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Concentrations and Estimation of the Annual Effective Doses of Natural Radioactivity Uptake through Ingestion of Virgin and Extra Virgin Olive Oil

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Abstract: This study assesses the natural radioactive concentrations in virgin olive oil in Egypt. The presence and concentrations of naturally occurring radioactive materials (NORM) in 26 olive oil samples collected from different locations in Saini were conducted using two techniques: gamma spectroscopy using a high purity germanium detector (HPGe) and inductively coupled plasma-mass spectroscopy (ICP-MS). Because of their very low concentrations, the radioactivity of NORM was below the detection limits of the HPGe detector for the time of measurement, On the other hand ICP-MS results showed the concentrations of natural radioactive elements such as uranium and thorium in some examined samples. The concentrations of uranium and thorium elements were converted into specific activities using conversion factors recommended by the International Atomic Energy Agency (IAEA). The internal effective dose for individuals from the consumption of olive oil was estimated based on the calculated radionuclide contents and using dose coefficients given by the International Commission on Radiological Protection (ICRP). It was found the olive oil samples are considered radiologically safe for human consumption and don't contribute to any health problems.

Keywords: Olive oil, ICP-MS, annual effective dose, Uranium, Thorium.

1 Introduction

Naturally occurring radionuclides of terrestrial origin (also called primordial radionuclides) are present in varying levels in all media in the environment as well as in the human body itself. Only those radionuclides with half-lives comparable to the age of the Earth and their decay products are present in significant quantities in these materials. These radioactive radionuclides are present in the soil, water, and air, and their abundance changes depending on the geological and geographical features of the origin. ^{238}U and ^{232}Th series, as well as ^{40}K, are the major sources of natural external irradiation of the human body. These radionuclides are also found in the body and irradiate the various organs with alpha and beta particles, as well as gamma rays, as a result of the consumption of foods and water [1]. Due to their presence in soil and phosphate fertilizers, primordial radionuclides and their progeny are Transmitted by water from the soil to olive tree roots, olives, and olive oil. It is necessary to measure the radionuclide concentrations of olive oil samples to assess potential radiation doses, and, if necessary, to take steps to protect consumers from radiation exposure [2]. One of the most prominent ingredients included in the diet is extra virgin olive oil (EVOO), which is characterized by its health benefits[3]. Olive oil is a fat that is widely used in pharmaceuticals, cosmetics, and cooking. Olive oil is popular in cooking due to its cholesterol-lowering effect. Unlike animal fats, which have a cholesterol effect on humans [4]. uranium (^{238}U), thorium (^{232}Th), radon (^{222}Rn), and (^{220}Rn) contents were detected in various locally produced olive oil samples were collected in rural areas of Morocco by using CR-39 and LR-115 type II solid-state nuclear track detectors (SSNTDs) [2]. uranium (^{238}U), uranium (^{234}U) were detected by using high purity germanium (HPGe) and also Polonium (^{210}po) using Passivated Implanted Planar Silicon (PIPS) detectors in a

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CANBERRA alpha spectrometer in various locally produced olive oil samples collected in New Zealand [5]. Gross beta radioactivity measurements were detected by the Krieger method using a proportional gas flow counter (Ortec Telenet Systems) and also the activity concentration of ^{40}K was detected by using high purity germanium (HPGe) in various locally produced olive oil samples collected in Turkey [6]. The activity concentration of ^{40}K was detected by using high purity germanium (HPGe) in various locally produced olive oil samples collected in the KSA and Palestine [7]. Egypt is one of the largest producers and consumers of olive oil. However, there has been no report, to our knowledge, on the natural radioactive concentrations in Egyptian olive oil. The main objective of this study was to determine the natural radioactive concentrations of olive oil consumed in Egypt and estimate potential radiation doses since olive oil is an essential component in our daily life.

2 Materials and Methods

2.1 Sample Collection

26 samples of virgin and extra virgin olive oil 10 samples of them were collected locally from different locations in Sinai, as it is famous for being the largest producing and exporting city for olive oil. 16 samples of them were purchased from the most widely accepted and most frequently consumed brands from Egyptian supermarkets, and their origin is Saini. The naturally radioactive concentrations in olive oil deserve importance the study because several studies indicated that soils in the Sinai region have high background radiation and the naturally radioactive concentrations exceed the permissible values such as [8,13]. since no studies have been conducted to estimate the natural radioactive concentrations in Egyptian olive oil Especially that obtained from olive trees grown in Sinai.

2.2 Experimental Design

2.2.1 Gamma Spectroscopy

Gamma peak analysis was performed on 150 ml individual samples of olive oil using Broad energy HPGe gamma spectroscopy BE3830 model with cryostat CP5-PLUSE- SL and iPA-Sl preamplifier. FWHM: 450 eV at 5.9 keV, FWHM:750 eV at 122keV, FWHM, 1900 eV at 1332.5 keV) with a relative efficiency of 30% and shielded with Lead to minimize background. The detector is operated by Geine 2000 software and calibrated mathematically using LABSOCS based Monte Carlo software. Samples were counted for 86400 seconds.

2.2.2 ICP-MS

0.5 g of olive oil samples were mixed with 5 ml of HNO3 and 5 ml of H2O2 then boiled for 10 h in covered beakers on a hot plate (until brown vapors stopped). All samples were filtered and transferred into 15 ml volumetric tubes and filled to the mark with deionized water. Prior to analysis, samples were split into two equal volume sets. The first set had the original samples preparation dilution; while, in the second set samples were further serial diluted up to 300 times the original concentration. All samples were analyzed for the presence of uranium and thorium by an iCAP triple-quadrupole inductively coupled plasma mass spectrometry system from Thermo Scientific Inc. and the Thermo Scientific software program Qtegra Intelligent Scientific Data Solution.

3 Results and Discussion

No data were obtained from the gamma peak search analysis after subtraction of the ambient background after 24-hour count for individual samples.

Table 1: Concentrations of investigated Uranium and Thorium in olive oil samples understudy in ppm± SD.

	Natural Radio Active Element	
Label	^{238}U [ppm]	^{232}Th [ppm]
1	0.23±0.01	0.13±0.01
2	0.05±0.01	N.D.
3	0.29±0.01	0.06±0.01
4	0.1±0.00	N.D.
5	0.05±0.00	N.D.
6	0.05±0.00	N.D.
7	0.04±0.00	N.D.
8	0.51±0.01	N.D.

9	0.03±0.00	N.D.
10	0.1±0.01	0.7±0.17
11	0.06±0.00	0.07±0.01
12	0.05±0.00	0.01±0.01
13	0.03±0.00	0.02±0.01
14	0.05±0.01	N.D.
15	0.06±0.01	N.D.
16	0.02±0.00	N.D.
17	0.02±0.00	N.D.
18	N.D.	N.D.
19	0.04±0.00	N.D.
20	0.01±0.00	N.D.
21	0.25±0.00	N.D.
22	0.02±0.00	N.D.
23	0.05±0.00	0.39±0.01
24	0.02±0.00	N.D.
25	0.01±0.00	N.D.
26	0.04±0.00	N.D.

As shown in table1, some variations in concentrations of Uranium and Thorium in investigated samples; U was detected in most of the samples but varied from one sample to another. Several studies have determined the radioactivity contents of various foodstuffs in different parts of the world. In the Morocco study, the uranium and thorium contents were higher than our values [2]. The variations in the uranium and thorium concentrations could be due to differences in the geological location of the plants and the radiochemical composition of the soils in which olive trees are grown. The variations in the uranium and thorium concentrations could be due to differences in the geological location of the plants and the radiochemical composition of the soils in which olive trees are grown. It is necessary to assess the natural radioactive concentrations, specially the uranium and thorium contents. Most uranium is eliminated within a few days of ingestion and never enters the bloodstream. The small fraction that is absorbed into the bloodstream (0.2 to 5%) is deposited preferentially in bone (about 22%) and kidneys (about 12%), with the rest being dispersed throughout the body (12%) and eliminated. Most of what goes to the kidneys leaves within a few days (in urine), while that deposited in the bone can remain for many years. Only a small fraction of thorium inhaled or ingested in food and water is absorbed into the bloodstream, and most of it is eliminated within a few days. Internally deposited thorium in the general population comes primarily via gastrointestinal absorption of food or water. Through the intestines, about 0.02 to 0.05% of the quantity ingested is absorbed into the bloodstream. About 70% of the amount entering the blood is retained in bone, where it has a biological half-life of about 22 years, 4% is retained in the liver, where it has a biological half-life of 700 days, and 16% is uniformly distributed to all other organs and tissues of the body, where it has a biological half-life of 700 days. The majority of the remaining 10% is excreted immediately. Thorium is deposited primarily on mineral bone's endosteal surfaces and only slowly redistributes throughout the bone volume [14] Large amounts of these radionuclides deposited in specific organs will influence and deteriorate a person's health by weakening the immune system, causing numerous diseases, and ultimately raising mortality rates [15]. The investigation of the level of natural radionuclides in the environment specially in foodstuffs is important wherever elevated radiation background exists. Therefore, measuring natural radiation background levels has increased as a result of the recognition that nature is one of the largest sources of radiation exposure for the general public [16].

Specific activity in (Bq/kg) can be calculated from measured radioelement concentrations of uranium (in ppm) and thorium (in ppm) using conversion factors given by the International Atomic Energy Agency [17] ; where 1 ppm U= 12.35 Bq/kg from U238 and 1 Th ppm = 4.06 from Th 232 Bq/kg.

As shown in table 2, Radioactivity concentrations for all samples were carried out using conversion factors to evaluate the internal dose. Many researchers have studied the existence of radionuclides and the annual ingestion effective dose in different foodstuffs used in Egypt using different techniques. Such as radon concentrations and effective radium content were measured by using the CR-39 polymer track detector. Also, the annual effective dose was determined for different types of household foods used in Egypt [18]. The annual effective dose was calculated from the corresponding radon concentration using Solid State Nuclear Track Detectors (CR-39) of different types of salt collected from different locations of local markets in Egypt [19]. The concentrations of ^{137}Cs and ^{40}K in some Egyptian foodstuffs were measured by using HPGe detectors [20]. The concentration of ^{210}Po was measured using the alpha spectrometry technique and the effective ingestion dose was determined in different foodstuffs of plant origin purchased from markets in Qena City, Upper Egypt [21]. the levels of naturally occurring radionuclides, Cs-137 were measured by using HPGe- detector also annual effective dose was determined in herbal plants used in Egypt [22]. natural uranium-238 and thorium -232 together with their decay products were determined as well as K-40. Also, Cs-137 radiation level was determined in different environmental samples of plants in the Inshas Region and the Area Nearby in Egypt [23]. Radionuclides ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were measured by using HPGe detector and the annual effective dose was determined in foodstuffs in the Alexandria Region, Egypt [24]. It is worth noting that this is the first study of natural radionuclides in olive oil obtained from Sinai in Egypt.

The radiation-induced health effects linked with radionuclide consumption in the body are proportional to the overall dose provided by the radionuclides while resident in the various organs, according to estimates. The ingestion of radionuclides through food is one of the most important pathways, accounting for a significant portion of the average radiation doses to the body's numerous organs, which was evaluated by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)[25]. Therefore, it is very important to know the effective dose resulting from exposure to natural radionuclides such as ^{238}U and ^{232}Th in olive oil which is a significant part of a person's diet.

Table 2: specific activity (Bq/kg) of ^{238}U and ^{232}Th in olive oil samples**.**

	Natural Radio Active Element	
Label	^{238}U [Bq/kg]	^{232}Th [Bq/kg]
1	2.8±0.12	0.53±0.04
2	0.61±0.12	N.D.
3	3.56±0.12	0.25±0.04
4	1.23±0.00	N.D.
5	0.61±0.00	N.D.
6	0.61±0.00	N.D.
7	0.49±0.00	N.D.
8	6.27±0.12	N.D.
9	0.36 ±0.00	N.D.
10	1.2±0.12	2.9±0.69
11	0.73 ±0.00	0.28±0.04
12	0.61 ±0.00	0.04±0.04
13	0.36±0.00	0.08±0.04
14	0.61±0.12	N.D.
15	0.73±0.01	N.D.
16	0.24±0.00	N.D.
17	0.24±0.00	N.D.
18	N.D.	N.D.
19	0.49±0.00	N.D.
20	0.12±0.00	N.D.
21	3±0.00	N.D.

Table 2: (continued: specific activity (Bq/kg) of ^{238}U and ^{232}Th in olive oil samples**.**

	Natural Radio Active Element	
Label	^{238}U [Bq/kg]	^{232}Th [Bq/kg]
22	0.24±0.00	N.D.
23	0.61±0.00	1.6±0.04
24	0.24±0.00	N.D.
25	0.12±0.00	N.D.
26	0.49±0.00	N.D.

The annual effective dose from ingestion (Dint(ing)) can be calculated using the expression [26].

Dint(ing) (pSv/y) = CR × IR × IT × DCFing,

Where;

Dint(ing) is the annual effective dose due to ingestion of radionuclides through food (pSv y−1)

CR is measured activity concentration of ingested radionuclide R in Bq/g,
IR Inadvertent Ingestion rate g/h,
IT Time of Ingestion rate h/y,
DCFing is the ingestion dose conversion factor in Sv/Bq, as shown in table3
Assuming that average consumption of olive oil is 20g/day [28, 29], the calculated annual doses of ^{238}U and ^{232}Th are shown in table4.

Table 3: Effective Dose Coefficient for Ingestion (Sv/Bq) [27].

Nuclide	$t_{1/2}$	fi	Adult
238U	4.468×10^{9}	0.02	4.5×10^{-8}
^{232}Th	1.405×10^{1}	0.0005	2.3×10^{-7}

Table 4: the annual effective dose due to ingestion of radionuclides through food Dint(ing) (pSv y−1) for adults.

	Natural Radio Active Element	
Label	^{238}U [pSv y−1]	^{232}Th [pSv y−1]
1	0.0197	0.0017
2	0.0033	N.D.
3	0.0230	0.0008
4	0.0099	N.D.
5	0.0033	N.D.
6	0.0033	N.D.

Table 4: (continued): the annual effective dose due to ingestion of radionuclides through food Dint(ing) (pSv y−1) for adults.

	Natural Radio Active Element	
Label	^{238}U [pSv y−1]	^{232}Th [pSv y−1]
7	0.0030	N.D.
8	0.0329	N.D.

9	0.0026	N.D.
10	0.0066	0.0126
11	0.0033	0.0008
12	0.0033	0.0002
13	0.0023	0.0003
14	0.0033	N.D.
15	0.0066	N.D.
16	0.0016	N.D.
17	0.0013	N.D.
18	0.0000	N.D.
19	0.0030	N.D.
20	0.0010	N.D.
21	0.0197	N.D.
22	0.0016	N.D.
23	0.0033	0.0042
24	0.0016	N.D.
25	0.0010	N.D.
26	0.0033	N.D.

Since Dint(ing) determines how unsafe an individual's exposure to radiations. it is obvious from the determined values of Dint(ing) values, shown in table4, for 238U and 232Th in olive oil samples are much less than the Worldwide average annual effective dose value of 300 µSv.y-1 as reported in United Nations Scientific Committee on the Effect Atomic Radiations (UNSCEAR) [25] and may not have diverse health effects if used for long periods of time.

4 Conclusions

Two different techniques have been used to determine the concentrations of 238U and 232Th in olive oil samples in Egypt, HPGe and ICP-MS. The HPGe requires more than 24 hours to efficiently detect the gamma activity for the very low concentrations of 238U and 232Th. On the other hand, the ICP-MS technique was able to efficiently and rapidly determine the concentration of ions238U and 232Th. If used for quality assurance in the olive oil production process, we recommend the application of the ICP-MS technique because of its advantage of being sensitive and rapid. The variation in our concentrations may be due to geological and environmental factors. The annual effective doses from the Egyptian olive oil samples are safe in terms of the radiological hazard, as per international standards.

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