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Spatial distribution and lifetime cancer risk due to naturally occurring radionuclides in soils around tar-sand deposit area of Ogun State, southwest Nigeria

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HIGHLIGHTS

- Five different minerals were identified and characterized as major and minor minerals.
- Spatial distribution of ^{238}U, ^{232}Th and ^{40}K radionuclides in the study area were investigated.
- The mean activity concentrations of the radionuclides were in the order: $^{40}K > ^{232}Th > ^{238}U$.
- The tar-sand soil samples from the present study area are radiologically hazardous and pose a greater carcinogenic risk to the local population.

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ABSTRACT

Anthropogenically enhanced naturally occurring radionuclides ^{238}U, ^{232}Th and ^{40}K present in the tar-sand soil samples were measured by gamma ray spectrometer using a highly shielded NaI (Tl) detector with the aim of evaluating the environmental radiological hazards. Mineralogical analysis and characterization was carried out using Fourier transform infrared (FTIR) spectroscopy and X-ray diffraction (XRD) techniques. The results obtained showed that the average activity concentrations of ^{238}U, ^{232}Th and ^{40}K ranges from BDL↔76.00 ± 12.00, BDL↔204.48 ± 13.02 and BDL↔755.6 ± 40.15 $Bqkg^{-1}$ respectively. Eleven other radiological and hazard parameters were estimated to know the complete hazardous nature of the samples. The values obtained for radionuclides and all its associated radiological and health hazard parameters were all higher than the world's average set by UNSCEAR. Five different minerals were identified and characterized as major and minor minerals. The mean radionuclides activity concentrations, radium equivalent and dose rate of measured radionuclides were compared with other literature values. RESRAD computer code was used to calculate the probability of excess lifetime cancer incurred by dwellers of the tar-sand deposit site, the level of which was determined to be 2.3×10^{-3} $\left(\frac{23}{10000}\right)$. Spatial distribution of natural radionuclides in the study area was also investigated for radioactivity disequilibrium. Multivariate statistics were applied to establish the effect of mineralogy on radionuclides activity. Therefore, the tar-sand soil samples are considered to pose serious radiological hazard and cancer risk to human through the multiple routes of exposure and significant soil remediation action need to be taken for future use of the soils.

1. Introduction

Radionuclides contamination of soil pose serious threats to the environment and can damage human health through various absorption pathways such as direct ingestion, dermal contact, diet through the soil–food chain, inhalation, and oral intake (Lu et al., 2011). However various human activities such as tar sand mining and processing can release dangerous inorganic pollutants

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(radionuclides) into the soil, thereby elevating their level in the soils. Peasant farmers cultivate legumes, vegetables and other plants on the contaminated soil around these tar sand deposit area. Consequently, due to the soil-solute interaction which could lead to the transport of specific pollutants into groundwater and soil-to-plant transfer processes, human are exposed to these pollutants. Exposure to these harmful naturally occurring and anthropogenic enhanced radionuclide materials through human activity is mostly regarded as undesirable at every level due to their harmful effect on human health and the environment (Ademola et al., 2015). Moreover, human health risk assessment of these pollutants is an important environmental impact indicator of the tar sand mining sites. Hence, the need to evaluate the level of these harmful radionuclides materials in tar sand deposit in order to protect and find better approach of reducing health and radiological risk to the local populations is indispensable. Therefore, the primary objectives of the present study were: (i) to evaluate the activity concentration of naturally occurring radionuclides ^{238}U, ^{232}Th and ^{40}K in the representative soil samples from tar sand deposit area of Ogun State, Southwest Nigeria (ii) to explore the current extent of local radionuclides pollution in the tar sand soils in the study area (iii) to evaluate the spatial distribution, excessive lifetime cancer risk and other radiological hazard indices from the selected tar sand soil samples through multiple human exposure pathways to the general public and (iv) to identify the main radionuclides pollution sources and establish the relationship among the radionuclides and its associated radiological parameters using multivariate statistical analyses (Correlational, PCA and Cluster analysis). These provide a baseline data for future monitoring, management, and control of radionuclides pollution of the tar sand deposit sites in Nigeria.

2. Materials and methods

2.1. Description of the study area

Onikitinbi village (study area) is located within latitude 06°40′0″N and 06°50′0″ North of the Equator and longitude 06°10′0″E and 04°20′0″ East of the Greenwich Meridian (Fig. 1). The study area falls within the eastern part of Dahomey basin. The Dahomey basin (Benin) which covers much of the continental margin of Gulf of Guinea, extending from Volta-delta in Ghana in the west to the Okitipupa ridge in Nigeria in the east (Klemme, 1975). The basin is marginal pull-apart (Klemme, 1975) which develop in the Mesozoic due to the separation of Africa from America plate in the Mesozoic era (Burke et al., 1971; Whiteman, 1982). Occurrence of seepage of bitumen and tar sand deposits over the Okitipupa ridge in the Dahomey basin provided the initial impetus for oil exploration in Nigeria. The eastern Dahomey basin in the Nigeria sector, contain extensive wedge of cretaceous to recent sediment, up to 3000 m which thicken towards the offshore. The basin has been of much geological interest as a result of the reported occurrence of bitumen, tar sands, limestone, quartz, glass sand and phosphate (Nton, 2001). It is noteworthy that exploration activity for hydrocarbon commenced in Nigeria in this basin in 1908. Recently, due to increased government incentives to prospectors and re-evaluation of data gathered from such unsuccessful attempts there is a resurgence of interest in exploration activity in the eastern Dahomey basin particularly bitumen exploration.

2.2. Soil samples collection and preparation

A total of fifteen (15) randomly selected tar sand soil samples were obtained from the sampled locations using a hand-driven soil auger. The samples were taken within 0–15 cm depth and taken to the laboratory in a labeled polythene bags. The sample locations were recorded in degree-minute-second (latitudinal and longitudinal position) using a hand-held Global Positioning System (GPS) (Model GARMIN GPS-13). Samples were air-dried at laboratory temperature in order to avoid cross contamination, pulverized by grinding before made to pass through a 2 mm mesh sieve. Five hundred grams (500 g) of each sieved soil samples was subsequently weighed using an analytical weighing balance with a precision of ± 0.01 g and packed into a plastic marrinelli beaker. These samples were safely conveyed to National Institute of Radiation Protection and Research (NIRPR), University of Ibadan, Ibadan, Southwest Nigeria. At the laboratory, the sample plastic were sealed hermetically and externally with adhesive tape for about 5 weeks to ensure that the parent and daughter nuclides in the tar-sand soil samples were at secular equilibrium between radium and its gaseous decay progenies. At the end of the in-growth period of the samples in the laboratory, the samples were subjected to γ-ray spectroscopy counting in accordance with Isinkaye and Farai (2008).

2.3. Radioactivity measurements

2.3.1. System used for measurements

Activity concentration ^{238}U, ^{232}Th and ^{40}K were measured by gamma-ray spectrometry using NaI (TI) detector. The counting system used in the determination of the natural radionuclide contents of the soil consists of 760 mm × 760 mm NaI (TI) detector (Model Bicron) coupled to Canberra 10 multi-channel analyzer with adequate lead shielding which reduced the background by a factor of about 95%. The spectrometer was tested for its linearity and calibrated for energy and efficiency using the well calibrated standard gamma source obtained from an International Atomic Energy Agency (IAEA), laboratories, Vienna, Austria (Ademola et al., 2008). Efficiency is the measure percentage of radiation at a given detector coming from the overall yield that is emitted from the source into a solid angle of usually 4π in the photo-peaks (Hossain et al., 2012). Accuracy of efficiency calibration of detector is necessary to obtain the high precision measurements with radioactive samples. The resolution of the detector is 8% at 0.662 MeV of ^{137}Cs. This resolution is capable of distinguishing the gamma ray energies of interest in the study (Jibiri et al., 2014). All the samples were counted for 36,000 s in order to obtain good statistics for uranium, thorium daughter products and ^{40}K. Also measurements were repeated at intervals for quality assurance purposes as well as to ascertain the stability of the measuring system. The background radiation due to the naturally occurring radionuclides in the environment around the detector was measured by using an empty plastic container; the empty plastic container was measured in the same manner as the soil samples for the same counting time of 36 000s (10 h). The background spectrum was subtracted from the measured spectra to obtain the net radionuclides activities. The background, reference sample and the soil samples were measured under the same conditions (Jibiri et al., 2014). Three regions of interest in the spectrum were identified. These were centered on the three characteristic photo peaks at approximately 1.460 MeV for (^{40}K), 1.760 MeV for (^{214}Bi) and 2.614 MeV for (^{208}TI) in the samples was obtained. These were used for evaluating the activity levels of ^{40}K, ^{238}U and ^{232}Th series respectively. The activity concentration (A_c) of ^{238}U, ^{232}Th and ^{40}K in Bqkg^{-1} were obtained using the relation in equation (1) below (Amrani and Tahtat, 2001):

$$\text{Sample activity}(A_C)\left(\text{Bq } kg^{-1}\right) = \frac{C_i}{\varepsilon(E)P_\gamma\,(E)tm} \quad (1)$$

where C_i is the net peak area after subtraction of background of the