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Measurement of radioactivity levels of liquid milk samples in Kütahya, Turkey

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► ***Short report***

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ABSTRACT

Background: Milk is a fundamental part of the human daily diet, so radioactivity concentrations in milk must have been controlled regularly. In this study, radiation activities were measured in 12 ultra-high temperature (UHT) milk samples collected in Kütahya, Turkey, in August 2021. ***Materials and Methods***: Radium (Ra-226), thorium (Th-232) and potassium (K-40) values were determined by using the Canberra 3x3 NaI (Tl) gamma spectroscopy method in 8 samples, and Radon (Rn-222) activity measurements have been conducted in 12 samples using the E-perm method. ***Results***: Radium (Ra-226), thorium (Th-232) and potassium (K-40) levels are measured as < MDA (2.28 Bq/kg), < MDA (2.53 Bq/kg), 40.43 ± 2.25 to 53.31 ± 2.30 Bq/kg, respectively. Radon (Rn-222) concentrations were determined by the E-perm system between 0.46 ± 0.02 and 2.01 ± 0.05 Bq/l with an average of 1.11 ± 0.03 Bq/l. ***Conclusions***: Annual ingested radiation doses were calculated using radioactivity levels for different age groups as a result of milk consumption. Ingestion dose levels are below the worldwide average values given in the UNSCEAR report. It has been observed that the results are below the safety limits.

INTRODUCTION

All radionuclides, artificial or natural, impact human life, and it is necessary to evaluate radionuclides in the view of radiologically. There are several pathways, for instance, ingestion of foods, consumption of water and air breathing to be transferred radioactive to living organisms. Man-made radioactivity contributes an additional dose to the environment, while the main contribution is caused by natural radioactivity. The mean effective dose is about 2.4 mSv annually as a result of natural radioactivity [1]. The earth's crust is made up with heavy elements, which are transported by geological phenomena to the earth's surface, so these heavy elements are the origin of natural radioactivity. These elements, which are known as naturally occurring radioactive material, are uranium (U-238), thorium (Th-232), their decay products and potassium (K-40). They are found in every strata of the earth's crust and their concentrations in soil, sand and rocks depend on the geological specifications of the location and vary with geographical areas. The transfer of these elements into biological systems such as plants, animals, water and air contributes significantly to the ingestion dose. Finally, natural radioactivity reaches humans through the soil-grass-cow-milk pathway [2].

Milk contains protein, carbohydrates, fat, several vitamins and minerals, so it is a crucial part of the human daily diet. Ultra-high temperature (UHT) milk is the most consumed milk type because of its long storage life [3].

A total of 1.5 million tons of cow's milk were produced in 2021, in Turkey [4]. Milk and dairy products are consumed in the country and exported abroad.

There are some studies for measurements of radionuclide activity in milk samples around the world. A literature summary is presented in table 1.

Table 1. Some study results for radionuclide concentrations in milk reported around the world.

Location	Ra-226 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/kg)	Rn-222 (Bq/l)
Iraq [5]	-	-	-	7.53-4.56
Brazil [6]	0.005 - 0.048	0.012-0.153	-	-
Brazil [7]	0.004-0.5 (Bq/l)	-	-	-
Tunisia [80]	-	-	11.1 - 58.5	-
Bulgaria [9]	1.63 (Bq/l)	0.05 (Bq/l)	53.1 (Bq/l)	-
Vietnam [2]	1.45 - 2.45	0.60 - 1.21	341 - 387	-
Turkey [10]	3.2 (Bq/l)	1.1 (Bq/l)	31.3 (Bq/l)	-
Nigeria [11]	16.6	10.6	317.6	-
Iraq [12]	0.01, 0.19	-	-	-

In this study, Rn-222 and K-40 radioactivity levels of 12 UHT milk samples collected in Kütahya, Turkey in August 2021 were investigated using NaI(Tl) and E-Perm system. This study is important in terms of expanding the database, as the number of radiometric studies and the database are limited in the country.

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MATERIALS AND METHODS

Sample collection and preparation

The E-Perm system was used for determining Rn-222 concentrations and the NaI(Tl) gamma spectroscopy system was used for determining Ra-226, Th-232 and K-40 concentrations. 12 different brands of UHT cow milk were purchased from markets in Kütahya, in August 2021. Firstly, Rn-222 activities of 12 milk samples were measured. After that, only 8 of 12 brands with the same manufacturing date were found and bought in the markets to perform gamma spectroscopy measurements. This situation was caused by pandemic conditions at the time, and products from these brands temporarily ran out during sampling. Samples were brought to the laboratory and labeled.

Experimental

Radon measurements

136 ml volume milk samples were used for radon measurements with the E-Perm method. A short-term electret (ST) was placed in the S-chamber to measure Rn-222 in milk. This chamber was placed just below the cover of the jar so that it didn't come into contact with the milk to be measured. Rn-222 transferred from the milk in the jar enters the S-chamber and performs radioactive decay. The initial voltages of electrets were measured. The cover of the jar was tightly closed to prevent the escape of radon. The chambers were exposed to Rn-222 for three days. Finally, the electrets were removed from the S-chambers and final voltages were noted. Two experiments were performed simultaneously for each sample and the results were averaged [13].

Gamma spectroscopy measurements

For gamma spectroscopy measurements, 8 of 12 milk samples with a 1 liter volume were transferred to 1000 ml marinelli beakers to keep for 30 days to provide radioactive equilibrium between U-238, Th-232 and their daughter products. The covers of beakers were tightly closed [14].

3×3 inch NaI(Tl) gamma spectroscopy system, Canberra with DSA-1000 multichannel analyzer equipped with Genie-2000 spectroscopy software, located at Nuclear Physics Laboratory at Dumlupınar University, Kütahya, was used to measure the radioactivity levels of milk samples.

The detector is shielded with 1 cm of copper and 5 cm of lead to reduce the background radiation. It has a 6.8% resolution for the 1332 keV gamma ray of cobalt (Co-60) source.

A soil sample with a density of 1.3 g/cm^3 in a 1000 ml marinelli beaker prepared by Turkish Energy Nuclear and Mineral Research Agency Nuclear Energy Research Department (TENMAK-NÜKEN) was used as energy and efficiency calibration source. The activities of the source are 22 ± 3; 28 ± 3 and 375 ± 60 Bq/kg for Ra-226, Th-232 and K-40, respectively.

The energy values used to detect the K-40, Ra-226 and Th-232 in this study are 1460 keV (K-40), 1764 keV (Bi-214) and 2614 keV (Tl-208), respectively [14]. Samples and background were counted for 20000 s, and background measurements were repeated daily using ultra-pure water.

Ra-226, Th-232 and K-40 activities of the sample were calculated as given in the literature (14). The self-absorption correction factor was used as given in Jodlovski [15].

MDA (Minimum Detectable Activity) was calculated as 6.94, 2.28 and 2.53 Bq/kg for K-40, Ra-226 and Th-232, respectively, for the live time of 20000 s [14].

Radon calculations

RnC is the average Rn-222 concentration in the air inside the jar measured by the E-Perm system. RnC was calculated with equation 1 in the units of pCi/l:

$$RnC = \left[\left(\frac{(IV-FV)-(0.067 \times TA)}{\left(A+B\times\left(\frac{IV+FV}{2}\right)\right)\times TA}\right) - (BG \times G)\right] \tag{1}$$

here, IV and FV are the initial and final electret voltages, TA is the analysis time in days, A and B are equal to 1.69776 and 0.0005742, respectively for SST type chamber configuration, BG is the background gamma radiation in the laboratory which is measured using a Ludlum 44-2 handheld detector, G is the gamma conversion constant.

Rn_{milk} is the dissolved Rn-222 concentration in the milk calculated by equation 2 in the unit of pCi/l:

$$Rn_{milk} = \frac{RnC \times e^{-\lambda \times TD} \times \left(\frac{V_A}{V_m}+OC\right) \times \lambda \times TA}{\left(1-e^{-\lambda \times TA}\right)} \tag{2}$$

In equation 2, TD is the delay time in days, V_A is the volume of air in the jar in liters, V_m is the volume of milk in liters, OC is Ostwald Coefficient which is 0.2593 at 20 °C, λ is the decay constant for Rn-222.

Lower Limit of Detection (LLD) was determined between 0.15 and 0.27 Bq/l for the radon concentration for 3 days measurements [13].

Annual effective ingestion dose for different age groups

Annual effective ingestion dose, D in the units of μSv/y, due to the consuming of milk can be calculated by equation 3:

$$D = A \times I \times DCF \tag{3}$$

Where A is the activity of the milk sample in units of Bq/kg, DCF is the dose conversion factor for radon as 3.5 nSv/Bq [1, 16]. DCF is taken as 42 nSv/Bq, 13 nSv/Bq, 6 nSv/Bq for potassium for infants, children and adults, respectively [2]. I is the annual intake of milk assuming 120, 110 and 105 kg for infants, children and adults, respectively.

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Statistical analysis

Statistical analysis was performed with R code version 4.2.2 and Microsoft Excel version Professional Plus 2016 for Windows. The p-values for Rn-222 and K-40 activity results were calculated as <0.05.

RESULTS

Radionuclide concentrations of milks sampled in August 2021 in Kütahya, are presented in Table 2. Ra-226 and Th-232 activities are below the MDA. K-40 activities vary between 40.43 ± 2.25 and 53.31 ± 2.30 Bq/kg with an average of 45.49 ± 2.27 Bq/kg. Rn -222 activities vary between 0.46 ± 0.02 and 2.01 ± 0.05 Bq/l with an average of 1.11 ± 0.03 Bq/l. The highest and lowest K-40 concentrations were found on Sample 4 and 6, respectively. The highest and lowest Rn-222 concentrations were found in Sample 3 and 7, respectively.

Table 2. K-40 and Rn-222 activities in milk samples.

Sample Number	K-40 (Bq/kg)	Rn-222 (Bq/l)
1	43.27 ± 2.29	1.11 ± 0.04
2	47.79 ± 2.28	0.61 ± 0.02
3	45.56 ± 2.27	2.01 ± 0.05
4	53.31 ± 2.30	0.96 ± 0.03
5	46.55 ± 2.27	0.92 ± 0.03
6	40.43 ± 2.25	0.92 ± 0.03
7	42.00 ± 2.25	0.46 ± 0.02
8	45.01 ± 2.27	1.15 ± 0.04
9	NM**	1.77 ± 0.05
10	NM**	0.59 ± 0.02
11	NM**	0.96 ± 0.03
12	NM**	1.79 ± 0.05
Average	**45.49 ± 2.27**	**1.11 ± 0.03**

One sample t-test was performed for K-40 and two sample t-test was performed for Rn-222 activities. Results were showed the p-value were <0.05. *MDA: Minimum detectable activity, **NM: Not measured.

Annual effective ingestion dose results are given in table 3. The average annual effective ingestion dose results caused by Rn-222 activities are calculated as 0.46 µSv/y, 0.43 µSv/y and 0.41 µSv/y for infants, children and adults, respectively. The average dose results from K-40 in milk samples are calculated as 229.27 µSv/y, 65.05 µSv/y and 29.61 µSv/y for infants, children and adults, respectively.

Table 3. Annual effective ingestion dose results for different age groups (DRn is the ingestion dose from radon activity and DK is the ingestion dose due to potassium activity in the milk samples).

	infant	child	adult	infant	child	adult
Sample Number	**D_{Rn} (µSv/y)**	**D_{Rn} (µSv/y)**	**D_{Rn} (µSv/y)**	**D_K (µSv/y)**	**D_K (µSv/y)**	**D_K (µSv/y)**
1	0.47	0.43	0.41	218.07	61.87	28.17
2	0.26	0.24	0.22	240.84	68.33	31.11
3	0.84	0.77	0.74	229.62	65.15	29.66
4	0.40	0.37	0.35	268.67	76.23	34.70
5	0.39	0.36	0.34	234.63	66.57	30.31
6	0.39	0.36	0.34	203.77	57.81	26.32
7	0.19	0.18	0.17	211.70	60.07	27.35
8	0.48	0.44	0.42	226.84	64.36	29.30
9	0.74	0.68	0.65	NC*	NC*	NC*
10	0.25	0.23	0.22	NC*	NC*	NC*
11	0.40	0.37	0.35	NC*	NC*	NC*
12	0.75	0.69	0.66	NC*	NC*	NC*
Average	0.46	0.43	0.41	229.27	65.05	29.61
(*NC: Not calculated)						

DISCUSSION

UNSCEAR reported the worldwide average Ra-226, Th-232 concentrations in milk as 5 Bq/kg, 0.3 Bq/kg, respectively [1]. In this work, Ra-226 and Th-232 activities in milk samples were measured below the MDA. There is no safety limit specified by any health organization for K-40 activity in milk in the literature. There is no correlation between Rn-222 and K-40 activities.

The Rn-222 activity results of milk samples measured in this study are lower than the results in Iraq [5] which were the only results of radon in milk. K -40 activity results given in this study are lower than those in Nigeria [11], Vietnam [2] and Bulgaria [9]. Our results are higher than the activity of Tunisia [8] and the another study performed in Turkey [10].

Average world-wide exposure from ingestion of foodstuff is 290 µSv/y for uranium, thorium series and potassium [1]. Intake of milk doses given in Table 3 are below than the world-wide average.

It has also been calculated ingestion dose caused by consuming milk for different age groups. It can be seen that infants were exposed to the highest dose; this is related to high consumption of milk.

Results show there is no significant contribution to the total radiation dose. Activity and ingestion dose results are below the world-wide average given by UNSCEAR.

CONCLUSION

The radioactivity content of UHT milk samples in Turkey was investigated by two different methods. E-Perm was used to determine Rn-222 concentration and NaI(Tl) gamma spectroscopy was used to determine Ra-226, Th-232 and K-40 concentrations in milk samples.

Turkey is one of the biggest producers and consumers of milk and milk products in the region, although there is insufficient data in this area related to radiometry. In view of this purpose, this study contributes to developing a database for radioactivity in UHT milk samples in Turkey. It can be referenced for future studies to evaluate radioactivity in milk in this region.

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