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Multivariate Pollution Risk Assessment of Natural Radionuclides and Heavy Metals in Topsoils of Nonwa, Rivers State, Nigeria

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Abstract

The health risk exposure to natural radionuclides and heavy metals in topsoils of crude oil polluted community was assessed with multivariate statistical tools. Six soil samples were collected from three sub-communities of Nonwa with hand auger to 20cm depth and analyzed for natural radionuclides using gamma-ray spectrometry and for heavy metals using atomic absorption spectrophotometer methods. Measured activity concentration of ²³⁸U, ²³²Th and ⁴⁰K are lower than world average values, except for ⁴⁰K in Nonwa-Tai community. The radiological hazard indices are lower than the safety limits except for indoor ELCR that was 14% higher than the safety standard in all the community soil while AGDE value in Nonwa-Tai community was 22% higher than safety limit implying residents' high exposure to organ diseases. The clark value characterized soils in Nonwa-Tai and part of Gbene-Nonwa as FELSIC minerals while soils in Nonwa-Uedume and part of Gbene-Nonwa as MAFIC minerals. The pollution hazard indices classified the soils as deficiency to minimal enrichment, none to moderate contamination, low to moderate degree of contamination, and no pollution to polluted soils. Cluster analysis grouped and classified heavy metals into clusters of their source similarity indicating petrogenic origin. PCA and CA confirmed that heavy metals are from agrochemical and petrogenic sources. Correlation analysis suggested that heavy metals are not responsible for increasing natural radioactivity level in topsoils. Lime applications to crude oil spilled areas were recommended to raise soil pH, reduce the toxicity of identified toxic metals to prevent groundwater pollution, crop bioavailability and bioaccumulation.

Keywords: Cluster Analysis, Correlation Analysis, Heavy Metals Pollution Health Risk, Principal Component Analysis, Radiological Hazards Health Risk

1 Introduction

Soil is an environmental repository that contains essential nutrients, acts as a sink for contaminants of potentially hazardous substances and as a pathway for contaminants' entry into the food chain which in the long run finds its route into human populations through ingestion, inhalation, and dermal contact [60,100]. The geochemical integrity of soils in areas or localities with intense industrial activity is constantly challenged by discharge of anthropogenic wastes into the environment and this negatively affects the inhabitants in those regions. The Niger Delta region located in the South-South zone of Nigeria exemplifies and represents such an environment that is challenged by anthropogenic waste discharge since it is seen as the country's economic nerve centre that hosts her dense petroleum refineries, palm-oil and fertilizer producing industries as well as manufacturing industries [42,103]. While boosting economic growth, these industries unwittingly and accidentally contribute to environmental decadence through crude oil spillages resulting from corroded/failed crude oil conveying pipelines, gas flaring stacks emitting effluents and particulate matter into the atmosphere that later settles/binds onto the soil, and in surface and underground waters. Besides, these emissions have the potential of enriching the soil with toxic or hazardous substances that ends up as contaminants in the soil, surface and underground water bodies [106].

Heavy metals and organic compounds like hydrocarbons are the primary constituents of crude oil and when spilled or its products are discharged into the environment, it would elevate the concentration levels of natural radionuclides and heavy metals resulting in a slight but significant increase in health hazard indices in affected areas, including annual effective dose equivalent and excess lifetime cancer risk [92]. [107] noted that assessment of radiological hazards is very necessary to ensure that the radiation exposure to the public is within the recommended safe limit. Thus, naturally occurring radioactive materials (NORMs) and potentially toxic metals (PTMs) from industrial processes of oil and gas exploration and fertilizer producing industries are among the contaminants that affects soil quality and enhance the concentration of natural radionuclides (such as ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K) and potentially toxic metals (like Pb, Cd, Cr, Ni and As) when released into the ecosystem [87-89,108,135]. The persistence bio-accumulative potential and the spectrum of carcinogenic and non-carcinogenic adverse health effects of these substances even at low levels of chronic exposures makes these substances of grave concern in the environmental studies [108].

The inhabitants of those communities within this industrial landscape are on the vanguard and forefront of this potential exposure to elevated levels of natural radionuclides and PTMs due to their heavy reliance on subsistence farming which exposes adults and children to direct contact with the local soils especially when children play in the soils that are reservoirs of these pollutants through dermal contact, accidental consumption of soil particles, and the consumption of food crops grown in contaminated soil that constitutes the primary exposure pathways [88]. While many researchers such as [13,65,90,80,109-111] have investigated either the metallic pollution or the radiological burden in parts of the Niger Delta, a comprehensive and integrated multivariate pollution risk assessment that simultaneously quantifies the synergistic threats of levels of natural radionuclides and potentially toxic heavy metals in the soils of heavily populated communities located in the economic nerve centre of

Nigeria's South-South geopolitical zone remains a yawning gap of knowledge. Utilizing multivariate statistical tools such as Principal Component Analysis (PCA), Cluster Analysis (CA), and Pearson Correlation Analysis (PCorA) would help to simplify, deconvolve and resolve complex pollution datasets, differentiate natural geogenic background variability from anthropogenic inputs, and identify whether the specific sources of contaminants are either petrogenic or lithogenic.

The study area, Nonwa and its sub-communities host numerous oil and gas facilities and have documented histories of crude oil spills due to systemic pipeline failures, illegal artisanal crude oil refining activities commonly referred to oil bunkering or Kpofire by the dwellers, and gas flaring activities which have contributed to the pollution matrix of the area and have left the area severely degraded [30,49]. The Federal Government of Nigeria is currently constructing her first indigenous crude oil export terminal at Nonwa community. The facility has the capacity of pumping 360,000 barrels per day (Bpd) of crude into a storage capacity of 750,000 barrels and would boost the country's crude export infrastructure [6]. This highlights the community as a significant and active hydrocarbon hub whose activities would increase the risk of hydrocarbon pollution in the area. Besides, there have been documented reports of illegal artisanal crude oil refining in bushes, creeks, and illegal tapping of crude oil on the Trans Niger Delta pipeline (TNP) in Nonwa sub-communities where crude oil has been siphoned for over 7 years, and loading two 33,000 litres tankers of crude oil every night thereby polluting the area with crude oil hydrocarbon [5, 7, 9-10]. The livelihoods of the inhabitants of the communities are agriculture, fishing and the use of soil for civil infrastructures like construction of residential dwellings and industrial buildings. This study therefore: (i) determines the activity concentration of natural radionuclides and the concentration levels of selected potentially toxic heavy metals in topsoils; (ii) evaluate the radiological hazards to human health due to exposure to natural radionuclides in soil; (iii) assess the human health risks associated with the potentially toxic heavy metals; and (iv) using multivariate statistical analysis to identify the potential sources of pollution. The findings of this research will serve as an important scientific guide for environmental monitoring, provide informed decision on public health policy and recommend remediation strategies on safeguarding the well-being of the vulnerable sensitive populations that would use the soil for construction of civil infrastructures of dwellings or that may directly ingest the soil particles or indirectly consume the soil pollutants through ingestion of foods or crops grown from the soil in these industrial communities located in the Niger Delta. This research would further provide a strong data-driven framework for radiological and heavy metal pollution risk assessment in the area.

2 Materials and Methods

2.1 Study Area

The community of Nonwa is one of the communities that consist of Ogoniland which is located in Tai Local Government Area of Rivers State, Nigeria with coordinates of Latitudes 04°44'28.33''N and 04°46'41.27''N of the equator and Longitudes 007°12'12.42''E and 007°13'18.79''E of the Greenwich Meridian (Figure1). It lies on the *Benin Formation* which is often referred to as the "Ogoni Sands" that consists of the

coastal plain sands that are unconsolidated and deposited by the Niger River and its tributaries [4]. The formation occurred during the tertiary period of the Oligocene-Pleistocene epoch while the receding water that deposited the coastal plain sands occurred during the Miocene-Pleistocene epoch in the Cenozoic era [61]. The geology of Ogoniland show evidence of shallow water depositional environments in the coastal plain sands (Qp) formation and the structural units such as natural levees, back swamp deposits and point bars that were identified within these sands indicate a variable depositional history [8]. All aquifers in Ogoniland are located within the Benin litho-stratigraphic unit that are highly porous coarse grained fresh water bearing sandy soils with few shale intercalations [91]. The *Benin Formation* also consists of multiple layers of clay, clay conglomerates, peat and lignite of variable thickness and texture [52]. [3] characterized the kaolin clay unit in Ogoniland as “Ogoni Clay” and were found very suitable for certain industrial uses. The area is unevenly flat lying with a wetland landscape and consists of terrestrial and marine environment dominated by mangrove forests and swamps. The climate of the area is defined by a short dry season and long wet season that spans from March to November with abundant annual rainfall. The mean rainfall ranges from 3000mm to 4500mm [47, 49, 93].

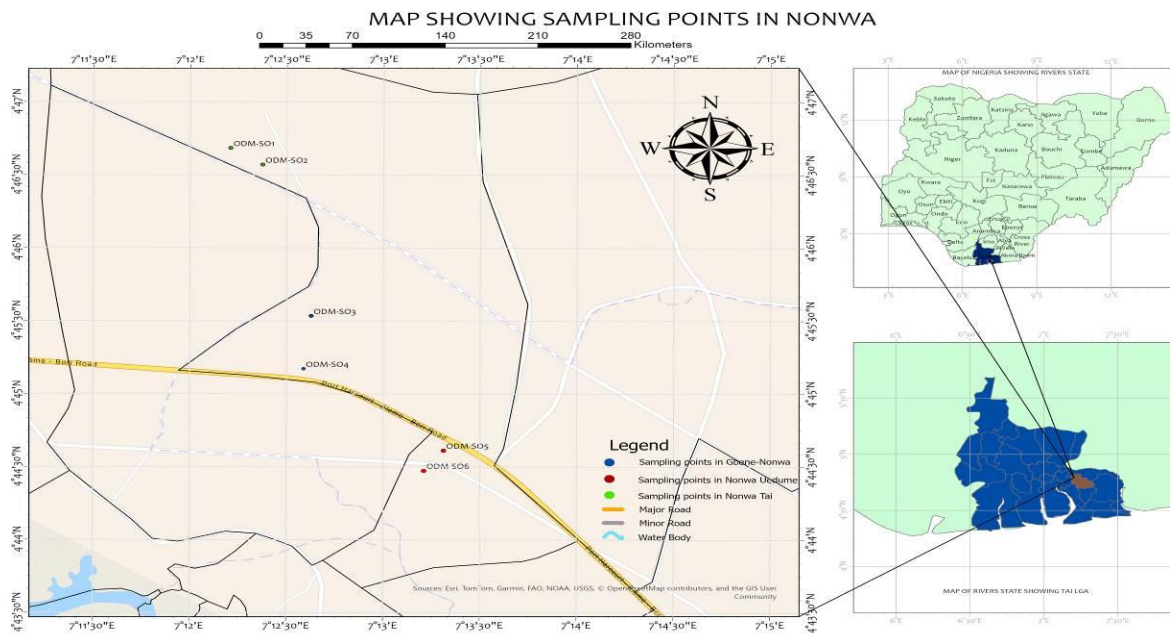


Fig. 1: Map of Rivers State Showing the Study Area and Soil Sampling Points

2.2 Data Acquisition

This study employed the purposive and stratified random soil sampling methods to collect soil samples according to international established experience by [70, 94-95]. The site in the study area consists of 3 sub-communities of Nonwa (namely Nonwa-Tai, Gbene-Nonwa and Nonwa-Uedume). These 3 sub-communities were split into sampling areas then were further divided into cells of 50 metres by 50 metres grids. A unique number was assigned to each grid block and specific sampling grid points or locations where samples were collected was randomly chosen by using a simple random generator (for instance choosing from N identical cards) to ensure unbiased

selection or randomization within defined boundaries of the study area. In each of the 3 sub-communities two (2) sampling points that are 30 metres apart was chosen as where soil samples will be collected such that 2 sampling points were each chosen at Nonwa-Tai, Gbene-Nonwa and Nonwa-Uedume communities giving a total of 6 sampling points. Five (5) cores were drilled into the soil within each of the cells in a zigzag pattern to a depth of 20cm as the international acceptable depth compromise to ensure adequate representation of the soil profile arising from alternate measures that are often based on deposition of radioactivity in soil per unit area assuming atmospheric fallout [88]. Thus five (5) soil samples were collected from the cores using a stainless steel soil hand auger and a spatula which have been thoroughly cleaned with acid, washed with detergent, then rinsed with distilled water before it was used to drill to the 20cm required depth. The soil samples that were collected from the various points of the drilled 5 cores were mixed thoroughly to give a composite sample in each grid of the cells. The thorough mixing of the soil samples was to ensure that a homogenous sample that is representative of the soil of the sampled area was obtained. The samples were stored in a clean aluminum foil enclosed in a blank polyethylene bag in order to prevent them from atmospheric humidity. All the samples were properly labeled in-situ. The communities and their respective coordinates where soil samples were collected are shown in Table 1.

Table 1: Co-ordinates of the Study Area

Community	Sampling Location	Co-ordinates	
		Northing	Easting
Nonwa-Tai	ODM-SO1	4°46'41.27"N	7°12'12.42"E
	ODM-SO2	4°46'34.15"N	7°12'41.86"E
Gbene-Nonwa	ODM-SO3	4°45'32.08"N	7°12'37.61"E
	ODM-SO4	4°45'10.27"N	7°12'35.36"E
Nonwa-Uedume	ODM-SO5	4°44'36.72"N	7°13'18.79"E
	ODM-SO6	4°44'28.33"N	7°13'12.69"E

2.2.1 Soil Sample Preparation and Counting for Radioactivity

Stones and organic materials were removed from the collected samples in the laboratory then soil samples were oven-dried at a temperature of about 105°C for several hours to remove excess moisture content. The dried samples were crushed into fine grains of <2mm grain size and then sieved through a 150µm mesh to remove any remaining debris and to homogenize the clay and mineral particles present. About 0.1 - 0.5kg of the representative portions of the crushed samples were weighed into a clean dry Marinelli beaker, then sealed with an adhesive tape and was left for 28 days to attain secular equilibrium between the long-lived parent radionuclide and their short-lived daughter radionuclides.

The samples were thereafter placed in the detector for a pre-set time to obtain spectrums for counting. The acquired spectrums from the counting were analyzed with software in order to obtain energies that correspond to the different radionuclides present in the sample. The activity of natural radionuclides of the collected soil samples were determined with a 76mm x 76mm Thallium activated Canberra Sodium Iodide [NaI(Tl)] detector at the Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Nigeria. Each of the samples was counted for 10 hours (36000

seconds) in the NaI[Tl] detector with an installed Maestro software and was coupled to a multichannel analyzer (MCA) model 803 through a preamplifier which was adequately shielded by Lead (Pb) to reduce the background radiation by about 95%. The NaI[Tl] has an energy resolution of 8% at 0.662MeV (^{137}Cs). The energy values upon which ^{238}U , ^{232}Th series and ^{40}K activities of the radionuclides measurements were to be based in the soil samples were initially determined. The analysis was done for four (4) weeks to ensure that the activity concentration of each radionuclide and their progenies in the soil samples were identified. The activities of ^{238}U was determined from the average activities of its progenies (^{214}Pb at 352Kev and ^{214}Bi at 609Kev) at gamma energy of 1.760MeV while that of ^{232}Th was determined from the average activities of its progenies (^{208}Ti at 583Kev and ^{228}Ac at 911Kev) at gamma energy of 2.615MeV and the activity concentration of ^{40}K was determined with only its gamma energy of 1.460MeV. The activity of the respective radionuclides in the soil samples was calculated after subtracting decay corrections. However, the background spectra that were measured under the same conditions for both the standard and sample measurements were used to correct the calculated sample activity concentrations. The specific activity concentrations of the counted radionuclides in the soil samples were estimated with the relationship adopted by [63] and [64]:

$$AC_{sp} \text{ (Bq/kg)} = \frac{C_n}{\epsilon P_\gamma M_s t} \dots \dots \dots 1$$

Where: AC_{sp} is the activity concentration of radionuclide in soil samples in Bq/kg; C_n is the count rate under each photo peak due to each radionuclide; ϵ is the detector efficiency for the specific gamma-ray energy; P_γ is the absolute transition probability of the specific gamma-ray; M_s is the mass of the soil sample (kg); and t is the counting time in seconds (s).

2.2.2 Soil Sample Preparation and Measurement for Physico-chemical Parameters and Heavy Metals

Clods and clumps were removed from the soil samples then broken into pieces. The samples were later dewatered by drying at 105°C for 48 hours. Thereafter samples were crushed in a mortar and sieved using stainless steel sieves with a mesh size of <2mm to remove large debris, plant roots and gravel size materials. The sieved samples were digested with a strong acid (such as HNO_3) to dissolve the samples and their inorganic contents in solution before subjecting them to heavy metal analysis. The concentration levels of lead (Pb), arsenic (As), zinc (Zn), copper (Cu), cadmium (Cd), chromium (Cr), manganese (Mn) and nickel (Ni) in the soils were determined with Atomic Absorption Spectrophotometer (AAS) method following recommended procedures by [21, 24, 58]. The AAS was fitted with background correction devices that employs the principle that atoms in the ground state absorbs light of wavelengths that are characteristic to each element when light is passed through the atoms in the vapour state[11, 22-23]. The soil pH and electrical conductivity (EC) was determined in-situ electronically by using the glass method with a standard calibrated pH meter. The procedure adopted was weighing about 0.02kg of soil samples in a beaker, and adding 100ml of water to it, then stirred gently and allowed to stand for 30 minutes before

introducing the pH meter into the soil-water suspension for 60 seconds. Readings were thereafter taken. The soil pH was taken in-situ because soil chemistry is sensitive to environmental changes [57, 128].

2.3 Evaluation of Radiological Hazard Indices and Heavy Metals Hazard Indices in Soil Samples to Human Health

2.3.1 Radiological Hazard Indices in Soil samples to Human Health

The radiological risk assessment of the health status of radiated and irradiated persons from the measured radionuclides in soil samples of the investigated communities was carried out using nine (9) radiological health hazard indices as recommended by [66, 97] and defined in [44] as follows:

(i) Radium Equivalent Activity (Ra_{eq}): Is a single quantity that assesses the combined effect of the radiation hazards from any material that contains ^{238}U , ^{232}Th and ^{40}K [83]. The radium equivalent activity was calculated using the relation by [96]:

$$Ra_{eq} \text{ (Bq/kg)} = A_u + 1.43A_{Th} + 0.077A_k \dots \dots \dots 2$$

Where A_u , A_{Th} and A_k are respectively the activity concentration of ^{238}U , ^{232}Th and ^{40}K in Bq/kg. The assumption applied in the above equation was that 10Bq/kg of ^{238}U , 7Bq/kg of ^{232}Th and 130Bq/kg of ^{40}K produced equal gamma dose as this will allow for the calculation of the percentage contribution of the three radionuclides to the gross activity in the sample [44, 88].

(ii) Absorbed Dose Rate (D): Measure the uniform distribution of soil radionuclides in the study area that was radiated or deposited in the air emitted from the soil radionuclides at 1.0m above the ground surface which was absorbed by individuals. This radiological quantity was computed by applying the formula given by [68 – 69]:

$$D \text{ (}\mu\text{Gy/h)} = 0.462A_u + 0.604A_{Th} + 0.0417A_k \dots \dots \dots 3$$

Where A_u , A_{Th} and A_k are respectively the activity concentration of ^{238}U , ^{232}Th and ^{40}K in Bq/kg.

(iii) Committed Outdoor and Indoor Annual Effective Dose Equivalent (AEDE): This hazard index measures both the stochastic and deterministic risks of the effects of the radiations from radionuclides in soil on the irradiated individuals living in an area. This study examines two exposure scenarios: (a) outdoor exposure, and (b) indoor exposure. The outdoor public exposures of individuals to radiations from radionuclides in soil ($AEDE_{(Outdoor)}$) refers to the total effective dose equivalent that an individual would receive strictly from being outdoors in 365 days or one (1) year due to ionizing radiation sources present in the outdoor environment from terrestrial gamma radiation emitted from naturally occurring radionuclides present in the soil and rocks [66 – 67, 97, 136]. The indoor annual effective dose equivalent ($AEDE_{(indoor)}$) refers to the total

effective equivalent dose that an individual would receive in one (1) year from radiation sources present in the indoor environment such as radiations emitted from radionuclides that are naturally present in building materials like concretes, bricks, stones and gypsum [97]. An individual can also be exposed to radiation indoor through inhalation of radioactive gases such as ^{222}Rn that emanates from the soil beneath the building and from building materials[66]. Another source is when the short-lived solid decay products of radon and thoron get attached to aerosol particles in the air and are inhaled; they are then deposited in the lungs delivering a significant radiation dose to the bronchial epithelium [66, 68 – 70, 96]. Due to the enumerated health risk of AEDE that irradiated individuals would experience both for the outdoor and indoor environment, the outdoor and indoor AEDE was computed using the relation by [96, 116]:

$$\text{AEDE (mSv/y)} = D \times T \times \text{OF} \times C \dots\dots\dots 4$$

$$\text{AEDE}_{(\text{Outdoor})} \text{ (mSv/y)} = D \times 8760 \times 0.2 \times 0.7 \times 10^{-6} \dots\dots\dots 5$$

$$\text{AEDE}_{(\text{Indoor})} \text{ (mSv/y)} = D \times 8760 \times 0.8 \times 0.7 \times 10^{-6} \dots\dots\dots 6$$

Where D is the absorbed dose rate in air, T is the average time an individual is exposed to radiation from radionuclides in one (1) year (i.e. 24 hours per day x 365 days = 8760hours/year), OF is the occupancy factor which is taken as 0.2 for outdoor and 0.8 for indoor, and C is the dose conversion factor taken as 0.7Sv/Gy.

(iv) Annual Gonadal Dose Equivalent (AGDE): The annual gonadal dose equivalent (AGDE) as a radiological metric estimates the radiation dose to the reproductive organs (or gonads) which a person would receive over one year from environmental radiation sources [12, 25, 96, 127]. The gonads also consists of the bone marrow and the bone surface cells and are considered as organs of interest by [66, 96, 97] due to their sensitivity to radiation because an increase in AGDE has been known to affect the bone marrow, causing destruction of the red blood cells that are then replaced by white blood cells. This situation results in a blood cancer called leukemia which is fatal [85]. The AGDE due to the activities of ^{238}U , ^{232}Th and ^{40}K for resident of Nonwa communities using the soil for building was evaluated with the equation [86, 97]:

$$\text{AGDE (mSv/y)} = 3.09A_{\text{U}} + 4.18A_{\text{Th}} + 0.314A_{\text{K}} \dots\dots\dots 7$$

Where A_{U} , A_{Th} and A_{K} are respectively the activity concentrations of ^{238}U , ^{232}Th and ^{40}K in Bq/kg for the soil samples.

(v) Excess Lifetime Cancer Risk (ELCR): The ELCR was used to estimate the probability or likelihood of an individual or a population contracting cancer due to exposure to a specific amount of ionizing radiation from radionuclides above and beyond the baseline or natural incidence of cancer [66, 96]. The word “excess”

particularly applies to the additional risk attributed solely to the radiation exposure [66]. The Excess lifetime cancer risk for this study was computed using the relation [66]:

$$\text{ELCR} = \text{AEDE} \times \text{DL} \times \text{RF} \dots \dots \dots 8$$

Where AEDE is the annual effective dose equivalent, DL is the average duration of life of an individual or members of the public which was estimated to be 70 years for Nigeria by [66, 119], and RF is the risk factor or the fatal cancer risk per sievert which was given by [66] as 0.05. This study examined ELCR for only indoor individuals since [96] and [97] assumed that most persons spend more time indoors (19 hours per day) than outdoors (5 hours per day) hence exposing indoor dwellers more to radiations from natural radionuclides in every 24 hours.

(vi) Activity Utilization Index (AUI): The activity utilization index is a regulatory decision index used to decide whether a particular material can be utilized in a product as building material or it must be treated as radioactive waste [70]. This index also measures the hazards or risks of external gamma radiation when an individual is standing next to or is being surrounded by the material such as living in a house made of brick or standing on a highly radioactive soil [70, 71]. [71 – 72] stated that for a material to be considered suitable for unconditional release or utilization, AUI must be less than or equal to 2. The AUI of soil in this study was computed with this relation [70 – 72, 116]:

$$\text{AUI} = \frac{A_U}{150} + \frac{A_{Th}}{100} + \frac{A_k}{1500} \leq 2 \dots \dots \dots 9$$

Where A_U , A_{Th} and A_k are respectively the activity concentration of ^{238}U (^{226}Ra), ^{232}Th and ^{40}K

(vii) Internal Hazard Index (H_{in}): The internal hazard index examines the potential risk from internal radiation caused by ingestion or inhalation of radionuclides, especially the hazards caused to humans or individuals when they inhale radon gas released from soil or building materials which leads to contracting lung cancer when inhaled and deposited in the epithelium of the lungs [70 – 72]. The internal hazard index therefore helps to limit the activity concentration of ^{226}Ra in a material such that the emitted radon gas does not accumulate to dangerous levels in a confined room [55]. The internal hazard index was computed using the relation [54]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \leq 1 \dots \dots \dots 10$$

Where A_{Ra} , A_{Th} and A_k are respectively the activity concentration of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K

(viii) External Hazard Index (H_{ex}): The external hazard index evaluates the external gamma radiation dose rate inside a structure to ensure that the gamma radiation dose rate does not exceed a defined safety limit [76, 120]. The acceptable safety limit value

for H_{ex} so that a material can be classified as acceptable is less than unity (<1) [55]. The external hazard index for this study was computed with the relation [76, 120]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \leq 1 \dots \dots \dots 11$$

Where A_{Ra} , A_{Th} and A_k are respectively the activity concentration of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K

(ix) Clark Value: The Clark value is the average elemental mass concentration of a radioisotope in the earth crust which is used as a baseline to determine if a material has been enriched or depleted in radionuclides arising from industrial processes [1, 143]. The Clark value is therefore seen as a geochemical concept that distinguishes between natural background radiation levels and the enrichment or depletion of radioactive materials through anthropogenic processes, which also serve as a baseline for identifying environmental contamination by NORM or radioisotopes such that measured concentrations of radionuclides above Clark value for uranium and thorium means accumulation of NORM [1, 20, 143]. The Clark value also provides the basis for regulatory decisions as regards radiation protection measures since it helps to assess whether the activity concentrations of radionuclides are of natural origin or have been enriched technologically through anthropogenic means. The clark value was computed using the relation by [1, 143]:

$$\text{Clark Value} = \frac{[\text{Th}]}{[\text{U}]} \dots \dots \dots 12$$

Where $[\text{Th}]$ and $[\text{U}]$ are respectively the elemental mass concentrations of thorium and uranium expressed in parts per million (ppm). However, the activity concentration values of uranium and thorium (in Bq/kg) were converted to ppm using the following relations [137]:

$$1\text{ppm eU} = 12.35\text{Bq/kg of } ^{238}\text{U} \dots \dots \dots 13$$

$$1\text{ppm eTh} = 4.06\text{Bq/kg of } ^{232}\text{Th} \dots \dots \dots 14$$

$$1\text{ppm \%K} = 313\text{Bq/kg of } ^{40}\text{K} \dots \dots \dots 15$$

2.3.2 Heavy Metal Pollution Indices in Soil Samples to Human Health

The quality of soil samples collected in the investigated communities was evaluated with five (5) heavy metals pollution hazard indices and was analyzed also with some statistical tools. The pollution hazard indices employed include:

(i) The enrichment factor (EF): This is an index that estimates the degree of contamination, distinguishes between natural and anthropogenic sources of heavy metals in soil or sediment and determines how much an environment has been modified, enriched or depleted with metals as a result of extraneous or anthropogenic activities [129]. [46] opines that $EF \approx 1.0$ implies that the metal is of crustal origin and $EF < 1.0$ denotes mobilization or depletion of metals (Table 2). [53] stated that $EF > 1.0$

implies that metal is of anthropogenic origin (Table 2). However, [130] and [132] adduced that $EF < 2$ implies deficiency to minimal enrichment while $EF > 40$ connotes extremely high enrichment (Table 2). The enrichment factor of the heavy metals in the soil samples were analyzed using the relation [48]:

$$\text{Enrichment Factor (EF)} = \frac{C_n(\text{sample})/C_{ref}(\text{sample})}{B_n(\text{control})/B_{ref}(\text{background})} \dots \dots \dots 16$$

Where $C_n(\text{sample})$ is the content of the examined metal in the examined (polluted) environment (Nonwa), $C_{ref}(\text{sample})$ is the content of the reference metal in the examined (polluted) environment (i.e Nonwa) = 206.7mg/kg, $B_n(\text{control})$ is the content of the examined metal in the unpolluted environment (i.e the control) = 98mg/kg, $B_{ref}(\text{background})$ is the background value of the reference metal in the reference environment (polluted). The $B_{ref}(\text{background})$ was taken from literature by [78]. Metals such as Scandium (Sc), manganese (Mn), aluminium (Al) and iron (Fe) have been commonly used as reference elements as recommended by [59]. However due to the reaction of Fe with As, Mn was used as the stable reference element in this study.

(ii)The Contamination factor (CF) is the ratio of the measured concentration of each metal in the soil sample to its baseline or background value [131]. It is expressed as [56]:

$$CF = \frac{\text{Concentration of trace metal in sample}}{\text{Background value}} \dots \dots \dots 17$$

Where the background value is the Niger Delta baseline value of the investigating metal in the investigating environment in question (Nonwa communities) and these values were extracted from literature by [78]. [56] and [74] classified the contamination levels on the basis of their intensities on the scale ranging from 1 to 6 (Table3).

(iii)The Geo-accumulation Index (I_{geo}) is a classification system for the degree of metal enrichment when compared to the background [126, 138]. It is expressed as:

$$I_{geo} = \log_2 (C_n/1.5 \times B_n) \dots \dots \dots 18$$

Where C_n is the concentration of the measured metal in the soil sample; B_n is the Background concentration of that metal in the earth crust. Since this study is investigating the metal pollution level in the Niger Delta region, the background concentration values of the investigated metal was taken from literature by [78]. The 1.5 in the formula is a factor that was used to account for the natural variations in background levels. The [56] and [138] I_{geo} classification scale (Table 4) was adopted for the classification of the metal contents in the soil samples in this study.

(iv)The Degree of Contamination (C_d) is the index that was used to assess the overall contamination by the various metals in the soil samples. It represents the summation of

all the contamination factors for the measured heavy metals and was obtained by applying the relation [74]:

$$C_d = \Sigma(CF) \dots \dots \dots 19$$

The [74] interpretation scale (Table 5) was used to interpret the degree of contamination of the investigated soils in the study area.

(v)The Pollution Load Index (PLI) is an index used to assess the total degree of contamination in soil or sediment samples and it presents a simple comparative means to assess the pollution status of soils or sediments [134]. It was calculated as the n^{th} root from the n-concentration factors that was obtained for all the heavy metals in the study area by using the formula [50].

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n} \dots \dots \dots 20$$

Where CF is the contamination factor, n is the number of metals. The PLI classification scale by [73] and [134] (Table 6) was used to classify the pollution load index for this study.

Table 2: Enrichment Factor (EF) Interpretation Scale

EF Value	Description of Modification Level	Soil/Sediment Quality	Source
> 1.5	Human Influence		[45, 82]
1.5-3.0	Minor Human Modification		
3.0-5.0	Moderate Human Modification		
5.0-10.0	Severe Human Modification		
> 10	Very Severe Human Modification		
<2		Deficiency to Minimal Enrichment	[130, 132]
2 - 5		Moderate Enrichment	
5 - 20		Significant Enrichment	
20 – 40		Very High Enrichment	
> 40		Extremely High Enrichment	

Table 3: Geo-accumulation (I_{geo}) Interpretation and Classification Scale [2]

I_{geo} Value	Class	Description
≤ 0	0	Practically uncontaminated
0 – 1	1	Uncontaminated to Moderately Polluted
1 – 2	2	Moderately Contaminated
2 – 3	3	Moderately to Strongly Contaminated
3 – 4	4	Strongly Contaminated
4 – 5	5	Strongly to Very Strongly Contaminated
> 5	6	Very Strongly Contaminated

Table 4: Contamination Factor (CF) Interpretation Scale

CF Intensity Value	Category/Contamination Level	Source
0	None Contamination/Pollution	[56]
1	None to medium Contamination/Pollution	
2	Moderate Contamination/Pollution	
3	Moderately to Strong Contamination/Pollution	
4	Strongly Polluted	
5	Strong to very Strong Pollution	
6	Very Strong Pollution	
< 1	Low Contamination Factor	[74]
$1 \leq CF < 3$	Moderate Contamination Factor	
$3 \leq CF < 6$	Considerable Contamination Factor	
$CF \geq 6$	Very High Contamination Factor	

Table 5: Degree of Contamination (C_d) Interpretation Scale [74]

C_d Value	Contamination Degree Description
$C_d < 8$	Low Degree of Contamination
$8 \leq C_d < 16$	Moderate Degree of Contamination
$16 \leq C_d < 32$	Considerable Degree of Contamination
$C_d \geq 32$	Very High Degree of Contamination

Table 6: Pollution Load Index (PLI) Interpretation Scale [73, 134]

PLI Value	Description/ Interpretation
$PLI > 1$	Polluted
$PLI < 1$	No Pollution (background levels Present)
$PLI = 1$	Baseline level of Pollution

2.3.3 Statistical Analysis of the Natural Radionuclides and Heavy Metals in the Soil Samples

Multivariate statistical tools such as the R-mode hierarchical cluster analysis (CA), principal component analysis (PCA) and pearson correlation analysis (PCorA) were employed for this study. CA was done on the original data set to group objects into clusters so that objects within a class are similar to each other, but different from those in other clusters [79]. The R-mode CA was specifically used to classify parameters into groups in this study based on their similarity with each other in order to examine and visualize the spatial groupings of measured variables of the physico-chemical and heavy metal parameters in soil, thus classifying them into clusters that presents the picture of the groups and their proximity [51, 79]. PCA was run to extract latent information from the physico-chemical and heavy metals data and group the measured elements into fewer groups for interpretation in order to examine the root or source of the soil quality parameters. This helps to know whether the source was due to geogenic (natural) or anthropogenic (human influenced) activities in the area [84]. After the utilization of the PCA, a normalized varimax rotation was applied to minimize the variances of the factor loadings across variables for each factor. Only factors with eigenvalues greater than or equal to 1.0 was accepted as possible sources of variance in the data [81]. The PCorA was performed to ascertain the relationship between the physico-chemical parameters/heavy metals and the natural radionuclides content in the soil. Moreover, other statistical tools such as mean, standard deviation (SD), and graphical visualization charts were used to present data for clearer understanding. Hence data values were reported as: mean $\pm$ SD and $p < 0.05$ for all comparisons. The statistical package for social sciences (SPSS) version 27 software was used for the data analysis.

3 Results and discussion

3.1 Assessment of Soils for Natural Radioactivity and Associated Radiological Health Risk Indices

The activity concentration of natural radionuclides in soil samples for ^{238}U , ^{232}Th and ^{40}K (Table 7) respectively ranged from 17.52 ± 4.50 to $32.82 \pm 4.21 \text{ Bq/kg}$, 25.82 ± 4.30 to $34.92 \pm 5.60 \text{ Bq/kg}$, and 122.56 ± 8.50 to $495.20 \pm 17.30 \text{ Bq/kg}$ with mean values of $26.05 \pm 4.13 \text{ Bq/kg}$, $29.73 \pm 4.85 \text{ Bq/kg}$ and $247.53 \pm 12.33 \text{ Bq/kg}$. These values are lower than the world average values of natural radionuclides concentration in soil for ^{238}U

(33Bq/kg), ^{232}Th (45Bq/kg) and ^{40}K (420Bq/kg) recommended by [97]. Nevertheless, the activity concentration of ^{40}K in Nonwa-Tai soil sample (ODM-SO1) was higher than the world average value of 420Bq/kg which may be attributed to either the presence of feldspar, clay minerals and mica that characterize the sedimentary formation of the Niger Delta or the application of phosphate fertilizers in farm soils to enhance crop yield in the area [87, 88, 92].

The radiological hazard indices used to assess public health risks to radioactivity exposure in soil are R_{eq} (range: 70.75 to 111.40Bq/kg, mean: 87.63Bq/kg), D (range: 32.07 to 52.52 $\mu\text{Gy/h}$, mean: 40.31 $\mu\text{Gy/h}$), outdoor AEDE (range: 0.04 to 0.06mSv/y, mean: 0.05mSv/y), indoor AEDE (range: 0.16 to 0.26mSv/y, mean: 0.19mSv/y), AGDE (range: 224.31 to 373.57mSv/y, mean: 282.50mSv/y), indoor ELCR (range: 0.56 to 0.91mSv/y, mean: 0.69mSv/y), H_{ex} (range: 0.19 to 0.29, mean: 0.23), H_{in} (range: 0.23 to 0.36, mean: 0.31) and AUI (range: 0.51 to 0.83, mean: 0.64). The mean values of these radiological hazard indices except for that of indoor ELCR are lower than the international recommended safety limits set by radiological bodies such as [66, 70, 96, 97, 125]. The implication of low values of radiological health hazard indices according to the above referenced international bodies is that using the investigated soil for various purposes (such as residential and agricultural purposes, construction of bricks and concretes, or as foundation soil) would not pose a significant radiological health risks arising from external gamma radiation exposure to people living or working in the vicinity of the investigated communities therefore is considered safe to use without the need of radiological intervention [112, 142]. However, the values of indoor ELCR in all the investigated communities are 14% higher than the recommended safety limit of 0.29mSv/y (Figure 2). From environmental radiological protection standpoint, indoor environments exhibits higher radiation levels than outdoor settings due to reduced ventilation and contributions from building materials (like bricks, concretes and soil) and it is based on the assumption that individuals spend more time indoors (19 hours within 24 hours) than outdoor (5 hours within 24 hours) therefore are more irradiated [67, 96, 136, 140]. Hence it follows that higher values of indoor ELCR recorded in this study imply that individuals spending much time indoors in the investigated communities have higher chances of developing cancer over a lifetime exposure of radiation levels emanating from the soil, and the values are high enough to cause death [66, 97]. The high value of about 22% above safety limit for AGDE recorded at Nonwa-Tai community in sample ID ODM-SO1 relates that radiations emanating from the use of soil in that community will pose serious threat to resident's reproductive organs, bone marrow and bone surface cells, and would eventually lead to individuals being diagnosed with Osteosarcoma, and Leukemia [66, 96]. The mean AUI value obtained in this study is lower than those obtained by [67, 113–115, 118, 136, 141]. The percentage contribution of ^{238}U , ^{232}Th and ^{40}K to the gross activity of the investigated soils was calculated from the parameters in the R_{eq} formula on the assumption that 10Bq/kg of ^{238}U , 7Bq/kg of ^{232}Th and 130Bq/kg of ^{40}K produced equal gamma dose [88]. The result (Figure 3) showed that ^{238}U , ^{232}Th and ^{40}K respectively constitute 30%, 48% and 22% to the gross activity of the soil samples with ^{232}Th contributing the highest percentage to the gross activity of the soil samples in the investigated communities of Nonwa. Due to the high value of absorbed dose rate recorded in Nonwa-Tai community soil (ODM-SO1), the

percentage contribution of each of the three natural radionuclides to the absorbed dose rate was evaluated and the result showed that ^{232}Th contributed the highest (43%) to the absorbed dose rate in Nonwa-Tai community soils (Figure 4).

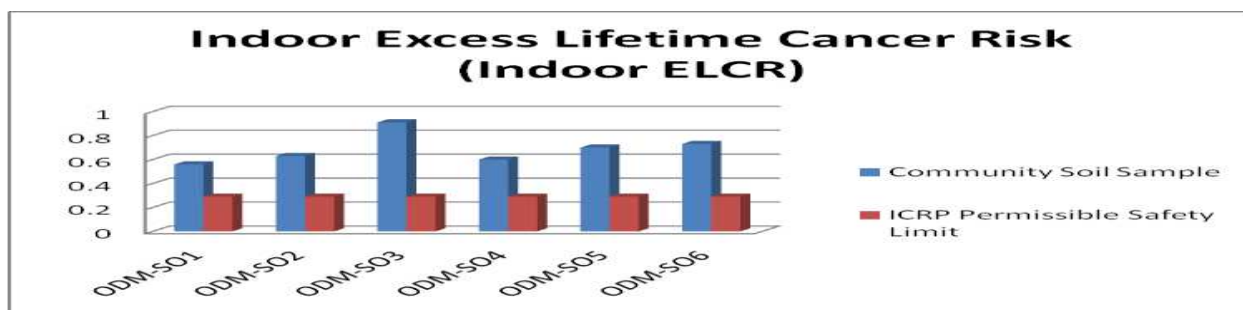


Fig. 2: Indoor Excess Lifetime Cancer Risk in Soil Usage Compared to ICRP (2012) Permissible Safety Limit

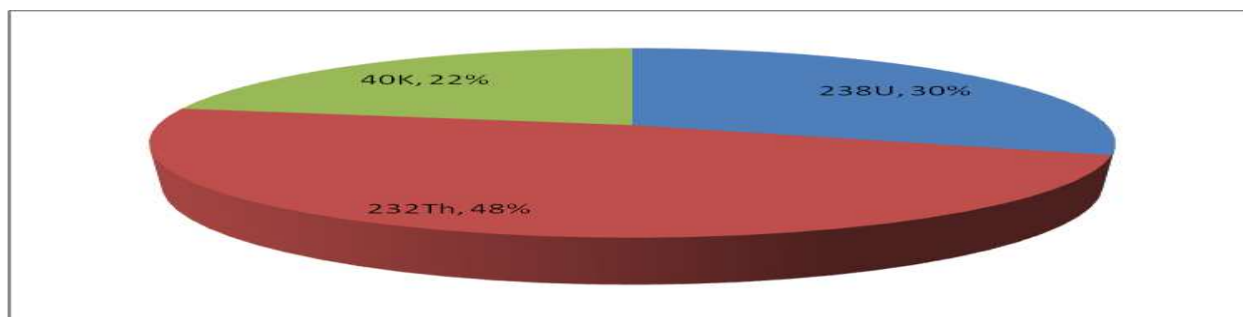


Fig. 3: Percentage Contribution of the three Natural Radionuclides to the Gross Activity of the Soil Samples in Nonwa Community

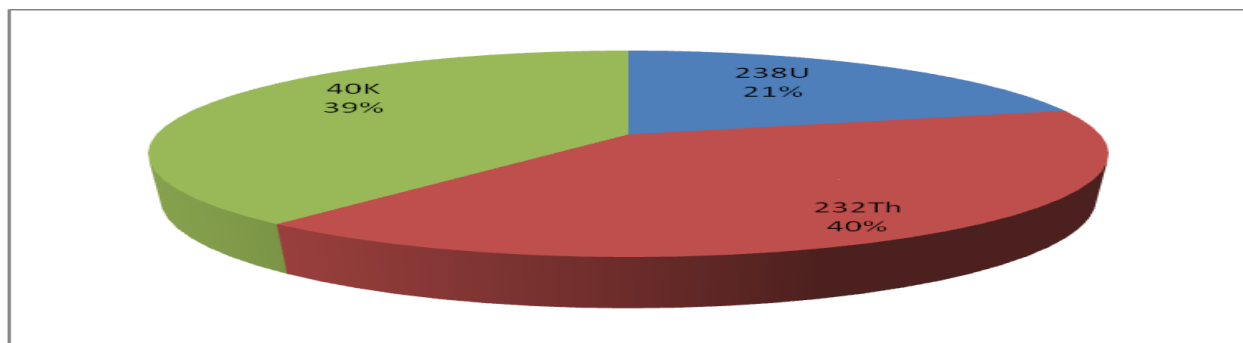


Fig. 4: Percentage Contribution of the three Natural Radionuclides to the Absorbed Dose Rate in Soil of Nonwa-Tai Community.

Table 7: Activity Concentration of Natural Radionuclides in the Community Soil Samples and the associated Radiation Hazard Indices

Community	Sample Location (Sub-Community)	Sample ID	Specific Activity (Bqkg-1)			Radiological Hazard Indices								
			²³⁸ U	²³² Th	⁴⁰ K	Ra(eq) (Bq/kg)	D (ηGy/h)	AEDE (Outdoor) (mSv/y)	AEDE (Indoor) (mSv/y)	AGDE (mSv/y)	ELCR (Indoor) (mSv/y)	Hex	Hin	AUI
Nonwa	Nonwa-Tai	ODM-SO1	23.34±3.40	34.92±5.60	495.20±17.30	111.40	52.52	0.06	0.16	373.57	0.56	0.29	0.36	0.83
		ODM-SO2	17.52±4.50	28.40±7.30	163.90±13.50	70.75	32.07	0.04	0.18	224.31	0.63	0.19	0.23	0.51
	Gbene-Nonwa	ODM-SO3	32.82±4.21	25.82±4.30	122.56±8.50	79.18	35.86	0.04	0.26	247.82	0.91	0.21	0.30	0.56
		ODM-SO4	18.72±5.60	32.43±6.20	220.30±11.60	82.05	37.43	0.05	0.17	262.57	0.60	0.23	0.28	0.59
	Nonwa-Uedume	ODM-SO5	31.29±3.65	27.52±3.40	260.80±13.40	90.72	41.94	0.05	0.20	293.61	0.70	0.24	0.33	0.66
		ODM-SO6	32.64±3.40	29.32±2.30	222.40±9.70	91.69	42.06	0.05	0.21	293.25	0.73	0.25	0.34	0.66
Mean			26.05±4.13	29.73±4.85	247.53±12.33	87.63	40.31	0.05	0.19	282.50	0.69	0.23	0.31	0.64
Minimum			17.52±4.50	28.82±4.30	122.56±8.50	70.75	32.07	0.04	0.16	224.31	0.56	0.19	0.23	0.51
Maximun			32.82±4.21	34.92±5.60	495.20±17.30	111.40	52.52	0.06	0.26	373.57	0.91	0.29	0.36	0.83
International Permissible Standards [70, 97, 117, 125, 133, 141]			33	45	420	370	59	0.07	0.41	300	0.29	≤ 1.0	≤ 1.0	≤ 2.0

3.1.1 Assessment of Enrichment and Depletion of Natural Radionuclides in Soil

The Clark value ranged from 2.39 to 5.26 with a mean of 3.75 (Table 8). Soil samples collected at Nonwa-Tai and parts of Gbene-Nonwa communities have higher Clark values above the world Earth crustal average value of 3.8 [121, 124]. [121] and [122] posited that high elemental concentration ratio of thorium to uranium ($[Th]/[U]$) in an area or region implies thorium enrichment or uranium depletion in source rock and that the rock type in the region would be more of FELSIC (Feldspar, SiO_2 and Al) minerals in composition than MAFIC (Mg, Fe, Ca) minerals arising from uranium loss due to weathering and geochemical process in uranium fractionation from thorium. Thus, soils and rocks in the community of Nonwa-Tai and some parts of Gbene-Nonwa consist of FELSIC minerals since they have higher Clark values than the standard world earth crustal value of 3.8. The FELSIC mineral composition in Nonwa-Tai and some parts of Gbene-Nonwa communities may have been responsible for the high radioactive potassium (^{40}K) values in sample ID: ODM-SO1 collected in Nonwa-Tai community. Conversely, soil samples with samples ID: ODM-SO3 collected at Gbene-Nonwa community, and ODM-SO5 and ODM-SO6 collected at Nonwa-Uedume community exhibited lower Clark values compared to the world Earth crustal average value of 3.8. [121-122] opined that low elemental concentration ratio of thorium to uranium ($[Th]/[U]$) in an area is an indication of thorium depletion or uranium enrichment in the source rock and that the rock type in the region would be more of MAFIC minerals in composition than FELSIC because the uranium precipitated from groundwater due to oxidation in an oxidizing environment which lead to economic abundance of uranium mineralization that creates a distinct geologic unit of radiological signature dominated by uranium clay chain with significant radiological hazards from radon gas. This further implies that a geologic unit with $[Th]/[U]$ value < 3.8 is a potential source of radon gas that has been reported by [123] as hazardous and unsafe when inhaled into the bronchial epithelium as it would result in chronic health effects like leukemia and lung cancer. The Clark values obtained in this study is higher than that obtained by [67] but lower than the value obtained by [117].

Table 8: Elemental Concentration of Natural Radionuclides in Soil Samples and the associated Clark Values

Sample Location (Community)	Sample ID	Elemental Concentration			Clark Value $[Th]/[U]$
		^{238}U (ppm)	^{232}Th (ppm)	^{40}K (%)	
Nonwa-Tai	ODM- SO1	1.89±0.28	8.60±1.38	1.58±0.06	4.55
	ODM- SO2	1.42±0.36	6.99±1.79	0.52±0.04	4.92
Gbene-Nonwa	ODM- SO3	2.66±0.34	6.36±1.06	0.39±0.03	2.39
	ODM- SO4	1.52±0.45	7.99±1.53	0.70±0.04	5.26
Nonwa-Uedume	ODM- SO5	2.53±0.30	6.78±0.84	0.83±0.04	2.68
	ODM- SO6	2.64±0.28	7.22±0.57	0.71±0.03	2.73
Mean		2.11±0.33	7.32±1.19	0.79±0.04	3.75
International Permissible Standards [97,121, 124]		2.67	11.08	1.34	3.8

3.2 Assessment of Soils for Heavy Metals and Associated Pollution Health Risk Indices

The mean concentrations values of heavy metals in the investigated soils (Table 9) are Pb (10.78mg/kg), As (0.50mg/kg), Zn (30.17mg/kg), Cu (6.00mg/kg), Cd (0.41mg/kg), Cr (14.54mg/kg), Mn (206.69mg/kg), and Ni (6.00mg/kg), but the order of heavy metals

abundances above the Niger Delta background value in a decreasing sequence is Mn>Zn>Cr>Pb. Soil quality in this study was assessed with some pollution hazard indices such as enrichment factor (EF), Geo-accumulation index (I_{geo}), contamination factor (CF), degree of contamination (C_d), and pollution load index (PLI) as summarized in Table 10 and Table 11. The range and mean values of EF for the heavy metals in soil samples are Pb (range: 0.47 to 1.12, mean: 0.88), As (range: 0.03 to 1.12, mean: 0.67), Zn (range: 0.78 to 1.48, mean: 1.18), Cu (range: 0.42 to 1.11, mean: 0.80), Cd (range: 0.37 to 3.65, mean: 1.55), Cr (range: 0.91 to 1.69, mean: 1.28), Mn (range: 0.58 to 0.95, mean: 0.76), and Ni (range: 0.37 to 0.67, mean: 0.55). Applying EF classification scale by [45, 82, 130, 132] as summarized in Table 2, the soils in the 3 investigated communities of Nonwa-Tai, Gbene-Nonwa and Nonwa Uedume are classified as deficiency to minimal enrichment of heavy metals due to human influence. The range and mean values of I_{geo} for heavy metals in soil are Pb (range: -0.68 to 0.59, mean: 0.17), As (range: -6.49 to -1.12, mean: -2.46), Zn (range: -0.70 to 0.23, mean: -0.14), Cu (range: -1.64 to -0.23, mean: -0.77), Cd (range: -3.75 to -0.46, mean: -2.27), Cr (range: -0.57 to 0.33, mean: -0.10), Mn (range: 0.10 to 0.81, mean: 0.47), and Ni (range: -1.27 to -0.41, mean: -0.73). Adopting [2] classification intensity and interpretation scale (Table 3) the investigated soil is classified as zero geo-accumulation intensity and practically uncontaminated with heavy metals. However, the negative values of I_{geo} for Pb, As, Zn, Cu, Cd, Cr and Ni indicates that although these heavy metals could have been deposited in the soil from industrial discharge and other sources of human influenced activities such as crude oil spill, gas flaring and artisanal oil refining occasioned by human influence, yet their concentration in the soil are still within the natural rock weathering/leaching background in the environment.

Table 9: Physico-chemical parameters and heavy metals content in Soil Samples

Community	Sample Location/ Sample ID	pH	Ec ($\mu\text{S}/\text{cm}$)	Heavy Metals (mg/kg)							
				Pb	As	Zn	Cu	Cd	Cr	Mn	Ni
Nonwa-Tai	ODM- SO1	5.20	195	13.35	0.50	36.95	5.21	0.12	14.36	199.56	5.82
	ODM- SO2	5.35	185	11.97	0.55	37.76	3.14	0.12	16.85	157.66	7.34
Gbene-Nonwa	ODM- SO3	4.36	139	7.13	0.75	19.83	6.72	0.65	10.31	257.56	5.34
	ODM- SO4	4.63	155	5.67	0.02	26.89	4.87	0.52	13.78	167.66	4.04
Nonwa-Uedume	ODM- SO5	5.15	197	12.95	0.83	36.52	7.78	0.10	12.72	220.51	7.23
	ODM- SO6	4.26	198	13.62	0.35	23.08	8.31	0.98	19.22	237.20	6.25
Maximum		5.35	198	13.62	0.83	37.76	8.31	0.98	19.22	257.56	7.34
Minimum		4.26	139	5.67	0.02	19.83	3.14	0.10	10.31	157.66	4.04
Mean		4.83	178	10.78	0.50	30.17	6.00	0.41	14.54	206.69	6.00
Control		5.53	114	5.78	0.35	12.07	3.56	0.13	5.38	128.14	5.21
Niger Delta Background[78]		5.99	35.50	6.05	1.2	21.50	6.5	0.90	10.20	98	6.5
World Average [62, 98, 99, 101]		7	<4000	27	6.83	70	38.9	0.41	59.5	488	29

Table 10: Enrichment Factor (EF) and Geo-accumulation Index (I_{geo}) of heavy metals in Soil Samples

Community	Sample ID	Pb		As		Zn		Cu		Cd		Cr		Mn		Ni	
		EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}
Nonwa-Tai	ODM- SO1	1.09	0.56	0.67	-1.85	1.45	0.19	0.69	-0.90	0.45	-3.49	1.27	-0.09	0.74	0.44	0.53	-0.75
	ODM- SO2	0.98	0.40	0.74	-1.71	1.48	0.23	0.42	-1.64	0.45	-3.49	1.49	0.14	0.58	0.10	0.67	-0.41
Gbene-Nonwa	ODM- SO3	0.59	-0.35	1.00	-1.26	0.78	-0.70	0.89	-0.54	2.42	-1.05	0.91	-0.57	0.95	0.81	0.49	-0.87
	ODM- SO4	0.47	-0.68	0.03	-6.49	1.06	-0.26	0.65	-1.00	1.94	-1.38	1.22	-0.15	0.62	0.19	0.37	-1.27
Nonwa-Uedume	ODM- SO5	1.06	0.51	1.12	-1.12	1.43	0.18	1.04	-0.33	0.37	-3.75	1.12	-0.27	0.82	0.59	0.66	-0.43
	ODM- SO6	1.12	0.59	0.47	-2.36	0.91	-0.48	1.11	-0.23	3.65	-0.46	1.69	0.33	0.88	0.69	0.57	-0.64
Mean		0.88	0.17	0.67	-2.46	1.18	-0.14	0.80	-0.77	1.55	-2.27	1.28	-0.10	0.76	0.47	0.55	-0.73

Table 11: Contamination Factor (CF), Degree of Contamination (C_d) and Pollution Load Index (PLI) of heavy metals

Community	Sample Location/Sample ID	Contamination Factor (CF) in Soil Sample								Degree of Contamination (C_d)	Pollution Load Index (PLI)
		Pb	As	Zn	Cu	Cd	Cr	Mn	Ni		
Nonwa-Tai	ODM- SO1	2.21	0.42	1.72	0.80	0.13	1.41	2.04	0.89	9.62	0.89
	ODM- SO2	1.98	0.46	1.76	0.48	0.13	1.65	1.61	1.13	9.20	0.86
Gbene-Nonwa	ODM- SO3	1.18	0.63	0.92	1.03	0.72	1.01	2.63	0.82	8.94	1.01
	ODM- SO4	0.94	0.02	1.25	0.75	0.58	1.35	1.71	0.62	7.22	0.59
Nonwa-Uedume	ODM- SO5	2.14	0.69	1.69	1.20	0.11	1.24	2.25	1.11	10.43	1.02
	ODM- SO6	2.25	0.29	1.07	1.28	1.09	1.88	2.42	0.96	11.24	1.20
Mean		1.78	0.42	1.40	0.92	0.46	1.42	2.11	0.92	9.44	0.93

The negative I_{geo} range values for As (-6.49 to -1.12) and Cd (-3.75 to -0.46) implies that the concentration of As and Cd in the investigated soil is low since [2] noted that $I_{geo} = -2$ means that the soil contains approximately 75% less metal than the natural baseline. The I_{geo} classification and description of the investigated soil as practically uncontaminated agrees with the EF classification of the soil as deficiency to minimal enrichment by heavy metals. The CF range and mean values of soil samples are: Pb (range: 0.98 to 2.25, mean: 1.78), As (range: 0.02 to 0.69, mean: 0.42), Zn (range: 0.92 to 1.76, mean: 1.40), Cu (range: 0.48 to 1.28, mean: 0.92), Cd (range: 0.11 to 1.09, mean: 0.46), Cr (range: 1.01 to 1.88, mean: 1.42), Mn (range: 1.61 to 2.63, mean: 2.11), and Ni (range: 0.62 to 1.13, mean: 0.92). Since all the eight (8) investigated heavy metals in the soil samples have an overall CF range from 0.02 to 2.63 (Table 11), [74] classification scale as expressed in Table 4 classified the investigated soil as moderate contamination factor while [56] classified the soil as none contamination/pollution to moderate contamination/pollution. The range and mean value of degree of contamination (C_d) of the soil samples are: (range: 7.22 to 11.24, mean: 9.44) while the range and mean value of pollution load index (PLI) of the investigated soils are: (range: 0.59 to 1.20, mean: 0.93) (Table 11). Applying the classification and interpretation scale by [74] the investigated soils are classified as low degree of contamination to moderate degree of contamination while [73, 134] described the PLI of the investigated soils as no pollution to polluted soils (Table 6).

However, since the investigated communities are in the Niger Delta region which has been described by several authors like [14-17] as an environmentally sensitive region, the Niger Delta background values for the investigated metals were extracted from the works of [78] and were used for this study. Consequently, it was observed that the mean values of heavy metals like Pb, Zn, Cr and Mn were respectively 78%, 40%, 42%

and 110% higher than the Niger Delta background values of the heavy metals (Table 9 and Figure 5). The high recorded values of these heavy metals above the Niger Delta background values implies that they are of petrogenic source and may pose serious health risk to members of the public who consume crops grown in the polluted soil since the mobile metals will bio-accumulate in the crops through soil-to-crop transfer. But when compared to [62] world average values, the heavy metals values in the investigated soils were found lower by 60% for Pb, 92% for As, 57% for Zn, 84% for Cu, 75% for Cr, 58% for Mn, 79% for Ni and 0% for Cd. The mean concentrations values of heavy metals in the investigated soils (Table 9) are Pb (10.78mg/kg), As (0.50mg/kg), Zn (30.17mg/kg), Cu (6.00mg/kg), Cd (0.41mg/kg), Cr (14.54mg/kg), Mn (206.69mg/kg), and Ni (6.00mg/kg), but the order of heavy metals abundances above the Niger Delta background value in a decreasing sequence is Mn>Zn>Cr>Pb>Cu, Ni>As>Cd.

3.3 Statistical Analysis for Natural Radionuclides, Physico-chemical Parameters and Heavy Metals in Soil Samples

Cluster Analysis (CA) was carried out on all the sample sites to group and classify them into clusters and to visualize the measured variables of the soil sample data sets in the groupings by generating a dendrogram using correlation similarity measure of hierarchical cluster analysis (HCA). The obtained dendrogram (Figure 6) distinguished three (3) main clusters that closely relate to the group of heavy metals and physico-chemical parameters as follows: (i) Cd, Mn and Cu; (ii) Zn, pH, Ni, Ec and As; and (iii) Pb and Cr. Cluster 1 consists of 3 metals Cd, Mn and Cu suggesting that these metals are from same crude oil and its by products (petrogenic) source since Cu and Cd are deposits of particulate matter from gas flaring and waste sludge from artisanal crude oil refining [28-30]. However, Mn is negatively correlated with Cd and Cu implying that Mn content in the polluted environment was due to its chemical behavior in soil because in oxygen-rich (oxidizing) conditions, the insoluble oxides formed by Mn strongly adsorb Cd and Cu and then pull them out of solution [31-32]. Arising from the fact that oxygen is depleted in waterlogged rain-fed farmlands, known to be a very common attribute of farmlands in the Niger Delta region, the soil bacteria ends up using manganese oxides as their oxygen source to breathe, thus reducing manganese to soluble Mn^{2+} [32]. Therefore negative correlation of Mn with Cd and Cu implies that Mn leaches into groundwater as it dissolves and then releases the trapped Cd and Cu into topsoil where crop grows to poison the food chain. Cluster II comprises Zn, pH, Ni, Ec and As suggesting that they all originated from the same pollution sources of crude oil, gas flaring, water from artisanal refining, and agrochemicals that were used from nearby farmlands, and rusty metallic pipes/drums used for illegal artisanal refining. However, the negative correlation of As from other parameters in this cluster implies that the As may have come from a different source like refining catalyst waste or a specific pesticide drift that independently varies from crude oil sludge that deposits the Zn and Ni. Hence, As levels drops when alkalinity and the concentrations of other metals increases with the implication that under more acidic or reduced conditions it has a high risk in the topsoils than the other metals. Cluster III consists of both Pb and Cr suggesting that both metals share identical pollution sources (such as gas flaring particulate emission, crude oil burns and sludge from artisanal crude oil refining and weathered crude from oil spillage). The cluster further showed that Pb and Cr bind

strongly to soil organic matter and clay that accumulate in the topsoil instead of leaching away into groundwater under non-acidic pH levels [33, 34]. Moreover, Pb and Cr co-occurrence implies that there is a chronic ongoing deposition from crude oil combustion and discharged waste stream signaling a petro-industrial toxicity that highlights human health hotspots that may trigger neurotoxic and carcinogenic diseases making it a serious public health concern.

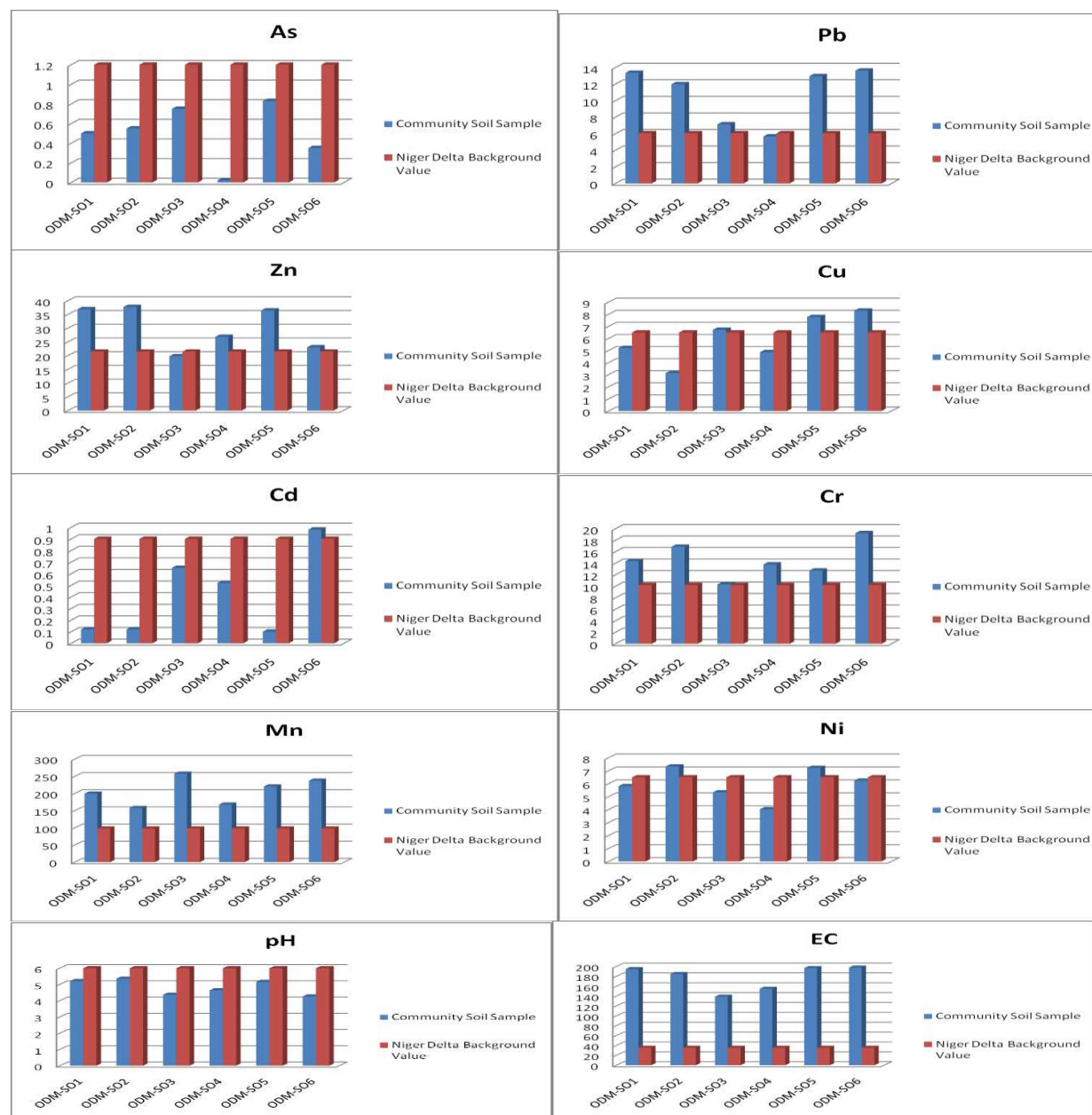


Fig. 5: Measured Heavy Metals and Physico-chemical Parameters Compared to the Niger Delta Background Value in Soil Samples in the Study Area

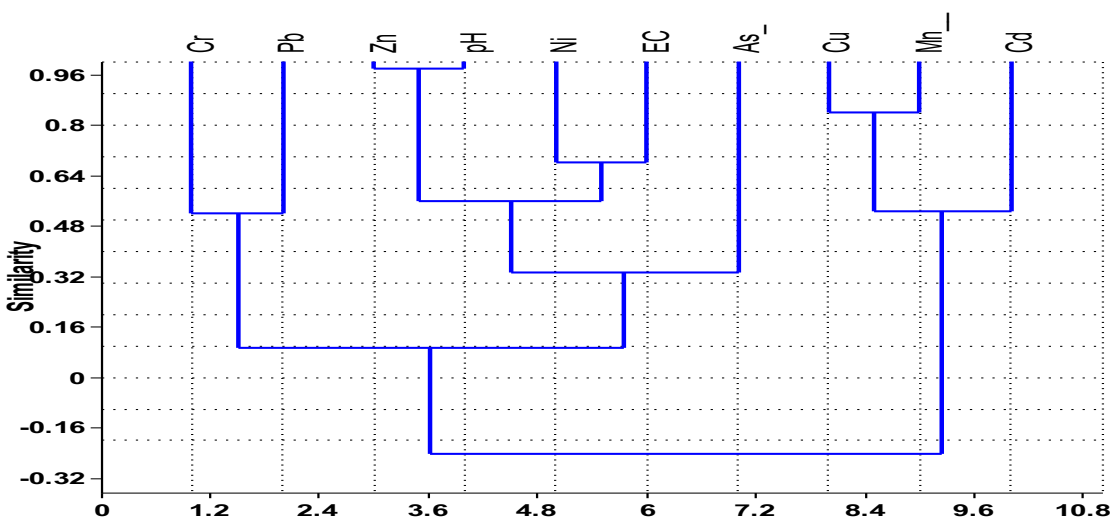


Fig. 6: Dendrogram of hierarchical cluster analysis for the physico-chemical Parameters and heavy metal in the soil samples

The Principal Component Analysis (PCA) which was run to extract information about the source of heavy metals and physico-chemical parameters considered eigen values ≤ 1.0 or approximately equal to 1.0 as acceptable possible sources of the variance in the data set. Three principal factors (PC1, PC2, and PC3) respectively accounted for 43.97%, 27.14% and 20.71% of the variance with 91.83% cumulative variance of all variables (Table 12). The communality showed that about 99% of the variations in As was explained by the factor model for the 3 factors which further explained variations in As, Zn, pH, Cd, Ec and Mn. The result also revealed that about 80% of the variation in Ni, Cr, Pb and Cu was explained by the factor analysis model for the 3 factor components.

Table 12: Varimax Normalized Factor Loading Matrix of the Heavy Metals Distribution and Physico-chemical Parameters in Soil Samples

Parameters (Variable)	Principal Component (PC)			Communality	Proportion of the total variation experienced by the 3 factors
	PC 1	PC 2	PC 3		
Cd	0.895	-0.346	0.222	0.970	
pH	-0.873	0.465	-0.035	0.979	
Cu	0.827	0.324	-0.124	0.804	
Zn	-0.812	0.557	0.135	0.988	
Mn	0.810	0.167	-0.517	0.951	
Ni	-0.151	0.915	-0.112	0.873	
EC	-0.102	0.854	0.477	0.967	
Pb	-0.244	0.022	0.871	0.819	
Cr	0.157	0.360	0.826	0.836	
As	0.011	0.613	-0.788	0.997	
Total				9.183	9.183/10 = 0.918
Eigen Value	4.397	2.714	2.071		
% Total Variance	43.971	27.144	20.713		
Cummulative %	43.971	71.115	91.828		

The proportion of the total variation obtained by the 3 factors was 0.918 which gives the total communality value of 9.183 providing an overall assessment of the PCA model performance. PC 1 strongly correlated positively with Cd, Cu, and Mn, but has a negative pH and Zn loading which explains 43.97% of the variance. These associations strongly suggests that the variables have strong relationship with gas flaring particulates and waste sludge from artisanal refining and could be said to be from these petrogenic sources. This agrees with cluster I, cluster II and cluster III of the cluster analysis results thus confirming same anthropogenic source. It should be noted that PC 1 would increase with increase in Cd, Cu and Mn scores and would decrease in scores of pH and Zn since decrease in pH and Zn concentration would increase concentration of Cd, Cu and Mn in topsoil of the investigated communities (Table 9). PC 2 showed high positive loadings and correlation with pH, Cu, Zn, Ni, Ec, Cr and As but showed negative loading with Cd accounting for 27.14% of variance (Table 12). The high positive loadings of pH, Ec, Cu, Zn, Ni, Cr and As defines crude oil and artisanal refining signatures. This is because a rise in Ec and pH may have occurred due to components of acid rain (like sulphuric and nitric acids) from gas flaring that leached alkaline cations from soils as well as artisanal refining which increases the soil salinity due to deposition of caustic chemicals, ash and brine wastes. Ni and Cr directly points to crude oil spillage, burning and refining residues since Ni is a classic crude oil marker [102, 105]. Moreover, Cu, Zn and As are used as crude oil refining additives to serve as catalysts and corrosion inhibitors. They are also produced as leachates from pipes and used in illegal refining and rusting metal drums [35-37]. Therefore high positive loadings of pH, EC, Cu, Zn, Ni, Cr and As strongly suggests that they originate from the same petroleum processing pollution environment. Negative loading of PCA with Cd means that as Cd decreases in concentration crude oil pollution increases which strongly suggest a geochemical separation signature as a source since Cd is rich in fertilizers and would accumulate in soil when applied to enhance crop growth in areas less affected by the crude oil thereby creating an inverse relationship outside the petroleum plume. Studies have shown that soil pH decreases in crude oil polluted environment and that at low pH Cd is very mobile [43]. Therefore in crude oil zone that is of low pH, Cd is not captured instead it precipitates as insoluble carbonates or hydroxides while in zones less in hydrocarbon pollution that is used in farming, pH is higher and Cd stays detectable and soluble [38-41]. This explains the negative loading of Cd in PC 2. PC 2 agrees with the results of cluster II confirming gas flaring, wastes from illegal artisanal crude oil refining and rusting metallic drums used for that purpose, and agrochemicals used in nearby farmlands as possible sources of pollution in the area. PC 3 showed high positive loadings with EC, Zn, Pb, Cr and Cd but negative loadings with pH, Cu, Mn, Ni and As accounting for 20.71% of variance. The result of PC 3 gives a clear picture of two distinct pollution sources in the study area viz: (i) farming lands treated with fertilizers consisting of high pH, Cu, Mn, and Ni; and (ii) refining and spill zones consisting of high EC, Pb, Cr, Cd and low pH. The near spill, refining and gas flaring sites are classified as polluted sites because the cluster consists of the positive loading metals and EC group that comprised of high EC, Zn, Pb, Cr and Cd with low pH, Cu, Mn, Ni and As representing petrogenic pollution source. The negative relationship between pH and the heavy metals increases the solubility and toxicity of Pb, Cd and Cr

which will allow these heavy metals become more bioavailable to groundwater and crops, and may also activate mechanism for heavy metal poisoning [18, 19, 26, 27]. Conversely, farmlands treated with fertilizers exhibited elevated levels of Cu, Mn and Ni resulting in healthy farm soils because the applied fertilizers are essential plant nutrients. The negative loadings of these metals suggest that they are depleted in polluted environments and may have either bound to organic matter in the crude oil or may have precipitated out of the crude oil [77]. Furthermore, negative loading of As suggests that it was generated from agrochemicals in farming areas and not from crude oil hence indicating that unpolluted farm soils have As while sites used for and affected by crude oil refining have lower As. The negative pH loading implies that the refining by-products in the environment (such as sulphuric and organic acids) are strongly acidic thereby lowering soil pH. This pH drops in an environment with worst pollution scenario, but remains normal where soil is clean. PC 3 agrees with results of cluster I, II and III in the performed cluster analysis thus confirming same source.

The Pearson Correlation Analysis (PCorA) was incorporated in this study to understand the relationship between heavy metals/physico-chemical parameters (Pb, As, Zn, Cu, Cd, Cr, Mn, Ni, pH, EC) and natural radionuclides (^{238}U , ^{232}Th , ^{40}K) concentration in the topsoil of the investigated communities. This approach was necessary in order to know the magnitude of the association between the sets of parameters and the direction of the association. The result (Table 13) found that ^{238}U have negative correlation with As and Ni and negligible positive correlation with Pb, Zn, Cu, Cd, Cr and Mn which was considered as not significant. ^{232}Th show negative correlation with Cu, Cd, Cr, Mn and ^{238}U , but is positively correlated with Pb, As, Zn and Ni which was also considered negligible and insignificant. ^{40}K has significant correlation with ^{238}U ($r=0.904$, $P<0.05$ at 81.72% variance), negative correlation with As, Cd, Cr, Ni and ^{232}Th , yet positively correlated insignificantly with Pb, Zn, Cu and Mn. The significant positive correlation between ^{40}K and ^{238}U strongly suggest that the chemical properties of ^{238}U mainly influenced ^{40}K contents in the top soil and not the radioactive decay of ^{238}U since studies by [75,104,139] describes uranium as a substance that exhibits dual characteristics as both a heavy metal and a radioactive element. Hence different chemical properties of ^{238}U and ^{232}Th are the reason for their different behavior in the environment as observed in Table 13 in the non- correlation between their measured concentrations. However, Copper (Cu) had a positive significant correlation with manganese ($r=0.839$, $p<0.05$) which is an indication that an increase in copper deposition in the topsoil of the crude oil polluted environment would also lead to an increase in manganese deposit. Cadmium had a negative significant correlation with zinc ($r= -0.908$, $p<0.05$ at 81.72% variance) which explains that the high concentration of Cd level above the Niger Delta background value in the topsoils of Nonwa-Uedume community at ODM-SO6 (Table 9) has a petrogenic source and could cause serious ecological human health risk due to its toxic effect. While the high concentration of Zn above the Niger Delta background value in the topsoils of Nonwa-Tai (ODM-SO1 and ODM-SO2) and Gbene-Nonwa (ODM-SO4) was from agricultural sources in the use of fertilizers applied to farmlands to improve crop yield. Similarly, pH exhibited a significant positive correlation with Zn ($r=0.976$) at $p<0.01$ and $p<0.05$ with 95.26% variance, but showed a high negative correlation with Cd ($r = -0.962$) at $p<0.01$ and $p<0.05$ with

92.54% of variance. This implies that soil pH accounted for 95.3% of Zn that is present in the polluted soil and that at high pH Zn is in solution in the organic complexes from the crude oil. The strong negative correlation of pH with Cd implies that Cd behavior in soil was explained by 92.5% of soil pH and further explaining why cadmium toxicity risks in soil and plant uptake is reduced or plummets.

Table 13: Pearson Correlation between Natural Radioactivity and Heavy Metals/Physico-chemical Parameters in Soil Samples

	Pb	As	Zn	Cu	Cd	Cr	Mn	Ni	²³⁸ U	²³² Th	⁴⁰ K	pH	EC
Pb	1.0												
As	-0.693	1.0											
Zn	0.368	0.219	1.0										
Cu	-0.114	0.287	-0.465	1.0									
Cd	-0.102	-0.368	-0.908*	0.528	1.0								
Cr	0.519	-0.411	0.144	-0.035	0.267	1.0							
Mn	-0.597	0.506	-0.612	0.839*	0.524	-0.293	1.0						
Ni	-0.151	0.669	0.577	0.067	-0.422	0.327	0.013	1.0					
²³⁸ U	0.289	-0.307	0.046	0.033	0.005	0.021	0.060	-0.423	1.0				
²³² Th	0.023	0.112	0.390	-0.494	-0.553	-0.321	-0.554	0.167	-0.694	1.0			
⁴⁰ K	0.330	-0.034	0.379	0.071	-0.329	-0.055	0.018	-0.121	0.904*	-0.507	1.0		
pH	0.213	0.286	0.976**	-0.577	-0.962**	0.003	-0.623	0.546	-0.023	0.492	0.297	1.0	
EC	0.495	0.136	0.641	0.173	-0.304	0.643	-0.146	0.680	0.178	-0.235	0.422	0.479	1.0

*. Correlation is significant at the 0.05 level (2-tailed).

** . Correlation is significant at the 0.01 level (2-tailed).

4 Conclusion

The measured activity concentration of ²³⁸U, ²³²Th and ⁴⁰K are lower than the world average values recommended by [97], except for the activity concentration of ⁴⁰K in Nonwa-Tai community soil sample whose value was higher than the world average which could be attributed to either the fertilizers applied to farmlands to enhance crop yield or to the presence of feldspars, clay minerals and mica that characterize the sedimentary formation of the region. The radiological hazard indices used to assess public health risk are lower than the international acceptable safety limits. However, the indoor ELCR was about 14% higher than the permissible safety standard in all the community soil samples while the AGDE value at Nonwa-Tai community soil was about 22% higher than the acceptable safety limit set by [97] implying high residents' exposure to diseases in their reproductive organs, bone marrow and bone surface cells. The clark value characterize the soils in Nonwa-Tai and some part of Gbene-Nonwa communities as consisting of FELSIC minerals while soils in communities of Nonwa-Uedume and parts of Gbene-Nonwa (soil sample ODM-SO3 as consisting of MAFIC minerals occasioned by the ongoing illegal artisanal crude oil refining in the area. This study further revealed that the MAFIC and FELSIC mineral compositions of the investigated soils are dominated by uranium clay chain which was considered a potential source of radon gas and would be radiologically harmful to the bronchial epithelium when inhaled. The pollution hazard indices that were used to assess soil quality classified the investigated soils in the area as deficiency to minimal enrichment, none to moderate contamination, low to moderate degree of contamination, and no pollution to polluted soils due to human influence. Cluster analysis grouped and classified heavy metals into clusters of their source similarity and revealed that they are of petrogenic and anthropogenic origin. PCA confirmed the CA results that heavy

metals are from crude oil spill, gas flaring particulates, used agrochemical from nearby farmlands, crude oil burns and sludges from artisanal refining and further revealing signatures of refining additives/leachate from rusted metal pipes and drums used for illegal crude oil refining. The performed correlation analysis suggested that heavy metals such as Pb, As, Zn, Cu, Cd, Cr, Mn and Ni are not responsible for increasing natural radioactivity level of ^{238}U , ^{232}Th and ^{40}K in topsoils of the investigated area, but that the radioactivity level of ^{40}K may have come from clay minerals, feldspars and mica that are dominated in the uranium clay chain geologic unit of the Niger Delta region. Lime application to crude oil spilled areas is recommended to help raise soil pH in order to immobilize the identified toxic metals as this would reduce their toxicity to prevent groundwater pollution, crop bioavailability and bioaccumulation. Phyto-remediation or chelate amendment or soil washing remediation strategies are also recommended to manage mobility of Zn and the co-variance of Pb and Cr in the investigated soils. It would be good too to stop gas flaring and artisanal oil refining since they are the primary control points of the metals.

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Author contributions

All authors contributed equally to the preparation of this manuscript. Thompson Chinedum Irunkwor developed the research framework and prepared the manuscript. Bright Atalor, Dandy Dumbiri Nmorsi, Harriet Osegbe and Pascal Ifeanyi Maduaka performed the experimental setup and analyzed the results. Bright Olisemeka Oranyelu performed the geoinformatics. Kingsley Chijioke Ogboi, Ngozi Chinyere Eze, Ejiro Newton Omoko and Azuka Ocheli further analyzed the results and improved on the theoretical aspects of this paper. All authors revised and approved the manuscript.

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Data Availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Ethics approval and consent to participate

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Consent for publication

Not applicable.

Competing Interests

The authors declare no competing interests.

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