Araştırma Makalesi **BAUN Fen Bil. Enst. Dergisi, 19(2), 169-176, (2017)**

DOI: 10.25092/baunfbed.340608 **J. BAUN Inst. Sci. Technol., 19(2), 169-176, (2017)**

Radioactivity concentrations of the milk and dairy products

Yusuf AĞUŞ*

TAEK, Sarayköy Nükleer Araştırma ve Eğitim Merkezi, 06983, Saray, Ankara, Turkey.

Geliş Tarihi (Recived Date): 30.05.2017
Kabul Tarihi (Accepted Date): 15.09.2017

Abstract

Natural and artificial radioactivity concentrations of the white cheese, whey powder, milk powder, labaneh, kaskhaval cheese and milk were determined via gamma spectrometric method. For this purpose, p-type 110% relative efficiency coaxial HPGe detector was used. Average ^{40}K radioactivity concentrations of the white cheese, whey powder, milk powder, labaneh, kaskhaval cheese and milk were found to be 63.1 ± 7.1, 392.6 ± 40.2, 482.1 ±38.6, 152.0 ±12.1, 48.7 ± 5.8 and 52.2 ± 4.2 Bq/kg respectively. ^{137}Cs radioactivity concentrations were found to be below MDA. Except the milk powder ^{232}Th radioactivity concentrations were found to be below MDA. ^{232}Th radioactivity concentrations of the milk powder samples were found to be ranged from 3.2 ± 0.3 - 9.8 ± 0.8 Bq/kg. ^{226}Ra radioactivity concentrations were ranged from 5.9 ± 0.5 - 8.3 ± 0.7 Bq/kg for whey powder, 5.1 ± 0.5 – 9.8 ± 0.8 Bq/kg for milk, 4.6 ± 0.5 - 9.9 ± 1.9 Bq/kg for white cheese, 3.5 ± 0.3 - 16.0 ± 1.5 Bq/kg for milk powder, 4.2 ± 0.3 - 11.1 ± 1.0 Bq/kg for labaneh.

Keywords: *Gamma spectrometry, radioactivity concentration, milk, dairy products.*

Süt ve süt ürünlerinin radyoaktivite konsantrasyonları

Özet

Beyaz peynir, peynir altı suyu tozu, süt tozu*, labne, kaşar peyniri ve sütün doğal ve yapay radyoaktivite konsantrasyonları gama spektrometrik metot ile belirlendi. Bu amaçla, p-tipi %110 rölatif verimli eş eksenli HPGe dedektör kullanıldı. Beyaz peynir, peynir altı suyu tozu, süt tozu, labne, kaşar peyniri ve sütün ^{40}K ortalama radyoaktivite konsantrasyonları sırasıyla 63.1 ± 7.1, 392.6 ± 40.2, 482.1 ±38.6, 152.0 ±12.1, 48.7 ± 5.8 ve 52.2 ± 4.2 Bq/kg dir. ^{137}Cs radyoaktivite konsantrasyonları MDA altında bulunmuştur. Süt tozu dışında ^{232}Th radyoaktivite konsantrasyonları MDA altında*

* Yusuf AĞUŞ, yusufagus06@gmail.com, http://orcid.org/0000-0003-1436-9928

bulunmuştur. Süt tozu ^{232}Th radyoaktivite konsantrasyonu 3.2 ± 0.3 - 9.8 ± 0.8 Bq/kg aralığında bulunmuştur. ^{226}Ra radyoaktivite konsantrasyonları peynir altı suyu tozu için 5.9 ± 0.5 - 8.3 ± 0.7 Bq/kg, süt için 5.1 ± 0.5 – 9.8 ± 0.8 Bq/kg, beyaz peynir için 4.6 ± 0.5 - 9.9 ± 1.9 Bq/kg, süttozu için 3.5 ± 0.3 - 16.0 ± 1.5 Bq/kg, labne için 4.2 ± 0.3 - 11.1 ± 1.0 Bq/kg aralığında bulunmuştur.

Anahtar Kelimeler: *Gama spectrometri, radyoaktivite konsantrasyonu, süt, süt ürünleri.*

1. Introduction

Human being may exposure to radiation in various ways [1]. Inhalation, digestion and external exposures are the main ways. Radionuclides in the foodstuff pass to the human body via digestion. Therefore the knowledge of radioactivity concentrations and radionuclides in the foodstuff are important for determination of the effects of radiation due to both natural and artificial radioactivity. The knowledge of radioactivity concentrations and radionuclides in the foodstuff are important for some certain reasons. The main reason is the assessment of public exposure dose rates, health risks and keeping reference data records of the environmental radioactivities.

Main radionuclides in the foodstuffs are naturally occurring radionuclides and artificial radionuclides. ^{238}U series , ^{232}Th series and ^{40}K are naturally occuring radionuclides (NORM) and sources of the radiation on earth. ^{137}Cs is artificial radionuclide and has 30.05 years half-life [2,3]. As a result of the nuclear weapon tests and nuclear accidents ^{137}Cs occurs. Depending on geological and geographical conditions, the natural environment radioactivity changes and appears at various levels in the soil of different region in the world. Although the activity levels of naturally occurring radionuclides generally at low levels, activity concentrations of natural radionuclides in soil may reach elevated levels with industrial activities [4]. Additionally, due to the effect of fertilizers, the natural radionuclide activity concentrations of soils and plants are higher in cultivated farming areas [5].

Natural and artificial radionuclides in soil pass into plants. Initial and significant step of radionuclide transfer from soil to the plant in the food chain is root uptake [6]. As a part of the food chain, radionuclides pass to the livestock through eating plants [7]. Radionuclide in the livestock passes to the milk and dairy products. Therefore determination of the level of radioactivity concentrations in milk and dairy products is important to ensure consumer safety.

Milk and dairy products are important components of human diet. According to the Turkish Statistic Institute (TÜİK) milk, cheese, milk powder and whey powder production in Turkey is 1.325.548, 631.085, 128.868 and 668.145 MT at 2014 respectively [8]. Determination of radioactivity concentrations will provide meaningful information that can contribute to the knowledge of population exposure and to the keeping reference data record. According to European Council Regulation No 737/90 for the reported radionuclides, the limit values for ^{137}Cs activity are 370 Bq/kg for milk and dairy products [9]. Additionally, the annual effective ingestion dose level is 1 mSv/y for the public.

The aim of this work is to determine natural and artificial radioactivity concentrations of the milk and dairy products. For this purpose, natural and artificial radioactivity concentrations of the milk and dairy product samples which all of those produced in Turkey were measured.

2. Materials and methods

2.1. *Sampling and sample preparation*

Different brand milk and dairy samples which all of those produced in Turkey were purchased from local markets. Samples were separately labeled and brought into the laboratory. All samples except milk samples were dried in an oven at a temperature of 105 °C. The dried samples were powdered and homogenized in the laboratory. Adequate dried samples and milk samples were put into cylindrical plastic analysis containers which has 6x5 cm (diameter x height) geometry. Samples weighed and sealed with parafilm to prevent the escape of radon gas. ^{226}Ra is usually determined using the most intensive gamma transitions of its progenies (^{214}Pb and ^{214}Bi). Therefore step was necessary to avoid disequilibrium problems between ^{226}Ra and its progenies before the measurements. Then each sample was measured and the values were given in Bq/kg dry weight for dried samples and milk samples.

2.2. *Measurement*

Radioactivity measurements were performed by using a gamma spectrometer, which p-type HPGe detector and equipped with DSA-1000 multichannel analyser, 110% relative efficiency, 85:1 Peak/Compton ratio and 1.08 keV and 1.95 keV energy resolutions at 122 keV of ^{57}Co and 1332.5 keV of ^{60}Co, respectively. Using photopeaks of ^{241}Am (59.5 keV), ^{137}Cs (661.6 keV) and ^{60}Co (1173.2 , 1332.5 keV) radionuclides energy calibration was done before the measurements. Two different certified radioactive standards were used for efficiency calibration. For the milk samples 79830-839 liquid certified radioactive standard were used. The 79829-839 coded commercially available volume standart source was used to determine the other dairy samples radioactivity concentrations. These certified radioactive standards contain 13 radionuclides that have the energy range of 59.5–1836.1 keV. Before the measurement background spectrum was taken. After measurements and subtraction of the background the activity concentrations were determined. Measurement times for the samples were 85000 s.

The activity concentration of ^{226}Ra was calculated from 295.2, 351.9 keV gamma-ray energies of ^{214}Pb and 609.3 keV of ^{214}Bi. Due to overlapped peaks of ^{235}U (185.7keV) and ^{226}Ra (186keV) radionuclides in the spectrum, 186 keV photopic of ^{226}Ra was not preferred for calculations. The activity concentration of ^{232}Th was calculated from 338.4 and 911.2 keV of ^{228}Ac and 583.2 keV gamma-ray energies of ^{208}Tl. The activity concentration of ^{40}K was determined by using its own energy of 1460.8 keV. Activity concentration of ^{137}Cs was calculated from 661.7 keV photopic energy of ^{137m}Ba radionuclide, which is the daughter product of ^{137}Cs. A sample spectrum is shown in Figure 1.

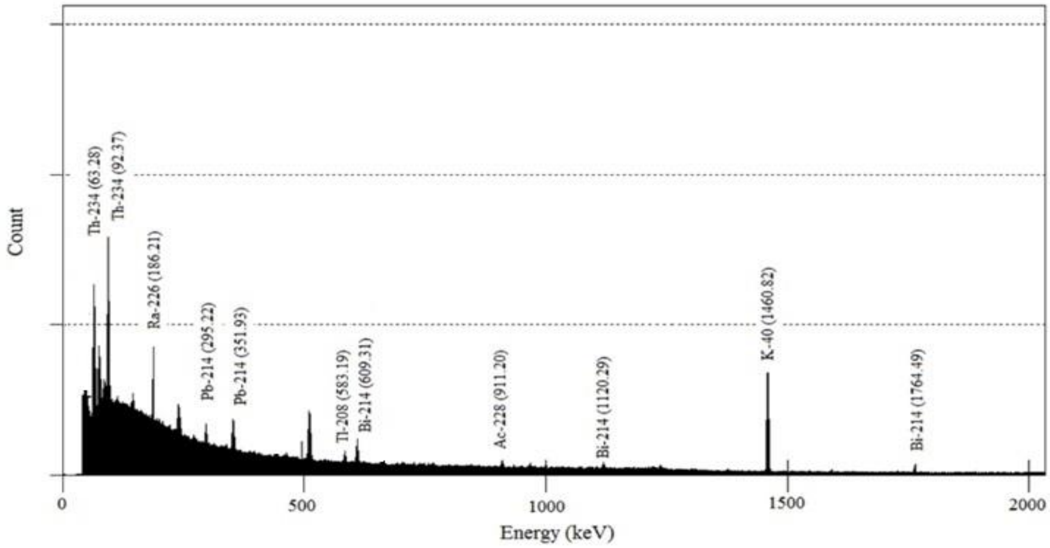

Figure 1. A typical gamma spectrum of the dairy product.

The activity concentrations of the samples are calculated by the following formula:

$$A = \frac{N}{\varepsilon \cdot \gamma \cdot t \cdot m} \tag{1}$$

where N corresponds to the net peak area of gamma-ray energy, ε denotes the absolute efficiency, γ is the gamma-ray yield per decay, t and m denotes the counting time and sample mass, respectively [10]. The relative combined standard uncertainty of the activity concentration is given by the following formula [10]

$$u_{c(A)} = \frac{\sigma_{(A)}}{A} = \sqrt{\left(\frac{\sigma_{(m)}}{m}\right)^2 + \left(\frac{\sigma_{(N)}}{N}\right)^2 + \left(\frac{\sigma_{(\gamma)}}{\gamma}\right)^2 + \left(\frac{\sigma_{(\varepsilon)}}{\varepsilon}\right)^2} \tag{2}$$

Minimum detectable activity (MDA) calculations were performed by the following formula [11]:

$$MDA = \frac{(1.64)\cdot\sigma_n}{\varepsilon \cdot P \cdot t \cdot w} \tag{3}$$

where σ_n stands for standard deviation, ε is absolute efficiency, P is the emission probability, t and w denote measurement time and weight of the dried sample, respectively.

3. Results and discussion

Natural and artificial radioactivity concentrations were shown in Table1-6. Less than (<) sign indicates MDA value. ^{40}K average radioactivity concentrations of the white cheese, whey powder, milk powder, labaneh, kaskhaval cheese and milk were found to be 63.1 ± 7.1, 392.6 ± 40.2, 482.1 ±38.6, 152.0 ±12.1, 48.7 ± 5.8 and 52.2 ± 4.2 Bq/kg, respectively. ^{137}Cs radioactivity concentrations were found to be below MDA. Except the milk powder ^{232}Th radioactivity concentrations were found to be below MDA. ^{232}Th radioactivity concentrations of the milk powder samples were found to be ranged from 3.2 ± 0.3 - 9.8 ± 0.8 Bq/kg. ^{226}Ra radioactivity concentrations were ranged from 5.9 ± 0.5 - 8.3 ± 0.7 Bq/kg for whey powder, 5.1 ± 0.5 – 9.8 ± 0.8 Bq/kg for milk, 4.6 ± 0.5 - 9.9 ± 1.9 Bq/kg for white cheese, 3.5 ± 0.3 - 16.0 ± 1.5 Bq/kg for milk powder, 4.2 ± 0.3 - 11.1 ± 1.0 Bq/kg for labaneh.

Froidevaux et al. [12] found that radioactivity concentration of the ^{40}K ranged from 1.31 ± 0.2 - 2.15± 0.3 Bq / g for the cheese produced in different regions of Western Europe. Froidevaux et al. [12]. found that radioactivity concentrations of the ^{137}Cs under detection limit. Gomaa et al. [13] found that radioactivity concentration of the ^{40}K for the Egypt cheese was 58 Bq/kg. Gomaa et al. [13] found that radioactivity concentration of the ^{137}Cs was 1.7 Bq/kg. Lavi et. al. [14] found that radioactivity concentration of the ^{40}K for Israeli cheese were 44.8 Bq/kg.

Lavi et. al. [14] found that radioactivity concentration of the ^{137}Cs for Israeli milk ranged from 54.2-55.3 Bq/kg. Othman et al. [15] found that radioactivity concentration of the ^{40}K for the Syria milk was 290 ± 60 Bq/kg. Melquiades et al. [16] found that radioactivity concentration of the ^{137}Cs for the Brazilian milk ranged from 0.01-1.32 Bq/L. In that study, Melquiades et al. [16] found that radioactivity concentration of the ^{40}K for the Brazilian milk ranged from 48 ±1-55 ±2 Bq/L. Lavi et. al. [14] found that radioactivity concentration of the ^{40}K for Israeli milk powder ranged from 450.8-472.3 Bq/kg.

Table 1. Radioactivity concentrations of the whey powder (Bq/kg).

Sample No	**^{226}Ra**	**^{232}Th**	**^{40}K**	**^{137}Cs**
1	<3.2	<3.1	381.1 ± 30.5	<1.9
2	<3.1	<3.5	292.6 ± 26.3	<2.1
3	6.9 ± 0.6	<4.2	354.2 ± 28.4	<2.4
4	8.3 ± 0.7	<3.9	348.2 ± 27.8	<2.3
5	5.9 ± 0.5	<4.3	587.1 ± 52.4	<1.9
6	<3.0	<4.1	460.8 ± 39.5	<1.8
7	6.6 ± 0.6	<3.7	416.3 ± 32.6	<2.2
8	5.9 ± 0.5	<3.8	337.4 ± 27.3	<1.9
9	<3.1	<4.2	370.1 ± 30.2	<2.3
10	<2.9	<4.1	378.1 ± 31.1	<1.9

Table 2. Radioactivity concentrations of the milk (Bq/kg).

Sample No	**^{226}Ra**	**^{232}Th**	**^{40}K**	**^{137}Cs**
1	<2.6	<2.3	49.5 ± 5.1	<2.2
2	9.8 ± 0.8	<1.9	38.6 ± 3.9	<2.4
3	<3.1	<2.6	50.1 ± 5.1	<2.5
4	<2.9	<2.8	49.1 ± 4.9	<2.3
5	5.1 ± 0.5	<2.4	47.5 ± 4.8	<1.9
6	<3.6	<2.4	57.9 ± 5.3	<2.4
7	5.2 ± 0.5	<2.5	34.1 ± 3.5	<1.9
8	<3.2	<2.1	59.5 ± 5.4	<2.1
9	9.1 ± 0.8	<2.2	50.9 ± 5.2	<2.3
10	<3.8	<1.9	84.5 ± 7.6	<2.2

Melquiades et al. [16] found that radioactivity concentration of the ^{40}K for the Brazilian milk powder from two different region mean values were 475, 489 Bq/kg. Melquiades et al. [16] found that radioactivity concentration of the ^{137}Cs for the Brazilian milk powder from two different region range were 5.1-7.3 and 7.0-11.2 Bq/kg.

Table 3. Radioactivity concentrations of the white cheese (Bq/kg).

Sample No	**^{226}Ra**	**^{232}Th**	**^{40}K**	**^{137}Cs**
1	8.8 ± 0.9	<2.4	49.9 ± 5.1	<2.0
2	<2.3	<2.6	60.3 ± 5.8	<2.3
3	4.6 ± 0.5	<2.3	86.7 ± 8.9	<2.6
4	<2.4	<2.1	70.3 ± 7.1	<2.1
5	13.1 ± 1.2	<1.8	53.2 ± 5.4	<1.9
6	16.2 ± 1.5	<2.5	65.6 ± 6.5	<2.1
7	<2.5	<2.4	87.1 ± 8.9	<2.4
8	19.9 ± 1.9	<1.9	54.1 ± 5.5	<2.4
9	<2.6	<1.9	36.1 ± 3.5	<2.3
10	13.8 ± 1.4	<2.2	67.8 ± 6.9	<2.4

Table 4. Radioactivity concentrations of the kashkaval (Bq/kg).

Sample No	**^{226}Ra**	**^{232}Th**	**^{40}K**	**^{137}Cs**
1	<2.1	<2.2	61.4 ± 6.2	<2.1
2	<1.9	<1.8	63.1 ± 6.4	<2.0
3	<2.3	<2.5	69.3 ± 7.1	<2.4
4	<2.4	<2.6	40.7 ± 4.2	<2.5
5	<2.6	<2.1	46.1 ± 4.8	<2.6
6	<2.7	<2.3	40.2 ± 4.1	<2.4
7	<2.5	<1.9	20.6 ± 2.0	<1.9
8	<2.4	<2.5	55.4 ± 5.3	<2.6
9	<2.3	<2.4	46.5 ± 4.5	<2.4
10	<2.6	<2.6	43.8 ± 4.3	<2.1

Table 5. Radioactivity concentrations of the milk powder (Bq/kg).

Sample No	**^{226}Ra**	**^{232}Th**	**^{40}K**	**^{137}Cs**
1	13.1 ± 1.2	4.3 ± 0.4	513.5 ± 52.1	<2.4
2	8.3 ± 0.7	9.8 ± 0.8	515.1 ± 52.3	<2.3
3	3.7 ± 0.4	6.2 ± 0.6	515.0 ± 52.3	<2.5
4	16.0 ± 1.5	3.2 ± 0.3	658.4 ± 66.2	<2.1
5	8.8 ± 0.9	3.8 ± 0.3	565.5 ± 57.5	<2.3
6	5.5 ± 0.5	3.3 ± 0.3	596.7 ± 58.7	<2.0
7	6.6 ± 0.6	3.9 ± 0.3	401.4 ± 39.1	<1.9
8	3.9 ± 0.3	6.5 ± 0.6	279.8 ± 26.5	<2.2
9	3.5 ± 0.3	3.6 ± 0.3	522.8 ± 51.1	<2.4
10	3.7 ± 0.3	4.1 ± 0.4	252.7 ± 25.1	<.21

Table 6. Radioactivity concentrations of the labaneh (Bq/kg).

Sample No	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
1	4.5 ± 0.4	<2.1	111.3 ± 12.1	<1.9
2	9.3 ± 0.8	<2.3	147.6 ± 15.1	<2.1
3	5.0 ± 0.5	<1.7	157.3 ± 16.1	<2.0
4	10.1 ± 1.0	<1.5	128.1 ± 13.2	<1.9
5	9.1 ± 0.8	<1.9	170.4 ± 18.1	<2.3
6	4.2 ± 0.3	<1.8	168.8 ± 17.2	<1.8
7	9.5 ± 0.9	<2.3	170.8 ± 18.3	<2.3
8	5.0 ± 0.4	<2.4	156.3 ± 16.7	<1.6
9	11.1 ± 1.0	<1.9	175.4 ± 19.2	<2.2
10	5.3 ± 0.5	<2.2	133.5 ± 14.2	<1.8

4. Conclusion

In this study, natural and artificial radioactivity concentrations of the white cheese, whey powder, milk powder, labaneh, kaskhaval cheese and milk were determined via gamma spectrometric analysis. ^{40}K radionuclide activity concentrations are in accordance with other studies. ^{137}Cs radionuclide activity concentrations were found to be below the MDA. In the EU, the limit for radiocaesium was established at 370 Bq/kg for milk and dairy product. According to European Council Regulation No 737/90 for the reported radionuclides ^{137}Cs activity values are not hazardous for public health.

It is very important to determine the level of radioactivity concentrations in milk and dairy products to ensure consumer safety. The obtained result provides useful information to carry out dose assessment due to ingestion of these products.

Acknowledgement

This study was done as routine activities of Radioactivity and Analytic Measurement Department of SANAEM / TAEK.

References

[1] United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR), **Exposures from natural sources**, UNSCEAR, (2000).

[2] Gilmore G.R., **Practical Gamma-ray Spectrometry**, 2nd ed, Sussex, ENGLAND, Wiley, (2008).

[3] Nucléide–Lara, **Library for gamma and alpha emissions** http://www.nucleide.org/ Laraweb/, (02.03.2017).

[4] IAEA, International Atomic Energy Agency, **Naturally occurring radioactive materials (NORM IV)**, IAEA-TECDOC-1472, Vienna, (2005).

[5] Ahmed, F., Impact of fertilizers on background radioactivity level in two newly developed desert areas, **Radiation Effect and Defects in Solids**, 165, 1, 65–71, (2010).

[6] IAEA, International Atomic Energy Agency, **Quantification of Radionuclide Transfer in Terrestrial and Freshwater Environments for Radiological Assessments**, TECDOC 1616, Vienna, (2009).
[7] Kalac, P.A., Review of edible mushroom radioactivity, **Food Chemistry**, 75, 29–35, (2001).
[8] TUIK (Türkiye İstatistik Kurumu), http://www.tuik.gov.tr, (13.12.2016).
[9] European Council Regulation No 737/90.
[10] IAEA- International Atomic Energy Agency, **Guide Quantifying Uncertainty in Nuclear Analytical Measurements**, IAEA-TECDOC-1401, Vienna, (2004).
[11] Currie, L.A., Limits for qualitative detection and quantitative determination, **Analytical Chemistry**, 40, 3, 586-693, (1968).
[12] Froidevaux, P., Geering, J.J., Pillonel, L., Bosset, J.O. and Valley, J.F., ^{90}Sr, ^{238}U, ^{234}U, ^{137}Cs, ^{40}K and $^{239/240}Pu$ in Emmental type cheese produced in different regions of Western, **Europe Journal of Environmental Radioactivity**, 72, 287–298, (2004).
[13] Gomaa, M.A., Abdel-Fattah, A.T., Essa, M.W. and El-Shinawy, R.M.K., Radioactivity in foodstuffs in Egypt, **Applied Radiation and Isotopes**, 46, 6/7, 607-608, (1995).
[14] Lavi, N., Golob, G. and Alfassi, Z.B., Monitoring and surveillance of radio-cesium in cultivated soils and foodstuff samples in Israel 18 years after the Chernobyl disaster, **Radiation Measurements**, 41, 78–83, (2006).
[15] Othman, I., Yassine. T., Natural radioactivity in the Syrian environment, **The Science of the Total Environment,** 170, 119-124, (1995).
[16] Melquiades, F.L. and Appoloni. C.R., Radiation of powdered milk produced at Londrina, PR, Brazil, **Radiation Physics and Chemistry**, 61, 691–692, (2001).