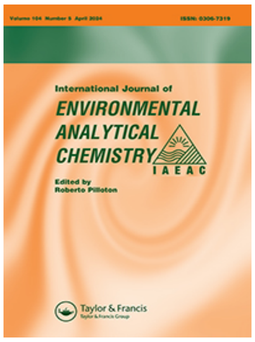

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Ingestion exposure and committed health risk of natural radioactivity and toxic metals in local rice sold in Enugu urban markets

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ABSTRACT

Rice (*Oryza sativa*) is an important source of human internal exposure to radionuclides and heavy metals because, worldwide, a large fraction of the population consume rice as their daily basic diet. In this study, the levels and correlation of natural radioactivity (^{232}Th, ^{226}Ra and ^{40}K) and heavy metals (Pb, Ni and Cd) in local rice sold in Enugu urban markets, a southeastern part of Nigeria, were examined. Possible health implications were also evaluated. The mean activity concentrations were estimated to be 235.81 ± 12.93, 54.29 ± 8.08 and 63.70 ± 3.93 Bqkg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th respectively. Obtained values were higher than values reported in the literature for rice in other locations. The estimated committed effective doses for ^{226}Ra and ^{232}Th exceeded the global ingestion dose average of 0.12 mSvy,$^{-1}$ whereas that of ^{40}K is below the 0.17 global average. The average concentrations of the metals were estimated to be 0.41, 3.70, and 0.02 mgkg^{-1} for Pb, Cd and Ni, respectively, with only Ni having an average concentration below the threshold food safety limit of FOA/WHO. Only the concentrations of Cd were of significant levels with their health risk indices exceeding the tolerable reference levels for both children and adult. Only the pairs, ^{232}Th-Ni and Pd-Cd, correlated significantly ($p < 0.05$) which implies common sources. Multivariate principal component analysis indicated common natural sources for ^{226}Ra, ^{232}Th and Ni in local rice, possibly of lithogenic and paedogenic in nature. It is believed that the results of this study will be valuable to the radiological and toxicological food safety and policy framework of WHO/FAO in Nigeria and the rest of the world.

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Natural radionuclides; heavy metals; local rice samples; correlation; ingestion; Enugu urban

1. Introduction

All foodstuffs contain detectable quantity of naturally occurring radionuclides belonging to the series thorium (^{232}Th), uranium (^{238}U) and non-series potassium (^{40}K), as well as heavy metals, like iron (Fe) nickel (Ni), zinc (Zn), copper (Cu), lead (Pb), arsenic (As), cadmium (Cd), chromium (Cr), etc. [1–3]. These elements are released into the environment from both natural

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Supplemental data for this article can be accessed here.

and anthropogenic activities. Their deposition in the cultivated soil is majorly responsible for the contamination of the food crops due to transfer from the soil to the different components of crops via roots. The practices of irregular application of fertilisers, pesticides, herbicides and irrigation water together with mechanical cultivations have continued to further contaminate the food crops with unprecedented levels of heavy metals and radionuclides [1,2]. Owing to the probable adverse effects on the entire food chains and human health, these practices over the years have drawn the attentions of scientists/researchers in the field of environmental monitoring and health risk assessment. Evidently, some studies have reported quite significant levels of naturally occurring radionuclides and toxic metals in various food crops [1–21].

Humans receive their own portion of these elements through consumption of the food and water, thus making the ingestion pathway the main route of human exposure to natural radionuclides and heavy metals. The human being has increase need for food supply to provide energy and other essential nutrients for life processes. Since food is essential for life sustenance, the habitual consumption can thus result in deposition of radionuclides in the internal organs with varying radiological doses. Rice (Oryza sativa), being a major staple food consumed daily by more than half of the world population and as an important source of energy, vitamins and essential elements, has been noted to be a major source of radionuclides and heavy metals to humans [1,2,19,20,22]. Paddy rice has a world range cultivation and owing to its importance as a major staple food in many countries and potential source of transfer, it has attracted global attention in radiological and toxic metal assessment [1,2,5–7,19–27]. Some of these studies have shown significant level of concentrations of these contaminants in the various rice grains investigated.

Though some metals like Fe, Zn, Cu, and Ni, within recommended limits, are required as essential micronutrients for biochemical and metabolical processes, whereas metals like Cd, Hg, Pb, Cr and As have no beneficial relevance; they are only known to be toxic and carcinogenic even at low concentrations [2,23]. In addition to being carcinogenic, these metals are known to induce a variety of disorders in humans, such as nausea, abdominal pain, anaemia, delirium, memory loss, renal dysfunction, hypertension, skin lesions and keratosis [28,29]. Similarly, ^{238}U and ^{232}Th and their progenies are highly radiotoxic [27]. When ingested through dietary intake, these radionuclides can accumulate in critical organs and subject them to radiation doses, thereby altering their functions and inducing some morphological and biochemical changes [1,3,27]. The lungs and kidneys are usually the deposition sites for ^{238}U and ^{226}Ra; ^{232}Th may be accumulated in either lungs, liver, or bones, while ^{40}K is present everywhere in the body but is mostly accumulated in the muscles [3,27]. In effect, the immune system of the body can become weak against various kinds of diseases with increased mortality rates [3].

In Nigeria, rice finds acceptance in the diets of both the rich and poor, thus its cultivation in virtually all of Nigeria’s agro-ecological zones [22,30]. The closure of the Nigerian border against foreign rice importation by the government has increased its demand in market places, thus increasing its annual production rate from 5.5 million tons in 2015 to 5.8 million tons in 2017, with a 5% projected increase every year [31,32]. Due to the cultivation practices, milling and packaging, there has been low-level acceptance among Nigerians on some locally grown rice products marketed in the country [33]. Due to this, studies on locally grown rice in Nigeria are mainly focused on cultivation practices, marketing strategies and nutritional values, thus neglecting the issues of contaminations by both radionuclides and toxic metals and the related health issues posed by ingestion. Evidence from the literature search conducted indicates that radionuclides and heavy metals contamination and health risk assessment of Nigerian locally grown rice are rarely reported to the best of our knowledge. The available studies [6,7,10,15,21,22,24,26,30,33–38] were done on freshly harvested rice, which does not represent the final state of the rice for consumption, since alteration (addition or reduction) in the radionuclides and heavy metal contents can occur during parboiling and milling processes. Also, exposure to atmospheric dust at the point of sales in market places can result in further contamination of the rice grains. None of these studies incorporated a multi-facet assessment of both natural radionuclides and heavy metal concentrations and their possible correlation with the local rice.

From the foregoing, it is imperative for continuous monitoring of radionuclides and heavy metal contaminants in rice, up to the point of sale. No representative data on natural radioactivity levels in local rice sold in Enugu urban markets that estimate the ingestion committed dose as well as their correlation to toxic heavy metals are available. Therefore, this present study was designed to assess the levels of natural radionuclides (^{232}Th, ^{226}Ra and ^{40}K) and toxic heavy metals (Pb, Ni and Cd) in local rice at the point of sale in various markets in Enugu urban areas and to evaluate the daily intake and committed effective dose via ingestion. Also, statistical correlations were carried out in order to evaluate possible inter-relationships between radionuclides and heavy metal pairs with the intent of establishing possible sources of contamination. It is expected that the generated data from this study will be valuable reference data to the radiological and toxicological food safety, quality control and policy framework of radiation agencies and WHO/FAO in Nigeria and the rest of the world.

2. Materials and methods

2.1. Sample collection and preparation

Enugu, the state capital of Enugu State in the southeastern part of Nigeria lies between latitudes 06°30′ and 06°40′N and longitudes 07°20′ and 07°35′E and covering a total landmass area of about 145.8 Sq km. Local rice sold in the Enugu urban areas are cultivated and processed in rural communities of the state from where they are transported down for marketing. Such of those notable communities are Ugbawka, Adani, Owo and Ameachi-Idodo, as well as neighbouring communities in Ebonyi and Anambra States. Local rice trading within the Enugu urban markets is prominent among food stuff traders.

For the purpose of this study, samples of local paddy rice grains were bought from five major markets within Enugu metropolis. The markets are located in Abakpa (A), Emene (E), Garriki (G), New Haven (N) and Ogbete (O). In each location, three local rice samples were purchased from different sellers in order to have a good representation of the location. Samples were packaged in polythene bags and properly labelled to avoid any mix-up as well as cross contamination and thereafter transported to the laboratory for processing.

At the laboratory, rice grain samples were washed with double-distilled water, spread on aluminium foil and left for 24 hours to get dried under normal laboratory conditions. Samples were later oven dried (DHG–9030) at 75°C until constant weight was achieved and thereafter were reduced to fine powder using Binatone electronic blender (Model: BRG-451). Then, 250 g of each sample was packaged into a radon-impermeable cylindrical plastic containers, measuring 7.6 cm by 7.6 cm, which were selected based on the space allocation of the detector vessel (geometry). To allow for radioactive secular equilibrium between parent and daughter nuclides and to prevent radon-222 escaping, the packaging containers were triple sealed and left for 30 days.

2.2. Gamma spectroscopy analysis of natural radionuclides content in rice samples

The analysis was carried out using a 76 mm × 76 mm NaI (Tl) detector crystal coupled to a photomultiplier tube (PMT) which was accessed at the Center for Energy Research and Training, Ahmadu Bello University, Zaria. The assembly has a preamplifier incorporated into it and a 1.0 kilovolt external source. Data acquisition was done with a multichannel analyser coupled to a PC equipped with MAESTRO software that matches gamma energies to a library of possible isotopes. The cylindrical plastic containers housing the samples were put to fit the geometry of the detector. To minimise the effects of background and scattered radiation, the detector was enclosed in a 6 cm lead shield with cadmium and copper sheets lining. Since the accuracy of the analytical system depends on its calibration, adequate energy and efficiency calibrations of the system were made using standard gamma energy sources of Cs-137 and Co-60. These were done with the amplifier gain that gave 72% energy resolution for the 661.16 keV of Cs-137 counted for 30 minutes. The International Atomic Energy Agency (IAEA) standard reference materials, RGK-1 for ^{40}K, RGU-1 for ^{226}Ra (Bi-214 peak) and RGTh-1 for ^{232}Th (Ti-208 peak) were used to check for the calibration. The background spectrum measured under the same conditions as the reference and experimental samples was used for correction of the sample spectra. An energy range of 0 to 3000 keV was set for the system to accommodate the energy range of interest of the present study. Both the background and samples were counted for 29,000 seconds. The activities of ^{226}Ra (from 1764.0 keV of ^{214}Bi), ^{232}Th (from 2614.5 keV of ^{208}Ti) and ^{40}K (1460.0 keV) were evaluated from the spectral energy window of 1620–1820 keV, 2480–2820 keV and 1380–1550 keV, respectively. The peak area of each energy in the spectrum was used to compute the activity concentrations in each sample by the use of the following equation [39];

$$C = \frac{C_n}{C_{fk}} \tag{1}$$

Where, C is the activity concentration of the radionuclides in the sample in $\mathrm{Bqkg^{-1}}$, C_{fk} is the calibration factor of the detector and C_n is the count rate (counts per second) defined as; $C_n = \mathrm{Netcount/livetime}$.

2.3. Analysis of heavy metals concentrations in local rice samples

The concentrations of lead (Pb), nickel (Ni) and cadmium (Cd) in the rice samples were determined by the method of atomic absorption spectrophotometry (AAS) after dry ashing according to the method of the American Public Health Association (APHA) [40]. Here, 2 gram of samples were ashed in a furnace at 550°C for 3 hours under gradual increase. Twenty (20) mL of 20% H_2SO_4 was added, boiled for 45 minutes and further diluted with 30 mL of deionised water and filtered with filter paper. The solutions were then analysed for Pb, Ni and Cd concentrations by aspiration into Varian FS240AA flame atomic absorption spectrophotometer (Agilent Technologies) with air acetylene flame as fuel source. The metals were analysed at wavelength of 217.3 nm (Pb), 232.0 nm (Ni) and 228.8 nm (Cd). The AAS slit width was 0.2 nm for all the metals. Validation of the system was made by stock standard solution of the metals prepared from their salt. A series of working standard metal solutions in the optimum concentration range were freshly prepared by diluting the single stock element solutions with distilled water containing 1.5 ml of concentrated nitric acid per litre. A calibration blank was prepared using all the reagents except for the metal stock solutions. Calibration curves of each metal were obtained by plotting the absorbance of standards versus their concentrations. Triplicate analyses of each sample were carried out with the blank and standard solutions analysed after every three samples in order to ascertain the reproducibility, accuracy and precision, obtained at above 95% of confidence. The detection limit (mg/L) of the system was obtained to be 0.003 for Pb and 0.002 for Ni and Cd. All reagents employed were of the analytical grade. All glass wares employed were thoroughly washed with distilled water.

2.4. Evaluation of radionuclides ingestion daily intake (D_{int}), committed effective dose (CED) and lifetime cancer risk (LCR)

The detection of radionuclides and quantification of their activity concentrations in food matrix provide little or no information on the radiological risk posed to consumers of the foods. Therefore, in an effort to gain an in-depth knowledge of the radiological risk associated with the ingestion of radionuclides via the local rice, the radionuclides daily intake (D_{int}), committed effective dose (CED) and lifetime cancer risk (LCR) were estimated using Eq. 2, 3 and 5, respectively, as follows [1,17,27]:

$$D_{int} = \frac{A_c \times A_{cr}}{D_{yr}} \tag{2}$$

where, D_{int}, A_c, A_{cr} and D_{yr} stand for the daily intake of radionuclides (Bq/day), measured activity concentrations of the examined radionuclides (Bq/kg), annual consumption rate of rice in Nigeria (= 26.35 kg/year/person) [7,41] and the total number of days available in a year (365 days), respectively.

$$CED = A_c \times A_{cr} \times D_{cf} \tag{3}$$

where CED is the committed effective dose in $mSvy^{-1}$, A_c is the measured activity concentration of the examined radionuclides in rice grains, and A_{cr} is the average annual consumption rate of rice. The parameter, D_{cf} represents the dose conversion factor (Sv/Bq) of the respective radionuclide, extracted from the International Commission on Radiological Protection (ICRP) [42]. Since the D_{cf} is age dependent, adult dose (>17 years) conversion factors of 2.8×10^{-7} Sv/Bq for ^{226}Ra and 2.3×10^{-7} Sv/Bq for ^{232}Th and 6.2×10^{-9} Sv/Bq for ^{40}K were used.

The total CED due to the combination of the radionuclides is therefore the sum of the individual radionuclide CED, that is:

$$\text{Total CED} = \sum \left(A_c^r \times A_{cr}\right) D_{cf}^r \tag{4}$$

where A_c^r and D_{cf}^r are the activity concentration and dose conversion factor of radionuclide r.

The lifetime cancer risk associated with the radionuclides in the analysed local rice of the present study was evaluated using the prescription of the US Environmental Protection Agency (USEPA) [43] as follows:

$$\text{Lifetime cancer risk} = A_{int} \times A_{lt} \times R_{rc} \tag{5}$$

where A_{int} is the annual intake of examined radionuclides (Bq/y), A_{lt} is the average lifetime (70 years) expectancy in Nigeria [39] and R_{rc} represents the morbidity risk coefficients (Bq^{-1}) of radionuclide of interest expressed as the probability of radiogenic effects per unit intake of activity averaged over all ages and sexes [43]. The adopted ingestion risk coefficients R_{rc} obtained from USEPA [43] are 9.56×10^{-9} Bq^{-1} for ^{226}Ra and 2.45×10^{-9} Bq^{-1} for ^{232}Th. For this risk assessment, ^{40}K was not considered since it is an essential intracellular cation that is under homoeostatic regulation by the body [44].

2.5. Evaluation of heavy metals ingestion average daily intake (ADI), Hazard quotient (HQ), Total hazard index (THI) and cancer risk (CR)

For the non-carcinogenic risk, the average daily intake (ADI), hazard quotient (HQ) and total hazard index (THI) of the heavy metals were evaluated, while for the carcinogenic risk, the cancer risk (CR) index was evaluated using the risk models of the United States Environmental Protection Agency (USEPA) [45,46], which have been applied by several authors.

$$\text{ADI} = \frac{C_m \times \text{IR}}{\text{BW}} \tag{6}$$

Where C_m is the heavy metal concentration in the studied rice grain (mg/kg), IR is the consumption rate of rice (kg/day) and BW is the body weight (kg) of the exposed individual.

The hazard quotient (HQ) and total hazard index (THI) were calculated using Eq. 7 and 8, respectively.

$$HQ = \frac{ADI}{RfD} \tag{7}$$

$$THI = \sum HQ_i \tag{8}$$

where ADI is the average daily intake (mg/kg/day) of the specific metal and RfD indicates the reference oral ingestion dose of the metal (mg/kg/day). The RfD of 0.0035, 0.02 and 0.001 for Pb, Ni and Cd, respectively, were obtained from the USEPA Integrated Risk Information System (IRIS) [46]. The RfD gives an approximation of daily dose or exposure that is not likely to cause any harm [45]. This means that exposures below the RfD are unlikely to produce adverse health effect. Above this value, an exposed individual may be at risk of any deleterious effect. Values of $HQ < 1$ and $THI < 1$ indicate that the non-carcinogenic risk of the metals is within the safe limits, implying that any exposed individual may not experience any noticeable adverse health effect. Consequently, values of HQ and THI greater than 1 mean that the limit of acceptance is exceeded and there is a possibility that a noncancerous effect may occur with a probability that tends to increase as THI increases.

To evaluate the carcinogenic or cancer risk (CR) posed by a specific metal, the ADI was multiplied with the corresponding ingestion cancer slope factor (SF) of the metal [19,25]. The total cancer risk (TCR) of a group of metals is thus the addition of the individual CR.

$$\mathrm{CR} = \mathrm{ADI} \times \mathrm{SF} \tag{9}$$

$$TCR = \sum CR_i \tag{10}$$

where SF is the ingestion slope factor of metal, which is 0.0085 for Pb, 1.7 for Ni and 15.0 for Cd [19,25]. For regulatory purposes, CR and TCR values can be classified as: values $<10^{-6}$, meaning that the cancer risk is of no significant health risk; values ranging from 10^{-6} to 10^{-4}, meaning that the risk is within tolerable/acceptance level; while values $>10^{-4}$, meaning that the cancer risk is unacceptable and of great significant health risk [45].

2.6. *Statistical analysis*

In order to statistically describe the distributions and variations of the radionuclides and heavy metal concentration in the local rice, descriptive statistics of the generated data, which include minimum (Min.) and maximum (Max.) values, arithmetic mean (AM), standard error (SE) of the mean and standard deviation (SD) were computed. A one-way ANOVA comparison with post-hoc Tukey test was also performed to statistically compare the most significant concentrations of the radionuclides and metals. Statistical significance was considered at $p \leq 0.05$ levels. Furthermore, multivariate statistics involving Pearson correlation (PC), principal component analysis (PCA) and hierarchical cluster analysis (HCA) were carried out to examine the inter-relationship and connectivity between the radionuclides and heavy metals in the local rice. Correlation coefficients ≥ 0.5 were considered to be the most significant. PCA by Varimax rotation with Kaiser Normalisation was preferred in simplifying the component factor loading. Component factors with eigenvalues greater than 1 were extracted and only loadings ≥ 0.5 were considered significant. Agglomerative HCA using Ward's method and squared Euclidean distance was employed for grouping the variables into small homogenous clusters on the basis of similarity. Microsoft Excel 2013 and the Statistical Package for Social Sciences (SPSS, version 22) were employed for the statistical analysis.

3. Results and discussion

3.1. *Activity concentrations of natural radionuclides in local rice and associated daily intake and ingestion effective dose*

The measured activity concentrations (including the analytical uncertainty) of ^{40}K, ^{232}Th and ^{226}Ra in examined rice grains are presented in Table 1 while Figure 1 shows the levels and variations. As indicated, the computed mean activity concentrations (± the standard error) were estimated to be 235.81 ± 12.93 Bqkg^{-1}, 54.29 ± 8.08 Bqkg^{-1} and 63.70 ± 3.93 Bqkg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th respectively. The result shows that the radionuclides were present and detected in all the samples in significant amount, but their levels in the rice as shown in Figure 1 indicates uneven distribution which varied considerably within and between the sample locations. The maximum concentration of 292.74 ± 0.16 Bqkg^{-1-1} for ^{40}K was obtained in the sample from Emene (E1), while the minimum value of 135.99 ± 0.21 Bqkg^{-1} was

Table 1. Measured concentrations of radionuclides and heavy metals in local rice samples.

		Activity concentrations of natural radionuclides (Bqkg^{-1})			Concentrations of heavy metals (mgkg^{-1})		
S/No.s	Sample ID	^{40}K	^{226}Ra	^{232}Th	Pb	Cd	Ni
1	A1	290.38 ± 0.41	81.18 ± 4.51	59.76 ± 3.58	0.85	4.53	BD
2	A2	242.87 ± 0.47	93.36 ± 6.07	92.01 ± 2.44	1.45	5.63	0.10
3	A3	135.99 ± 0.21	96.12 ± 5.27	87.45 ± 3.70	BD	0.55	BD
4	E1	292.74 ± 0.16	89.49 ± 5.83	39.40 ± 4.09	0.80	8.70	BD
5	E2	261.85 ± 0.36	48.14 ± 8.07	72.46 ± 0.94	BD	0.83	0.03
6	E3	225.82 ± 1.20	26.81 ± 8.55	44.59 ± 3.15	BD	0.55	BD
7	G1	221.20 ± 0.14	28.36 ± 5.43	40.34 ± 3.11	BD	0.70	BD
8	G2	275.15 ± 0.19	48.82 ± 5.15	64.95 ± 2.75	0.68	0.70	0.05
9	G3	178.89 ± 0.07	44.90 ± 4.23	56.19 ± 1.81	BD	3.35	BD
10	N1	155.08 ± 0.54	15.18 ± 4.19	62.56 ± 1.42	1.07	22.83	BD
11	N2	194.98 ± 0.42	20.65 ± 5.67	66.45 ± 1.77	BD	3.78	0.05
12	N3	275.20 ± 0.82	78.94 ± 4.19	66.65 ± 6.49	BD	0.50	0.03
13	O1	258.85 ± 1.01	95.04 ± 6.59	77.73 ± 8.30	0.63	1.50	BD
14	O2	289.04 ± 1.11	30.64 ± 4.75	64.68 ± 3.15	BD	1.20	0.02
15	O3	239.17 ± 1.20	16.78 ± 0.92	60.32 ± 1.61	0.73	0.13	BD
Mean ± standard error		235.81 ± 12.93	54.29 ± 8.08	63.70 ± 3.93	0.41 ± 0.13	3.70 ± 1.51	0.02 ± 0.01

BD: Below detection

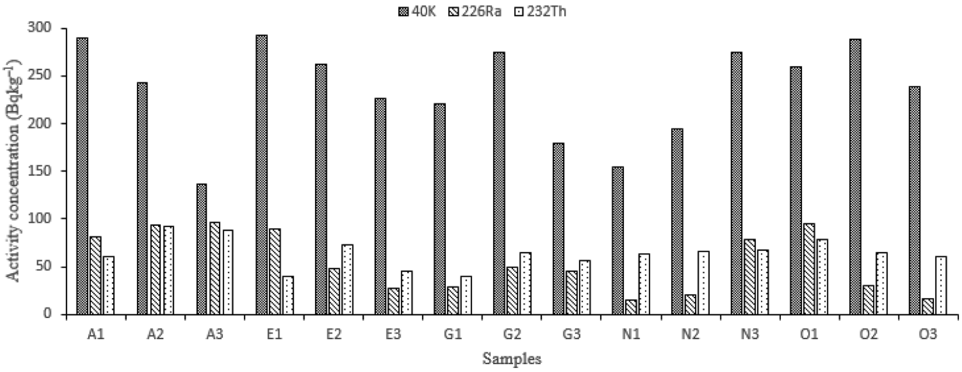

Figure 1. Activity concentration of radionuclides in local rice samples.

measured in sample A3 from Abakpa. The minimum and maximum concentrations for ^{226}Ra, which are 15.18 ± 4.19 Bqkg^{-1} and 96.12 ± 5.27 Bqkg,$^{-1}$ respectively, were gotten from samples in New Haven (N1) and Abakpa (A3), respectively. A minimum concentration of 39.40 ± 4.09 Bqkg^{-1} and a maximum of 92.01 ± 2.44 Bqkg^{-1} for ^{232}Th were obtained in samples E1 and A2 from Emene and Abakpa locations, respectively. In general, the results indicated uneven variation in the activity concentrations of the radionuclides in the various locations. The observed non-uniformity levels and variations of the radionuclide activities in the rice can be attributed to the varied nature of the geological compositions and background abundance of radionuclides in the cultivated soils, soil physicochemical properties, type and quantity of fertiliser applied during cultivation, soil-to-rice transfer ability, metabolic features of the rice plant species, agricultural practices at different farming areas [1,22,27,37,47–49].

As indicated in Figure 1, the concentrations of ^{40}K measured in each sample are statistically significant ($p < 0.05$) in excess of those of ^{226}Ra and ^{232}Th. More so, it was noted that 60% of the samples show higher levels of ^{232}Th than ^{226}Ra but no significant difference ($p > 0.05$) exist between them. The mean activity concentrations of the radionuclides decreased in this order of ^{40}K > ^{232}Th > ^{226}Ra with ^{40}K contributing about 66.7% to the total radioactivity content of the investigated rice. The higher levels of ^{232}Th measured in some of the samples may be ascribed to the radionuclide background concentration in the soils where they were grown. Since the locations of the cultivated soils and the background activity concentrations of the radionuclides in the soils are not known, this claim cannot be further substantiated in this present study. Nevertheless, recent studies show enhanced background activity concentrations of radionuclides in some rice farms of neighbouring state [22,37]. No doubt, high level of ^{40}K in foodstuffs is mainly connected to the practices of fertiliser applications by farmers in a way to revitalise soil nutrients for better crop yields [37]. Since fertilisers are products of phosphate rocks, they contain high contents of ^{40}K [50], of which their addition to the soil may result in higher transfer of the radionuclides to food crops. Also, since ^{40}K is very much distributed in soil with higher solubility and mobility in soil-root system than ^{238}U and ^{232}Th, and coupled with the fact that it is an essential macronutrient for plant growth, it is easily absorbed by plant roots and translocated to various parts [51]. Furthermore, soil physicochemical characteristics have been reported to favour the mobilisation and migration of potassium into plant [52,53]. Therefore, the results of the present study that indicate higher concentrations of ^{40}K than the other nuclides in the sampled rice are very much in tandem with similar studies conducted in other locations [1,3,4,17,21,22,27,48,54,55].

At present, no valid data on the world average or recommended limit of radionuclides in rice are available. However, the current study shows higher levels of ^{232}Th and ^{226}Ra when compared with data collated by the United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR) [44] for similar grains in some parts of Asia and European countries. Also, as shown in Table 2, conscious efforts were made to compare the activity concentrations of the present study with values measured in rice and other food crops reported by various authors. The concentrations of ^{40}K in the present study are of higher magnitude than values reported in rice grains of other locations, however, are contained in the range of concentrations measured in other food items like Potato [17], Red beans [6], Cowpea [9], wheat grains [14], maize and wheat [56], Yam and Cassava [54]. It can be observed that the activity concentrations of ^{232}Th and ^{226}Ra measured in this study are of higher magnitude as compared to studies listed in Table 2 and those mentioned in UNSCEAR [44]. From the foregoing, it can be concluded that the radioactivity content of the local rice available in Enugu metropolis is of higher magnitude than what is obtainable in other locations, which may be signifying a radiological threat. This observation can be attributed to the geological characterisation and parent materials of the soil where the rice plant were cultivated as well as the nature of water sources for irrigation and the probable use of fertilisers to boost soil nutrient which in most cases leads to contamination of the cultivated soil and later transported to plants through soil–root interaction. In this regard, the background concentrations of the radionuclides in the soil of all the paddy farms found in Enugu state and other neighbouring states where the local rice is cultivated need to be thoroughly investigated to unravel the cause of the high concentration of these radionuclides in the rice. Earlier studies had already indicated an elevation of these radionuclides concentrations above the world average in some paddy soils [22,37,57,58] and surface water sources [59] in neighbouring communities of Ebonyi state where large-scale rice farming activities are prominent.

Table 2. Comparison of radionuclides concentrations in local rice grains with other locations.

			Natural radionuclide concentration (Bqkg^{-1})			
S/N	Location	Sample	^{226}Ra	^{232}Th	^{40}K	Reference
1	Enugu, Nigeria	Rice	54.29	63.70	235.81	Present study
2	Sungai Besar, Malaysia	Rice	9.1 ± 1.8	20.6 ± 3.1	114.6 ± 6.6	[1]
3	Kampung Sakan, Malaysia	Rice	7.2 ± 2.0	11.6 ± 1.9	76.5 ± 5.7	[1]
4	Ekiti, Nigeria	Rice	–	10.5 ± 2.13	74.46 ± 5.54	[6]
5	Jigawa, Nigeria	Rice	1.7 ± 0.2		90 ± 4	[21]
6	Accra, Ghana	Rice	4.72 ± 2.17	4.33 ± 2.33	104.36 ± 10.22	[9]
7	Dhaka, Bangladesh	Fragrance rice	1.32 ± 0.30	–	6.57 ± 1.78	[27]
8	Dhaka, Bangladesh	Boiled rice	1.01 ± 0.27	–	4.02 ± 1.74	[27]
9	Alexandra, Egypt	Rice	< 0.32	–	13.97 ± 2.18	[5]
10	Port-Harcourt, Nigeria	Cassava	–	11.4 ± 3.3	426.9 ± 33.8	[54]
11	Port-Harcourt, Nigeria	Yam	–	8.4 ± 2.6	227.0 ± 27.3	[54]
12	Port-Harcourt, Nigeria	Cocoyam	–	7.1 ± 2.3	195.8 ± 25.8	[54]
13	Southwest, India	Rice	3.07 ± 0.02	34.3 ± 11.3	120.8 ± 2.1	[53]
14	Qassim, Saudi Arabia	Rice	0.4	0.2	138	[60]
15	Republic of Serbia	Wheat plant	0.3–2.3	<0.1	210–410	[56]
16	Republic of Serbia	Maize plant	0.6–6.8	–	240–400	[56]
17	India	Wheat grain	0.12 ± 0.05	0.04 ± 0.03	202 ± 3.7	[14]
18	Accra, Ghana	Cowpea	5.32 ± 2.31	5.43 ± 2.33	344.00 ± 18.55	[9]
19	Kano, Nigeria	Red beans	–	3.5 ± 1.85	256.97 ± 12.65	[6]
20	Enugu, Nigeria	Potato	–	1.14 ± 0.42	526.39 ± 51.40	[17]
21	Sio Paulo, Brazil	Rice	<0.11	–	14.7	[61]
22	Kuwait	Rice	–	0.48 ± 0.10	48.60 ± 18.34	[62]

The variation of the present study with those of other studies can be ascribed to the geological locations and the background abundance of radionuclides in cultivated soil. With areas of high background abundance, elevated levels of radionuclides can be expected in food crops. Studies by Jibiri et al. [41] and Arogunjo et al. [7] demonstrated higher activity levels of radionuclides in food crops sampled from high background areas of Jos than this present results. Specifically, the reported values of thorium and uranium nuclides by Arogunjo et al. [7] in spinach, cucumber, cocoyam, yam, Irish potato and sweet potato from the Jos environment are of 2 to 10 magnitude higher than the values measured in local rice in the present study. Their results clearly indicated the impact of high background abundance on food items due to tin mining in the area.

The presence of radionuclides in food items may result in radiation doses to the internal organs, which can linger for a long time after the intake [63]. To perfectly understand the intake of the radionuclides and associated committed effective dose via the local rice, the daily intake (D_{int}), committed effective dose and lifetime cancer risk were estimated. A summary of the results is presented in Table 3 (complete detailed results are available in Table S1 of supplementary material). Since ^{40}K is an essential intracellular cation that is under homoeostatic regulation, it was not considered for the lifetime cancer risk.

The average daily intake of ^{232}Th, ^{226}Ra and ^{40}K were estimated to be 4.60, 3.92 and 17.02 Bq day,$^{-1}$ respectively. The estimated daily intake of ^{232}Th and ^{226}Ra were about two to three magnitude higher than values estimated in different brands of rice available in Bangladesh [27], rice cultivated in different parts of northern and western regions of Malaysia [1] and in potato consumed in Enugu [17]. Differences in concentrations and consumption rates of a given food species account for this noticeable difference in the radionuclide intake. The average committed effective dose was evaluated to be 0.04, 0.40 and 0.39 mSvy^{-1} for ^{40}K, ^{226}Ra and ^{232}Th, respectively (see details in Table S1 of supplementary material). The committed effective dose for ^{226}Ra and ^{232}Th exceeded the global

Table 3. Radionuclides daily intake and committed effective dose.

	Daily intake of radionuclides (Bq/day)			Committed effective dose (CED) (mSv/y)				Lifetime cancer risk (LCR)		
	^{40}K	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	Total CED	^{226}Ra	^{232}Th	Total LCR
Min.	9.82	1.10	2.84	0.02	0.11	0.24	0.49	2.7×10^{-4}	1.8×10^{-4}	5.5×10^{-4}
Max.	21.13	6.94	6.64	0.05	0.71	0.56	1.29	1.7×10^{-3}	4.2×10^{-4}	2.1×10^{-3}
Ave.	17.02	3.92	4.60	0.04	0.40	0.39	0.83	9.6×10^{-4}	2.9×10^{-4}	1.2×10^{-3}
SE	0.93	0.58	0.28	0.00	0.06	0.02	0.07	1.4×10^{-4}	1.8×10^{-5}	1.5×10^{-4}
SD.	3.62	2.26	1.10	0.01	0.23	0.09	0.29	5.5×10^{-4}	6.9×10^{-5}	5.9×10^{-4}

ingestion dose average of 0.12 mSvy^{-1} for both uranium and thorium series nuclides, whereas that of ^{40}K is below the global 0.17 mSvy^{-1} for non-series potassium as contained in the report of UNSCEAR [44]. It can be noticed that the ingestion effective dose resulting from ^{40}K intake in the local rice is lower than those of ^{232}Th and ^{226}Ra despite it having higher concentration in the rice, implying the essential status of potassium as an intracellular cation that is under homoeostatic regulation, therefore not of much concern to human internal dose. The corresponding average lifetime cancer risk of 9.6×10^{-4} and 2.9×10^{-4} for ^{226}Ra and ^{232}Th falls below the global range of~ 10^{-3} for radiological risk of the general public [1]. The suggestion here is that the intake of the radionuclides through the local rice may result in radiation doses higher than the global average and may linger for a longer time. However, the probability of radiogenic cancer morbidity incidence in exposed individuals, either throughout their lifetime or for a short period of time, is not feasible. Although it is somehow difficult at this point to arrive at a valid conclusion on radionuclides intake and dose estimation, a logical conclusion is often made since the food items from which such estimations are made may still be modified during cooking leading to a reduction or addition of radionuclide contents [41]. The use of several cooking ingredients as well as the process of heating could either be an additional contributor or an inhibitor to the bioavailability of the radionuclides for intake [7].

3.2. Concentrations of Pb, Cd and Ni in local rice and associated ingestion non-carcinogenic and carcinogenic risks

The last three columns of Table 1 show the measured concentrations of Pb, Cd and Ni, respectively, in the examined local rice. The levels and variations of the metals are presented in Figure 2. The result shows that Cd was detected in all the samples, whereas detectable levels of Pb and Ni were found in only seven and six samples, which account for 46.7% and 40.0%, respectively, of the total samples. The result indicates no significant variation in the metal concentrations across and within the sample location, especially between Pb and Ni. The levels of the metals depict the accumulation ability of the rice plant in relation to the various soil properties. The content of Cd in the rice was in the range of 0.13 to 22.83 mgkg^{-1} having a mean of 3.70 mgkg^{-1} while that of Pb and Ni both ranged from below detection (BD) to 1.45 mgkg^{-1} and BD to 0.01 mgkg^{-1} respectively. The mean concentration for Pd and Ni was recorded to be 0.41 mgkg^{-1} and 0.02 mgkg^{-1} respectively. The highest concentrations of Ni and Pb were obtained in samples collected from Abakpa locations, whereas that of Cd, which is 22.83 mgkg^{-1}, was measured in rice N1 collected from New Haven. Ni and Pb fall below the detectable limits for most samples obtained from Emene, Garriki and Ogbete, indicating the non-presence or insignificant

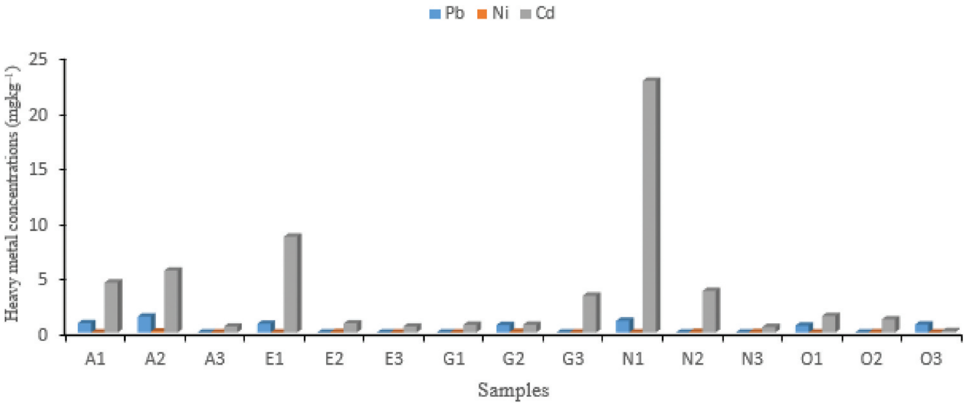

Figure 2. Concentration levels of Pb, Ni and Cd in Enugu local rice samples.

levels of the metals in the rice from these areas. Meanwhile, Cd levels were >0.05 mgkg^{-1} in those areas. The high concentration of Cd in the rice samples especially that of N1 can be attributed to several factors like location and contamination of the cultivated soil where the rice were grown, chemical fertilisers and pesticides applied during cultivation as well as sources of water for irrigation [15,30]. Metal-laden wastes from industries disposed on open land are also major anthropogenic sources of metal contamination in soil [28,29]. Substantial level of Cd has been reported in some rice paddy fields located in Enugu State [24,26] and neighbouring Ebonyi State [30,33] where most of the local rice sold in the Enugu urban markets are grown. Furthermore, the rice displayed in an open space within the market environment, without proper shading, are not devoid of depositions from environmental dust particles, which can lead to contamination [15]. The noticeable variation in the levels of the examined metals in the rice of the same location can be ascribed primarily to the geographical locations of cultivated soil as well as soil physicochemical properties [20,30]. Other factors are farming practices employed by farmers, type and amount of agrochemicals, quality of water for irrigation, and handling and processing of the crops.

Only Ni recorded concentrations below the threshold food safety limit of FOA/WHO [64], thus the values of Pb and Cd in the rice suggest a potential risk in terms of food safety. Generally, heavy metals are regarded as important constituents of food contamination due to their ability to persist, accumulate, and become toxic in living organisms when consumed. Cadmium has been reported to be a major contaminant in rice [65,66]. Normally, Ni is present at low level in soil, and its transfer to rice and other edible crops has been examined to be very low ranging [30,67]. The low concentration of Ni in the rice indicates that its level in the cultivated soils has not reached the point of proportionate accumulation in the rice plants. Although Ni is believed to play a significant physiological role as a cofactor in the absorption of iron from the intestine, however, it can induce some health complications like lung inflammation when present at elevated levels in the body system [68]. Cadmium and lead are carcinogenic in nature and have no biological or physiological significance in the human body, that even at the level of <1 µgkg^{-1}, it can be very toxic with severe detrimental effects on human health [67]. The measured concentrations of Pb and Cd in the rice grains could accumulate the metals in vital organs of the body upon consumption, which can result in severe effects to the organs. These effects arise from the fact that the metals react with some enzymes in the body, thereby altering their catalytic activities [12]. Cadmium, as a potential carcinogen, is reported to be a cause of some of the diseases relating to cardiovascular, blood, kidney, bone, as well as the nervous system [28].

Many studies on heavy metals in rice have been reported in different parts of the world with the results also indicating variations of the investigated metals in rice at those locations. Exception to Cd that seems significantly higher in this study, the levels of the metals are comparatively in range with published data for rice cultivated in Ezillo paddy fields in Ebonyi, Nigeria [30], rice available in the markets of Lagos and Ogun State, Nigeria [15], rice grains available in some cities of Bangladesh [20], in rice cultivated in Hainan Island, China [19], in rice cultivated in Wenling, China [69], in rice from different parts of Iran [2,8]. Average Pb concentrations of 3.19 $mgkg^{-1}$ in rice cultivated in industrial area of Bangladesh [16], 61.17 $mgkg^{-1}$ in rice marketed in Owerri, Nigeria [10], 17.13 $mgkg^{-1}$ and 11.5 $mgkg^{-1}$ in rice grown in some parts of India [13] and Iran [70], respectively, are significantly higher than the average concentrations reported in this present study. Moreover, Ni average values of 5.00 $mgkg^{-1}$ [20], 0.415 $mgkg^{-1}$ [19], 0.72–0.79 $mgkg^{-1}$ [71] and 0.90 ± 0.51 [26] from other locations are higher than this current study.

Summary of the results for the heavy metals daily intake (Table 4) and the non-carcinogenic and carcinogenic health risk indices (Table 5) of the heavy metals are presented here (Complete details of the results are available in Table S2 and S3 of the supplementary material). The mean ADI of the metals due to consumption of the local rice was estimated to be 4.09E–04, 1.80E–05 and 3.66E–03 mg/kg/day for Pb, Ni and Cd, respectively, for adult group and 1.91E–03, 8.60E–05 and 1.71E–02 mg/kg/day, respectively, for children. The ranking of the metals' ADI is in this order of Cd > Pb > Ni in both children and adult. All three metals have higher ADI values in children than the adult group. The total ADI of Pb and Ni in both groups were lower than the daily oral reference dose of 0.0035 and 0.02 mg/kg/day [46]. Also, the mean HQ values of Pb and Ni, which were estimated as 1.17E–01 and 9.23E–04, respectively, for adult and 5.46E–01 and 4.31E–03, respectively, for children are less than 1. This indicates that the daily intake of Pb and Ni into the body through the local rice may not pose any significant risk to human health. On the contrary, the ADI of Cd exceeded its 0.001 mg/kg/day daily reference dose [46], and the mean HQ values estimated as 3.66 in adult and 17.07 in children are significantly in excess of unity, implying that both categories of individuals are subject to any plausible non-carcinogenic health risk of Cd through local rice consumption in Enugu. The average HI for adults and children were estimated to be 3.78 and 17.62, with Cd as the major contributor; it contributed about 96.8% to the total HI in both categories of individuals. Based on this, the contributions of Ni and Pb to any non-carcinogenic health effects

Table 4. Daily intake of metals through consumption of local rice.

	Children daily intake of metals (mg/kg/day)			Adult daily intake of metals (mg/kg/day)		
	Pb	Ni	Cd	Pb	Ni	Cd
Min	0.000	0.000	6.00E–04	0.000	0.000	1.29E–04
Max	6.79E–03	4.62E–04	1.05E–01	1.43E–03	9.90E–05	2.26E–02
Ave	1.91E–03	8.60E–04	1.71E–02	4.09E–04	1.80E–04	3.66E–03
SE	5.90E–04	3.50E–04	6.95E–03	1.26E–04	7.00E–05	1.49E–03
SD	2.29E–03	1.35E–04	2.69E–02	4.90E–04	2.90E–05	5.77E–03

Table 5. Hazard quotient, hazard index and cancer risk of heavy metals in both children and adults.

		Hazard quotient				Cancer risk			
		Pb	Ni	Cd	Total hazard index	Pb	Ni	Cd	Total cancer risk
Children	**Min**	0.000	0.000	0.600	1.563	0.00	0.00	9.00E−03	9.00E−03
	Max	1.912	0.023	105.372	106.783	5.70E−05	7.85E−04	1.58E+00	1.58E+00
	Ave	0.546	0.004	17.071	17.622	1.60E−05	1.46E−04	2.56E−01	2.56E−01
	SE	0.169	0.002	6.946	7.039	5.00E−06	5.90E−05	1.04E−01	1.04E−01
	SD	0.653	0.007	26.903	27.263	1.90E−05	2.29E−04	4.04E−01	4.04E−01
Adult	**Min**	0.000	0.000	0.129	0.335	0.000	0.000	2.00E−03	2.00E−03
	Max	0.410	0.005	22.580	22.882	1.20E−05	1.68E−04	3.39E−01	3.90E−02
	Ave	0.117	0.001	3.658	3.776	3.00E−06	3.10E−05	5.50E−02	5.50E−02
	SE	0.036	0.000	1.489	1.508	1.00E−06	1.30E−05	2.20E−02	2.20E−02
	SD	0.140	0.001	5.765	5.842	4.00E−06	4.90E−05	8.60E−02	8.60E−02

accruing from the local rice intake in Enugu metropolis are not yet significant. The cancer risk values of Ni and Pb in both groups are within the acceptable range of 10^{-6} to 10^{-4} [46], indicating that the metals carcinogenic effects on the consumers are acceptable. The cancer risk of Cd with values of 0.256 for children and 0.0549 for adult is outside the acceptable range and very significant than those of Pb and Ni. This implies that there is a high risk of Cd induced carcinogenic effects in high rice-consumers among Enugu inhabitants with children being more likely to be susceptible groups.

3.3. Statistical correlation and inter-relationship of radionuclides and heavy metals in local rice

This section is committed to identifying the interplay between radionuclides and/or heavy metals. The null hypothesis of no significant difference between the parameters was first assumed. Firstly, basic descriptive statistics of the radionuclides and heavy metal concentrations in the examined local rice were presented (Table 6).

The values of % coefficient of variance (CV) reveal that the distributions of Pb, Ni and Cd within the rice samples exhibited extremely high variance (above 100%), unlike ^{40}k and ^{232}Th, which vary moderately (21 to ≤50%) and ^{226}Ra, which shows slightly higher variance (51 to ≤100%). As revealed by Kolmogorov-Smirnov and Shapiro–Wilk tests of normality, a nearly normal distribution ($p \geq 0.05$) was maintained by ^{40}K and ^{232}Th, whereas the distributions of ^{226}Ra and that of the metals deviated from normality ($p < 0.05$). This is evident from the histogram curves shown in Figure 3. The histogram curves (Figure 3) show the relatively long tail positive skewness and flat peak distributions of the metals, which are opposite to those of the radionuclides, implying asymmetric characteristics for the metals. The observed differences in variance and distribution nature between the radionuclides and metals in the local rice may be attributed to their levels of abundance available for uptake in the cultivated soil where they were grown, which is also controlled by the geology of the locations and soil characteristics, as well as planting practices and plant species.

The inter-relationship and connectivity of both radionuclides and heavy metal pairs in the local rice were analysed using Pearson correlation coefficient, principal component analysis (PCA) and hierarchical cluster analysis (HCA). Table 7 presents the Pearson correlation coefficient matrix between the radionuclides and heavy metals. Figure 4 shows the correlation scatter plots between the radionuclides and heavy metal pairs. Summary of the component factor loadings of the PCA and dendrogram of clusters are

Table 6. Basic descriptive statistics of measured concentrations of natural radionuclides and heavy metals in local rice samples.

	Radionuclides (Bq kg^{-1})			Heavy metals (mg kg^{-1})		
Descriptive statistics	^{40}K	^{226}Ra	^{232}Th	Pb	Ni	Cd
Minimum	135.99	15.18	39.40	BD	BD	0.13
Maximum	292.74	96.12	92.01	1.45	0.10	22.83
Median	242.87	48.14	64.68	0	0	1.20
Mean	235.81	54.29	63.70	0.41	0.02	3.70
Std error of mean	12.93	8.08	3.93	0.13	0.01	1.51
Std deviation	50.09	31.30	15.22	0.50	0.03	5.83
% coefficient of variance (CV)	21.24	57.65	23.90	119.58	156.67	157.59
Skewness	−0.74	0.20	0.12	0.70	1.79	2.87
Kortosis	−0.48	−1.76	−0.11	−0.76	3.26	9.12

shown in Table 8 and Figure 5 respectively. The PCA by Varimax rotation with Kaiser normalisation gave three component factors with each explaining variance of 33.903%, 25.847% and 19.509%, respectively, giving a total variance of 79.259%. The cluster dendrogram shows three clusters with only ^{40}K in cluster 1, ^{226}Ra and ^{232}Th in cluster 2, while the metals (Pb, Ni and Cd) were grouped in cluster 3.

As shown in Table 7, no significant correlation exists between the radionuclide pairs. Only ^{232}Th-Ni pair ($r = 0.546$) was significantly correlated ($p < 0.05$) among the radionuclide-heavy metal pairs, which implies common sources for ^{232}Th and Ni in the local rice. A significant correlation ($p < 0.05$) was also found between the pair Pd-Cd, which can be due also to common geogenic origin and geochemical affinity in the cultivated soil and identical transfer behaviour from soil to rice [16,30,72]. Aside from these two pairs, no other significant correlation exists between the radionuclides and heavy metals. The non-

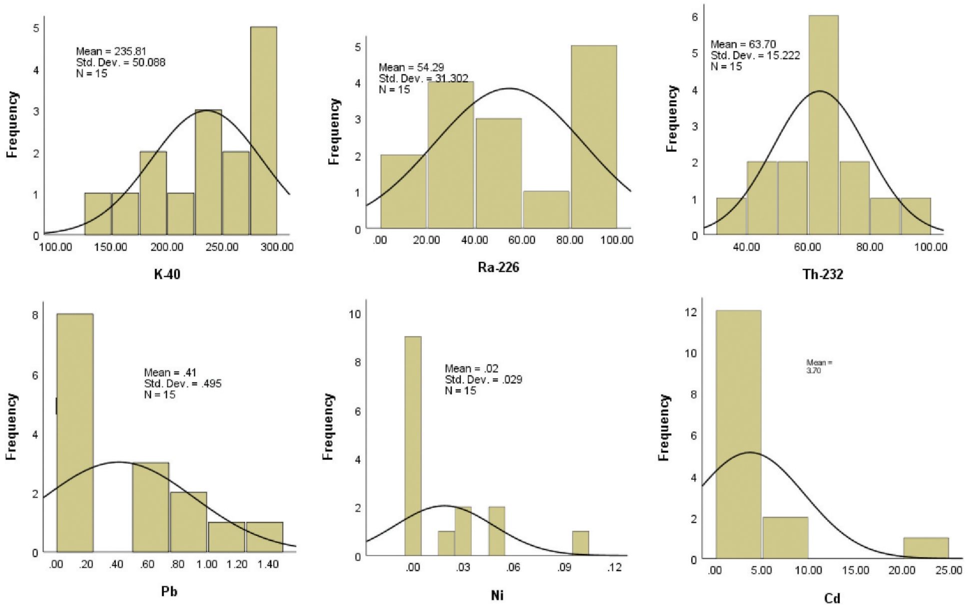

Figure 3. Histogram frequency curves for radionuclides and heavy metals distributions in local rice samples.

Table 7. Pearson correlation coefficient matrix of radionuclides and heavy metals in local rice samples.

	^{40}K	^{226}Ra	^{232}Th	Pb	Ni	Cd
^{40}K	1					
^{226}Ra	0.248	1				
^{232}Th	− 0.194	0.447	1			
Pb	0.171	0.278	0.230	1		
Ni	0.178	0.159	0.546*	0.311	1	
Cd	−0.320	−0.150	−0.075	0.543*	−0.077	1

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

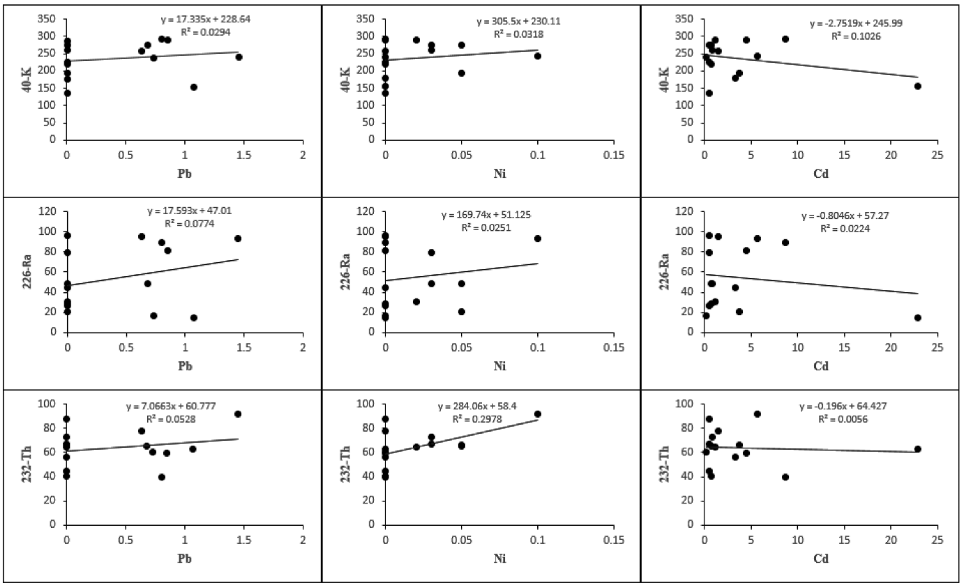

Figure 4. Correlation scatter plot of radionuclides and heavy metal pairs in local rice samples.

correlation of ^{226}Ra with the metals has been proposed in other studies [72]. Though ^{40}K naturally abounds in the earth crust, its non-correlation with other nuclides and the metals may be that its source in the local rice is largely from fertilisers employed during planting and not from nature. From the PCA results shown in Tables 8, ^{40}K is the only significant loading in component 3 and the only member of cluster 1 of the HCA dendrogram (Figure 5). This further shows that the main source and transfer characteristics of ^{40}K from soil to the local rice are different from the others. As observed from Tables 8, ^{323}Th and ^{226}Ra, as well as Ni, were significantly loaded into component 1, and also both nuclides are the members of cluster 2 of the HCA. This grouping of ^{232}Th and ^{266}Ra is somehow expected since ^{232}Th and the parent of ^{226}Ra (that is ^{238}U) have common lithogenic and paedogenic occurrence in nature and both possess similar transfer behaviours [73], though they differ in their solubility. Meanwhile, Ni was also significantly loaded into component 1, therefore upholding the correlation relationship between the pair ^{232}Th-Ni, affirming a common source. Cluster 3 with Pb, Ni and Cd may be affected by their high variance coefficients owing to their low range of concentrations in the local rice, suggesting a natural factor responsible for their distribution within the

Table 8. PCA rotated factor loadings of radionuclides and heavy metals.

Variables	Communality (initially at 1.00)	Rotated factor component		
		Component 1	Component 2	Component 3
^{40}K	0.927	0.008	−0.053	0.961
^{226}R	0.525	0.612	0.027	0.387
^{232}Th	0.906	0.916	−0.009	−0.256
Pb	0.908	0.322	0.860	0.254
Ni	0.573	0.743	0.091	0.111
Cd	0.917	−0.170	0.888	−0.315
% of variance explained		33.903	25.847	19.509
Cumulative %		33.903	59.750	79.259

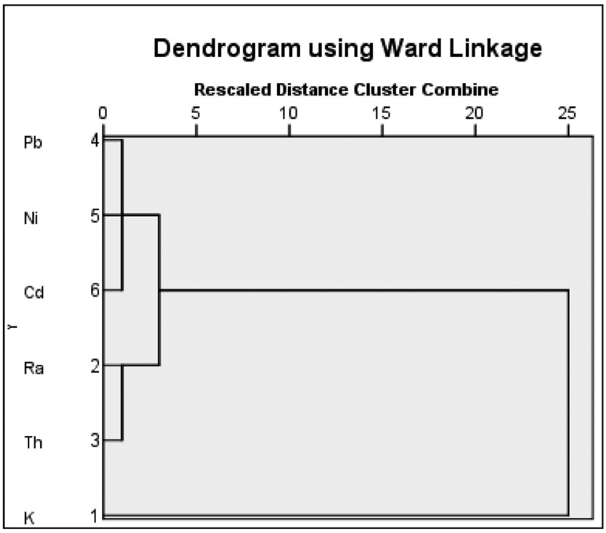

Figure 5. Hierarchical cluster dendrogram of radionuclides and heavy metals in local rice samples.

samples. As observed, however, clusters 2 and 3 were joined together at a slightly higher level, which may represent the earlier correlations between the pair ^{232}Th-Ni and component grouping of ^{226}Ra, ^{232}Th and Ni in component 1, this possibly implying a source common to them.

4. Conclusion

Rice presents an important source of human internal exposure to radionuclides and potentially toxic metals. Local rice produced within the Nigerian agro ecological zones has not received much attention in terms of food safety and health risk assessment. In this study, the activity concentrations of ^{40}K, ^{232}Th and ^{226}Ra and the committed effective dose together with the concentrations of selected heavy metals (Pb, Ni and Cd) in local rice sold in Enugu urban areas have been determined. As a result of the investigation, ^{40}K, ^{232}Th and ^{226}Ra in all investigated local rice samples were present in appreciable amount but vary considerably within and between the sampling locations with mean values of 235.81 ± 12.93, 54.29 ± 8.08 and 63.70 ± 3.93 Bqkq^{-1} for ^{40}K, ^{226}Ra and ^{232}Th, respectively.

It was found that ^{40}K contributed about 66.7% to the total radioactivity content of the investigated local rice sold in Enugu. The obtained concentrations in the local rice were found to be higher than values reported in similar studies involving paddy rice from other locations of the world, signifying that the radioactivity content of the local rice might present some radiological threats. The mean concentrations of the metals were as follows: 3.70 $mgkg^{-1}$ for Cd, 0.41 $mgkg^{-1}$ for Pb and 0.02 $mgkg^{-1}$ for Ni, with only that of Ni going below the threshold food safety limit of FOA/WHO. The levels of Cd in this study seem significantly higher as compared to similar studies, while those of Ni and Pb are comparatively in range with published data. Only the average daily intake, hazard quotient, hazard index and cancer risk index of Cd were of significant, exceeding the tolerable reference levels for both children and adult. The radiological committed effective dose owing to ingestion of ^{226}Ra (0.40 $mSvy^{-1}$) and ^{232}Th (0.39 $mSvy^{-1}$) exceeded the global ingestion dose average of 0.12 $mSvy^{-1}$ for both uranium and thorium series nuclides, whereas the corresponding lifetime cancer risk of 9.6×10^{-4} and 2.9×10^{-4} respectively fall below the global range of 10^{-3} for radiological risk, suggesting that cancer morbidity incidence owing to ingestion by the local rice consumers in Enugu urban areas is not feasible. Only ^{232}Th-Ni and Pd-Cd pairs were significantly correlated ($p < 0.05$) which implies common sources. The results generated from this study will be valuable to the radiological and toxicological food safety and policy framework of WHO/FAO in Nigeria and the rest of the world. More so, it is recommended that the background natural radioactivity of natural radionuclides in soils of rice paddy fields in Enugu state and other neighbouring states where the local rice are grown be investigated in future studies to ascertain their sources and transfer to the paddy plants in order to substantiate the evidence of relative high activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K reported in this study.

Disclosure statement

The authors have declared that no financial or any other personal interest exists between them in regard to this study.

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