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Assessment of Natural Radioactivity and Heavy Metal Accumulation in Selected Edible Fruit Nuts from Lagos and Ogun State Markets, Nigeria

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ABSTRACT
Edible fruit nuts are highly valuable to humans for nutritional, economic, medicinal, and environmental reasons, hence are in high demand in our local markets. This study assesses the levels of naturally occurring radionuclides and heavy metals in six commonly consumed edible fruit nuts—peanuts, cashew nuts, walnuts, date nuts, tiger nuts, and kola nuts—sourced from major markets in Lagos and Ogun States, southwestern Nigeria. The analysis aimed to determine the potential radiological and toxicological health risks associated with their consumption. Gamma-ray spectrometry was employed to evaluate the activity concentrations of radionuclides (^{40}K, ^{226}Ra, and ^{232}Th), while heavy metal content, including lead (Pb), cadmium (Cd), chromium (Cr), nickel (Ni), cobalt (Co), copper (Cu), zinc (Zn), arsenic (As), and mercury (Hg), was determined using atomic absorpti on spectrometry. Results revealed that the activity concentration of ^{40}K in all samples exceeded the UNSCEAR (2000) threshold of 412 Bq/kg, while ^{226}Ra and ^{232}Th remained below permissible limits. The internal hazard index, radium equivalent, absorbed dose rate, and annual effective dose values were also below internationally recommended levels, suggesting no immediate radiological health risks. For heavy metals, Ni and Cr showed relatively higher concentrations, especially in tiger nuts and kola nuts, though still within acceptable limits. Essential elements like Zn and Cu were present in moderate amounts, indicating nutritional value alongside trace-level contamination. Despite the safety of these nuts for human consumption, the presence of environmentally toxic metals such as Ni and Cr underscores the need for continuous monitoring. The study concludes that while current contamination levels pose no significant health risk, proactive regulation and periodic food safety evaluations are essential to safeguard public health.

Keywords:
Heavy metals,
Radioactivity,
Fruit nuts,
Health risks,
Food safety.

INTRODUCTION

Naturally occurring radioactive materials (NORMs) are integral components of our environment, present in the earth's crust, atmosphere, water, food, and even the human body (Al-Khawlany et al., 2018). These radionuclides originate from two principal sources: cosmogenic, resulting from interactions of cosmic rays with the Earth's atmosphere, and terrestrial, derived from the primordial isotopes embedded in geological materials (Khalid et al., 2024). Among terrestrial radionuclides, uranium (^{238}U, ^{235}U), thorium (^{232}Th), and potassium (^{40}K) are the most significant contributors to natural background radiation. These isotopes and their decay products, such as radium (^{226}Ra, ^{228}Ra), emit gamma radiation (Ershov, 2024) detectable through spectrometric analysis and are relevant to human health due to their potential bioaccumulation and radiotoxicity. Uranium and thorium are present in varying amounts in soils and rocks and can enter the food chain through uptake by plants. Potassium-40 (^{40}K), on the other hand, is an essential nutrient and the most abundant natural radionuclide in the human body (Komisova et al., 2023). While these radionuclides have natural origins, human exposure can be enhanced by agricultural practices, mining, and consumption of contaminated foodstuffs. Of particular concern is ^{226}Ra, which mimics calcium in biological systems and tends to accumulate in bones.

Parallel to radioactive contamination, the presence of heavy metals in the environment—both naturally occurring and anthropogenic—raises additional health concerns. While some heavy metals such as Zn, Cu, and Fe are essential in trace amounts, others like Pb, Cd, Hg, and As are toxic even at low concentrations (Kerna et al., 2024). The ingestion of food contaminated with these metals can lead to adverse effects on various organ systems, including renal, neurological, and skeletal damage, especially with chronic exposure.

However, several research studies in this area have it that Radiation detection relies on instruments such as gas-filled detectors (ionization chambers, proportional counters, and Geiger-Müller counters), scintillation detectors, and semiconductor detectors (Cember & Johnson, 2009; Knoll, 2000). These devices convert the interaction of gamma photons with detector materials into electrical signals, primarily via photoelectric effect and Compton scattering. An essential requirement is that the detector's response be proportional to the energy deposited by the radiation (IAEA, 1989). Gamma-ray spectrometry is widely used for environmental radioactivity assessment, providing energy spectra that enable radionuclide identification. For heavy metal analysis, techniques such as Inductively Coupled Plasma Mass Spectrometry (ICP-MS) allow multi-elemental detection with high sensitivity, broad dynamic range, and rapid throughput (Thomas, 2008). This enables the trace-level analysis of heavy metals in food matrices, and previous studies have reported heavy metal contamination in foodstuffs. Nnorom and Ewuzie (2013) found essential metals in roasted cashew nuts, with no toxicological risk. Conversely, Oti (2015) reported elevated levels of Pb, Cd, and As in ceremonial fruits. Emurotu (2017) showed that tiger nuts contained essential nutrients and were free from toxic metals. Blair and Lamb (2017) analyzed U.S. peanuts for heavy metals and pesticides, highlighting the need for routine monitoring to ensure food safety.

Food safety is an essential aspect of public health, particularly with the increasing concern over the presence of naturally occurring radioactive materials (NORMs) and heavy metals in agricultural produce. Nuts are nutrient-rich foods widely consumed for their health benefits, but they may accumulate harmful elements from the environment, including radionuclides like ^{40}K, ^{238}U, and ^{232}Th, as well as heavy metals such as lead (Pb), cadmium (Cd), chromium (Cr), and nickel (Ni). In Nigeria, where fruit nuts like peanuts, cashew nuts, walnuts, date nuts, tiger nuts, and kola nuts are commonly consumed, it is critical to assess their radiological and toxicological safety.

This study investigated the concentrations of selected radionuclides and heavy metals in six types of fruit nuts sourced from two major markets in Lagos and Ogun States. Gamma spectrometry and atomic absorption spectrometry techniques were employed to evaluate the radiological risk and heavy metal contamination, with a view to determining the potential health risks associated with their consumption.

MATERIALS AND METHODS

Study Area

The study was conducted in two southwestern states (Lagos and Ogun) in Nigeria. Two major markets were patronized for the fruit collection.

Sample collections, preparation, and measurement

Six fruit nut samples, including peanuts (PN), cashew nuts (CN), walnuts (WN), date nuts (DN), tiger nuts (TN), and kola nuts (KN), were obtained from the two markets. The sample preparation began with the nuts weighing for wet weight, and then they were sun-dried for about three weeks. The samples were reweighed several times until a constant weight was maintained. The dry-weighed fruit nuts were pulverized using a grinding machine to obtain fine-grained nut samples and homogeneity, after which they were tightly sealed in MARINELLI beakers to fit the sodium iodide detector counting chamber. These beakers were weighed, packaged, appropriately labeled, and sealed to prevent the loss of radionuclide progeny. The sealed samples were reweighed to obtain the net weight and left for 30 days to achieve secular equilibrium.

Radionuclide analysis using gamma spectrometry was carried out on the nut samples with a sodium iodine detector, and the activity measurements were performed. 0.5g of the pulverised samples were digested using HNO_3 and Hydrogen peroxide for the heavy metal analysis. The analysis of the heavy metals [lead (Pb), Cadmium (Cd), Chromium (Cd), Nickel (Ni), Cobalt (Co), Copper (Cu), Zinc (Zn), Arsenic (As), Mercury (Hg)] was carried out using Atomic Absorption Spectrometry technique. The elemental analysis of the trace elements was carried out at the National Nuclear Research Institute (NNRI) Chemistry Department, Ghana Atomic Energy Commission (GAEC) AAS Laboratory, while the Radioactivity measurements were carried out at the Radiation Protection Institute, GAEC.

The Digestion Protocol for Nut Sample Using SINOE JUPITA-A Microwave Acid Digestion

0.5g of nut sample was weighed into a previously acid-washed, labeled 100 ml polytetrafluoroethylene (PTFE) Teflon bomb. 7 mL of concentrated nitric acid (HNO_3, 65%) and 1 mL of hydrogen peroxide (H_2O_2, 30%) were added to each sample in a fume chamber. The samples were then loaded on the microwave carousel. The vessel caps were secured tightly using a wrench. The complete assembly was microwave irradiated for 30 minutes using the SINEO JUPITA-A microwave digestion program. After digestion, the Teflon bombs mounted on the microwave carousel were cooled in a water bath to reduce internal pressure, and the volatilized material was allowed to re-stabilize. The digestate was made up to 20ml with double distilled water and assayed for the presence of Zinc (Zn), Lead (Pb), Copper (Cu), Cadmium (Cd), Nickel (Ni), Cobalt (Co), Chromium (Cr), Arsenic (As) and Mercury (Hg) using VARIAN AA 240FS-

Atomic Absorption Spectrometer in an acetylene- air flame

Determination of Activity Concentrations of Natural Radionuclides (^{40}K, ^{232}Th and ^{238}U)

The activity concentrations of natural radionuclides were obtained from Equation 1.

$$A_c = \frac{C_n}{\varepsilon(E)P_\gamma m} \quad (1)$$

Where ε(E)is the efficiency of detecting gamma rays of energy E by the detector, C_n is the net count rate under the photo peak corresponding to the gamma energy, P_γ is the photon emission probability of the gamma rays of energy E emitted by the radionuclide of interest, A_c is the activity concentration of the radionuclide in the sample and m is the mass of the sample.

Absorbed and observed dose rate

The mean activity concentrations of ^{238}U, ^{232}Th, and ^{40}K were converted into dose rate based on the conversion factor given by UNSCEAR (2000)

$$D = (0.462A_u + 0.604\, A_{Th} + 0.0417\, A_k)\, nGyh^{-1} \quad (2)$$

Where D is the absorbed dose rate (nGyh^{-1}), and A_U, A_{Th}, and A_k are the activity concentrations (Bq/kg) of ^{238}U, ^{232}Th, and ^{40}K respectively while 0.462, 0.0417, and 0.604 nGyh^{-1} per Bqkg^{-1} were the conversion factors that correspond to ^{238}U,^{40}K and ^{232}Th.

Hazard index

The external hazard index (H_{ex}) was used to measure the external hazard due to the emitted gamma radiation and the internal hazard index (H_{in}) were calculated using the equation from Tufail et al. (1992):

$$H_{ex} = \frac{A_u}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \leq 1 \quad (3)$$

$$H_{in} = \frac{A_u}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \leq 1 \quad (4)$$

Where A_U, A_{Th}, and A_K are the specific activities of ^{238}U, ^{237}Th, and ^{40}K, in Bq kg^{-1}, respectively.

RESULTS AND DISCUSSION

Table 1: Activity concentration of Natural radionuclides in the fruit nut samples

Sample ID	^{40}K (Bq.kg^{-1})	^{226}Ra (Bq.kg^{-1})	^{232}Th (Bq.kg^{-1})
DN	1465.2	0.34	BDL
CN	1451.66	1.02	2.48
KN	1462.36	0.69	3.85
TN	1669	0.11	32.74
WN	1240	4.67	2.09
PN	1650	7.83	4.46
MEAN ±σ	1489.7±156.84	2.44±3.13	7.6±13.24
UNSCEAR (2000)	412	45	32

The activity concentrations of the natural radionuclides present in the fruit nut samples used in this study are presented in Table 1. The values ranged from 1240 Bq/kg in WN to 1669 Bq/kg in TN, with a mean value of 1489.7 Bq/Kg for ^{40}K, 0.34 Bq/Kg in DN to 7.83 Bq/kg in PN, with a mean value of 2.44 Bq/kg for ^{226}Ra, and 2.09 Bq/kg in WN to 32.74 Bq/kg, with a mean value of 7.6 Bq/kg for ^{232}Th. The mean activity concentration of the natural radionuclides reveals values below recommended permissible limits in ^{226}Ra and ^{232}Th according to UNSCEAR (2000), while the mean activity concentration of ^{40}K is more than the threshold value of 412 Bq/Kg. Many studies have identified ^{40}K as the most dominant source of radioactivity in the samples studied, such as Perker (2023), who worked on Brazil nuts. This finding highlights the natural occurrence of potassium in food and its contribution to background radiation. However, the report of Ademola and Morakinyo (2020) on some nuts in south-western, Nigeria, Inuyomi et al., (2019) on commonly consumed nuts in Ile-Ife, south-western, Nigeria, were in contrast to this, who found all natural radionuclides' activity concentrations below the world average.

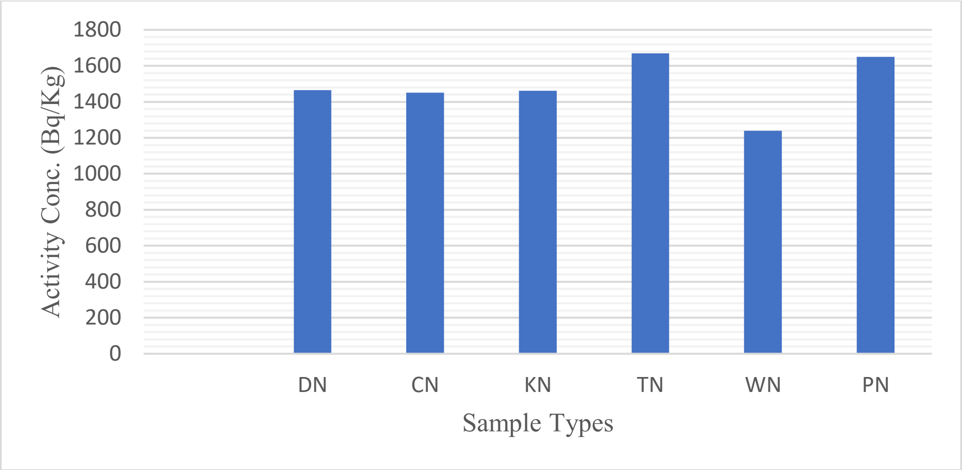

Figure 1: Activity concentration of ^{40}K in fruit nut samples

The activity concentrations of ^{40}K in fruit nut samples analysed are presented in Figure 1. Walnut (WN) had the lowest value of 1240 Bq/Kg, and the values for cashew nut, kola nut, and date nut were within the same range of 1451.66, 1465, and 1465.2 Bq/Kg, respectively. While peanut and tiger nut showed a little rise in value with 1650 and 1669 Bq/Kg, respectively.

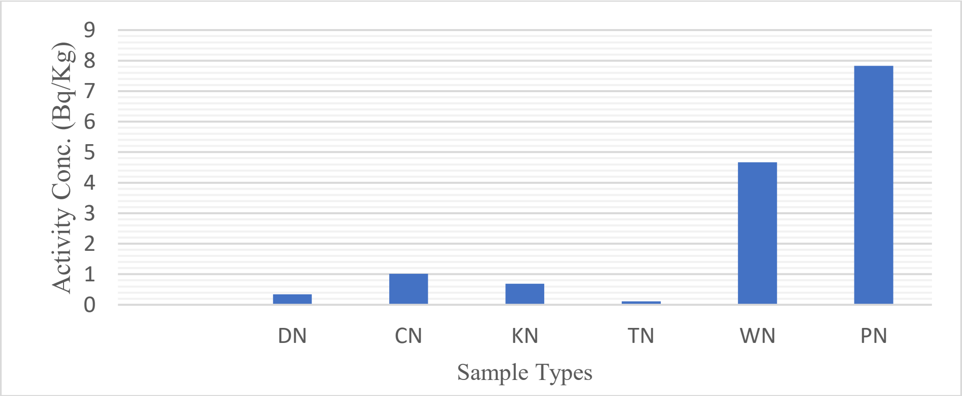

Figure 2: Activity concentration of ^{226}Ra in fruit nut samples

^{226}Ra activity concentrations in the fruit nuts samples were generally the lowest when compared with the other natural radionuclides examined. The activity concentration values ranged from 0.11 Bq/Kg to 7.83 Bq/Kg as shown in Figure 2. Tiger nut and peanuts had the lowest and the highest values, respectively. This corroborates the findings of Perker (2023), Ademola and Morakinyo (2020), and Inuyomi et al., (2019) on different nuts, including peanuts, cashew nut, et.c with values of ^{226}Ra activity concentrations far below the world's average value.

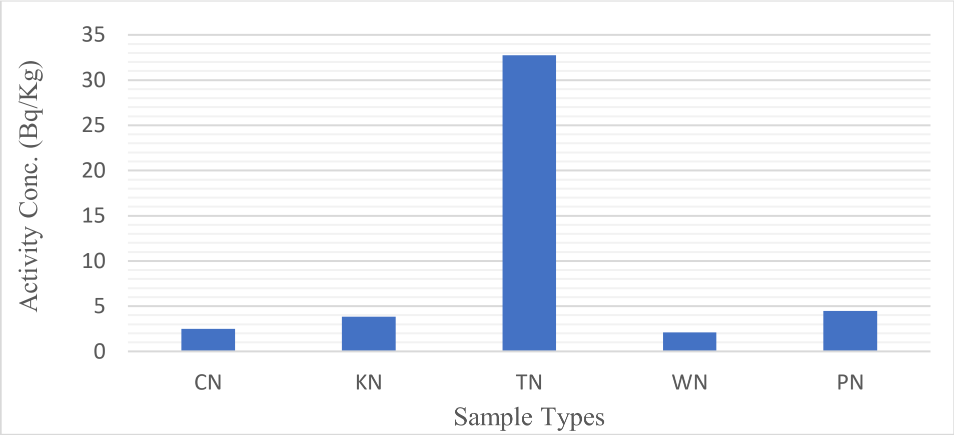

Figure 3: Activity concentration of ^{232}Th in fruit nut samples

Figure 3 reveals the activity concentration of ^{232}Th in fruit nut samples. Tiger nuts (TN) exhibited the highest activity concentration with a value of 32.74 Bq/Kg among all the samples analyzed. While Cashew nuts (CN), Kola nuts (KN), Peanuts (PN), and Walnuts (WN) all recorded ^{232}Th activity concentration values less than 5 Bq/kg. However, Date nut (DN) was not captured at all because it was below the detection limit of the detector. In all, these activity concentrations below the world average align with the report of Perker (2023), Ademola and Morakinyo (2020), and Inuyomi et al., (2019).

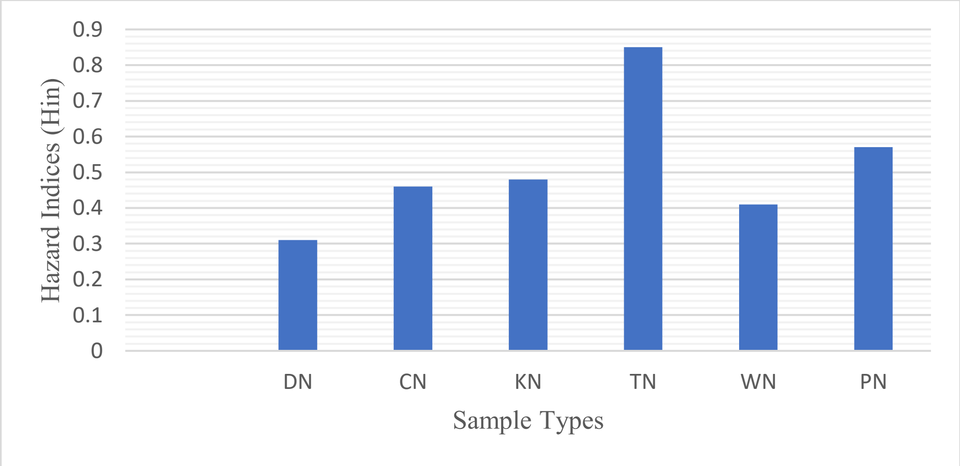

Figure 4: Hazard Index (H_{in}) in fruit nut samples

The internal hazard index H_{in} of the fruit nuts are presented in Figure 4. They all showed values less than 1 with the highest value of 0.85 from tiger nut and the lowest from date nut with 0.31 hazard index. Reports on radiological assessments on nuts have shown internal hazard index of less than 1(Ademola and Morakinyo, 2020; Inuyomi et al., 2019).

Table 2: Radium equivalent, absorbed dose, and annual effective dose in fruit nut samples

Sample	Raeq Bq.kg-1)	Absorbed dose rate (nGyhr-1)	Annual Effective Dose (mSvyr-1)
DN	113.16	1.8296	0.0045
CN	116.34	1.245	0.0031
KN	118.79	0.9654	0.0024
TN	175.44	0.6233	0.0015
WN	103.14	0.3177	0.0008
PN	141.25	0.3328	0.0008
Mean± σ	128.02±24.1010	0.8856±0.5360	0.0022±0.0013
UNSCEAR (2000)	370	60	1

The Radium equivalent, absorbed dose rates, and annual effective dose calculations from the selected fruit nuts, if consumed, are presented in Table 2.
Radium equivalent values range between 103.14 Bq/kg and 175.44 Bq/kg with a mean value of (128.02±24.1010) Bq/kg, the Absorbed dose rate ranged from 0.3177 $nGyhr^{-1}$ to 1.8296 $nGyhr^{-1}$ with a mean value of (0.8856± 0.5360) $nGyhr^{-1}$, and the Annual effective dose ranged from 0.0008 $mSvyr^{-1}$ to 0.0045 $mSvyr^{-1}$ with a mean value of (0.0022±0.0013) $mSvyr^{-1}$. Having all these values less than the permissible threshold points to the safe consumption of these nuts. This is in line with the submission of Inuyomi et al., (2019).

Table 3: Heavy metal concentration in Fruit nut samples

Sample ID	Pb	Cd	Cr	Ni	Co	Cu	Zn	As	Hg
KN	0.04	0.08	1.76	2.55	0.2	2.05	1.52	0.04	0.04
PN	0.04	0.08	0.72	1.45	0.2	1.07	1.04	0.04	0.04
WN	0.04	0.08	2.23	2	0.2	2.4	1.73	0.04	0.04
DN	0.04	0.08	0.43	0.04	0.2	0.48	0.04	0.04	0.04
CN	0.04	0.08	0.72	2.84	0.2	0.43	2.32	0.11	0.04
TN	0.04	0.08	2.63	3.39	0.2	0.72	0.49	0.09	0.04
STD QC	5.002	2.001	2.002	5.003	5	5.001	0.501	0.038	0.04
MEAN	0.04	0.08	1.42	2.05	0.2	1.19	1.19	0.06	0.04

Heavy metal concentration is shown in Table 3, Pb, Cd, Hg and Co had a constant value of 0.04 mg/kg, 0.08 mg/kg, 0.04 mg/kg and 0.2 mg/kg respectively for all samples, Cr metal concentration ranged from 0.43 mg/kg to 2.63 mg/kg, Ni ranged from 0.04 mg/kg to 3.39 mg/kg, Cu concentration value ranged from 0.43 mg/kg to 2.40 mg/kg, Zn ranged from 0.04 mg/kg to 2.32 mg/kg, while As ranged from 0.04 mg/kg to 0.11 mg/kg.

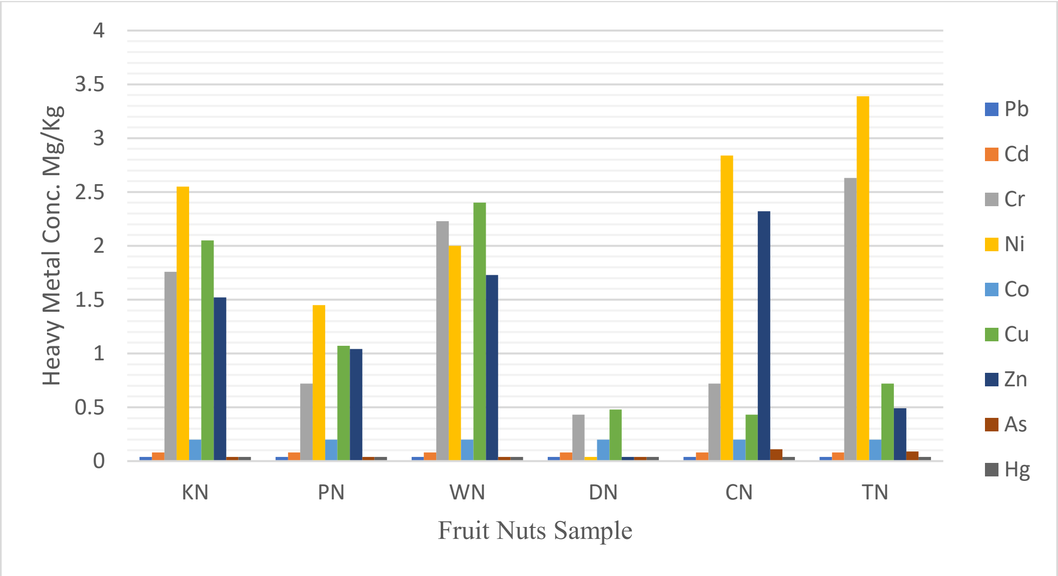

Figure 5: Fruit nuts sample with heavy metal concentration

Heavy metal concentration analysed in all six fruit nut samples is presented in Figure 5. Ni is seen to have the highest concentration values in tiger nut, cashew nut, peanut, and kola nut. Although it was present in walnuts as one of the significant heavy metals, it was not the highest in concentration. In date nut, the sixth sample, its presence is so insignificant with a concentration value of 0.04 mg/kg. While Ni may have minor biological roles in trace amounts, excessive exposure to it is harmful and should be monitored with environmental and health safety regulations. Cr is seen next noticeable in three nuts: tiger nut, walnut, and kola nut, though with concentration values less than 3 mg/kg. Chromium (Cr) is a heavy metal that serves a dual mandate in trace amounts, could be biologically important, and could be toxic and carcinogenic. This dual nature makes Cr of both nutritional interest and environmental concern. Both Ni and Cr are classified as environmentally toxic heavy metals whose exposure poses significant health risks (Kerna et al., 2024).

Zn is also noticeable in five nuts: Walnut, cashew nut, peanut nut, tiger nut and kola nut. Though a heavy metal but essential for health in trace amounts and is only toxic when in excess, while Cu has higher concentrations in three nuts: kola nut, peanut, and walnut, above other nuts. Cu is essential for human health in trace amounts, though a heavy metal chemically and environmentally, but toxic in excess, making it of biological and environmental significance.
The presence of heavy metals and minerals in nuts have been assessed by some researchers, such as Ademola and Morakinyo (2020), who reported the presence of Fe, Zn, Cu, and Cr in some nuts in south-western Nigeria. While Bielecka et. al (2021) reported the presence of minerals like Pb and Hg in peanuts, and Markiewicz-Żukowska et al (2022) reported and mentioned the presence of Zn in some nuts as being a crucial mineral for health.

CONCLUSION

The study revealed varying levels of natural radionuclides and heavy metals in all the fruit nut samples analyzed. Among the radionuclides, ^{40}K was the most prominent, with concentrations exceeding the UNSCEAR (2000) reference level, though ^{226}Ra and ^{232}Th remained below permissible limits. The internal hazard index (Hin), radium equivalent, absorbed dose rate, and annual effective dose for all samples were found to be below internationally recommended safety thresholds, indicating no significant radiological health risk from the consumption of these nuts. In terms of heavy metals, nickel (Ni), chromium (Cr), and copper (Cu) showed relatively higher concentrations compared to others, though all were below the safety limits provided in quality control standards. However, the presence of environmentally toxic metals such as Ni and Cr, even in trace amounts, highlights the need for continuous monitoring due to their potential health risks over prolonged exposure. It is recommended that regular monitoring, public awareness, stricter environmental regulations, and expanded research on nuts are essential to address contamination risks. Long-term dietary exposure assessments should be conducted, and agencies like NAFDAC and SON must enforce safety standards to protect consumers from harmful levels of radionuclides and heavy metals in food products.

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