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Radon (^{222}Rn) Concentration in Fresh and Processed Coconut Water Using a RAD7 Detector

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ABSTRACT

Coconut water has several uses that are beneficial to humans, mainly because of its nutritional and medicinal properties. In this study coconut water samples were collected from the fresh mature coconuts and from packaged processed coconut water to measure the radon concentrations in them. The results were used to estimate the annual effective radiation dose to the general adult population from ingestion of coconut water. The samples were analyzed for radon concentrations using the Durridge RAD-7 radon detector. The results of measured radon concentrations for the fresh mature coconut were ranged from 381.10 $Bq\cdot m^{-3}$ to 1716.80 $Bq\cdot m^{-3}$. The results for packaged processed coconut samples ranged from 68.08 $Bq\cdot m^{-3}$ to 556.85 $Bq\cdot m^{-3}$. The mean values for the samples were (1081.40 and 222.78) $Bq\cdot m^{-3}$ for the fresh and processed samples, respectively. The estimated mean annual effective doses (AED) due to ingestion of coconut water were 0.41 and 0.08 $\mu Sv\cdot y^{-1}$ for fresh and processed samples, respectively. These values are significantly lower than the safe limits recommended by the World Health Organization and the European Commission of 0.1 $mSv\cdot y^{-1}$ and the maximum contamination level (MCL) of 11,000 $Bq\cdot m^{-3}$ by the United States Environmental Protection Agency and so do not constitute a risk to consumers of coconut water.

1. INTRODUCTION

Radon (^{222}Rn) is a naturally occurring radioactive noble gas that is the daughter product of radium-226. Every nucleus of ^{222}Rn eventually decays to Lead (^{210}Pb). Radon-222 has a half-life of 3.824 days and emits alpha particles during its radioactive decay.

Radon that is inhaled and ingested in the body is thought to be quickly eliminated from the body. Ingested radon is removed from the body via exhalation. However, the radon that is absorbed in the body is done so through dissolving in the blood in the gastrointestinal tract and other tissues [1]. Of the noble gases, radon has the highest solubility ratio for adipose tissue compared with blood, which is the major tissue of deposition for radon in the body. In addition, the major radiation dose in the body is due to the inhalation of the progeny of radon [2]. The comparatively longer half-life of radon-222 results in a greater buildup of this heavy radioactive gas in enclosed spaces. Emitted alpha particles resulting from the decay of radon-222 are responsible for DNA damages and other cellular injuries in the respiratory tract. This damage may result in lung disease and lung cancer from "chromosomal abnormalities, double-strand DNA breaks, and the production of reactive oxygen species, leading to carcinogenesis" [3]. Lung cancer deaths from smoking remain the leading cause of cancer deaths in the United States [4] thus qualifying radon-induced lung cancer as the second leading cause of an estimated 21,000 cancer deaths in the United States [5]. Additionally, radon has been associated with Alzheimer's disease deaths in the United States [6].

Radon has been classified as a Group 1 carcinogen by the International Agency for Research on Cancer (IARC). This classification renders radon as known to be carcinogenic based on evidence in human and animal studies. A similar classification of radon carcinogenicity has been given by the Agency for Toxic Substances and Disease Registry (ATSDR) but not by the US Environmental Protection Agency (EPA) [7].

The natural existence of radioactivity in the earth's environment has as a logical consequence the presence of the radioisotopes or their progenies in the food supply and the radioactive elements existing in the blood and bones. The worldwide annual average effective dose of radioactivity received from natural background radiation is approximately 2.4 mSv. A little over half of this amount (1.4 mSv) comes from progenies of radon and thoron, with 1.24 mSv from radon progeny [8]. It may be assumed that the vectors for the plant uptake of radon are from radionuclides in the soil or water and deposition on the leaves, to seed and subsequently to the coconut water and into the food supply [9-12].

Human and animal studies have indicated that even low to moderate radiation exposure may increase the long-term risk of cancer. The organ receiving the highest dose of total effective radon dosage due to ingestion is the stomach [13]. Hence, stomach cancer's highest risk contributor is radon due to ingestion [1].

A review of the published literature by Das [14] indicated that several forms of malignant and non-malignant cancers were correlated with radon levels. High levels of radon in ground water that is ingested is absorbed in the gastrointestinal tract. The United States Environmental Protection Agency (USEPA) has estimated that 11% of the stomach cancers may be attributed to high radon concentration ingested in water. Other cancers such as those of the blood, pancreas, and liver also show correlations to both inhaled and ingested radon [14, 15]. Although radon is removed rapidly from the blood, it is removed less rapidly from adipose tissue due to its high solubility in this kind of tissue and the longer time spent in the body may facilitate damage due to higher decay rates for radon and its progenies. The incidence of stomach cancer and/or mortality due to radon shows a higher risk in females than males, indicative of a gender dependent factor [14, 16].

Copious studies have been done on radon concentration in drinking water [12, 17-21]. Radon concentration in other food sources has been investigated such as for milk [16, 22, 23], fruits [24], juices and teas [22, 25-27], and certain foods [28-30].

Coconut water, the liquid (or endosperm) of the coconut, (*Cocos nucifera* L.) has been used for a variety of purposes including hydration, as an intravenous fluid [31], and fermented beverage rich in probiotics [32], and as a source of flavonoids which have anti-cancer anti-aging, anti-diabetic, anti-mutagenic anti-bacterial, anti-HIV and anti-inflammatory effects properties [33]. Coconut water may also aid in weight loss, as a means of shrinking and eliminating kidney stones, storage medium for avulsed teeth, and skin care [34]. As a fat free and zero cholesterol product, it has many health benefits as it is comprised of electrolytes, vitamins, sugar, protein, antioxidants, minerals, and dietary fiber [35].

Its effects on the body may serve to regulate hypertension by its introduction of high levels of potassium causing vasodilatation and the improvement in endothelial function [36, 37]. The consumption of coconut water has been increasing. For example, between 2010 and 2014, the USA consumption of coconut water grew from over 38,000 to 300,000 tons as consumers are becoming more aware of the nutritional and medical benefits of the coconut [38] so that new efforts are being implemented in certain parts of the coconut-producing regions to increase production and processing [39]. Thus, given the benefits and the increasing consumption of coconut water, it is important to assess if the levels of radon present in fresh and packaged coconut water pose any potential radiological risks to humans.

2. MATERIALS AND METHOD

Nine fresh mature coconuts samples were obtained from Mexico and Florida (USA). Fourteen packaged coconut water samples (11 different samples) were purchased from different stores in the locality. The packaged samples showed packaging information for Brazil, the Philippines, Thailand, and Vietnam. Care was taken to ensure that only packaged coconut waters whose ingredients were labeled "100% coconut water" (some with added ingredients) were chosen. The samples selected were also based upon their "bottled by" or "manufactured by" date so that only the freshest samples were being tested. Additionally, "best by" date for consumption was also a selection criterion since these dates (which are printed on the packages) typically represent a consumption date that is 12 months from the production date and the time frame when the product is best-tasting and most nutritious [40]. The coconut water samples from the fresh mature coconuts were extracted through the "soft eye" portion of the three "eyes" that are located at the base of the coconut. The coconut was then inverted into a short-tipped funnel lined with gauze (to filter particulates) that was inserted into the 40 ml sample bottles. At least two 40 ml samples (sub-samples) were collected from each coconut allowing spillage which was necessary to remove any bubbles from the samples. The processed coconut water samples were poured from their containers very quickly into 250 ml bottles and 40 ml bottles, checked for air bubbles, and quickly capped. For the sample P4; one 250 ml and two 40 ml sub-samples were made and for sample P13, only one 40 ml bottle was made. Except for two fresh coconut water samples, F3 and F4, all samples were immediately analyzed after they were bottled to minimize radon decay in the samples. Concentration values for samples F3 and F4 were corrected using the information in the RAD H_2O manual for decay correction [41].

The radon concentrations in the samples were measured with a Durridge RAD7 radon detector and the RAD H_2O Radon in Water Accessory (www.durridge.com) [41]. See **Figure 1**. The RAD7 is a solid-state alpha detector that has been widely used in numerous studies for over a decade [42-44]. The RAD7 measures the alpha particle resulting from the ^{222}Rn progeny (mainly, ^{218}Po; $t_{1/2}$ = 3.1 min) decay which are

Figure 1. A typical setup of the Durridge RAD7 radon detector with the attached RAD H_2O accessory showing a 250 ml sampling bottle and tube of the DRIRITE desiccant.

electrically attracted to a silicon alpha detector. By counting the decay of ^{218}Po, the radon concentration is assessed. The RAD H_2O accessory was used to measure the radon concentration in water over a range of 4 - 750,000 $Bq \cdot m^{-3}$ [41].

Prior to each test, the RAD7 was dried out using the Laboratory Drying Unit (LDU). In some cases, this was done overnight and for a period greater than 12 hours. A closed loop system was formed between the LDU and RAD7 unit. This method brought the humidity within the RAD7 to be as low as possible and resulted in starting humidity readings inside the detector of 2% to 4%. The relative humidity registered inside the detector was usually less than 10% after each run. The desiccant used to dry the gas in the LDU was regenerated indicating calcium sulphate granules made by W.A. Hammond Drierite Company (www.drierite.com).

The measurement process using the combination of the RAD7 and RAD H_2O forms a closed-loop system whereby the sample is aerated through air pumped through the outlet port of the RAD7 and bubbled through a glass frit [41]. This closed-loop aeration extracts radon within five minutes. This process is greater than about 90% efficient in extracting radon from the coconut water sample. The measurement cycle time for a sample is 30 minutes. At the end of the cycle a printout is obtained with the mean, standard deviation, minimum and maximum values for the measured radon concentration. For multiple sub-samples of a given sample, the detector was purged until the relative humidity was below 6% and the next sub-sample is analyzed.

3. RESULTS AND DISCUSSION

The values obtained for the radon concentrations in fresh coconut water and package coconut water samples are given in **Table 1** and **Table 2**, respectively. The mean values shown in the tables for a single sample are the average of the values for each of the four counting cycles in the measurement process. The uncertainty is one standard deviation. Values in the table that are starred represent the average of the mean values for each sub-sample and the uncertainties were calculated using the method of quadrature. This method of assessment was correct only because the sub-sample sizes were the same. The radon concentration values in the fresh coconut water samples ranged from 381.10 $Bq \cdot m^{-3}$ to 1716.80 $Bq \cdot m^{-3}$ with a grand mean value of (1022.38 ± 246.56) $Bq \cdot m^{-3}$. For the packaged coconut water samples, the radon concentration values range was 68.08 $Bq \cdot m^{-3}$ to 556.85 $Bq \cdot m^{-3}$ with a grand mean value of (153.62 ± 56.05) $Bq \cdot m^{-3}$. **Figure 2** shows the contrast between these mean values. This difference is reasonable given that processed coconut water is taken through a series of processes which aerate the coconut water and release

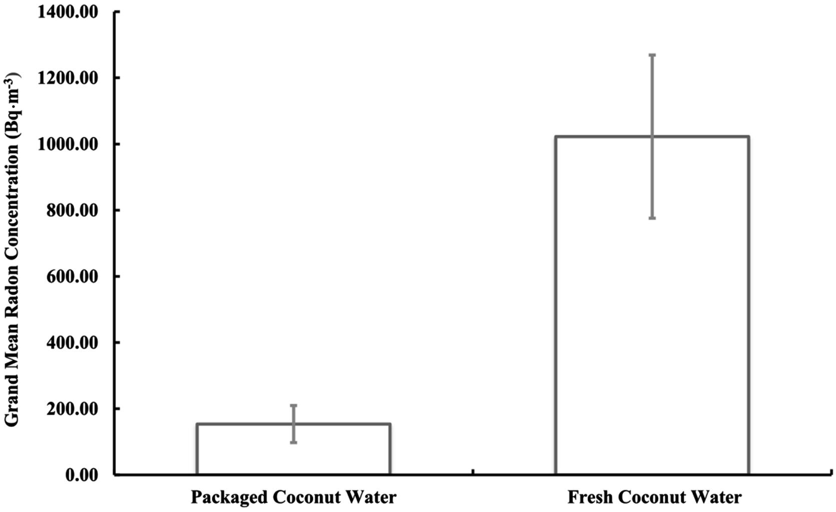

Figure 2. Comparison of the average radon concentration between the packaged and fresh coconut water samples used in this study.

Table 1. The concentrations of radon gas in fresh coconut water used in the current study.

Sample No.	Sample Code	Radon Concentration ($Bq \cdot m^{-3}$)	
		Mean	Uncertainty
1	F1	1292.23*	219.87
2	F2	954.60	190.55
3	F3	1166.63*	410.44
4	F4	976.31*	244.29
5	F5	1335.70	366.30
6	F6	381.10	220.15
7	F7	1335.70	366.30
8	F8	1716.80	653.05
9	F9	573.50*	196.11

*These values are the average of the means for at least two sub-samples of the same sample.

Table 2. The concentrations of radon gas in processed coconut water used in the current study.

Sample No.	Sample Code	Radon Concentration ($Bq \cdot m^{-3}$)	
		Mean	Uncertainty
1	P1	410.70	111.74
2	P2	306.73	116.18
3	P3	153.00*	42.85
4	P4	416.50*	139.48
5	P5	68.08	39.22
6	P6	136.16*	46.11
7	P7	112.85*	40.71
8	P8	163.05*	24.01
9	P9	187.96	142.64
10	P10	206.83*	83.50
11	P11	187.96*	40.67
12	P12	421.80	421.80
13	P13	94.17*	28.72
14	P14	112.85*	37.57

*These values are the average of the means for at least two sub-samples of the same sample.

even more radon in the environment [45]. Since no prior radon concentration results are available for coconut water, a qualitative comparison may be made from studies of radon in water. Akar *et al.* [46] reported lower values of radon concentration in tap water versus well water and Cho *et al.* [47] observed a 16.0% - 98.9% decrease in the radon concentration between raw water and treated water in their study of bottled water.

The large uncertainty values shown were due primarily to the low counts obtained for each sample. Poisson's statistics describes nuclear decay, and the uncertainty is the square root of the number of counts, n. However, the relative uncertainty means that the uncertainty is divided by the number of counts, n, the results being proportional $n^{-1/2}$. Hence if n decreases, the uncertainty increases and vice versa. In this study, the counting times were not extended for the samples. The counting periods were determined by the protocols Wat-40 (for the 40 ml samples) and Wat250 (for the 250 ml samples) [41]. The mean annual effective dose (AED) was estimated from the radon concentrations of the coconut water samples.

The radon concentration measured in this work was well below the recommended levels set by the World Health Organization (WHO) [8, 20], the European Commission (EC) [48, 49], and the United States Environmental Protection Agency (USEPA) [50]. Furthermore, as there are no prior coconut water studies to compare the values obtained in this study, it may be of interest to compare them to those for mineral water. The results for these comparative values are given in **Table 3**.

To estimate the annual effective dose (AED) for ingestion, Equation (1) was used [51-54].

$$ADE_{\text{ing}} = C_{\text{Rn}} \times R \times D_C \times T \times 1000 \tag{1}$$

where ADE_{ing} is the annual effective dose for ingestion in mSv/y, C_{Rn} is ^{222}Rn concentration in coconut water in Bq/l, R is the weighted estimate of coconut water consumption in l/d, T is the amount of time in days scaled to a year (d/y), and D_C is the effective dose coefficient for ingestion in Sv·Bq^{-1}. For ingestion, the following parameters were used; $A_{\text{DC}} = 3.50 \times 10^{-9}$ Sv·Bq^{-1} [1], R is assumed to be 0.30 l/d for adults. The time, T, is assumed to be 183 days to accounts for the fact that it is established at this time the actual consumption rate for coconut water for any of the age groups. The multiplicative factor of 1000 is the conversion from Sievert (Sv) to milli-Sievert (mSv). The values for the ADE for both the fresh and packaged coconut water are shown in **Table 4**. The value of 0.30 l/d is based on the 2016 datum for the USA importation of 288,537 metric ton of coconut water and assuming 10% of adult population consume coconut water.

Table 3. **Comparison of radon concentration in coconut water and mineral water.**

Place	Radon Concentration (Bq·l^{-1})		References
	Range	Mean Value	
Jeddah City, Saudi Arabia	5.56 - 14.87	9.92	[55]
Ratchaburi, Thailand	-	17 ± 0.9	[56]
Krakow, Poland	1.30 - 8.32	-	[57]
Egypt	0.93 - 6.89	-	[58]
Lubin, Poland	0.009 - 0.388*	0.0994*	[59]
Arar City, Saudi Arabia	5 - 10	-	[50]
Kuwait	1.02 - 6.05	2.97 ± 1.44	[60]
Algeria	2.6 - 14	7 ± 4	[61]
Mexico and Florida (fresh coconut water)	0.38 - 1.72	1.02$^{\$}$	This work
Various markets (packaged coconut water)	0.04 - 0.57	0.15$^{\$}$	This work

*Values were converted from mBq·dm^{-3} to Bq·l^{-1}. $^{\$}$Values are the grand means.

Table 4. **Annual effective dose to adults from ingesting fresh and processed coconut water used in the current study.**

Sample No.	Sample Code	Annual Effective Dose ($\mu Sv \cdot y^{-1}$)	
		Fresh	Processed
1	P1	-	0.16
2	P2	-	0.12
3	P3	-	0.07
4	P4	-	0.03
5	P5	-	0.22
6	P6	-	0.03
7	P7	-	0.04
8	P8	-	0.01
9	P9	-	0.03
10	P10	-	0.07
11	P11	-	0.10
12	P12	-	0.04
13	P13	-	0.16
14	P14	-	0.03
15	P15	-	0.03
16	F1	0.59	
17	F2	0.37	
18	F3	0.53	
19	F4	0.18	
20	F5	0.51	
21	F6	0.15	
22	F7	0.51	
23	F8	0.66	
24	F9	0.22	

The range of the estimated values for the annual effective dose to adults of radon in coconut water is 0.15 to 0.66 μSv/y for the fresh coconut water and 0.01 to 0.22 $\mu Sv \cdot y^{-1}$ for the packaged coconut water. These values are well below the recommended safe level of 0.1 $mSv \cdot y^{-1}$ established by the WHO and the European Council.

4. CONCLUSIONS

In this work measured radon concentration was presented for fresh and packaged coconut water. Also, the health impact through the consumption of coconut water from these sources was calculated. This work represents baseline data for radon concentration and the annual effective dose to humans from radon in coconut water. As may be assumed, the concentration is lower in the packaged coconut water versus the fresh coconut water. The data supported this assumption as the mean concentration data of the fresh coconut water samples was significantly higher than that for packaged coconut water samples, 1.07 $\text{Bq}\cdot\text{l}^{-1}$ versus 0.21 $\text{Bq}\cdot\text{l}^{-1}$, respectively. The radon concentrations for all samples were also much lower than the minimum contaminant level allowed by the USEPA of 11 $\text{Bq}\cdot\text{l}^{-1}$. None of the sample values were outside the range set by UNSCEAR (4 - 40) $\text{Bq}\cdot\text{l}^{-1}$.

The impact on the health of consumers of coconut water was assessed by estimating the annual effective dose received by ingesting fresh and packaged coconut water. The values for the AED ranged from 0.15 to 0.66 $\mu\text{Sv}\cdot\text{y}^{-1}$ and 0.01 to 0.22 $\mu\text{Sv}\cdot\text{y}^{-1}$ for the fresh coconut water and the packaged coconut water, respectively. These low dose values are also well below the recommended value of 0.1 $\text{mSv}\cdot\text{y}^{-1}$, so consumers are not at risk for the development of cancer because they consumed coconut water.

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CONFLICTS OF INTEREST

The authors declare no conflicts of interest regarding the publication of this paper.

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