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DETERMINATION OF RADIONUCLIDES IN SELECTED SAMPLES OF CIGARETTE, SHISHA AND SHISHA CHARCOAL MOSTLY USED IN KANO STATE, NIGERIA

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ABSTRACT
Shisha and Cigarette smokers suffer from deadly diseases. These could be as a result of the presence of quantities of radioisotopes from uranium and thorium-decay series which are radioactive and carcinogenic and pose toxic effects to living organisms. Increasing consumption of Shisha in Kano deemed it necessary to investigate those radioisotopes in products sold in the city markets. 30 samples were obtained, 10 each from Cigarette, Shisha and Shisha charcoal products and evaluated using Gamma Spectroscopy for radioisotopes. The average values of the activity concentration of ^{40}K, ^{238}U and ^{232}Th, absorbed dose rates, annual effective doses, radium equivalent activity indexes and excess life-time cancer risks of Shisha products were found to be 258.12±18.40 Bq kg^{-1}, 8.44±1.94 Bq kg^{-1}, 5.45±0.63 Bqkg^{-1}, 17.37 nGy h^{-1}, 4,975.80 µSv yr^{-1}, 34.55 Bq kg^{-1}, 17.42×10^{-3}, and that of Cigarette products to be 567.60±40.68 Bq kg^{-1}, 14.38±3.32 Bq kg^{-1}, 7.40±0.72 Bqkg^{-1}, 34.95 nGyh^{-1}, 894.32 µSv yr^{-1}, 68.67 Bq kg^{-1}, 3.13×10^{-3}, and that of Charcoal products to be 1129.14±80.40 Bq kg^{-1}, 16.93±3.79 Bq kg^{-1}, 7.90±0.77 Bqkg^{-1}, 59.53 nGy h^{-1}, 15,185.39 µSv yr^{-1}, 114.03 Bq kg^{-1}, and 53.15×10^{-3} respectively. Most of the values of these parameters were found to be higher than the recommended limit by UNSCEAR (2000) and WHO therefore posing serious health risks to smokers.
Keywords: Radiological Impacts, Radionuclides, Shisha, Cigarette, Kano.

INTRODUCTION

The fertilizers that tobacco farmers use to increase the size of their tobacco crops contain the naturally-occurring radionuclide radium and its decay products. As the plant grows, the radium from fertilizer along with naturally-occurring radon in the soil and rocks, transfer into and on the plant and later included in tobacco products made from these plants. Cigarettes made from this tobacco still contain these radioactive elements. The radioactive particles settle in smokers' lungs, where they build up as long as the person smokes. (EPA 2017)

In several studies, inhalation of some naturally occurring radionuclides via smoking has been considered to be one of the most significant causes of lung cancer. (Akinyose *et al.* 2018)

Tobacco contains minute amounts of radiotoxic elements such as ^{210}Pb, ^{210}Po and ^{238}U which are inhaled via smoking.

In many countries, cigarette smoking has been identified as a major serious health issue and contributor to the high mortality and morbidity rate of both smokers and passive smokers. Some surveys clarified that the content of certain chemicals especially Cadmium in fats, (Jha 2020), blood (El- Agha *et al*, 2002) and livers of tobacco smokers are much higher than those of non- smokers. Studies have shown that every 3000 non-smoking adults die of lung cancer as a result of breathing second-hand smoke from other's cigarettes. (Jha 2020)

It is reported that both Cigarette and Shisha Smokers have several complaints in common, including asthma, respiratory infections, shortness of breath, high blood pressure, increased blood sugar levels and sleep disturbances were similar in the 2 groups. (Husain *et al.,* 2016) produced evidence suggesting that shisha smoking is not safer than cigarette smoking.

In Nigeria, shisha smoking has rapidly become increasingly popular in major cities. Factors mediating this sudden trend are variable, including smoking for pleasure, smoking for its stimulating effect, experimentation, or perceived safety compared with cigarette smoking (Adams, 2016).

The vogue is rapidly pervading society and is commonly practiced by university undergraduates, adolescents and the older population in restaurants and hotels and at Social gatherings. (Adams, 2016) The research used Gamma Spectroscopy to investigate the presence of radionuclides in the samples of Cigarettes, Shisha and Shisha Charcoal obtained from Kano markets.

MATERIALS AND METHODS

Sample Collection

Ten (10) samples each of different brands of Shisha, Shisha coal and cigarette were randomly purchased from the local markets in Kano State. The ten different brands of cigarettes were coded as C_1, C_2, C_3, C_4, C_5, C_6, C_7, C_8, C_9 and C_{10} respectively. Also, the ten samples of Shisha were coded as S_1, S_2, S_3, S_4, S_5, S_6, S_7, S_8, S_9 and S_{10} respectively. Finally, the ten samples of Shisha coal were coded as H_1, H_2, H_3, H_4, H_5, H_6, H_7, H_8, H_9 and H_{10} respectively.

2.2 Sample Preparation

All covers, papers and filters were removed from the samples. The Cigarette and Charcoal samples were then grinded into a fine powdered form. 200g of each sample was neatly packed in well labelled polythene bags were air-dried for 48 hours under normal ambient temperature to remove the moisture content. The powder samples were then filled into cylindrical plastic containers. The sample containers were then sealed hermetically with adhesive tape and stored for 24 days before gamma ray analysis is performed to allow them to attain radioactive equilibrium (Sroor et al., 2001). The sample-filled containers were marked individually with identification parameters e.g., C_1, C_2, date of preparation and net weight. All samples were sealed throughout the analysis.

1.3 Gamma Spectrometer Instrumentation

The Gamma spectrometric analysis of the samples was achieved using a 3" × 3" sodium iodide [NaI(TI)] detector (Model 802 series, Canberra Inc. USA) which was accessed at the environmental radiation unit of the National Institute of Radiation Protection and Research (NIRPR), University of Ibadan, Nigeria. The gamma counting detector was enclosed in a 10 cm thick lead shield adequate to reduce the external background radiation sources by about 95%. The spectrum acquisition and processing were made possible by coupling the detector output to an ORTEC Multi-Channel Analyzer (MCA) alongside a PC equipped with Genie 2000 gamma energy evaluation software. Before gamma counting of the samples, efficiency and energy calibrations of the detector was carried out for the various energies of interest in the selected sample geometry in accordance with the existing procedures outlined by the International Atomic Energy Agency. (Livens, 1990)

RESULTS AND DISCUSSION

The activity concentration of the radionuclides detected in the samples are presented in Tables 1,2 and 3 and illustrated in Figures 1,2 and 3. All the radionuclides detected and quantified came from the naturally-occurring ^{238}U and ^{232}Th decay series, as well as non-series ^{40}K.

The specific activity concentration of ^{40}K, ^{238}U and ^{232}Th ranged from 681.10±49.04 to 1335.06±94.89 $Bqkg^{-1}$ (with an average of 1129.14±80.40 Bq kg^{-1}), 0.56±0.15 to 34.11±7.42 Bq kg^{-1} (with an average of 16.93±3.79 Bq kg^{-1}) and 0.73±0.07 to 14.04±1.37 Bq kg^{-1} (with an average of 7.90±0.77 $Bqkg^{-1}$) respectively for Shisha Charcoal samples, and from 329.39±23.68 to 767.07±55.12 $Bqkg^{-1}$ (with an average of 567.60±40.68 Bq kg^{-1}), 2.17±0.58 to 28.46±6.24 Bq kg^{-1} (with an average of 14.38±3.32 Bq kg^{-1}) and 1.62±0.16 to 14.04±1.37 Bq kg^{-1} (with an average of 7.40±0.72 $Bqkg^{-1}$) respectively for Cigarette samples, and from 26.68±1.10 to 333.87±23.92 $Bqkg^{-1}$ (with an average of 258.12±18.40 Bq kg^{-1}), 0.75±0.21 to 15.11±3.55 Bq kg^{-1} (with an average of 8.44±1.94 Bq kg^{-1}) and 1.61±0.16 to 12.13±1.18 Bq kg^{-1} (with an average of 5.45±0.63 $Bqkg^{-1}$) respectively for Shisha samples.

Table 1: Activity Concentration of Radionuclides and Radiological Impact (Bq kg^{-1}) in Cigarette Samples

SAMPLE CODE	K-40 (Bq/Kg)	U-238 (Bq/Kg)	Th-232 (Bq/Kg)	D (nGy h^{-1})	E (μSv y^{-1})	Raeq (Bq kg^{-1})	ELCR (× 10^{-3})
C1	635.05±45.50	28.46±6.24	2.01±0.20	41.03	414.57	80.23	1.45
C2	555.74±39.81	16.28±3.72	3.67±0.36	33.08	507.88	64.32	1.78
C3	329.39±23.68	13.82±3.30	4.03±0.39	22.65	528.27	44.95	1.85
C4	463.91±33.24	10.94±2.57	14.04±1.37	33.02	1580.77	66.74	5.53
C5	767.07±55.12	2.17±0.58	12.70±1.24	40.89	1377.80	79.40	4.82
C6	668.65±47.77	19.69±4.42	8.58±0.84	42.36	1057.34	83.45	3.70
C7	605.01±43.35	10.01±2.45	1.62±0.16	31.01	245.46	58.91	0.86
C8	534.39±38.46	10.28±2.53	1.90±0.19	28.34	276.87	54.15	0.97
C9	501.71±35.89	18.47±4.18	13.63±1.33	37.84	1588.91	76.59	5.56
C10	615.03±44.00	13.66±3.16	11.85±1.15	39.30	1365.28	77.96	4.78
Mean	567.60±40.68	14.38±3.32	7.40±0.72	34.95	894.32	68.67	3.13

Table 2: Activity Concentration of Radionuclides and Radiological Impact (Bq kg^{-1}) in Shisha Samples.

SAMPLE CODE	K-40 (Bq/Kg)	U-238 (Bq/Kg)	Th-232 (Bq/Kg)	D (nGy h^{-1})	E (μSv y^{-1})	Raeq (Bq kg^{-1})	ELCR (× 10^{-3})
S1	322.07±23.06	11.61±2.60	2.89±0.28	20.63	3330.53	40.54	11.66
S2	305.08±21.83	14.27±3.10	7.04±0.69	23.66	8029.83	47.83	28.10
S3	247.63±17.79	7.32±1.68	5.17±0.50	16.90	5880.91	33.78	20.58
S4	250.60±17.60	11.60±2.60	10.50±1.02	22.23	11924.94	45.91	41.74
S5	328.4±23.53	15.11±3.55	1.61±0.16	21.75	1902.64	42.70	6.66
S6	26.68±1.10	3.94±1.01	BDL	2.94	25.97	5.99	0.09
S7	333.87±23.92	3.66±0.95	1.80±0.95	16.80	2058.99	31.94	7.21
S8	188.08±13.61	0.75±0.21	BDL	8.25	7.91	15.23	0.03
S9	267.92±19.20	4.373±1.07	12.13±1.18	20.60	13728.23	42.35	48.05
S10	310.87±22.32	11.75±2.63	2.48±0.24	19.98	2868.01	39.23	10.04
Mean	258.12±18.40	8.44±1.94	5.45±0.63	17.37	4975.80	34.55	17.42

Table 3: Activity Concentration of Radionuclides and Radiological Impact (Bq kg^{-1}) in Shisha Charcoal Samples

SAMPLE CODE	K-40 (Bq/Kg)	U-238 (Bq/Kg)	Th-232 (Bq/Kg)	D (nGy h^{-1})	E (μSv y^{-1})	Raeq (Bq kg^{-1})	ELCR (× 10^{-3})
B1	1157.591±82.32	22.94±5.15	10.74±1.05	65.70	16786.35	127.43	58.75
B2	681.10±49.04	0.56±0.15	BDL	28.86	7373.73	53.00	25.81
B3	1207.79±85.87	13.50±3.07	1.60±0.16	57.93	14801.12	108.79	51.80
B4	1143.23±81.38	14.59±3.36	0.73±0.07	55.20	14103.60	103.66	49.36
B5	1297.51±92.37	21.53±4.80	1.17±0.11	65.15	16645.83	123.11	58.26
B6	1201.07±85.50	13.61±3.15	14.04±1.37	65.21	16405.66	126.17	57.42
B7	1133.61±80.66	22.09±4.80	11.42±1.11	64.71	16533.41	125.71	57.87
B8	1007.53±71.85	5.81±1.49	3.23±0.32	46.95	11995.73	88.01	41.99
B9	1335.06±94.89	34.11±7.42	0.73±0.07	72.27	18464.99	137.95	64.63
B10	1126.89±80.16	20.53±4.54	27.40±2.66	73.36	18743.48	146.48	65.60
Mean	1129.14±80.40	16.93±3.79	7.90±0.77	59.53	15185.39	114.03	53.15

Absorbed Dose Rate

The results of the absorbed dose rates D (nGy h^{-1}) in the air at 1m above the ground are calculated using equation (1),

$$D=C_U A_U+C_{Th}A_{Th}+C_K A_K \quad (1)$$

Where A_U, A_{Th}, A_K are the radioactivity concentration in Bqkg^{-1} and C_U, C_H, and C_K are dose conversion factors which are 0.462, 0.604 and 0.042 for ^{238}U, ^{232}H and ^{40}K respectively. (UNSCEAR 2000)

The values ranged between 28.86 to 73.36 nGy h^{-1} with an average of 59.53 nGy h^{-1} for Charcoal samples, and 22.65 to 42.36 nGy h^{-1} with an average of 34.95 nGyh^{-1} for Cigarette samples, and 2.94 to 23.66 nGy h^{-1} with an average of 17.37 nGy h^{-1} for Shisha samples. All the calculated values of the absorbed dose rate for Cigarette and Shisha samples were lower when compared to the recommended limit of 57 nGy h^{-1} (UNSCEAR 2000), however, about 70% of the Charcoal samples were higher.

Annual Effective Dose

Annual effective dose E (μSv yr^{-1}) was calculated using equation (2),

$$E = 0.75 \times A(Bq\ kg^{-1}) \times M\ (kg\ y^{-1}) \times DCF \quad (2)$$

E is the annual effective dose for cigarette smoke

A is the activity concentration of radionuclide

M is the consumption rate per year and DCF is the standard dose conversion factor.

The most recent dose conversion coefficients for the case of inhalation of cigarettes for adults are 2.9×10^{-6}, 4.5×10^{-5}, and 2.1×10^{-9} Sv Bq^{-1} for ^{238}U, ^{232}H and ^{40}K respectively (ICRP 119, 2012), and 5.0×10^{-7}, 1.1×10^{-4}, 2.1×10^{-9} Sv Bq^{-1} for ^{238}U, ^{232}H and ^{40}K respectively for Shisha.

The average mass of one (1) fresh tobacco per stick of cigarette is 0.86g.

Therefore, the annual consumption rate of consuming one (1) stick of cigarette daily were estimated to be 3.171 kg y^{-1} and 13.688 kg $y^{-1}$for Shisha.

The values of the annual effective dose ranged from 7,373.73 to 18,743.48 μSv yr^{-1} with an average of 15,185.39 μSv yr^{-1} for Charcoal samples, 245.46 to 1,588.91μSv yr^{-1} with an average of 894.32 μSv $yr^{-1}$for Cigarette samples and 7.91to13,728.2 μSv yr^{-1} with an average of 4,975.80 μSv $yr^{-1}$for Shisha samples. This dose was high when compared with the average worldwide exposure to natural radiation sources which is 2400 μSv y^{-1} and especially the part due to inhalation which is 1260 μSv y^{-1} (UNSCEAR 2000).

Radium Equivalent Activity Index

Radium equivalent activity index Raeq (Bq kg^{-1}) was calculated using equation (3),

$$Raeq = A_U + 1.43A_H + 0.077A_K \quad (3)$$

Where A_U, A_{Th} and A_K are the radioactivity concentration in Bq$kg^{-1}$of ^{238}U, ^{232}Th and ^{40}K respectively.

The values of the radium equivalent activity index, Raeq (Bq kg^{-1}) for Charcoal samples ranged between 53.00 and 146.48 Bq kg^{-1} with an average of 114.03 Bq kg^{-1}, 44.95 to 83.45 Bq kg^{-1} with an average of 68.67 Bq kg^{-1} for Cigarettes samples and from 5.99 to 47.83 Bq kg^{-1} with an average of 34.55 Bq kg^{-1} for Shisha samples. These values were found to be lower than the recommended limit of 370 Bq kg^{-1}(UNSCEAR 2000), and hence do not pose a serious health risk.

Excess Life Time Cancer Risk

Excess lifetime cancer risk (ELCR) was calculated using the below equation (4):

$$ELCR = AEDE \times DL \times RF \quad (4)$$

Where AEDE is the annual equivalent dose equivalent, DL is the average duration of life (estimated to 70 years), and RF is the Risk Factor (Sv^{-1}), i.e., fatal cancer risk per Sievert. For stochastic effects, ICRP uses RF as 0.05 for the public.(Oluyide et al., 2019)

The estimated values of the excess life-time cancer risk ($\times 10^{-3}$) for Charcoal samples ranged between 25.81 and 65.60 with an average of 53.15. Similarly, it ranged from 0.86 to 5.56 with an average of 3.13 for cigarette samples. Likewise, the excess lifetime cancer risk ELCR ($\times 10^{-3}$) for Shisha samples ranged between 0.09 to 48.05 with an average of 17.42 (Khater et al., 2008).

Since Shisha is consumed when burnt by its Charcoal, therefore the summation of both parameters of Shisha and Charcoal are absorbed concurrently, represented in table (4).

Table 4: Mean Activity Concentration of Radionuclides and Radiological Impact (Bq kg^{-1}) of Samples

PARAMETERS	CIGARETTE	SHISHA	CHARCOAL	SHISHA + CHARCOAL	Recommended Limit
K-40 (Bq/Kg)	567.60±40.68	258.12±18.40	1129.14±80.40	1387.26±98.8	400
U-238 (Bq/Kg)	14.38±3.32	8.44±1.94	16.93±3.79	25.37 5±73	35
Th-232 (Bq/Kg)	7.40±0.72	5.45±0.63	7.90±0.77	13.35±1.4	30
D (nGy h^{-1})	34.95	17.37	59.53	76.90	57
E (μSv y^{-1})	894.32	4975.80	15185.39	20161.19	1260
Raeq (Bq kg^{-1})	68.67	34.55	114.03	148.58	370
ELCR ($\times 10^{-3}$)	3.13	17.42	53.15	70.57	0.2

2. CONCLUSION

The results from the study show that the activity of Shisha products is higher than that of the Cigarettes, and Charcoal is higher than both. Moreover, all the products analysed are not safe for consumption due to being above the recommended limits by WHO.

As such it is recommended that concerned organizations and the government should put effort to discourage the consumption of Shisha.

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