Monitoring of Environmental Radioactivity in Foodstuff in Fiji

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Abstract: This study was conducted to measure natural environmental radioactivity level and to compute the presence of radionuclides in the foodstuff consumed in Fiji. It is the first attempt in Fiji to investigate the presence of radionuclides and to evaluate their concentration in commonly used local and imported food items. The present study concentrates on seven vegetables, three fruits and two nuts. NaI(Tl) detector system was used to measure radioactive nuclides. The presence of radionuclides: 241 Am, 137 Cs, 60 Co, 40 K and 235 U have been detected. It has been observed that the range on these radioactive nuclides lies in the range of 0.08 – 6.0 Bq. Kg⁻¹, 0.07 – 22 Bq. Kg⁻¹, 10.1 – 47 Bq. Kg⁻¹, 47 – 210 Bq. Kg⁻¹ and 0.019 – 29 Bq. Kg⁻¹, respectively.

Keywords: Natural radioactivity level, radioactive nuclides in foodstuff, Cesium, Potassium, Cobalt, Uranium, Americium

1. Introduction

Natural and artificial radioactivity is present everywhere in our surrounding like oceans, rivers, soils, rocks, vegetables, fruits and animals as well as human body tissues (Castro et al., 2012). It has been reported that humans and our environment are continuously exposed to these types of radiation of which 81% can be attributed to natural radiation and 19% comes from artificial sources (Mazzilli et al., 2002). The artificial radioactivity comes from the testing of nuclear weapons, nuclear accidents and radiological accidents that raised concern in the public resulting in considerable research activities conducted over the last several decades in computing and measuring radioactivity content in soil, air and water (UNSCEAR, 2008).

The natural radioactive nuclides consists of terrestrial long-lived radionuclides like ${}^{40}K$, ${}^{238}U$, ${}^{232}Th$, ${}^{235}U$ and cosmogenic radionuclides ${}^{3}H$ and ${}^{14}C$. It has been noted that the discharge of large amounts of radioisotopes into the environment are able to affect food items such as vegetables, fruit & animal feed through deposits from the air, contaminated rainwater or by falling onto the surface of said items. Radioactivity in water can also accumulate in rivers and the sea and can be deposited on fish and seafood. The presence of radioactive material in our environment can also be integrated infood as it is taken up by plants, seafood or ingested by animals (Mazzilli et al., 2002).

However the major nuclear accidents can introduce different kinds of radionuclides but some are very short-lived and others do not readily transfer into food. Radionuclides generated either by nuclear installations or nuclear accidents that could be significant for the food chain include; radioactive hydrogen (³H), carbon (¹⁴C), technetium (⁹⁹Tc), sulphur (³⁵S), cobalt (⁶⁰Co) strontium (⁸⁹Sr and ⁹⁰Sr), ruthenium (¹⁰³Ru and ¹⁰⁶Ru), iodine (¹³¹I and ¹²⁹I), uranium (²³⁵U) plutonium (²³⁸Pu, ²³⁹Pu and ²⁴⁰Pu), caesium (¹³⁴Cs and ¹³⁷Cs), cerium (¹⁰³Ce), iridium (¹⁹²Ir), and americium (²⁴¹Am) (Mahmood et al., 2013).

The iodine-131 is of intermediate concern because it is distributed over a wide land area, water and on crops and it is rapidly transferred from contaminated feed into milk. However, iodine-131 has a relatively short half-live and will decay within a few weeks. In contrast, radioactive cesium which can also be detected early on, is longer-lived (Cs-134 has a half-life of about 2 years and Cs-137 has a half-life of about 30 years) and can remain in the environment for a long-time. Radioactive cesium is also rapidly transferred from feed to milk. Uptake of cesium into food is also of long-term concern (Kam et al., 2016).

Radioisotopes such as strontium and plutonium pose a more long term risk if exposed to the environment. The half-life of strontium-90 is approximately 29 years whereas that of plutonium is significantly longer depending on the isotope (Pu-238: 88 years, Pu-239: 24,100 years, Pu-240: 6564 years). Fortunately the immobility of both these radioisotopes in the environment means they will pose little immediate or medium-term impact on the global food trade market.

Several studies have been conducted worldwide to measure radioactivity in food stuff (Kam et al., 2016; Thair and Alaamer, 2008; Omar El Samad et al., 2012; Castro et al., 2012; Mahmood et al., 2013; Nkuba and Mohammed, 2014; Abojassim et al., 2014; Schooshtari et al., 2017;; Jibiri et al., 2007, Júnior et al., 2005; Kant et al., 2015; Fadol et at., 2016; Królak and Karwowska, 2010; Beňová et al., 206; Al-Dughmah and Qurashy, 2012; Anwiri et al., 2011; Hosseini et al., 2016). Fiji is an agricultural based country and the most common food consists of grains, yam, cassava, sorghum, maize, rice, vegetables and fruits. The objective of present study is to measure the radioactivity levels in foodstuff that are commonly consumed by people living in Fiji. Apart from these initial seven vegetables, three fruits and two nuts were chosen and the presence of radioactive nuclides in these items was measured.

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2. Methodology

Natural radioactivity levels were measured using a gamma spectrometer system (Bridgeport) which includes gamma multichannel analyzer equipped with NaI(Tl) detector of $(3"\times3")$ crystal dimension and is shown in Figure 1

The gamma spectra were analyzed using the **MAESTRO** data acquisition and analysis system. The detector had coaxial closed facing geometry and its technical specifications are tabulated in Table 1.



Figure 1: Block diagram of NaI(Tl) detector (adapted from EMORPHO User's Manual - Bridgeport Instruments, LLC, page 5, 2009)

Table 1: The Intrinsic Peak Efficiency of the NaI (7	(I)
Crystal versus Gamma Ray Energy of Radionuclides	$(\varepsilon_P).$

Radionuclides	Associated Gamma Ray Energy (keV)	Intrinsic Peak Efficiency (ε_P)			
$^{241}_{95}Am$	59.50	0.80			
60 27 Co	1,332.50	0.055			
¹³⁷ ₅₅ Cs	661.70	0.15			
$^{40}_{19}K$	1,460.80	0.055			
^{99т} 43Тс	140.50	0.65			
$^{235}_{92}U$	185.70	0.55			
²⁰⁸ 81 81	2614.78	0.03			
²²⁶ ₈₈ Ra	241.00	0.40			

An energy calibration for this detector was performed with a set of standard γ -ray 0.25- μ Ci active ¹³⁷Cs and 1.0- μ Ci ⁶⁰Co sources. In this study, the activity concentration of ⁴⁰K was determined directly from the peak areas at 1460 keV. The Gamma transition lines of ²¹⁴Bi recoreded by the NaI(Tl) detector at 1765 keV were used to determine activity concentration of radioisotope in the ²³⁸U-series. Simiralry, the activity concentrations of radioisotope in the ²³²Th-series were calculated using the chracteristic gamma transition lines of ²⁰⁸Tl (2614 keV). The counting time for the present study was 64800 seconds and kept fixed for each sample.

Activity of Radionuclide

The Activity Concentration (A_c) of radionuclides (Bq/kg) in foodstuff was evaluated using Equation (1) taken from Abojassim et al., 2014:

$$A_{c} = \frac{C - BG}{\varepsilon \% M t I_{\gamma}}$$
(1)

Where A_c is the specific activity in (Bq/kg), C is the area under the photo peaks; ϵ % represents Percentage of energy efficiency. I_xis the percentage of gamma-emission probability of the radionuclide under consideration, t is counting time in (Sec.), M is mass of sample in (kg) and BG is background counts in the photo peak.

Sample Collection and Preparation

The samples of vegetables and fruits were purchased from different sellers at the Lautoka vegetable and fruit market where more than 90% of population of Fiji living in this town and its surrounding suburbs purchase their food. The measured samples were first washed with tap water and then with distilled water, and peeled when necessary.

Radioactivity Measurements

To qualitatively categorize the counts of nuclear nuclides in food stuff and to quantitatively compute their activities, all prepared samples of food stuff were measured using gamma-ray spectroscopy system as shown in Figure 1 and 2 using NaI(Tl) detector for 64,800 seconds. The equal counting time for background and sample measurements was chosen to minimise the uncertainty in the net counts. The spectrum of each sample was analysed and identification of unknown radionuclides was carried out by considering their peak centroid energies. The centroid energies of the peaks from the spectrum were compared with the reference gamma-ray energies obtained from the nuclear data (nuclear data tables shown to me). The radionuclides contained in samples were identifies and the areas under the peaks then determined the activity concentrations of each nuclide.

The number of counts under the full-energy peak areas (corrected for back ground peak areas), the counting time, the absolute full-energy peak efficiency for the energy of interest and gamma ray emission probability corresponding to the peak energy are used for the calculation of the concentration of a particular nuclide in the measured sample. Equation (1) discussed above has

been used to evaluate the activity concentration A_c .

3. Results and Discussion

Twelve foodstuff samples were collected out of which, one (Brazil Nuts) was an imported item and remaining eleven are locally grown representing the presence of radioactivity in Fijian soil. The measured activities of gamma in these items are tabulated in Table 2. The activity of the sample is given in Bq.Kg⁻¹ and the measurement errors are presented in Table 2.

Our preliminary results demonstrate that the gamma ray activity ranges from 346 ± 20 to 18.1 ± 0.8 for $\frac{^{226}}{^{88}}Ra$ Bq. Kg⁻¹; 210 ± 10 to 47 ± 2 for $\frac{^{40}}{^{19}}K$ Bq. Kg⁻¹; 29 ± 1 to 0.089 ± 0.03 for $\frac{^{235}}{^{92}}U$ Bq. Kg⁻¹; 210 ± 10 to 47 ± 2 for $\frac{^{40}}{^{19}}K$ Bq. Kg⁻¹; 22 ± 1 to 0.070 ± 0.004 for $\frac{^{237}}{^{55}}Cs$

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Bq. Kg ⁻¹ : 12.1 ± 6.0 to 0.086 ± 0.004 for $\frac{20}{8}$		$Bq. Kg^{-1}$	and 3.4 ± 0.2	to	0.089 ± 0.03	for	$^{99m}_{43}Tc$	
Bq. Kg ⁻¹ ; 6.0 ± 0.3 to 0.077 ± 0.006 for	or	$^{241}_{95}Am$	Bq. Kg ⁻¹ .					

Table 2: Specific activity of vegetables (v1 – V6), Fruits (F1 - F3) and Nuts (N1 – N2) commonly consumed in Fiji. The symbol '- 'in this table indicates negligible and/or zero value for the specific activity per unit mass of sample

Samuela	Specific Activity (<i>BqKg</i> ⁻¹)							
Sample	$^{241}_{95}Am$	60 27 Co	¹³⁷ ₅₅ Cs	$^{40}_{19}K$	^{99т} 43Тс	$^{235}_{92}U$	²⁰⁸ 81Ti	²²⁶ 88Ra
Cassava	(0.46 ± 0.02)	-	-	-	(2.19 ± 0.08)	(1.58 ± 0.06)	-	-
Ginger	(0.54 ± 0.03)	-	-	-	(0.129 ± 0.006)	(0.49 ± 0.02)	-	-
Layalaya	(0.43 ± 0.02)	(21.879 ± 0.002)	-	-	(0.91 ± 0.04)	(0.97 ± 0.05)	-	-
Bitter Gourd	(1.20 ± 0.06)	(47 ± 2)	-	-	-	-	-	-
Bottle Gourd	(0.077 ± 0.006)	-	-	-	-	-	-	-
Kumala	-	-	-	-	(0.089 ± 0.03)	(1.47 ± 0.05)	(3.5 ± 0.1)	-
Lady Fingers	-	-	-	-	-	(0.019 ± 0.001)	-	-
Banana	(6.0 ± 0.3)	(10.1 ± 0.5)	(0.070 ± 0.004)	(89 ± 4)	(0.155 ± 0.07)	(2.9 ± 0.1)	(0.86 ± 0.04)	(18.1 ± 0.8)
Lemon	(1.07 ± 0.05)	-	-	(59±3)	(0.038 ± 0.02)	-	-	-
Watermelon	-	-	-	(47 ± 2)	-	-	-	-
Brazil Nuts	-	-	(22 ± 1)	(210 ±10)	(3.4 ± 0.2)	(29 ± 1)	(12.1 ± 0.6)	(346 ± 20)
Peanuts	-	(14.1 ± 0.7)	-	-	(1.23 ± 0.06)	(0.94 ± 0.04)	(1.86 ± 0.09)	-
Minimum	(0.077 ± 0.006)	(10.1 ± 0.5)	(0.070 ± 0.004)	(47 ± 2)	(0.089 ± 0.03)	(0.019 ± 0.001)	(0.86 ± 0.04)	(18.1 ± 0.8)
Average	(1.40 ± 0.07)	(23.3 ± 0.8)	(11.0 ± 0.5)	(101 ± 5)	(1.03 ± 0.06)	(4.7 ± 0.2)	(4.6 ± 0.2)	(182 ± 10)
Maximum	(6.0 ± 0.3)	(47 ± 2)	(22 ± 1)	(210 ± 10)	(3.4 ± 0.2)	(29 ± 1)	(<i>12.1</i> ± 0.6)	(346 ± 20)

The radioactivity in the foodstuff grown in Fiji is less than that of the imported Brazil nuts. Brazil nuts are about a thousand times more radioactive than most common foods and if one was to consume 226 g, it would result in a dose of 0.5 mrem. Literature states that deep roots which form the Brazil nut trees are excellent at accumulating trace metals in the soil, which results in nuts that containto 7000 pCi Kg⁻¹ of $^{226}_{88}Ra$ (David 2016). Fortunately, the body has no need of radium, so even the most avid Brazil nut fan will excrete the isotope before it can do any harm (https://newatlas.com/radiation-explained-food-sources-danger/46233/).

The results presented in Table 2 show the presence of natural radioactive nuclide ${}^{40}_{19}K$ in banana $(84 \pm 4 \text{ Bq kg}^{-1})$; lemon $(59 \pm 3 \text{ Bq kg}^{-1})$; watermelon $(47 \pm 2 \text{ Bq kg}^{-1})$; and Brazil nuts $(210 \pm 10 \text{ Bq kg}^{-1})$. Once again, the contents of ${}^{40}_{19}K$ in

Fiji grown foodstuff is less than that of the food item imported in the country. The measured ${}^{40}_{19}K$ contents for banana grown in Fiji are less than that reported in literature that might be due to the difference in soil composition and the organic growth (non-use of chemical fertilizers) of this crop in the country.

There was a Fukushima nuclear accident on March 11, 2011 which could introduce artificial radioactivity in environment. A comprehensive investigation of the possible effect of this nuclear disaster was conducted by Environmental Radioactivity Studies and Monitoring Department (SESURE) in 2013 for French Polynesia and reported the absence of radiological impact in New Caledonia and Polynesia (report PRP-ENV?SESURE/2013-08). The present study also noted the presence of very small amount of $^{237}_{55}Cs$ in our foodstuff indicating that the effect of Fukushima nuclear accident might be minimal in this region of the globe. However, further investigation is warranted to quantify the impact of the Fukushima disaster on the Pacific countries.

The data reported in this study provides the building blocks to establish the environmental radioactivity baseline levels useful to detect and screen any suspected contamination or abnormal concentration resulting from any unforeseen accidental situation.

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The present project would be extended to include more vegetables and fruits; dairy products; imported food items; fish and meat. It is important to calculate the annual effective dose equivalent resulting from the exposure to natural radioactivity that is the part of a person's diet.

4. Conclusions

Radionuclide concentration in foodstuff consumed by the peoples living in Fiji was conducted and it has been noted that the ${}^{237}_{55}Cs$ concentration is in low levels. The presence of nuclides due to the natural radioactivity is well below the permissible radiation dose (see Table 3).

Table 3: Annual Maximum Permissible Dose limits adapted from (https://ehs.research.uiowa.edu)

mrem	rem	
5,000	5	Whole Body Dose Equivalent (Head, trunk, active blood-forming organs & reproductive organs)
50,000	50	Whole Body Shallow Dose Equivalent (Skin of the whole body)
15,000	15	Lens of Eye Dose Equivalent
50,000	50	Extremities (Hands, forearms, feet and ankles)

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