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MODELING THE GEOSPATIAL DISTRIBUTION AND SPATIAL VARIABILITY OF NATURALLY OCCURRING RADIOACTIVE MATERIALS ON FARMLANDS

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ABSTRACT

The surge in anthropogenic activities following industrialization compromises the nature of the environment, specifically through alterations of the activities of Naturally Occurring Radioactive Materials (NORMs). This research focused on modeling the spatial distribution of NORMs in farmlands. The soil was sampled from the study area and the selected NORMs ^{238}U, ^{232}Th, and ^{40}K were determined. The Kriging method of interpolation (ordinary Kriging) was also employed to model the activity of the radionuclides at the unsampled sites in the study area in the estimation of the unsampled points. The radiological analysis of the samples revealed the activity concentration of 3.49±1.07, 1.32±0.68 and 6.43±4.18 Bq/kg for the target radionuclides, ^{238}U, ^{232}Th, and ^{40}K, respectively, concentrations which were identified to be lower, compared to the average world limits 35, 30 and 400 Bq/kg for these radionuclide species. Using the kriging technique, the predicted map produced activity concentrations of the radionuclides which slightly deviated from the measured values, as well as showing a strong concentration gradient of the radionuclides across the study area, with high radioactivity occurring at the upper section of the area. The distribution map for ^{238}U showed high activity of the radionuclide from the northern to the central part of the study area while the southern section had relatively lower concentrations. Similarly, the spatial distribution for the activity of ^{232}Th and ^{40}K was varied, with higher concentration in the northern part of the study area, also, the greatest activity for ^{232}Th was shown in the central part of the study. As it relates to the measuring and monitoring of radioactivity in the environment, the study improved both theoretical and practical understanding of quality assurance. This will ensure that measurement findings can be compared in the future and that methods and processes are standardized at local levels.

Keywords: Activity Concentration, Monitoring, Ordinary Kriging, Spatial Distribution, Spatial Variability.

INTRODUCTION

As the world advances through urbanization and industrialization, food safety is gradually emerging as a challenge due to the soil quality compromise. While several factors have been identified as causes, one culprit of major concern is naturally occurring radioactive materials (NORMs) originating from terrestrial sources. Processes such as volcanic eruptions and mineral weathering account for natural sources of these radioactive materials in the environment (Inigo *et al.*, 2018). Notwithstanding, the rise in anthropogenic activities including fertilizer application, mineral extraction, and processing activities can result in higher levels of NORMs in agricultural soils (Mathuthu *et al*., 2016; Lecomte *et al*., 2019; Paiva *et al*., 2019), resulting in the destruction of soil health as well as presenting adverse health conditions in human through consumption of contaminated food.

Several research including studies by Ahmed et al. (2005) and Mathuthu et al. (2016) have identified NORMs in farm soils and phosphate fertilizers, as well as their emission via mining activities. Mekongtso et al. (2016) conducted an analysis to determine the concentrations of specific radionuclides in soil samples extracted from the core deposits of bauxite. Their investigation involved the quantification of the activity levels of 238U, 232Th, and 40K, resulting in values of 99 ± 69, 157 ± 67, and 671 ± 272 Bq/kg, respectively. Additionally, Faanu et al. conducted a study to evaluate the naturally occurring and human-induced radioactivity levels within soils situated near the Chirano gold mine in Ghana. The measurements revealed activity concentrations that amounted to 9.79 ± 5.39, 9.18 ± 7.06, and 237.40 ± 144.34 Bq/kg, respectively for ^{238}U, ^{232}Th, and ^{40}K.

Despite the presence of NORMs in the soil, according to Kritsananuwa *et al*. (2015) and Bezuidenhout (2019), their distribution and concentration are related to the geological and geographical factors, biogeochemical process, and physicochemical properties of the soil. Given the differences in geological and geographical factors among others which influence the availability of NORMs in the soil, these radioactive materials could accumulate presenting several hazardous impacts. To ensure a sustainable environment, there is a need to monitor the concentration and spatial variations of radioactive materials (Dindaroğlu, 2014). In determining the spatial variations of radionuclides, geostatistical analysis can be employed. One of the most employed geostatistical techniques is the kriging method, which has been used in research works by Dai *et al.* (2007) and Dragović *et al.* (2014). Geostatistical techniques involve analysis and estimation techniques to predict values of a variable dispersed in time and location.

In New Abirem and other catchment communities of the Akyem mines, the emergence of small-scale mining activities has accelerated resulting in several adverse impacts on the environmental status quo (Attiogbe *et al.,* 2017). These adverse effects include the destruction of water bodies, vegetation, and biotic components. Given that the community is partly an agrarian society, the application of phosphate and potassium sulfate fertilizers on farmlands, coupled with the increase in mining activities with little adherence to standards in the locality, the quality of farmland soils in the area and ultimately crops cultivated on the field are compromised. Though the activity of NORMs has been determined in communities around the mine concession (Faanu *et al.*, 2016), research on the radioactivity of farmlands in New Abirem has not been conducted. The aim of this study is to *i)* obtain radionuclide information on selected terrestrial NORMs: uranium (^{238}U) and thorium (^{232}Th) decay series and potassium (^{40}K) in the farmlands, and *ii)* model the distribution and spatial variation of these selected radionuclide species at the study area. While no prior data on the radionuclide activity of farmlands in the community exists, obtaining radionuclide information from the site could serve as baseline data for reference to future studies.

MATERIALS AND METHODS

Study area

The Birim North District of Ghana's Eastern Region has New Abirim as its administrative capital. Old Abirem, Akyemansa, and Ofoase all share a border with the town. This region is home to Newmont Cooperation's Akyem Gold Mine (see Figure 1). The evaluation is geographically confined to farmlands at New Abirem in the Birim North District. The district makes up less than one-tenth of the Eastern Region area, with an approximate land area of 1,250 km2. The Paleoproterozoic Birimian Supergroup and the underlying typical sedimentary Tarkwaian group encompass the research region of New Abirem (Attiogbe et al., 2017). Significant sections of these rocks, however, have been deposited as placer formations in a variety of streams and channels because of a succession of erosional episodes. The bulk of Ghana's rivers that drain Birimian rocks have placer gold deposits, commonly known as "alluvial gold". Near the river and floodplains channels of the Offin, Pra, Ankobra, Birim, and Tano Rivers, parts of which drain the study area, are also significant quantities of placer gold (Roy et al., 2018)

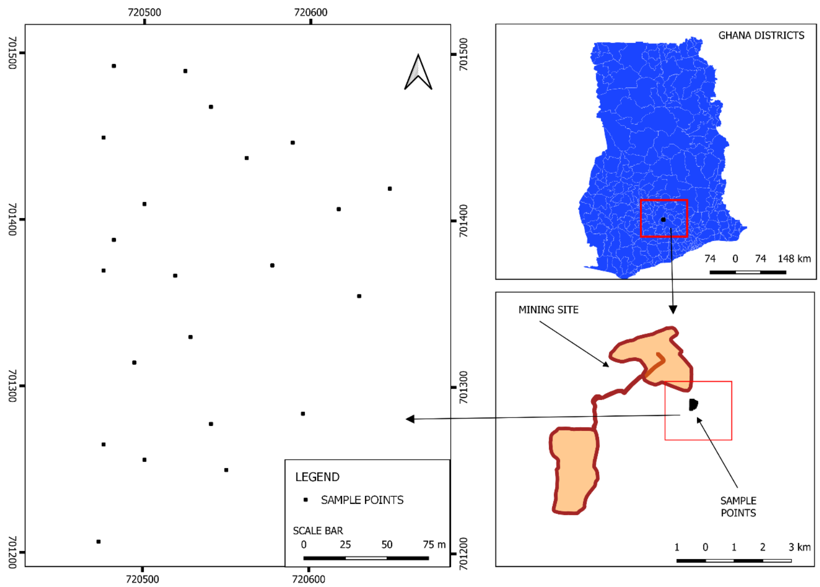

Figure 1: Map of Study Area and Locations of Sampling Stations.

Sample collection and preparation

At the study area, soil samples were obtained from 22 sampling stations which were selected using simple random samples from the chosen farmlands near the mine, as shown in Figure 1. At each sampling station averaging an area of $1m^2$, 4 core soil samples were collected from the top layer using a stainless-steel cylinder sampler and homogenized to obtain a representative sample. Samples from the sampling stations were collected into labeled ziplock bags, labeled, and transported to the laboratory. At the laboratory, the soil samples were prepped following the protocol by Vukašinović *et al*. (2010) with slight modification, by air-drying the soil samples for more than 24 hours, followed by screening using a sieve with a pore size of 0.25 mm. For each prepped sample from the sampling stations, 1 kg sample was weighed and put in an airtight polythene cylindrical container for more than 30 days to allow ^{238}U and its decay products to attain secular equilibrium before further gamma-ray measurements were taken (Bekelesi *et al*., 2017). An HPGe-ray spectrometry system was used to determine the amounts of ^{238}U, ^{232}Th, and ^{40}K.

The HPGe-ray spectrometry system used to test radioactivity was built on a p-type coaxial photon detector. The performance of the detector was greater than 32%. The spectrum was captured and processed using the software Gamma Vision (6.01) and a multichannel analyzer at energies ranging from 40 keV to 3 MeV. The entire detection device was contained under a 10 cm lead layer shield. Backgrounds were assessed and removed from the relevant photo peaks before and after all sample counts. The energy of the measuring system and efficiency calibrations were carried out using ^{238}U, ^{234}Th, and ^{40}K sources of the same size. The quantities of ^{238}U, ^{232}Th, and ^{40}K activity in soil samples were measured in Bq/kg dry weight.

The activity concentration of target metals in the samples was determined from the energy of 1001.0keV, 911.2keV, and 1460.8keV for ^{238}U, ^{232}Th, and ^{40}K, respectively. The analytical expression employed in the estimation of the activity concentrations in Bq/kg as presented in Equation 1:

$$A_{sp} = \frac{N_D}{P \times T_c \times \eta(E) \times M} \qquad \text{Equation 1}$$

Where A_{sp} = the activity concentration in Bq/kg, N_D = the net counts of the radionuclide in the samples, P = the gamma ray emission probability, T_c = the sample counting time, η(E) = the absolute counting efficiency of the detector system and M = the mass of the sample in kg (Augustine Faanu *et al*., 2016; Saadi, Azbouche and Benrachi, 2020).

Data Presentation and Geostatistical Analysis

Various software tools were used for data presentation and analysis. The descriptive statistical parameters of the data on the selected NORMs were determined using "R version 4.0.5 (2021-03-31)". To obtain prerequisite data for the distribution map construction, a global positioning system (GPS) was employed on the field to record the positions of the sampling stations. Geographic Information System (GIS) software was then employed to construct the radionuclide distribution map. The spatial geostatistical analysis was conducted using a Geostatistical Analyst.

Geostatistical Modeling

Geostatistics is based on the theory of a regionalized variable (Dragović *et al*., 2014), which shows spatial autocorrelation where observations close to each other are more alike than those that are far apart. The ordinary kriging (OK) method is the most commonly employed method among interpolation techniques for environmental situations (Dai *et al*., 2007). This study chose the OK method because of its advantages over other interpolation techniques. Ordinary Kriging (OK) is the top choice in geostatistics for estimating the value of a variable at unsampled locations using available data and the underlying spatial autocorrelation. What distinguishes OK is its extraordinary property of being the Best Linear Unbiased Estimator (BLUE). In essence, this means that

OK's predictions are not biased and provide the least variance among linear estimators. In practice, OK is the best method for providing unbiased and precise predictions when dealing with spatial data and its intrinsic spatial dependence patterns.

Geostatistical data modeling was done with the Geostatistical Analyst.

RESULTS AND DISCUSSION

The spatial distribution of the individual radionuclides

Using descriptive statistical methods, raw scores of the radionuclide concentrations were summarized in a more comprehensible form. The list of statistical data is presented in Table 1. The activity of ^{40}K was identified to contribute much to the radioactivity of these species in the study area with an average value of 6.43±4.18Bq/kg while ^{232}Th had the lowest concentration with an average of 1.32±0.68 Bq/kg. The activities range were 1.68-5.51Bq/kg, 0.52-3.39Bq/kg, and 3.20-17.11Bq/kg for ^{238}U, ^{232}Th, and ^{40}K, respectively. The concentrations of ^{238}U, ^{232}Th, and ^{40}K activity in our research were compared to documented ranges found in previous studies (26.90 Bq/kg, 71.8. Bq/kg, and 103.21 Bq/kg, respectively; as reported by Faanu et al.,2011; Faanu et al.,2016232Th and 40K in soil, rock, ore samples and gross alpha/beta analysis in water samples. The average absorbed dose rate in air at 1 m above sampling point using a radiation survey metre was determined to be 0.08 ± 0.02 μGyh−1 with a corresponding average annual effective dose calculated to be 0.093 ± 0.028 mSv. The average activity concentrations of 238U, 232Th, and 40K in the soil, rock, and ore samples were 65.1 ± 2.2, 71.8 ± 2.2 and 1168.3 Bqkg−1 respectively resulting in an average annual effective dose of 0.91 ± 0.32 mSv. The average Radium equivalent activity value was 257.8 ± 62.4 Bqkg−1 in the range of 136.6–340.2 Bqkg−1. The average values of external and internal indices were 0.7 ± 0.2 and 0.9 ± 0.2 respectively. The average gross alpha and gross beta activity concentrations in the water samples were determined to be 0.0032 ± 0.0024 and 0.0338 ± 0.0083 Bql−1 respectively. The total annual effective dose from the pathways considered for this study (gamma ray from the soil, rock and ore samples as well as doses determined from the gross alpha/beta activity concentration in water samples; and Moipone et al.,2021). The target radionuclides in these areas were found to be higher than levels identified in our study. This implies that the New Abirem mining community is less polluted, which could be attributed to the radionuclide concentration being less in the parent rock and or the mining activities engaged in at the New Abirem community is minimal. The individual, as well as mean activity concentrations of each radionuclide, were lower than the average world limit of 35, 30, and 400 Bq/kg for ^{238}U, ^{232}Th, and ^{40}K, respectively (UNSCEAR, 2000).

Table 1: Descriptive statistics of the activity concentration of target radionuclides in soil samples from farmlands at New Abirem, collected on 22nd October 2022

Statistics	^{238}U	^{232}Th	^{40}K
Mean	3.49	1.32	6.43
Standard deviation	1.07	0.65	4.18
Median	3.50	1.10	4.64
Minimum value	1.68	0.52	3.20
Maximum value	5.51	3.39	17.11
Range	3.83	2.87	13.91
Sum	76.79	28.98	141.41
Variance	1.15	0.42	17.45
Skewness	0.20	1.68	1.75
Kurtosis	-0.35	3.94	1.71

From the basic statistics, the symmetry and the degree of flatness of the distribution of the activity concentrations of the target radionuclides were assessed using skewness and kurtosis, respectively. As presented in Table 1, the value of skewness for the radionuclide species shows that the distribution of the activity concentration was positive, with the highest and lowest values recorded for ^{40}K and ^{238}U, respectively. The positive skewness of the activity concentrations of the target radionuclides is an indication that the soil samples that measured relatively lower activity concentrations were more than those that measured higher concentrations, in the order: ^{238}U > ^{232}Th > ^{40}K. Additionally, the leptokurtic nature of the activity concentrations of ^{232}Th and ^{40}K, shows a clustering of the activity concentration of these metals with a sharp peak, in contrast to the platykurtic nature of the activity concentrations of ^{238}U, pointing to the wider dispersion around the mean of the individual activity concentrations. Values of skewness and kurtosis close to zero obtained for ^{238}U showed the closeness to a perfect normal distribution by ^{238}U. However, high positive values obtained for ^{232}Th and ^{40}K deviated from a normal distribution, indicating that these species' datasets were asymmetrical and heavy-tailed. Using the Shapiro-Wilk test at an α-value of 0.05, the hypothesis that the data on each radionuclide was normally distributed was tested. The analysis revealed that the concentrations for ^{232}U were normally distributed with *p*-value = 0.52, as opposed to the *p*-values for ^{232}Th and ^{40}K, which were 0.01 and 0.00, respectively, suggesting that the activity concentrations of ^{232}Th and ^{40}K were not normally distributed, corresponding well with the conclusions from the skewness and kurtosis.

Digital distribution maps were plotted for the target terrestrial radionuclides from their activity concentrations for the study area. The distribution map for ^{238}U in Figure 2, showed high activity of the radionuclide from the northern to the central part of the study area while the southern section had relatively lower concentrations. Like Figure 2, the spatial distributions for the activity of ^{232}Th and ^{40}K were varied, with higher concentration in the northern part of the study area, evident from Figures 3 and 4. From Figure 3, the greatest activity for ^{232}Th was shown in the central part of the study. Figures 2 to 4 indicate that the structure of the parent rock northward and in the southern parts of the research territory may differ, with the northern section exhibiting high activity concentrations and likely containing parent rock with a high amount of these terrestrial radionuclides.

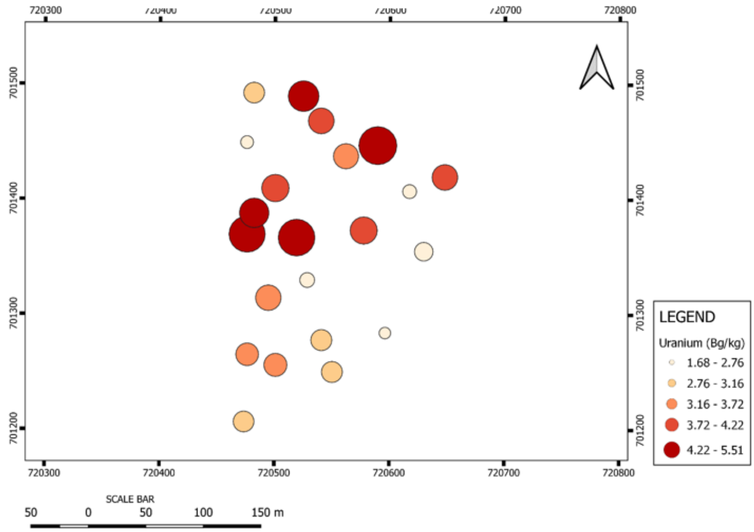

Figure 2: Map showing the spatial distribution of ^{238}U in soil samples from farmlands at New Abirem, collected on 22nd October 2022.

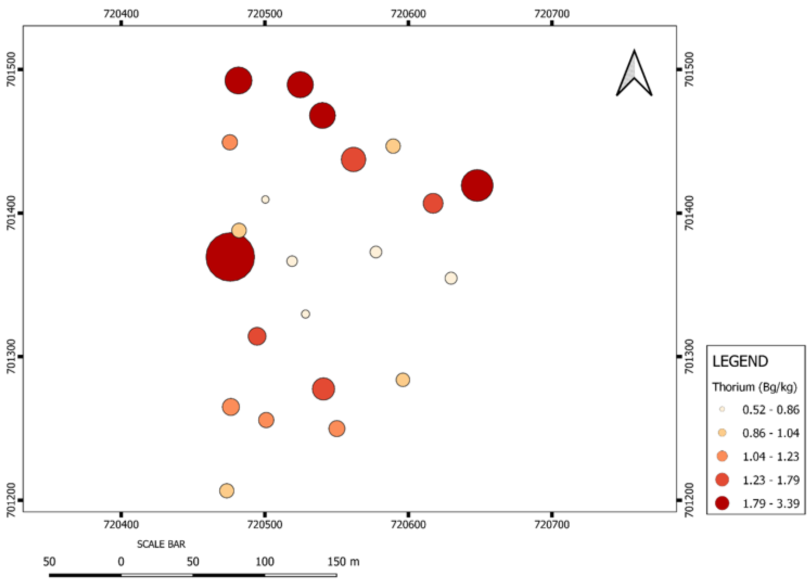

Figure 3: Map showing the spatial distribution of ^{232}Th in soil samples from farmlands at New Abirem, collected on 22nd October 2022.

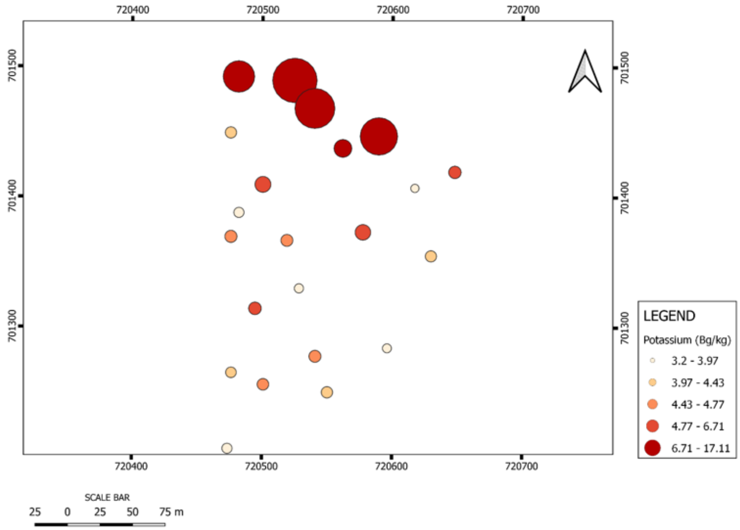

Figure 4: Map showing the spatial distribution of ^{40}K in soil samples from farmlands at New Abirem, collected on 22nd October 2022.

Spatial variability of radioactivity from predicted areas

Using the ordinary kriging technique, the concentrations of activity of the target radionuclides at unsampled points within the study area were estimated and presented in Figures 5-7. The predicted maps depicted in the figures showed a strong concentration gradient of the radionuclides across the site, an indication of a clear geographic variance among all the radioactive elements. According to the spatial variability map for ^{238}U in Figure 5, the concentration of activity is higher in soil and declines as one travels further from the northern portion of the research area, which may have been due to site fallout. Similar regional trends were observed in the spatial patterns for ^{232}Th and ^{40}K, where large quantities were found near the upper section of the study area. In comparison to the distance from the mining site, ^{40}K had a higher value than ^{238}U and ^{232}Th. Numerous factors, including intrinsic (soil formation factors, such as soil parent materials), and extrinsic factors (climate factors such as prevailing wind, and agronomic practices such as fertilizer application), may have had an impact on the regional variability of radionuclides in the soil (Kritsananuwat *et al.*, 2015; Adjirackor *et al*., 2017; Osman *et al.*, 2022). The predicted maps showed a strong resemblance to the distribution of the radionuclides from the sampled points as displayed in Figures 2-4. From the range of values displayed in Figures 2-4 in comparison to Figures 5-6, negative residuals were observed for the minimum values of ^{238}U and ^{232}Th, while positive residuals were recorded for the maximum values of these species. On the contrary, negative residuals were recorded for both the minimum and maximum values of ^{40}K, as shown in Figure 7. The deviation of $< \pm 2$ of predicted values from the minimum and maximum observed values of the target radionuclides suggests better prediction of the activities of ^{40}K, ^{238}U, and ^{232}Th at the study area by the model.

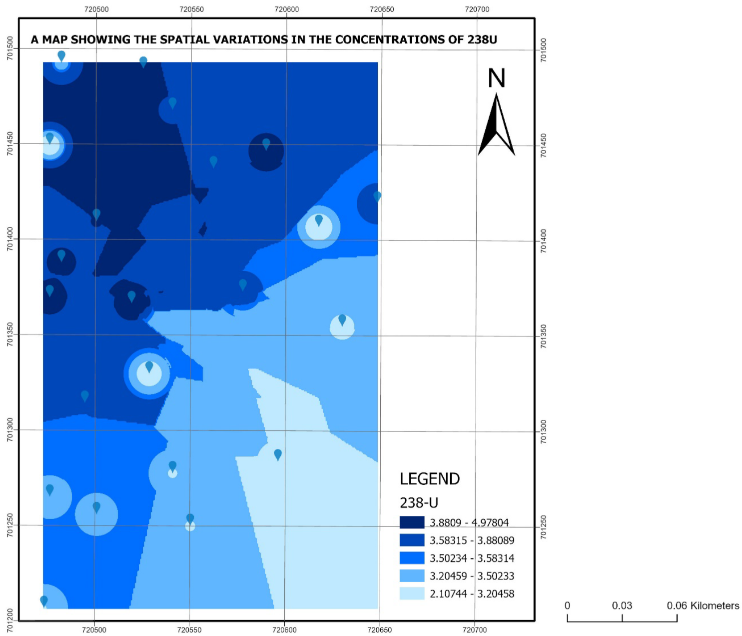

Figure 5: Map showing the spatial variations in the concentrations of ^{238}U in soil samples from farmlands at New Abirem, collected on 22nd October 2022.

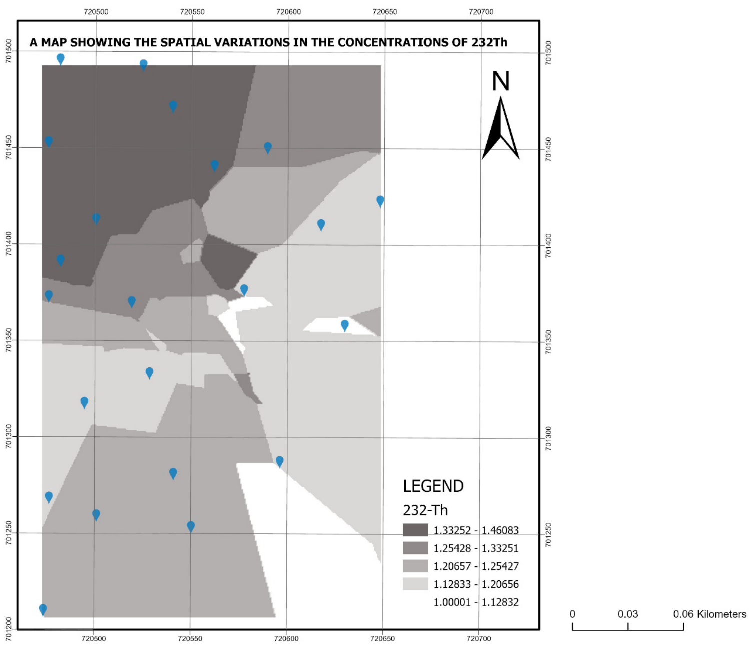

Figure 6: Map showing the spatial variations in the concentrations of ^{232}Th in soil samples from farmlands at New Abirem, collected on 22nd October 2022.

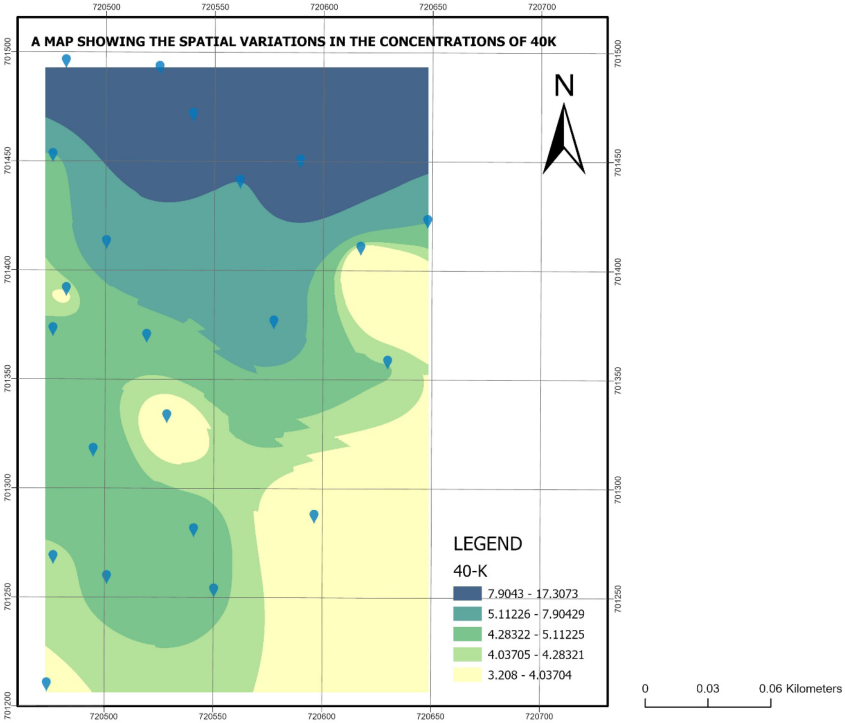

Figure 7: Map showing the spatial variations in the concentrations of ^{40}K in soil samples from farmlands at New Abirem, collected on 22nd October 2022.

CONCLUSIONS AND RECOMMENDATIONS

The study investigated the activity of terrestrial radionuclides - ^{238}U, ^{232}Th, and ^{40}K - in nearby farmlands near Akyem Concession at New Abirem. The mean activity concentrations of the target radionuclides, ^{238}U, ^{232}Th, and ^{40}K, measured from samples obtained from the study area were found to be below the average world limit of 35, 30, and 400 Bq/Kg, respectively. Based on the low radioactivity of the samples analyzed, the study area can be considered a low background radiation area, as such, results from this research can be employed as baseline data for studies conducted to assess the changes in the environmental radiation from anthropogenic sources.

The concentrations for ^{232}U were normally distributed, as opposed to those of ^{232}Th and ^{40}K, suggesting that the activity concentrations of ^{232}Th and ^{40}K were not normally distributed, corresponding well with the conclusions from the skewness and kurtosis

With the study area serving as farmland for crop cultivation as well as the continuous practice of mining close to the area, the development of effective management strategies to ensure the protection of the ecological integrity of the farmlands while safeguarding the health of farmers is recommended. Also, further studies should be conducted to ascertain the paths for the absorption of these radionuclides, particularly into the edible parts of the plants cultivated on this soil.

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