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Analysis of Natural and Anthropogenic Radionuclide Content in Palm Date Fruit of the United Arab Emirates: a Baseline Study.

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Abstract

The purpose of this study is to contribute to a wider effort of establishing environmental radiation baseline for the United Arab Emirates (UAE) before the startup of the country's first nuclear reactor in 2017. An investigation of gamma emitting radionuclides concentrations in palm dates grown in the UAE was performed. The palm date samples of 10 varieties originated from a several local commercial date palm farms of the UAE were collected and analyzed. The study targeted the naturally occurring radionuclides, such as ^{238}U, ^{232}Th, and ^{40}K, in addition to any potential anthropogenic radionuclides, such as ^{137}Cs and others. Gamma spectrometry revealed measured activity concentrations for ^{238}U (^{226}Ra), ^{232}Th (^{228}Ra) and ^{40}K in range between 0.61 to 0.80 Bq kg^{-1}, 0.10 to 0.23 Bq kg^{-1} and 191 to 362 Bq kg^{-1}, respectively, on dry weight basis and calculated activity concentrations on wet basis were found in range between 0.52 to 0.69 Bq kg^{-1}, 0.09 to 0.22 Bq kg^{-1} and 168 to 297 Bq kg^{-1}, respectively. No ^{137}Cs or other anthropogenic radionuclides could be detected in this study. All measurements were performed using coaxial HPGe detector with 40% relative efficiency quoted by the manufacturer. Efficiency calibration correction factors were calculated using Angle software package.

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Key words: Date fruit, ingestion, radioactivity, gamma-spectrometry, baseline study, UAE.

INTRODUCTION

In 2008 the United Arab Emirates (UAE) has launched a nuclear energy program with the aim of starting its first reactor in 2017 (Sung-yeop Kim et al. 2013). Before first reactor startup, it is essential for a country to collect complete environmental radiation baseline data for the purpose of monitoring effects of reactor operation on the environment and for mitigation of consequences of any potential accident. Literature survey revealed little to no environmental radiation data published for the UAE. Hence, the present study is focused on measuring radioactive isotope concentration in different varieties of date fruit originating from several local commercial date palm farms within the UAE. The results can be used as reference baseline data for any future study.

Environmental radioactivity originates from natural and anthropogenic sources. Natural radionuclides include ^{235}U, ^{238}U, ^{232}Th, their progeny, and primordial radioisotope of potassium ^{40}K. These naturally occurring radioactive materials (NORM) are long-lived with half-lives on the order of thousands of years. Anthropogenic radionuclides, like ^{137}Cs, ^{131}I, ^{90}Sr, ^{237}Np, ^{241}Am etc., are products of nuclear processes from industrial, medical and military applications and can be released into environment through controlled (regulated discharges) or uncontrolled releases like the Windscale accident in 1957, the Chernobyl accident in 1986 and the Fukushima-Daiichi accident in 2011 (IAEA, 2006; McGeoghegan et al., 2010; Richard Black, 2011). The presence of these anthropogenic radionuclides in environmental samples is an indicator of any previous nuclear accident or contamination events. Also some anthropogenic isotopes are present in the

environment due to the global fallout from the nuclear weapons testing in the second half of the 20th century.

Both natural and anthropogenic radionuclides can be found in terrestrial and aquatic food chains at different concentration levels and they have a potential of consequent transfer to humans through food consumption. Thus, the amount of internal radiation exposure to humans is directly related to the type and amount of food being consumed. Hence, there is an interest worldwide in measuring the internal radioactivity exposure that humans receive from the consumed food (IAEA, 1989 and 2006; ICRP, 1996; UNSCEAR, 2000). Literature search reveals numerous studies that investigate the natural radionuclide content in foods like date palm fruit, rice, cereals, fruits and vegetables, milk, beef, seafood, medicine plant, tea and coffee etc. (Hosseini et al., 2006; Franic et al., 2008; Saeed et al., 2011; Alrefae et al. 2012; Alrefae et al., 2013, Canbazoglu and Dogru, 2013, Alrefae et al., 2014; Alrefae. T., 2015).

The most prevalent agricultural product in the UAE is the date fruit which has become an integral part of the heritage and identity of the country. The UAE owns one third of the world's trade of dates, coming in first in exports and second in imports in 2009 (WAM, 2011). More than 44 million date palms that are growing in the UAE can be grouped into 199 different varieties and they produced 76,000 tons of fruit in 2011 (Chandrasekaran and Bahkali, 2013). Food and Agriculture Foundation (FAO) announced Liwa and Al Ain date palm oases as "Globally Important Agricultural Heritage Systems", for their importance as repositories of genetic resources (WAM Emirates news report, 2015). Furthermore, date fruit is widely consumed in other regions of the Middle-East, North Africa and some parts of Asia too. Hence, it was decided

in the present study to select date fruits as the focus of analysis as it is one of the important agricultural products in UAE. The goal of the present study is to measure radioactivity concentrations of natural and anthropogenic radionuclides in different varieties of palm dates collected from several local commercial farms to establish reference baseline.

MATERIALS AND METHODS

With assistance from the Federal Authority for Nuclear Regulation (FANR) and Al Foah Date Fruits Company, total of 22 date fruit batches were collected in a final ripen (Tamar) stage, from several local commercial palm date farms of 4 major date fruit cultivation locations of the UAE: Al Silaa, Liwa, Ghayathy and Sih Al Khair. The collection took place in three lots from August to October 2014, as this is the harvesting period for date fruits at their Tamar stages. Each batch represented a single sample and corresponded to a collection of dates from several trees of same variety and from a single palm date farm. Collected batches were of 10 different varieties namely Neghal, Dabbas, Rezaiz, Yardi, Khadi, Khalas, Bumaan, Shishi, Zahedy and Khanezi. All batches were delivered to the laboratory at Khalifa University in sealed clean plastic bags with appropriate labeling. Fruit were then kept in the refrigerator before sample preparation took place.

All palm dates underwent proper sample preparation following procedures of the IAEA, 1989 and Alrefae, 2015. First, fresh weight of all lots of samples was measured and recorded and then fruit were washed with distilled water and air dried. Then all samples were pitted and put into a conventional oven at 80°C for at least one to two weeks, depending on the moisture contents within the samples, for complete dryness. After drying, each sample was grinded into powder to achieve the required homogeneity before being transferred to a clean empty 1 L

Marinelli beakers (MB). Each MB's end cap was sealed with scotch tape to make them airtight. Prior to gamma counting, each sample was kept sealed for a period of 4 weeks to allow for the short-lived decay products of ^{226}Ra, from ^{238}U-series and ^{228}Ra, from ^{232}Th series to reach secular equilibrium between parent radionuclides and their progenies.

All samples were then measured on a gamma-ray counting system, a high-resolution mechanically-cooled p-type HPGe coaxial detector (EG&G ORTEC) coupled with a DSPEC Pro multichannel analyzer (MCA). To reduce the background radiation level the detector system was surrounded by a laminar shield with 100 mm thick lead and a 3 mm copper. The HPGe detector used in this study had an energy resolution of 1.85 keV FWHM at 1.33 MeV photopeak of ^{60}Co and 40% of relative detection efficiency. Energy calibration for the detector was performed using ^{152}Eu point source. The reference efficiency calibration was performed using a silicone resin matrix multinuclide (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, ^{203}Hg) 0.5 L MB calibration standard produced by Czech Metrology Institute. Efficiency calibration was corrected for each sample using ANGLE ver.3 software (Abbas et al. 2002; Jovanovic et al., 2010; Miller and Voutchkov, 2013). The software uses "efficiency transfer (ET)" principle. The ET factor is the ratio of the actual to reference efficiency at a given gamma-energy calculated based on the geometry and chemical composition of a sample and a reference source. Angle software input for the calibration standard and measured date samples is summarized in Table 1.

The samples were counted for a period of 86,400 s (24 hours) live time, to reduce statistical counting error. For spectrum analysis, ORTEC GammaVision software was used. The γ-ray energies of ^{212}Pb (238.63 keV), ^{208}Tl (583 keV), ^{228}Ac (911 and 969 keV) were used to

determine the concentration of ^{232}Th (^{228}Ra), and ^{214}Bi (609, 1120 and 1765 keV) and ^{214}Pb (295 and 352 keV) were used to determine the activity of ^{238}U (^{226}Ra). The ^{40}K and ^{137}Cs radionuclides were measured from their respective γ-ray energies of 1460 keV and 662 keV. In order to determine the background level in the environment around the detector, an empty 1 L MB was counted under the same conditions to determine the background counts. Several background counts were taken during the course of this study to verify consistency over time. No significant changes in background were observed, hence only one background spectrum (as a representative for all counts) has been used for background correction for activity calculation.

The activity concentration, A (Bq kg^{-1}) of each of the radionuclides in each date fruit sample was calculated using the formula (IAEA 1989):

$$A = \frac{N}{\varepsilon m t P_\gamma}$$

and the critical level for minimum detectable activity (CR_{MDA}) was calculated using the Critical Level ORTEC method of GammaVision software:

$$CR_{MDA} = \frac{2.33 \cdot S_b}{\varepsilon m t P_\gamma}$$

where,

- N is the net counts of the corresponding photopeak,
- ε and P_γ are the detector efficiency and the emission probability per disintegration, respectively, at each specific gamma line,
- m is the mass of the sample in kg,
- t is the live counting time in seconds, and 56
- S_b is the standard error in the net background count rate for the photopeak.

The CR_{MDA} values for all photopeaks are given in Table 2. CR_{MDA} values were calculated for the dry basis activity. Wet (fresh) weight basis activity concentration of date fruit was calculated by multiplying the dry basis activity concentration by a factor derived from the ratio of dry weight to fresh weight, for each date sample.

RESULTS AND DISCUSSION

The dry and wet basis radioactivity concentration data of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in pitted date fruit samples (edible flesh only) are shown in Tables 3 and Table 4. The total uncertainties in all data presented at 2-sigma standard deviation including counting statistics. Out of 22 samples, ^{238}U was found in 4 samples (Dabbas and Khalas type dates) above the average CR_{MDA} value. For ^{238}U, the maximum activity concentrations were found as 0.80 ± 0.74 Bq kg^{-1} (dry basis) and 0.69 ± 0.64 Bq kg^{-1} (wet basis), respectively, in one Dabbas type date sample collected from one garden of Ghayathy area and the minimum activity concentrations were found as 0.61 ± 0.79 Bq kg^{-1} (dry basis) and 0.54 ± 0.69 Bq kg^{-1} (wet basis), respectively, in a Khalas type date sample collected from one garden of Al Silaa area. The average activity for ^{238}U for samples with activities above CR_{MDA} were found as 0.66 ± 1.68 Bq kg^{-1} (dry basis) and 0.57 ± 1.29 Bq kg^{-1} (wet basis).

^{232}Th above CR_{MDA} value was found only in 3 samples on dry and wet basis (in Neghal, Shishi and Rezaiz type date). The maximum and minimum activity concentrations were found as 0.23 ± 0.13Bq kg^{-1} and 0.10 ± 0.11 Bq kg^{-1} in a Neghal type sample collected from one garden of Al Silaa area, for both dry basis (Tables 3 and 4). ^{232}Th in all other date samples were found below detectable limit (BDL).

^{40}K was detected in all date fruit samples ranging from 191 to 362 Bq kg^{-1} with an average 277 ± 38 Bq kg^{-1}, on dry basis and these values ranging from 168 to 283 Bq kg^{-1} with an average 239 ± 33 Bq kg^{-1}, on wet basis. The highest value was found in a sample of a Rezaiz type from one garden of Ghayathy area and the lowest value was found in a Khalas type sample collected from one garden of Al Silaa (Table 3 and 4).

It was observed that the measured ^{40}K activity concentration exceeds remarkably both ^{238}U and ^{232}Th. The frequent presence of ^{40}K and the infrequent presence of ^{238}U and ^{232}Th in all date fruit samples are due to their natural abundance within the UAE environment, mainly presence of these radionuclides in soil where the specific date palm trees grow. It can also be seen in Table 5 that the measured activity concentration data in present study are comparable to soil data from the UAE. Also it should be mentioned that the below detectable limit of ^{238}U and ^{232}Th in date fruits in this study do not indicate their total absence in the samples rather the background levels and the system CR_{MDA} could conceal minor photopeaks associated with these radionuclides. However, the activity concentrations of chosen radionuclides in this study are more or less comparable with other data on date fruits from other places of the world (Alrefae 2015).

CONCLUSIONS

The present study is the first publicly available in the UAE investigation of radioactivity concentration levels in palm date fruits. Long-lived natural and anthropogenic isotopes in palm dates of 10 varieties from 22 different local commercial palm date farms were analyzed using gamma spectrometry. ^{40}K was found in significant levels in all samples whereas ^{238}U and ^{232}Th were found only in few samples with negligible activity concentration levels. No ^{137}Cs or other

anthropogenic isotopes were detected in the present study. Hence, it can be concluded that the activity levels of the detected radionuclides mainly came from natural abundance that exists in UAE environment. Also, the activity concentrations in UAE palm dates were found comparable with activity levels in date fruits as well as with the other countries. The results of this work can be used as baseline reference data for any future investigation and will be included in the radiation baseline data for the UAE environment. However, for a more rigorous baseline assessment, alpha and beta emitting radionuclides should also be investigated as a future study.

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Table

Table 1. Container dimensions used for efficiency correction input for Angle software.

Dimension/Parameter	Calibration Standard Container	Sample Container
Container inside radius, mm	55	62
Marinelli cavity radius, mm	39	40
Marinelli cavity depth, mm	53	76
Marinelli upper bottom thickness, mm	2	2
Marinelli inner side thickness, mm	2	2
Marinelli lower bottom thickness, mm	2	2
Density, g/cm^3	0.985	0.525 - 0.726

Table 2. MDA values for the counting system for each chosen photopeaks for ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs, the corresponding background count and critical limits

Name of Radionuclide	Photopeak (keV)	Background rate (counts d^{-1})	CR_{MDA} [1] values (Bq kg^{-1})	
^{238}U decay series	295	130	0.134	
	352	352	0.115	
	609	322	0.129	
	1,120	12	0.136	
	1,765	95	0.558	
			0.214	**Avg.**
^{232}Th decay series	238	486	0.085	
	583	178	0.049	
	911	126	0.195	
	968	60	0.031	
			0.090	**Avg.**
^{40}K	1,460	407	1.212	
^{137}Cs	662	3	0.009	

[1] CR_{MDA} calculation was done using Critical Level ORTEC method and CR_{MDA} values shown here are the smallest concentration of radioactive material in a sample that yields a net count that will be detected with a 95% probability.

Table 3. Measured concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs (in Bq kg^{-1}, dry basis) in date fruits (flesh only) from different palm date gardens of United Arab Emirates.

Sample No	Location (no of garden)	Type	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	^{137}Cs (Bq kg^{-1})
1	Ghayathy (9)	Rezaiz	BDL	BDL	362 ± 11	BDL
2		Dabbas	0.80 ± 0.74	BDL	293 ± 9	BDL
3		Rezaiz	BDL	BDL	326 ± 10	BDL
4		Neghal	BDL	0.23 ± 0.13	303 ± 9	BDL
5		Dabbas	BDL	BDL	240 ± 7	BDL
6		Khadi	BDL	BDL	269 ± 8	BDL
7		Yardi	BDL	BDL	305 ± 9	BDL
8		Khalas	BDL	BDL	238 ± 7	BDL
9		Neghal	BDL	BDL	294 ± 9	BDL
10	Al Silaa (5)	Zahedy	BDL	BDL	283 ± 9	BDL
11		Shishi	BDL	0.10 ± 0.11	272 ± 8	BDL
12		Rezaiz	BDL	0.13 ± 0.11	330 ± 10	BDL
13		Zahedy	BDL	BDL	250 ± 8	BDL
14		Khalas	0.61 ± 0.79	BDL	191 ± 6	BDL
15	Liwa (7)	Bumaan	BDL	BDL	287 ± 9	BDL
16		Dabbas	BDL	BDL	273 ± 8	BDL
17		Rezaiz	BDL	BDL	270 ± 8	BDL
18		Bumaan	BDL	BDL	301 ± 9	BDL
19		Dabbas	0.61 ± 0.91	BDL	276 ± 8	BDL
20		Khanezi	BDL	BDL	201 ± 6	BDL
21		Khalas	0.61 ± 0.91	BDL	242 ± 7	BDL
22	Shih al Khor (1)	Neghal	BDL	BDL	281 ± 8	BDL

Note: BDL- Below Detection Limit; no in ( ) *indicates no of gardens; 2 sigma total uncertainties are presented in results.*

Table 4. Calculated activity concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs (in Bq kg^{-1}, wet basis) in date fruits (flesh only) from different date palm gardens of United Arab Emirates.

Sample No	Location (no of garden)	Type	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	^{137}Cs (Bq kg^{-1})
1	Ghayathy (9)	Rezaiz	BDL	BDL	283 ± 9	BDL
2		Dabbas	0.69 ± 0.64	BDL	253 ± 8	BDL
3		Rezaiz	BDL	BDL	288 ± 8	BDL
4		Neghal	BDL	0.22 ± 0.13	297 ± 9	BDL
5		Dabbas	BDL	BDL	214 ± 7	BDL
6		Khadi	BDL	BDL	224 ± 7	BDL
7		Yardi	BDL	BDL	255 ± 8	BDL
8		Khalas	BDL	BDL	198 ± 6	BDL
9		Neghal	BDL	BDL	253 ± 8	BDL
10	Al Silaa (5)	Zahedy	BDL	BDL	262 ± 8	BDL
11		Shishi	BDL	0.8 ± 0.09	220 ± 7	BDL
12		Rezaiz	BDL	0.09 ± 0.09	276 ± 8	BDL
13		Zahedy	BDL	BDL	224 ± 7	BDL
14		Khalas	0.54 ± 0.69	BDL	168 ± 5	BDL
15	Liwa (7)	Bumaan	BDL	BDL	254 ± 8	BDL
16		Dabbas	BDL	BDL	228 ± 7	BDL
17		Rezaiz	BDL	BDL	233 ± 7	BDL
18		Bumaan	BDL	BDL	260 ± 8	BDL
19		Dabbas	0.55 ± 0.81	BDL	247 ± 7	BDL
20		Khanezi	BDL	BDL	174 ± 5	BDL
21		Khalas	0.52 ± 0.69	BDL	206 ± 6	BDL
22	Shih al Khor (1)	Neghal	BDL	BDL	248 ± 7	BDL

Note: BDL- Below Detection Limit; no in ( ) *indicates no of gardens; 2 sigma total uncertainties are presented in results.*

Table 5. Comparison of measured ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs (in Bq kg^{-1}) in palm dates with other foodstuffs from other reports including palm dates and soil data from the UAE.

Sample type	Origin	Sample condition	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	^{137}Cs (Bq kg^{-1})	Reference
Palm dates	UAE	dry	0.61– 0.80	0.10 – 0.23	191-362	BDL	Present study
		wet	0.52- 0.69	0.11 – 0.23	168 - 283	BDL	
	UAE	dry	-	0.31 – 0.62	245 - 302	-	Tareq Alrefae (2015)
	Saudi Arabia		0.47 – 1.6	0.23 – 1.1	236 – 417	-	
	Tunisia		1.12	0.35	305	-	
	Jordan		-	0.34	293	-	
	Libya		-	0.38	283	-	
	Iran		0.80	0.53	296	-	
	Pakistan		-	0.38	249	-	
	India		1.90	0.72	276	-	
Soil	UAE	dry	9 – 35	1.39 – 5.35	45 – 215	-	Abdulla MAA, MSc Thesis (2015)