8].In this evaluation procedure, RESRAD uses 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. RESRAD 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 [8].

### 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 [15].

Input parameters to the model included 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 included 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 [15], while the area and thickness of the different zones were alsotaken into account.

The main exposures in this scenario included direct exposure to the radionuclides in soil, inhalation of dust, and ingestion of contaminated vegetables, water, and soil. Dietary information on vegetables, water, and soil were also considered through regulatory guidelines as set by theSouth African National Nuclear Regulator [16], since the information is not readily available with the Nigerian Nuclear Regulatory Authority.

Table 1: Principal Input Parameters for theRESRAD-OFFSITE Code

| Parameters | Value | Reference |
|---|---|---|
| Area of contaminated zone ($m^2$) | 10, 000 $m^2$ | Default |
| Density of contaminated zone ($g/m^3$) | 1.44 $g/m^3$ | [17] |
| Total porosity for contaminated zone | 0.43 | [15] |
| Hydraulic conductivity of contaminated zone (m/yr) | 1090 m/yr | [15] |
| [a]Soil-specific exponential b parameter | 5.3 | Default |
| Average annual wind speed (m/s) | 4.1 m/s | [18] |
| Precipitation rate (m/yr) | 1.0m/yr | [16] |
| Irrigation rate (m/yr) | 0.2 | Default |
| Runoff coefficient | 0.65 | [15] |
| Inhalation rate ($m^3$/yr) | 8059.2 $m^3$ /year | [17] |
| Exposure duration (yr) | 30 | Default |
| Soil ingestion rate (g/yr) | 37 g/yr | [17] |

[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**

Major mechanisms of radionuclides transport from potential primary contamination sources in the study area are the atmosphere,ground water sources, and surface water bodies. Air pollutants such as particulatematter from phosphate-rock dumps, water run-off from areas such as ore pilesand waste water, constitute a major source of NORMs pollution to the environment within the vicinity of the fertilizer industry.Radionuclides from these primary sources can also be transported to humans via the atmosphere,as windblow that could settle on farmlands or even residential buildings. They could also beleached into the underlying ground water aquifers or dissolved and drained through runoff into surface water bodies, thereby contaminating both soil and water sources. Thesummary of scenario description for Offsite Dose Assessment within the study area is given in Figure 2.

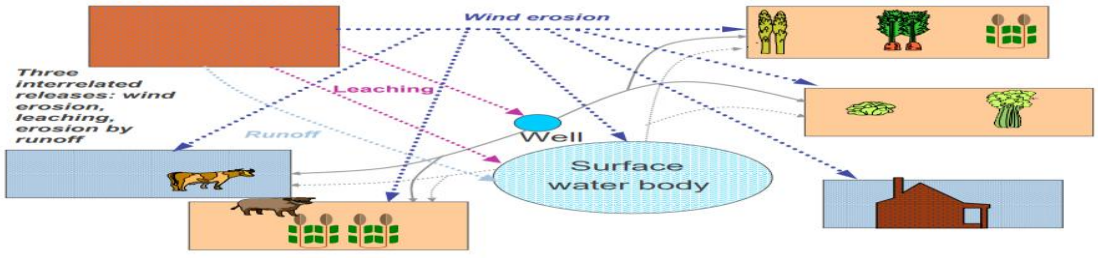


Fig. 2: Summary of Scenario Description for Offsite Dose Assessment[19].

3. Results and Discussion

3.1 NORM 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 as presented in Table 2. These values were all below the UNSCEAR world standardof 35$Bqkg^{-1}$, 30$Bqkg^{-1}$ and 400$Bqkg^{-1}$ for $^{238}U$, $^{232}Th$, and $^{40}K$ respectively[20],and were used as input parameters in the RESRAD-OFFSITE code to calculate the radiation dose, as well as the excess cancer risk. Figure 3 shows comparison of the mean activity concentration of the different radionuclide in different sample locations.

Table 2:Activity concentrations of $^{238}U$, $^{232}Th$, and $^{40}K$ ($BqKg^{-1}$)

| S/N | **Sample Code** | **$^{40}K$** | **$^{238}U$** | **$^{232}Th$** |
|---|---|---|---|---|
| 1 | $INDO_1$ | 24.23±2.77 | 8.97±2.29 | 0.40±0.05 |
| 2 | $INDO_2$ | 15.80±1.25 | 0.76 ±0.19 | 2.68 ±0.26 |
| 3 | $INDO_3$ | 25.89±2.75 | 3.26 ±0.89 | 4.28 ±0.43 |
| 4 | $INDO_4$ | 21.30±1.69 | 1.36 ±0.33 | 1.87 ±0.18 |
| 5 | $INDO_5$ | 52.26 ±4.09 | 1.18 ±0.32 | 0.40 ±0.04 |
| 6 | $INDO_6$ | 3.98±0.37 | 2.46 ±0.68 | 1.29±0.14 |
| 7 | $INDO_7$ | 60.33±5.99 | 5.46 ±1.36 | 0.70 ±0.09 |
| 8 | $INDO_8$ | 21.97 ±2.01 | 12.50±3.32 | BDL |
| 9 | $INDO_9$ | 13.42±1.23 | 3.96 ±0.99 | 0.60 ±0.07 |
| 10 | $INDO_{10}$ | 14.65±1.35 | 4.07±1.07 | 2.79±0.29 |
| 11 | $INDO_{11}$ | 27.79±2.60 | 6.31±1.52 | 0.37±0.04 |
| 12 | $INDO_{12}$ | 22.33±1.78 | 1.65 ±0.41 | 1.19±0.12 |
| 13 | $INDO_{13}$ | 71.97 ±6.52 | 6.46 ±1.64 | 0.76 ±0.09 |
| 14 | $INDO_{14}$ | 22.80 ±2.13 | 4.25±1.12 | 1.48±0.16 |
| 15 | $INDO_{15}$ | 27.71 ±2.52 | 2.56 ±0.70 | 1.67 ±0.18 |
| 16 | $INDO_{16}$ | 23.67 ±2.12 | 3.42 ±0.94 | 1.79 ±0.19 |
| 17 | $INDO_{17}$ | 40.26 ±3.12 | 1.37 ±0.33 | 1.57±0.15 |
| 18 | $INDO_{18}$ | 12.99±1.02 | 0.79 ±0.21 | 2.32 ±0.22 |
| 19 | $INDO_{19}$ | 17.58 ±1.78 | 4.18 ±1.12 | 2.33 ±0.25 |
| 20 | $INDO_{20}$ | 15.28±1.47 | 3.70 ±1.00 | 2.19 ±0.23 |
| 21 | $INDO_{21}$ | 2.91 ±0.28 | 3.07 ±0.80 | 1.04±0.11 |
| 22 | $INDO_{22}$ | 68.35±6.00 | 4.23 ±1.05 | 1.97±0.20 |
| | **Mean Value** | **27.61±2.49** | **3.91±1.01** | **1.53±0.16** |

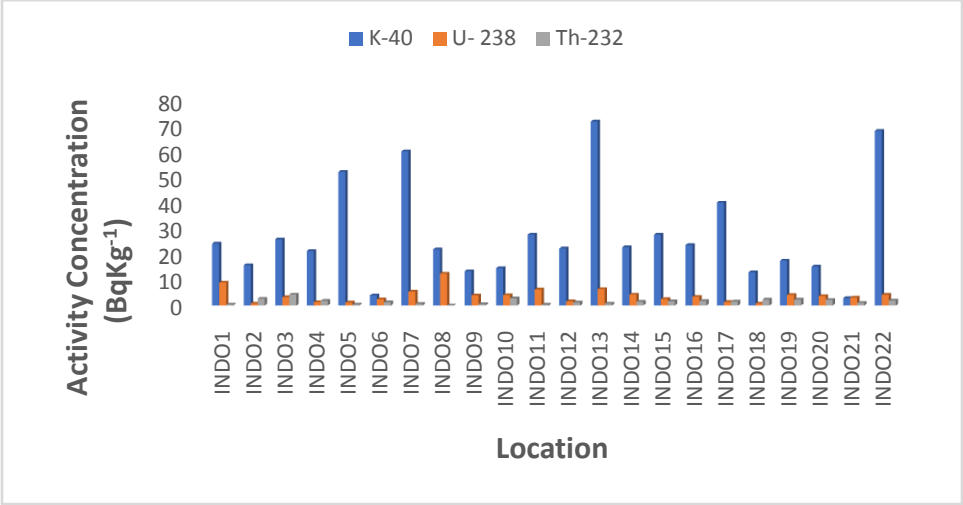


Fig. 3: Mean activity concentration of $^{238}$U, $^{232}$Th and $^{40}$K in soil.

### 3.2 **Offsite Dose Assessment**

The total effective doseequivalent (TEDE) in mSvy$^{-1}$predicted for an off-sitepublic exposure using theRESRAD-Offsite computer code version 4.0for a period of 100 yearsfor all pathways summed ispresented in Table3. Meanwhile, Figures 4 shows variation of TEDE with sample locations, while Figure 5 representsvariation 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.

Table 3: **Total effective dose equivalent (mSvy$^{-1}$) for all the pathways summed over a duration of 100 years.**

| Year | Total Effective Dose Equivalent(mSvy$^{-1}$) | | | |
|---|---|---|---|---|
| | Nuclides | | | Total |
| | $^{40}$K | $^{232}$Th | $^{238}$U | |
| 0 | 1.06E-05 | 1.17E-05 | 2.29E-06 | 2.46E-05 |
| 1 | 1.04E-05 | 1.24E-05 | 2.11E-06 | 2.49E-05 |
| 3 | 0.10E-05 | 1.39E-05 | 1.80E-06 | 2.29E-05 |
| 6 | 0.93E-05 | 1.61E-05 | 1.41E-06 | 2.68E-05 |
| 12 | 0.82E-05 | 1.91E-05 | 0.87E-06 | 2.82E-05 |
| 30 | 0.55E-05 | 2.13E-05 | 0.21E-06 | 2.70E-05 |
| 75 | 0.20E-05 | 2.13E-05 | 0.06E-06 | 2.33E-05 |
| 100 | 0.12E-05 | 2.11E-05 | 0.00 | 2.23E-05 |

According to the RESRAD, the maximum TEDE value from the study area during 100 years was found to be 2.82 x 10$^{-5}$mSvy$^{-1}$ (at year 12), while minimum TEDE value was 2.23 x 10$^{-5}$mSvy$^{-1}$ (at year 100) as

shown in Table 3. Figure 4 also indicated that for the various sample locations, TEDE value was lower at Indo 2, while it was higher at Indo 8.The highest dose in Indo 8 could be due to high level discharge of effluent from the fertilizer plant which is going on in the city, and might result to long-term stochastic effects to the public. Nonetheless, the dose from Indo 8 is within the commended ICRP public dosage boundary of 1 $mSvy^{-1}$, and all doses recorded from other study sites are also 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 with low external dose contribution [21].The maximum TEDE value of 2.82 x $10^{-5}mSvy^{-1}$is by far lower than the dose reported by Faanu*et al*., (2016)[22],with annual dose of 0.918 $mSvy^{-1}$ from mining in central region of Ghana, and also lower than the annual dose of 0.12 $mSvy^{-1}$ reported by Nwankwo *et al*., (2015)[23] from findings of public dose assessment around mining sites in Komu, Oyo State.


Fig. 4: Variation of TEDE with sample locations

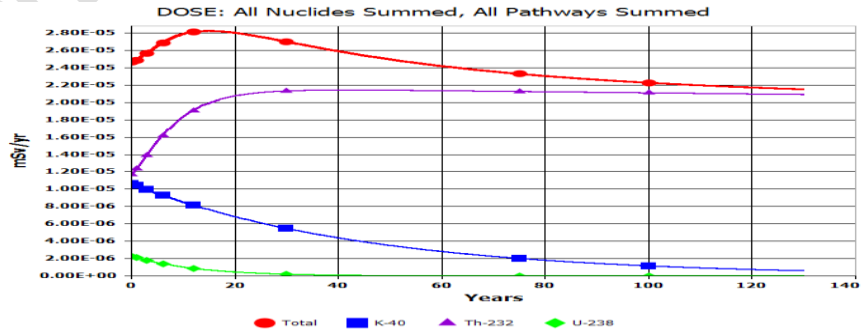


Fig. 5: Variation of TEDE with time (in Years)

### 3.3 Assessments of Excess CancerMorbidity Risk (ECMR)

The results of the radiation doseswere used by RESRAD-OFFSITE computer code version 4.0 to calculate the excess cancer morbidityrisks for all of the pathways summed over duration of 100 years, as shown in Table 4.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 proceeds 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 be3.52E-08 at year 6, while the minimum value was 2.37E-08 obtained at year 100.

Table 4: **Excess Cancer Risk for all the pathways summed over a duration of 100 years.**

| Year | Excess Cancer Risk | | | |
|---|---|---|---|---|
| | Nuclides | | | Total |
| | $^{40}K$ | $^{232}Th$ | $^{238}U$ | |
| 0 | 1.82E-08 | 1.55E-08 | 7.120E-10 | 3.44E-08 |
| 1 | 1.78E-08 | 1.62E-08 | 6.570E-10 | 3.46E-08 |
| 3 | 1.70E-08 | 1.74E-08 | 5.600E-10 | 3.50E-08 |
| 6 | 1.59E-08 | 1.89E-08 | 4.400E-10 | 3.52E-08 |
| 12 | 1.40E-08 | 2.06E-08 | 2.720E-10 | 3.48E-08 |
| 30 | 0.94E-08 | 2.18E-08 | 0.640E-10 | 3.13E-08 |
| 75 | 0.35E-08 | 2.18E-08 | 0.020E-10 | 2.53E-08 |
| 100 | 0.20E-08 | 2.17E-08 | 0.003E-10 | 2.37E-08 |

Figure 6 clearly showsthevariation of ECMR with time (in Years).Themaximum total ECMRprogressivelydecreased to 2.37E-08 at year 100.$^{232}$Th was driving the risk effect followed by $^{238}$U, and thenlastly$^{40}$K. The US Environmental Protection Agency (USEPA) considers acceptable, for regulatory purposes, a cancer risk in the range of E-06 to E-04[24]. Therefore, results obtained from the RESRAD-OFFSITE code has shown that the health risk from the study area is within acceptable levels according to international standards.

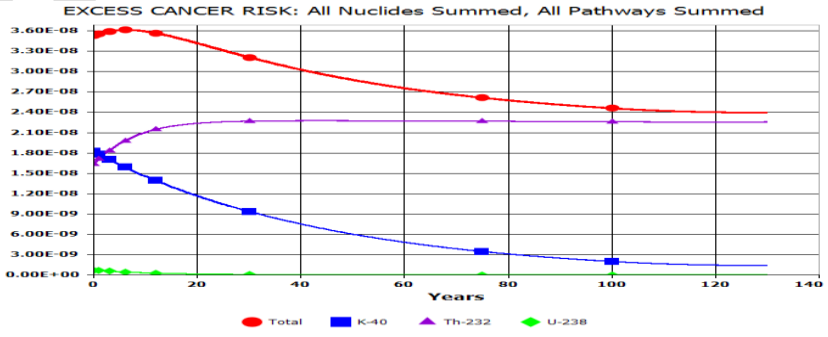


Figure 6: Variation ofECMRwith time (in Years)

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 from2.23E-05to 2.85E-05. The results of dose estimation from all the sample locations within the study area shows 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 orderto ensure that such exposure are kept below the dose limit of 1$mSvy^{-1}$ 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. 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.

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