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The natural radioactivity in some tropical fruit juices in Lapai metropolis by gross alpha and gross beta measurements

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1 ABSTRACT

The lack of adequate consumption of fruit has become a worldwide dietary concern since fruits play a pivotal role in attaining and maintaining good health. Assemblage of radionuclide's pollutant through ingestion in fruit juices may lead to some health hazards. One hundred and twenty juice samples obtained from eight different fruits; *Musa sapientum* (banana); *Mangifera indica* (mango); *Citrullus lanatus* (watermelon); *Musa paradisiaca* (plantain); *Carica papaya* (pawpaw); *Ananas comosus* (pineapple); *Citrus sinensis* (orange) and *Malus domestica* (apple) consumed in Lapai metropolis in Niger State, Nigeria, were analyzed for radionuclide contaminant by means of Alpha/Beta counting system (Canberra iMatic™). The average activity concentrations of gross α in the juice samples varied in a range of 1.96E+01 ± 3.77E+00 mBq/L in *Carica papaya* juice to the lowest of 4.39E-01 ± 4.73E-02 mBq/L in *Ananas comosus* juice with a corresponding average annual committed effective dose of 6.44E-01 mSv/year and 1.45E-02 mSv/year respectively. For the gross β, the average activity concentrations varied in a range of 5.10E+02 ± 3.90E+00 mBq/L for *Malus domestica* to 9.16E+00 ± 9.19E-01 mBq/L for *Ananas comosus* with a correspond average annual committed effective dose of 6.28E-01 mSv/year and 1.45E-02 mSv/year respectively. This variation may be attributed to geological factors of the region where the fruits are cultivated. The gross α/β activity values were low compared to the Food and Drug Administration (FDA) of 2004 limit of 100 mBq/L for gross α and 1000 mBq/L for gross β in beverages. These results show that the consumption of the fruit juices may not pose any significant radiological health hazards through ingestion to the consumers.

2 INTRODUCTION

The lack of adequate consumption of fruit has become a worldwide dietary concern since it play a pivotal role in attaining and maintaining good health. Fruits are crucial dietary components that can help reduce the risk for numerous chronic diseases, which, in many cases, have been shown to be initiated by long-term inflammation (Holt *et al.* 2009). According to the general standard for fruit juices and nectars, "fruit juices have the essential physical, chemical, organoleptic and nutritional characteristics of the fruit(s) from which it comes (CODEX 1989). Properly extracted juices are very similar to the fruit; they contain most substances, which are found in the

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original ripe and sound fruit from which the juice is made (EI, Hochman 2008; Lee *et al.* 2009 and Gerhaeuser 2008). Fruit juice is usually assumed to be free of chemical preservatives and other additives and only in a few cases, for example cloudy *Malus domestica* juice, the addition of ascorbic acid (vitamin C) is used for the prevention of browning. Fruit juices to some extent may contribute to internal radiation exposure (EI, Hochman, 2008) as a results radionuclides polluted; environments, soil, water, and surrounding atmospheric particles. The measurement of natural radioactivity in our physical environment allows the determination and assessment of population exposure to radiation. The occurrence of natural radionuclides in fruits may depend on the radioactive quality of the waters used in irrigation, nature of soil, the transfer rate of radionuclides (soil to fruits), humans activities in the area; such as fertilizers used in the agricultural processes. Considering the presence of natural radionuclides defined as nuclides which constantly emits radioactive particles such as alpha, beta and gamma and are detrimental to our health if the emission dose is high in juices. Radionuclides uptake can be influenced by the roots distribution within the soil profile, soil type, soil water status and characteristic rate uptake of fruit species Lasheen *et al.* (2008) and Jabbar *et al* (2009). There are large differences in the degree of translocation of radionuclides to fruits of different species, depending upon physiological characteristics of the plants (Njinga *et al.* 2013). The ingested radionuclides could be concentrated in certain parts of the body. Samat and Evans (2011) showed that uranium-238 accumulate in human lungs and kidney, thorium-232 in lungs, liver and skeleton tissues and potassium-40 in muscles. Depositions of large quantities of these radionuclides in particular organs will affect the health condition of the human such as weakening the immune system, induce various types of diseases and finally the increase in mortality rate (Njinga *et al.* , 2016). The decay of natural radionuclides when taken into body through ingestion may release several alpha and beta particles, which may be responsible for the damage living cells (WHO 2004 and IAEA 1996; 2005). It could lead to cell transformation, chromosome aberrations and mutation because of DNA damage (UNSCEAR 2000; 1993; 1988). The type and severity of the damages may depend on the quantity of the dose received. For individuals who receive more than the dose limit of 1 mSv/year, this may cause observable harm to organs or tissues (UNSCEAR 2000). Radiation hazard may include the modification of cells, in which case the damage in the viable cell is usually repaired and if this repair is imperfect, the modification will be transmitted to daughter cells, which may eventually lead to mutational events or cancer in the tissues or organs (Beir 2000 and Bernier *et al* 2001). The environmental radionuclides present the most risk to human health, so it is important to understand the transport, fate and effects of radionuclides moving through different types of fruit juices we consumed daily. This research is to quantify the presence of gross alpha and gross beta activity concentration in eight different types of fruit juices (*Malus domestica*, *Carica papaya*, *Citrus sinensis*, *Ananas comosus*, *Musa paradisiaca*, *Citrullus lanatus*, *Mangifera indica*, and *Musa sapientum*) consumed in Lapai LGA of Niger State, Nigeria. In this area, the major occupation is farming whereby all sorts of chemical fertilizers are used. The farmers also used various types of pesticides and the water used for irrigation run across tailings, refuge dumps etc and are highly polluted. The Food and Drug Administration (FDA) in the United State of America, protecting and promoting your health, release annually supporting document for guidance levels for radionuclides in domestic and imported foods (beverages). The documents cover the legislative definition of beverages and their exploitation, chemical contaminants, labelling and packaging (FDA, 2004). The current guidelines recommend that any beverages which could lead to an annual exposure to radiation of more than 0.1 mSv per year should be investigated further. The hypothesis of this study is to verify if the fruit juices produced locally from eight different fruits; *Musa sapientum* (banana)*; Mangifera indica* (mango)*; Citrullus lanatus* (watermelon)*; Musa paradisiaca* (plantain)*; Carica papaya* (pawpaw)*; Ananas comosus* (pineapple)*; Citrus sinensis* (orange) and *Malus domestica* (apple) had high radionuclide pollutant. This would be achieved through the measurement of the activity concentrations of gross alpha and gross beta in the eight aforementioned juice types in Lapai metropolis using gross Alpha/Beta counting system. This study also evaluate the committed annual effective dose for gross alpha and gross beta activity concentration and ascertain the radiological risk associated with the consumption of these different juice types. These evaluation were performed based on the ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{210}Pb and ^{210}Po of the Uranium series and ^{232}Th, ^{228}Ra and ^{228}Th of the Thorium series as the major alpha and beta emitting radionuclides linked to internal irradiation (Njinga *et al.* 2016).

3 MATERIALS AND METHODS

3.1 The sampling Area: Lapai is located on the latitude $9^0 24'0''$N to $8^0 9'0''$N and longitude $7^0 0'0''$E to $5^0 30'0''$E with the average elevation of 168 meters above the sea level. It has an area of 3,051 km² and a population of 110,127 at the 2006 census. Lapai as seen in Figure 1, share borders with Paikoro LGA and Gurara LGA to the North, Agaie LGA to the West, Kogi LGA to the South, and Abuja FCT to the East. Lapai, like other areas on the same latitude, is covered by two major rock formations; the sedimentary rocks to the south characterized by sandstones and alluvial deposits, particularly along the Niger valley and in most parts of Gulu, Muye and the eastern parts of Lapai town. These areas contain extensive flood plains of the River Niger and this has made the local government one of the largest and most fertile agricultural lands in the State. To the north of Lapai is the basement complex, characterized by out crops of the Migmatite-gneiss complex, the schist belts and the older granites of precambrian age, which can be found in the vast topography of rolling landscape. Such outcrops dominate the landscape in areas bounding Paikoro local government area in the northern part of Lapai local government.

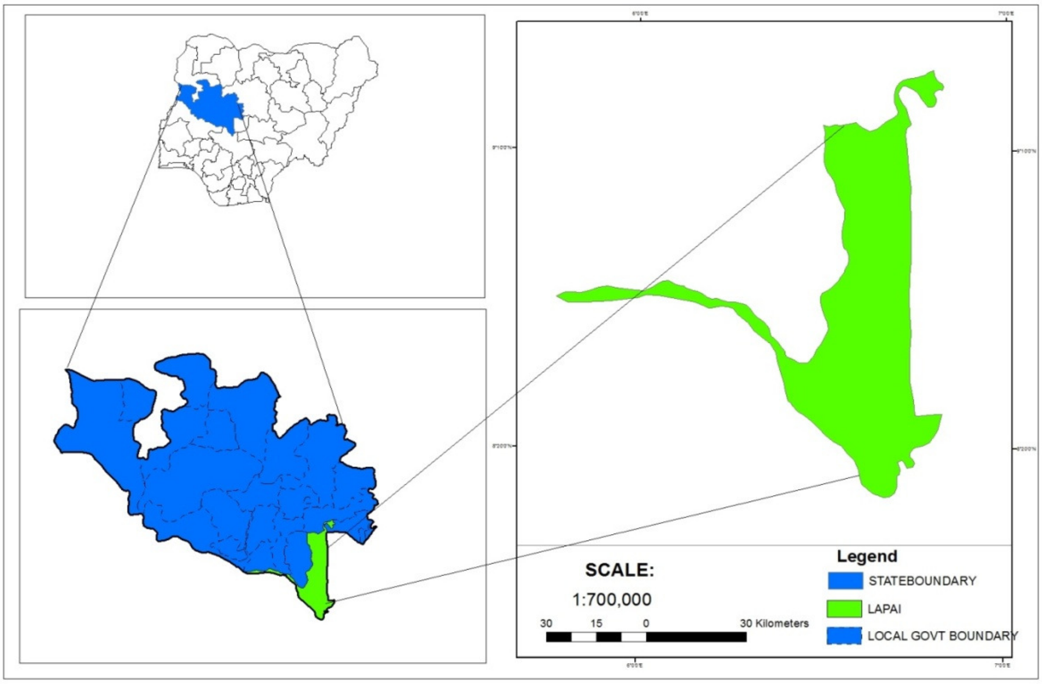

Figure 1: Map of the Study Area

3.2 Sample collection: One hundred and twenty (120) types of juices were taken as samples in Lapai LGA. The name of the samples juices and their number are as follows: *Malus domestica* (apple) (15), *Carica papaya* (pawpaw) (15), *Citrus sinensis* (orange) (15), *Ananas comosus* (pineapple) (15), *Citrullus lanatus* (watermelon) (15), *Mangifera indica* (mango) (15), *Musa paradisiaca* (plantain) (15), and *Musa sapientum* (banana) (15). This makes the total number of samples collected as One hundred and twenty (120).

3.3 Sample Preparation: One litre of each collected juice sample was first filtered through filter paper and transferred into 1 L beaker. Two millimetre of ultra-pure acid were added into 1L of filtered sample in order to maintain the radionuclides in the sample and to liberate dissolved metals from exchange sites on dissolved organic particles and let stay overnight. Each of filtered juice sample (300 ml) was transferred into 400 ml glass beaker and evaporated slowly to near dryness on the electrical hot plate placed in a fume chamber. The remaining fruit sample was transferred quantitatively in 47 mm stainless steel counting planchets. These were then heated to near dryness, stored into desiccators to allow them to cool into room temperature and to prevent them from absorbing moisture reading for counting in a low background Gas-less Automatic Gross Alpha/Beta counting system to determine the activity concentrations of gross alpha and gross beta present in each sample. The pH, salinity and total dissolved solids (TDS) of the juice samples were measured directly using Metrolin model 691 pH-meter for the pH and a portable (HACH) conductivity for salinity and TDS of juice samples.

3.4 The gas-flow-proportional counter: The gas-flow-proportional counter (eight-channel counter) in the Centre for Energy Research and Training (CERT), Ahmadu Bello University Zaria (ABU) was used. Each channel has a 450 μgcm^{-3} window thickness and 60 mm diameter. The detectors are operated within the radiation environment of < 101 $\mu radh^{-1}$. The system is connected to a microprocessor loaded with a spreadsheet program (Quattro-Pro) and graphic program (Multiplan) and is operated at a bias voltage ~1100 V, with P10 gas: argon-methane of 10 % (Onoja *et al.* 2004). ^{239}Pu and ^{90}Sr were used as the alpha standards with 24,110 years and 28 years half-life respectively. These standards were certified by CERCA LEA Laboratories in France. The plateau test was run in all the three operating modes for 1800 s for five cycles. The background radioactivity counting procedure were performed as reported elsewhere (Onoja *et al.* 2004) and the results indicated reproducibility in all the six adopted channels of the counter.

3.5 Analysis of Samples: The samples were counted according to the procedure selected during calibration of the equipment. The calibration was done using two standards sources: ^{241}Am source for Alpha and ^{90}Sr for beta, respectively. The counting efficiencies of alpha and beta were then 36.25% ± 2.18% and 73.16% ± 4.39%, respectively. The selected procedure was to allow the samples to be counted for 100 min each. This was done three times for each sample. The background reading of the detector for alpha and beta after counting three blank stainless steel planchets according to the procedure was 0.00148 Bq and 0.0026 Bq, respectively. The samples were analyzed for gross alpha and beta activity using an IN-20 model gas-flow proportional counter available at the Centre for Energy Research and Training, Ahmadu Bello University, Zaria, Nigeria. Each sample was counted three times and the mean used in computing the activity. The operational modes used for the counting were the α-only mode for the alpha counting and the β (+α) mode for the beta counting. The count rate of each sample was automatically processed by the computer using the equation below (Njinga *et al.* 2016);

$$A_{(\alpha,\beta)} = B_{(\alpha,\beta)} \times 60/T \quad 1$$

where $A_{(\alpha,\beta)}$ is the count rate (cpm) of alpha and beta particles $B_{(\alpha,\beta)}$ is the raw count of alpha or beta particle, T is the counting time (100 min).

The activity of each of the samples was calculated as;

$$C_{(\alpha,\beta)} = \left(A_{(\alpha,\beta)} - G_{(\alpha,\beta)}\right) \times \frac{U_{(\alpha,\beta)}}{H_{(\alpha,\beta)} \times S_{(\alpha,\beta)} \times V} \quad 2$$

where $C_{(\alpha,\beta)}$ is the alpha and beta activity (Bq/kg), $G_{(\alpha,\beta)}$ is the background count of alpha and beta particle, $U_{(\alpha,\beta)}$ is the unit coefficient of alpha and beta particle (1.67 x 10^{-2}) conversion factor from cpm to cps (1cps=1Bq), $H_{(\alpha,\beta)}$ is the channel efficiency for alpha or beta counting, $S_{(\alpha,\beta)}$ is the sample efficiency for alpha or beta counting and V is the sample mass.
However, the sample efficiency for the goat samples was computed using;

$$\varepsilon_{gs} = \frac{M_r}{M_i} \times 100 \quad 3$$

where M_r is the recovered mass after pellet was formed and M_i is the initial mass of the sample in powder form. The error associated with the sample activity was computed using;

$$E_r = \frac{\left[B + \frac{(100000)^2}{T_{bgd}} \times G_{(\alpha,\beta)}\right]}{100000} \times \frac{U_{(\alpha,\beta)}}{H_{(\alpha,\beta)} \times S_{(\alpha,\beta)} \times V} \quad 4$$

where *B* is the sample raw count, T_{bgd} is the background count time. The ingestion dose coefficients used to calculate indicative doses for naturally occurring alpha emitting radionuclides are published in ICRP (1991 and 2007). The committed annual effective dose for gross α or gross β were evaluated using the ingestion dose coefficient for adult (I_{DCA}) of 4.50E-08 Sv/Bq. The intake of the juice per year ((I_{JPY}) was assumed to be 730 L/year, the average annual alpha or beta committed effective dose for a particular juice type was calculated by averaging the individual annual committed effective doses contributed by the major alpha or beta emitters in the uranium-238 and thorium-232 series of the natural occurring radionuclides using equation 5 below (Tettey-Larbi *et al.* 2013);

$$E_{ing,J(\alpha/\beta)} = \frac{I_{JPY}}{V_{oJ} . N_R(\alpha/\beta)} \sum_{i}^{R(\alpha/\beta)} A_{\alpha/\beta} \times I_{DCA}(\alpha/\beta) \quad 5$$

where $E_{ing,J(\alpha/\beta)}$ is the average gross annual alpha or beta committed effective dose in the juice sample, $A_{\alpha/\beta}$ is the gross alpha or beta activity concentration in the juice sample in Bq/L, V_{oJ} is the volume of the juice samples used for the analysis, $N_R(\alpha/\beta)$ is the number of radionuclides considered as major alpha or major beta emitters in the uranium-238 and thorium-232 series of the natural radionuclides.

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4 RESULTS AND DISCUSSION

Tables 1 show the average pH, TDS, and salinity of juice samples obtained locally from various market within Lapai local government area (LGA). The activity concentrations of gross α and gross β in the juice types are shown in Tables 2 & 3 and their corresponding committed annual effective doses evaluated are shown in Figure 3.

Table 1: Physical parameters for Fruit juice samples

Specie name	Fruit Juice Types	Average Values of		
		pH	TDS (mg/L)	Salinity (ppm)
Citrus sinensis	Orange	6.41	4780	5.9
Ananas comosus	Pineapple	6.35	2010	2.1
Musa paradisiaca	Plantain	6.23	369	0.4
Citrullus lanatus	Watermelon	6.24	413	0.4
Mangifera indica	Mango	6.52	366	0.4
Musa sapientum	Banana	6.51	3550	3.9
Malus domestica	Apple	6.69	5220	5.3
Carica papaya	Pawpaw	6.99	7270	4.4

Tables 2: Gross alpha activity concentration in the eight different fruit juices

Juice Type	Number of sample	Gross alpha Activity Concentration (mBq/L)				
		Mean Val.	Max Val.	Min Val.	STDEV	Median
Citrus sinensis	15	3.21E+00	5.76E+00	1.33E+00	1.43E+00	3.52E+00
Ananas comosus	15	4.39E-01	1.93E-01	6.73E-02	4.73E-02	1.91E-01
Musa paradisiaca	15	1.16E+00	1.03E+00	1.50E+00	7.89E-02	1.16E+00
Citrullus lanatus	15	4.97E-01	9.25E-01	7.72E-01	1.61E-01	5.01E-01
Mangifera indica	15	2.09E+00	3.37E+00	8.00E-01	2.50E-01	2.13E+00
Musa sapientum	15	4.53E+00	7.32E+00	1.79E+00	1.01E+00	4.76E+00
Malus domestica	15	1.91E+01	2.73E+01	7.87E+00	2.07E+00	1.98E+01
Carica papaya	15	1.96E+01	2.93E+01	1.18E+01	3.77E+00	2.08E+01
ADF limit	-	1.00E+02	-	-	-	-

As observed in Table 2 and 3, the maximum activity concentrations of gross alpha in the locally produced juices varied from 1.93E-01 mBq/L to 2.93E+01mBq/L and for gross beta activity it ranged from 9.96E+00 mBq/L to 5.11E+02 mBq/L. The values of the minimum activity concentration for gross α ranged from 6.73E-02 mBq/L to 1.18E+01 mBq/L. The highest values of the gross α and gross β radioactivity concentrations were recorded in *Carica papaya* and *Malus domestica* juice types and the lowest recorded in *Ananas comosus* juice type. Since the cultivation areas of the fruits where the juice are extracted have slightly different geological formations, these slight variation in the activity concentrations of gross alpha and gross beta could be a result of the geological characteristics of the soils, the irregular distribution of the minerals in the surfaces of rocks or solids aquifers from which each of these plants are cultivated as well as the pH of each juice samples.

Table 3: Gross beta activity concentration in the eight different fruit juices

Juice Type	Number of sample	Gross beta activity concentration (mBq/L)				
		Mean Val.	Max Val.	Min Val.	STDEV	Median
Citrus sinensis	15	1.29E+02	1.19E+02	3.68E+01	2.44E+01	1.19E+02
Ananas comosus	15	9.16E+00	9.96E+00	2.98E+00	9.19E-01	9.23E+00
Musa paradisiaca	15	1.44E+02	1.66E+02	6.25E+01	2.71E+01	1.50E+02
Citrullus lanatus	15	5.12E+01	5.86E+01	3.02E+01	5.04E+00	4.98E+01
Mangifera indica	15	1.31E+02	1.31E+02	3.83E+01	1.45E+00	1.31E+02
Musa sapientum	15	1.17E+02	1.10E+02	8.23E+01	5.07E+00	1.18E+02
Malus domestica	15	5.10E+02	5.11E+02	3.82E+02	3.90E+00	5.11E+02
Carica papaya	15	3.29E+02	3.59E+02	1.58E+02	3.68E+01	3.49E+02
ADF limit	-	1.00E+03	-	-	-	-

This variation could also be because of radionuclides content in soil and bedrock system. As it is known, the gross alpha activity is presented by ^{238}U series and gross beta activity is presented by ^{40}K as well as ^{232}Th series (Yuce *et al.* 2009). Lapai, like other areas on the same latitude, is covered by two major rock formations; the Sedimentary Rocks to the south characterized by sandstones and alluvial deposits, particularly along the Niger valley and the eastern parts of Lapai town. These areas contain extensive flood plains of the River Niger and this has made the LGA one of the largest and most fertile agricultural lands in the state. To the north is the Basement Complex, characterized by out crops of the Migmatite-Gneiss Complex, the Schist Belts and the Older Granites of Precambrian age, which can be found in the vast topography of rolling landscape.

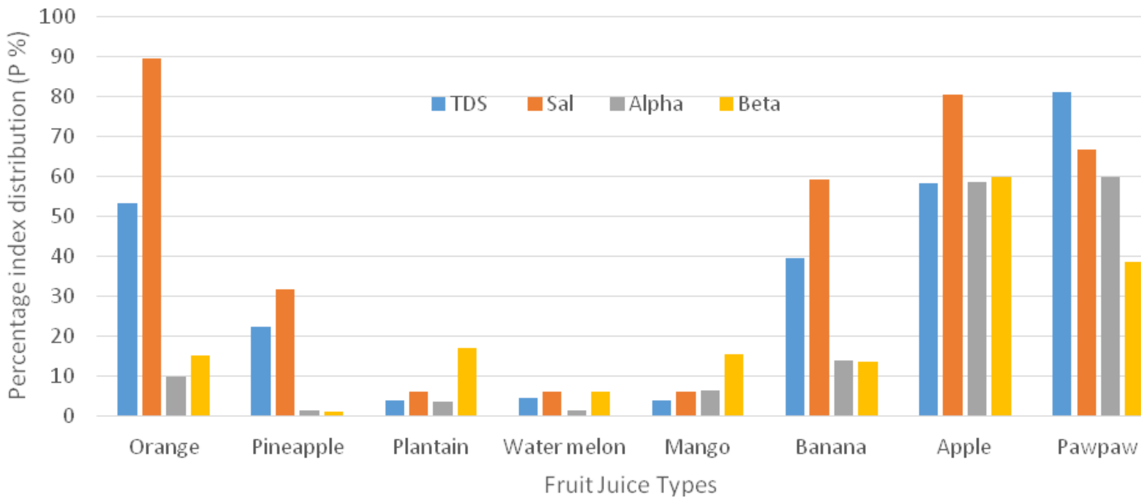

Figure 2: Comparison of the gross α and gross β activity concentrations with the TDS and the salinity for the juice samples. Each parameter was expressed as a percentage index, P (%)

As shown in Figure 2, the activity concentrations of the gross alpha and gross beta obtained in the present study was also compared with the salinity and the TDS. These parameters are given in Tables 1. Each of the parameters such as the salinity, the total dissolved solid, and the gross alpha and gross beta radioactivity were normalized to unity and expressed as percentage index, P (%). The TDS known as liquid quality parameters are assumed to have some effects on the gross α and gross β concentrations and hence radiological quality of the juice samples (Bonotto *et al.* 2009). Comparing the trends in the TDS and the salinity with the gross α and gross β activity concentrations, neither gross α nor gross β related to both TDS and salinity. This indicated that the radionuclide in a juice type is dependent on parameters that are difficult or impossible to properly evaluate. These parameters include the extent of large minerals surfaces and irregular distribution of large minerals in the surfaces of the rocks; soil (Bonotto *et al.* 2009).

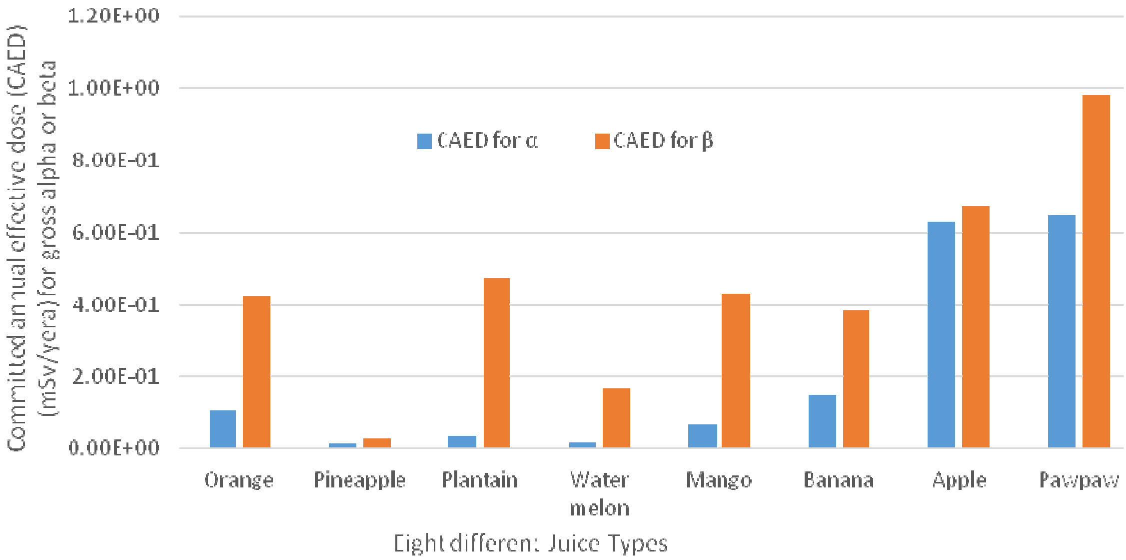

Figure 3: The committed annual effective dose (CAED) for gross alpha or gross beta in the juice types

As shown in Figure 3, the average annual committed effective dose for gross α activity concentrations in the juice samples varied in a range of 6.44E-01 mSv/year in *Carica papaya* juice to the lowest of 1.45E-02 mSv/year in *Ananas comosus* juice. The average annual committed effective dose for gross β activity varied in a range of 6.28E-01 mSv/year for *Malus domestica* to 1.45E-02 mSv/year for *Ananas comosus* juice types. As observed in Figure 3, the committed annual effective dose (CAED) corresponding for either gross alpha or gross beta showed great variation in eight different fruit juice types. The CAED for α and β in *Carica papaya* and *Malus domestica* juice samples were high compared to the other juices types while *Ananas comosus* recorded the lowest value for either gross α or gross β analysis. These high values of CAED recorded in *Malus domestica* and *Carica papaya* juices may be attributed to three major factors; - the water used during irrigation may be polluted with some level of radionuclides, - the fertilizers and soil content of radionuclides may be high, -method of preservatives (these produce are usually imported). The committed annual effective dose for gross α and gross β in the other juice types produced locally from *Mangifera indica* fruits, *Citrus sinensis* fruits, *Musa sapientum* fruits, *Musa paradisiaca*, *Citrullus lanatus* fruits, and *Ananas comosus* fruits where relatively low. These fruits are locally cultivated and based on the result, the water used in irrigation, the soil, fertilizer, and the industrial production processes appear to be relatively free of radionuclides contaminant. Based on the recommended screening levels for gross alpha and gross beta activity of 100 mBq/L and 1,000 mBq/L, respectively it was observed that all the juice samples had gross-α and gross-β values below the FDA (2004) screening levels for beverages. This indicated that consumption of the juices in the study areas may not cause any radiological health hazards to human due to ingestion and any action to reduce radioactivity might not be necessary. Based on these initial screening levels not exceeded the ADF health hazard limit, there is no need to evaluate the concentrations of individual radionuclides present in the juice samples.

5 CONCLUSION

The study considered public exposure in Lapai metropolis, Niger State, Nigeria due to natural radioactivity by accessing the activity concentrations of gross-α and gross-β in eight different fruit juices used in the surrounding communities of the study area. The ranged of the average activity concentrations of gross alpha and gross beta recorded in the juice sample types were 1.96E+01 ± 3.77E+00 mBq/L in *Carica papaya* to lowest of 4.39E-01 ± 4.73E-02 mBq/L in *Ananas comosus* and 5.10E+02 ± 3.90E+00 mBq/L in *Malus domestica* to 9.16E+00 ± 9.19E-01 mBq/L in *Ananas comosus* respectively. Similar trend in the distributions were observed in the committed annual effective dose for gross α and gross β. These variation may be attributed to geological factors including the geochemical characteristic of the region where the fruits are cultivated. A routine study is necessary to investigate the dependence of gross alpha and gross beta activity concentrations on these parameters. However, as revealed in this study, consumption of the juice types may not cause any radiological health hazards to human due to ingestion.

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