Radiation Dosimetry and Heavy Metal Assessment of Honey in Akure, Ondo State, Nigeria

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ORIGINAL RESEARCH

Abstract - Honey samples from the wild, apiary, and commercial outfits have been evaluated for the activity concentration of ^{40}K, ^{238}U, and ^{232}Th using gamma spectrometry, and for heavy metals using Atomic Absorption Spectroscopy. The mean committed effective doses ranged from 9 ± 6 for adults to 25 ± 12 $nSvy^{-1}$ for children (1 year) for the samples from the wild; 23 ± 14 for adults to 52 ± 25 $nSvy^{-1}$ for children (1 year) for the samples from the apiary; and 29 ± 19 for adults to 68 ± 41 $nSvy^{-1}$in children (1 year) for honey samples from commercial outfits. The samples collected from the wild and apiary had Cd and Cr below detection levels, while Pb, Cu and Zn have mean values of 7.5, 3.0 and 0.3 ppm and 3.8, 4.8 and 1.0 respectively. But for samples from commercial outfits, Cu, Cd and Pb were below detection limits while Cr and Zn have mean values of 23.0 and 11.0 ppm, respectively. The committed effective dose was below 0.29 $mSvy^{-1}$ recommended by UNSCEAR. The honey samples may not be of any radiological concern. However, health risks due to heavy metals cannot be ruled out.

Keywords: committed effective dose, excess lifetime cancer risk, gamma spectrometry, heavy metals, honey

1 Introduction

Honey is a complex natural sweetener produced by honey bees from the nectar of different plants and honeydews and stored in a honeycomb (Caridi *et al*., 2022; Xarchoulakos and Lasithiotakis, M. 2022). It is reported to be a source of food to ancient men (Crittenden, 2011). It is widely used and consumed in human diet because of its nutritious and medicinal properties (Borawska et al., 2013). Honey has been reported to be rich in antioxidants, antibacterial, antiviral and antifungal agents (Borawska et al., 2013; Xarchoulakos & Lasithiotakis, 2022), thus, it has found applications in medicine, cosmetic, human diet etc (Boryło et al., 2019; Dizman et al., 2020). During the process of producing honey, the bees gather not only the honeydews and nectars, but also pollens, plant resins, water as well as any pollutants that must have deposited or transferred via the roots to the plant materials (Bulubasa et al., 2021; Caridi et al., 2022). Given the distanced covered by honey bees and the huge number of flowers needed to produce honey, honey can used as a bio-indicator of the environmental pollutants (Boryło et al., 2019).

Natural radionuclides is ubiquitous in the earth crust, thus the exposure of human to radiation from natural sources is nonstop and inevitable (UNSCEAR, 2000a). Some human activities like farming, quarry and mining trigger the suspension of soil samples and dust particles in the air. Vehicle movement through untarred roads also raise soil dust which settle on nearby plants and flowers. The soil dust on plants and flowers from where the bees harvest the nectars and honeydews for the production of honey could lead to the contamination of the honey with natural radionuclides present in the soil. Heavy metal contamination has being, but has increased with industrial and technological development (Fakhimzadeh & Lodenius, 2000). Agricultural, industrial, transport and other anthropogenic activities have contributed significantly to the concentration of heavy metals in the soil, atmosphere and water (Ševeričienė et al., 2022). There have also been reports of heavy metal contamination in honey from different countries (Adugna et al., 2020; Akbari et al., 2012). Some

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of the heavy metal metals like Zn, Cu, Ni, Fe, Se and Mn are essential for human body metabolism at trace values, but become toxic when their respective tolerance levels are exceeded while other heavy metals like Pb, Cd,Cr and Al are considered toxic to human health (Kiliç Altun et al., 2017).

There is a massive media campaign on the elimination of refine sugar from human diet and substitution of the refined sugar with natural sweeteners like honey, stevia and date fruit. There has been therefore increase in the consumption rate of honey as a result of its nutritious and medicinal benefits (Arowosoge, 2018). As a result of the increasing demand for honey, wild nest harvesting is unable to meet up with the demand. Apiculture is thus on the increase to meet up with the increasing demand. (Ajibola et al., 2012)

In line with the goals of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) in the continuous estimation of radiation exposure to the human population (UNSCEAR 2000b), this work seeks to estimate the concentration of natural radionuclides and heavy metals in honey samples, estimate the committed effective dose from honey consumption due to natural radionuclides and lifetime cancer risk due to the consumption of honey.

2 MATERIALS AND METHODS

The study area Akure is located in Ondo State, Nigeria. It has a GPS reading 7.2571 °N and 5.2058 °E. Fifteen (15) honey samples were collected from the study area with 5 samples from the wild, 5 samples from apiary, and five samples from some grocery stores in Akure. The honey samples harvested from the wild were purchased from local farmers who harvest honey from the forest. The samples of honey collected from the wild and apiary were multifloral type while the samples collected from the grocery stores are not specified on the labels of the containers. The collected honey samples were poured into plastic container of diameter 30 cm and height 30 cm, each weighting 150 g. The samples were analysed for natural radionuclides (^{40}K, ^{238}U and ^{232}Th) and heavy metals (Zinc (Zn), Cadmium (Cd), Lead (Pb), Copper (Cu) and Chromium (Cr)).

The concentrations of the natural radionuclides were estimated using gamma spectrometry method. The energy calibration of the NaI(Tl) spectrometer was carried out using the RSS8 gamma source set traceable to Spectrum Techniques LLC, USA. It was accomplished by measuring the spectra of point sources emitting gamma-rays of known energies. The efficiency calibration of the detector was also carried out using a reference source consisting of known radionuclide activities: ^{40}K (578.4 $Bqkg^{-1}$), ^{238}U (20.9 $Bqkg^{-1}$) and ^{232}Th (10.47 $Bqkg^{-1}$). The source was prepared in a container that has the same geometry as the sample and counted for a period of 18000 s. Before the sample measurement, an empty container was counted for 18000 s, so as to determine the background gamma-ray distribution count. The sealed samples, after attaining secular equilibrium were each placed on the detector one after the other for analysis. Each sample was counted for the same period as that of the empty container. The following energy peaks were used in determining the concentration of the radionuclides: 1460.0 keV (^{40}K), 1764.5 keV of ^{214}Bi (^{238}U), and 2614.7 keV of ^{208}Tl (^{232}Th). The activity concentration A ($Bqkg^{-1}$) of each radionuclide in the sample was estimated using Equation 1 (Jibiri et al., 2007)

$$A = \frac{C_{net}}{P_\gamma \times \varepsilon \times m \times t} \qquad (1)$$

Where C_{net} is the net peak count for each radionuclide present in the sample after subtracting the background count from the gross count, $P\gamma$ is the absolute gamma ray emission probability of the identified radionuclide, ε is the obtained full energy peak efficiency for each identified radionuclide, m is sample mass and t is the counting time.

The committed effective dose E_{ing} (Svy^{-1}) due to the ingestion of honey was calculated using Equation 2 while Equation 3 gives the lifetime cancer risk which is the probability of developing cancer during lifetime (Adesiji & Ademola, 2019; Aladeniyi et al., 2021; Isinkaye & Emelue, 2015).

$$E_{ing} = \sum_s C_s A_{ing} D_s \qquad (2)$$

Where C_s is the activity concentration of a specific radionuclide ($Bqkg^{-1}$), A_{ing} is the amount of honey consumed (0.02 kgy^{-1}) (FAOSTAT, 2019) and D_s is the ingestion dose coefficients for specific radionuclide ($SvBq^{-1}$). The values for D_s used in this study for ages 1, 5, 10. 15

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years and adult for ^{40}K, ^{238}U and ^{232}Th were obtained from International Commission on Radiological Protection (ICRP), (2012).

$$ELCR = E_{ing}..LE..RF \quad (3)$$

Where E_{ing} is the committed effective dose (Svy^{-1}), LE is the life expectancy in Nigeria (55 years for males and 57 for females) (UNFPA, 2022) and RF is the fatal cancer risk factor (0.05 Sv^{-1}) ((ICRP. 1991).

The Atomic Absorption Spectroscopy (AAS) technique was employed to estimate the heavy metal concentration in the honey samples. The honey samples were digested with acid and the concentrations of Cu, Cd, Cr, Pb, and Zn were determined using a Buck Model 210 VGP Atomic Absorption Spectrophotometer. Digestion of samples was performed in FOSS TECATOR™ Digester. The digested samples were cooled to room temperature, then transferred into test tubes and examined for the presence of the heavy metals using VARIAN AA 240FS Flame Atomic Absorption Spectrometer. The fuel and oxidant used were acetylene and air, respectively.

3 RESULTS AND DISCUSSION

The activity concentration of ^{40}K, ^{238}U and ^{232}Th in $Bqkg^{-1}$ is presented in Table 1. The samples collected from the wild are labelled HON 1 – 5, samples collected from the apiary are labelled HONT 1 – 5, and samples collected from commercial outfits are labelled HONK 1 – 5. The activity concentration of ^{40}K, ^{238}U, and ^{232}Th ranged from 6.49 to 14.96 with a mean of 11.67 ± 3.63; 0.34 to 1.28 with a mean of 0.82 ± 0.39 and 0.29 to 2.96 with a mean of 1.47 ± 1.13 $Bqkg^{-1}$, respectively in the HON samples. The HONT samples ranged from 3.90 to 16.56 with a mean of 10.68 ± 5.01; 0.53 to 1.64 with a mean of 0.97 ± 0.44 and 1.55 to 8.65 with a mean of 4.53 ± 3.14 $Bqkg^{-1}$ respectively, for ^{40}K, ^{238}U and ^{232}Th. For the HONK samples, the activity concentration of ^{40}K, ^{238}U and ^{232}Th ranged from 1.83 to 33.68 with a mean of 18.77 ± 12.30; 0.40 to 2.15 with a mean of 1.68 ± 0.75 and 0.82 to 10.95 with a mean of 5.40 ± 3.80 $Bqkg^{-1}$, respectively. From the average activity of the radionuclides ^{40}K had the highest values followed by ^{232}Th with ^{238}U having the lowest values in all the samples. The honey samples sourced from the commercial stores recorded the highest mean values for the radionuclides. The concentrations of the radionuclides in the honey samples were much lower than the maximum permitted levels of radionuclides in foodstuffs (Varga, 2008).

Table 2 shows the mean committed effective doses that the public could incur from the consumption of honey samples stated according to age. The mean committed effective doses ranged from 9 ± 6 $nSvy^{-1}$ in adults to 25 ± 12 $nSvy^{-1}$ in children (1 year) for HON samples. In the HONT samples, the mean committed effective doses ranged from 23 ± 14 $nSvy^{-1}$ in adults to 52 ± 25 $nSvy^{-1}$ in children (1 year). The mean committed effective doses in the HONK samples ranged from 29 ± 19 $nSvy^{-1}$ in adults to 68 ± 41 $nSvy^{-1}$ in children (1 year). The values obtained for the committed effective dose were below 0.29 $mSvy^{-1}$ which is the total committed effective dose from the ingestion of natural radionuclide as recommended by UNSCEAR, (2000b) for the general public.

The mean ELTR (Table 3) for males in the HON samples ranged from 2.5×10^{-8} (1 year) – 6.9×10^{-8} (adult) and 2.6×10^{-8} (adult) – 7.1×10^{-8} (adult) for females. For HONT sample, ELCR ranges from 6.3×10^{-8} (1 year) – 14.3×10^{-8} (adult) for males and 6.6×10^{-8} (adult) – 14.3×10^{-8} (1 year) for females. While for HONK samples, ELCR ranged from 7.9×10^{-8} (1 year) – 18.8×10^{-8} (adult) for males and 8.2×10^{-8} (adult) – 18.8×10^{-8} (1 year) for females. It can be observed that the ELCR for females is higher for the males for all the samples. This could result from the LE for females being higher than the male in Nigeria. All the ELCR values obtained were less than 0.29×10^{-3} reported to be the world standard limit (Aladeniyi et al., 2021).

Table 4 shows the concentration of the detected heavy metals in part per million (ppm). The concentration of heavy metal of Cd and Cr were below detection level in both the HON and HONT samples. While the mean values for Cu, Pb and Zn are 3.0, 7.5 and 0.3 ppm and 4.8, 3.8 and 1.0 ppm respectively, for HON and HONT samples. For HONK samples, Cu, Cd and Pb were below detection level and the mean values for Cr and Zn were 23.0 and 11.0 ppm.

The mean value of Pb in HON and HONT samples exceed the European Union limit set at 3 ppm (Šerevičienė et al., 2022), thus there would be a need for caution to be applied in consuming these samples.

Table 5 compares the activity concentrations of the natural radionuclides in honey for the present study to the values obtained from other countries as reported in the literature. The average value of the activity

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concentration of the natural radionuclides for the honey samples sourced from commercial outfits are higher than the honey samples from the wild. A similar observation is shown from the values reported by Meli et al., (2020) and Meli et al., (2016) where the honey samples sourced from commercial outfits have higher activity concentrations than honey samples from apiaries. The activity concentration of ^{40}K in the honey samples was observed to have highest values for all the countries reported.

Table 1: Activity concentration of ^{40}K, ^{238}U and ^{232}Th in $Bqkg^{-1}$ in samples of honey

nple		J	'h
)N1	28±0.33	3±0.19	9±0.11
)N2	9±0.24	8±0.09	1±0.09
)N3	29±0.37	4±0.13	6±0.08
)N4	3±0.26	3±0.12	6±0.04
)N5	96 ± 0.33	4±0.04	4±0.07
:an ± STD	**11.67 ± 3.63**	**2 ± 0.38**	**7 ± 1.13**
)NT1	56 ± 0.33	4 ± 0.06	7 ± 0.07
)NT2	16 ± 0.32	3 ± 0.05	4 ± 0.06
)NT3	94 ± 0.28	4 ± 0.04	5 ± 0.05
)NT4	0 ± 0.17	4 ± 0.07	5 ± 0.07
)NT5	4 ± 0.23	8 ± 0.05	6 ± 007
:an ± STD	**68 ± 5.01**	**7 ± 0.44**	**3 ± 3.14**
)NK1	51 ± 0.35	3 ± 0.12	2 ± 0.04
)NK2	00 ± 0.50	0 ± 0.11	95 ± 0.15
)NK3	68 ± 0.55	5 ± 0.12	8 ± 0.12
)NK4	85 ± 0.40	0 ± 0.05	7 ± 0.09
)NK5	3 ± 0.13	4 ± 0.11	8 ± 0.08
:an ± STD	**77 ± 12.30**	**8 ± 0.75**	**0 ± 3.80**
ference Level[a]	**)0**	**)0**	**)0**

[a] (Varga, 2008)

Table 2: Committed effective dose (mean ± std) ($nSvy^{-1}$) due to natural radionuclides in honey samples

nple	ear	ears	years	years	ult
)N	± 12	± 9	± 7	± 6	6
)NT	± 25	± 20	± 17	± 15	± 14
)NK	± 41	± 30	± 24	± 20	± 19
:ference Level 5vy-1) [b]	9	9	9	9	9

[b](UNSCEAR, 2000b)

4 CONCLUSION

The activity concentration of natural radionuclides in honey samples collected from the wild, apiary and commercial outfits have been studied and the annual committed effective dose from ingestion of the honey samples estimated. Potassium had the highest contribution to the activity concentration and the consumption of honey possess no radiological concern as the committed effective was below the UNSCEAR recommended limit. Though there may be health risk as a result of high value of lead in the honey samples sourced from the wild.

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Table 3: Mean values of the life time cancer risk for male and female (10^{-8}) from the consumption of natural radionuclides in honey samples

Sample	1 year		5 years		10 years		15 years		Adult	
	Male	Female	Male	Female	Male	Female	Male	Female	Male	Female
HON	6.9±3.3	7.1±3.4	4.5±2.4	4.7±2.5	3.5±1.9	3.6±2.0	2.8±1.6	2.9±1.7	2.5±1.5	2.6±1.6
HONT	14.3±6.8	14.8±7.1	10.4±5.6	10.8±5.8	8.4±4.7	8.7±4.9	7.0±4.1	7.3±4.3	6.3±3.8	6.6±4.0
HONK	18.8± 11.4	19.5±11.8	13.3±8.3	13.8±8.6	10.6±6.7	11.0±6.9	8.8±5.6	9.2±5.8	7.9±5.1	8.2±5.3
Reference Level (10^{-3})[c]	0.29	0.29	0.29	0.29	0.29	0.29	0.29	0.29	0.29	0.29

[c] (Aladeniyi *et al*., 2021)

Table 4: Average concentration of heavy metals (ppm) detected in the samples of honey

Sample	Cu	Cd	Cr	Pb	Zn
HON	3.0	ND	ND	7.5	0.3
HONT	4.8	ND	ND	3.8	1.0
HONK	ND	ND	23.0	ND	11.0
Reference Level	5[a]	0.5[a]	-[a,b]	0.5[a]	25[b]

[a] (Ševerevičienė et al., 2022), [b] (Ru *et al*., 2013)

Table 5: Comparison of the concentration of natural radionuclide (Bqkg^{-1}) from the present work with values from other parts of the world as reported from literature

Country	Source of honey	^{40}K	^{238}U	^{232}Th	Reference
Nigeria	Wild	11.67 ± 3.62	0.82 ± 0.38	1.47 ± 1.13	Present work
Nigeria	Commercial	18.78 ± 12.30	1.68 ± 0.74	3.40 ± 3.80	Present work
Italy	Commercial	50.6 ± 46.3	0.023 ± 0.010	< 0.007	(Meli et al., 2020)
Central Italy	Apiary	28.1 ± 23.0	0.020 ± 0.010	< 0.007	(Meli et al., 2016)
Greek	Wild and Apiary	7.9 – 102.2	0.013 – 0.10	-	(Xarchoulakos and Lasithiotakis, 2022)
Kosovo (Europe)	Apiary	20.44 ± 0.69	-	1.28 ± 0.81	(Dizman et al., 2020)
Romania	Apiary	24.08 ± 4.37	BDL	1.51 ± 0.74	(Bulubasa et al., 2021)
Poland	Apiary	32.92 (2010) 31.13 (2011)	-	-	(Borawska et al., 2013)
Italy	Wild	16 -111	-	-	(Caridi et al., 2022)
Reference Level		1000	1000	1000	(Varga, 2008)

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