

## Radioactivity levels in some cow milks consumed in Eastern Black Sea Region of Turkey

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### Abstract

The radioactivity levels were determined in 12 different brands of cow milks consumed in Eastern Black Sea region of Turkey using a high-purity germanium detector. The mean activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs were 3.2, 1.1, 31.3 and 0.5 Bq.L<sup>-1</sup>, respectively. The values were comparable with the concentrations reported in the other countries. Due to consumption of cow milk, the radiological impact of them on the inhabitants was calculated by taking the annual intake into account through ingestion of aforementioned radionuclides. The estimated effective doses from milk were found to be 21.2 µSv year<sup>-1</sup> for <sup>226</sup>Ra, 6.1 µSv year<sup>-1</sup> for <sup>232</sup>Th, 0.1 µSv year<sup>-1</sup> for <sup>137</sup>Cs and 4.7 µSv year<sup>-1</sup> for <sup>40</sup>K. The calculated annual effective dose obtained from all measured radionuclides was much below the UNSCEAR recommended reference level of 200-800 µSv.y<sup>-1</sup> for all milk samples. The obtained result data will contribute to establish a baseline level of radioactivity in cow milk and help to develop future guidelines in the country for radiological protection of the population.

**Key Words:** Radioactivity, Cow Milk, Gamma Spectrometer

## 1. Introduction

Our environment possesses a lot of radiation and radioactivity sources. The earth and the atmosphere present different activity levels of natural radionuclides such as those of  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay chains as well as  $^{40}\text{K}$ . Additionally, nuclear industry, mining, crude oil and gas manufacturing have also contributed to the radionuclide inventory of the environment. All these nuclides, natural and artificial, have radiobiological significance because they can be incorporated into the human body from the food chain and also through inhalation of suspended dust in the air, constituting a direct route for internal exposure [1].

Therefore, the verification of the natural and artificial concentration levels in food supplies is an important parameter for evaluating abnormalities. In this sense, there is a need to measure the radioactivity levels in foods, particularly in milk which forms a major component in the human food chain because it is consumed by people of various ages and sexes, especially by infants [2]. Furthermore, milk is the first and the most only mainstay food ingested for a considerable of time in human consumption, providing calcium necessary for strong bones, proteins needed for brain development and tissue growth, vitamin A for normal vision, and vitamin D for absorption of calcium [3-6].

Radionuclides pass into cows and consequently in milk through cow's food. These radionuclides, when accumulated in food and milk, eventually affect humans, either directly or indirectly by entering the food chain through the plants or cows. Therefore, the establishment of radioisotope concentration in milk will provide meaningful information that can contribute to the knowledge of population exposure and to the setting up a regional baseline.

The objective of this study is to determine the level of radionuclide concentrations in the milk samples and to estimate the radiological implications on the population consuming this milk in the Eastern Black Sea region of Turkey.

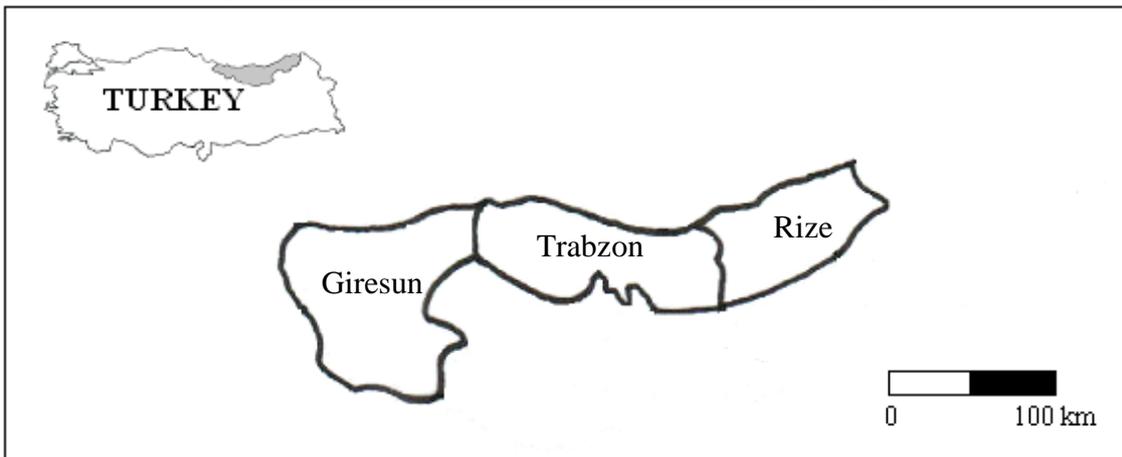
## 2. Materials And Methods

### 2.1. Sampling Area

The Eastern Black Sea region consists of deep running water valleys, mountains, steps and broken zones. All streams rising from this land flow into the Black Sea. Economic activities are based on agriculture and animal breeding, so industry has not yet developed sufficiently. According to 2007 census results, the population of the Eastern Black Sea region is approximately 2.6 million. While 1.4 million people live in cities, the rest live in villages. In the region, cow milk has been naturally consumed by the local people, and in the future, it may be bottled and transported to other parts of the country for consumption.

### 2.2. Sampling

The milk samples were collected from the provinces of Giresun, Trabzon and Rize, which are located in the Eastern part of the Black Sea Region of Turkey (Fig. 1).



**Figure 1.** Location of sampling sites indicating the Eastern Black Sea, Turkey

In order to measure the radioactivity in the milk samples, various samples were collected in 1-L-capacity polypropylene bottles. The bottles were cleaned using a procedure modified by Laxen and Harison [7]. The bottles were first soaked in 10% HNO<sub>3</sub> for 48 h, followed by deionized water for a few minutes, and finally rinsed three times with double-distilled, de-ionised water. The collected milk samples were acidified with concentrated nitric acid to pH 1 to break down the organic materials and to prevent loss of ions in the sample following binding to the container or precipitation.

Subsequently, the samples were slowly evaporated down to 100 mL volume in a furnace at 50°C and poured into cylindrical, polyethylene plastic containers (5.5 cm diameter and 5 cm height) for gamma activity analysis.

### 2.3. Instrumental

The activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in the milk samples were measured using a coaxial HPGe detector (Canberra, GC 1519 model). The relative efficiency of the detector is 15% and it has a resolution of 1.9 keV at the 1332 keV. The detector was shielded in a 10 cm thick lead well internally lined with 2 mm Cu foils. The detector output was connected to a spectroscopy amplifier (Canberra, Model 2025) [8]. The spectrum analysis was performed using Genie 2000 software from Canberra. A performance test using the certified reference sample (IAEA-375) was carried out to check the efficiency and energy calibration of the system. The activities of the standard were in accordance with their certified values within error margins not exceeding 10%. The quality assurance of the measurements was carried out by periodical efficiency and energy calibrations and repeating sample measurements. The sample counting time was 50.000 s to obtain gamma-spectra with good statistics. To determine the detector background, an empty container was counted in the same manner and in the same geometry as the samples. The background spectra were used to correct the net peak area of the gamma rays of the measured isotopes [9-10].

The activity concentrations of each sample for the radionuclides of interest were determined from their respective gamma lines or gamma lines emitted by their progenies. The gamma-energies lines of 351.9 keV ( $^{214}\text{Pb}$ ) and 609.3 keV ( $^{214}\text{Bi}$ ) were used to represent the concentrations for the  $^{226}\text{Ra}$  series, whereas 583.1 keV ( $^{208}\text{Tl}$ ) and 911 keV ( $^{228}\text{Ac}$ ) were used to represent the  $^{232}\text{Th}$  series.  $^{40}\text{K}$  activity determined from the 1460.8 keV emission gamma-ray lines and  $^{137}\text{Cs}$  activity was determined from the 661.6 keV emission gamma-ray lines.

The activity concentrations for the radionuclides in the measured samples were computed using the following relation;

$$A = \frac{C}{\varepsilon \text{PMt}} \text{ (Bq.L}^{-1}\text{)} \quad (1)$$

where  $A$  is the activity concentration of a radionuclide,  $C$  is the total net count of a specific gamma emission,  $\epsilon$  is the detector efficiency for the specific gamma emission,  $P$  is the absolute transition probability for that gamma emission,  $L$  is the volume of the sample and  $t$  is the counting time.

### 3. Results and discussion

The results of natural radioactivity in milks are presented in Table 1. The activity concentrations are expressed in  $\text{Bq.L}^{-1}$ . The mean specific activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  are 3.2, 1.1, 31.3 and 0.5  $\text{Bq.L}^{-1}$ , respectively. The  $^{226}\text{Ra}$  activity ranges between a minimum of 1.5  $\text{Bq.L}^{-1}$  and a maximum of 5.5  $\text{Bq.L}^{-1}$ . The  $^{232}\text{Th}$  activity lies between 0.1  $\text{Bq.L}^{-1}$  and 2.9  $\text{Bq.L}^{-1}$ . The activity of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  vary from <N.D. to 1  $\text{Bq.L}^{-1}$  and from 15.1 to 42.8  $\text{Bq.L}^{-1}$ , respectively. Measured  $^{137}\text{Cs}$  radionuclide is considered to be due to the atmospheric nuclear weapon tests conducted by several countries; however, some residual amount might be because of the Chernobyl accident, which occurred in 1986.

As shown in Table 1, the highest mean  $^{226}\text{Ra}$  and the lowest mean  $^{232}\text{Th}$  activity were determined in the milk samples taken from the province of Rize. While the highest mean  $^{232}\text{Th}$  activity was determined in the Trabzon province, the province of Giresun shows the lowest mean  $^{226}\text{Ra}$  activity. The mean lowest  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activities were determined in the milk samples taken from the province of Rize, but the mean highest  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activities were determined in the milk samples collected from the province of Trabzon.

If we compare our results with those of values in different countries, it is clear that  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  are high, while  $^{137}\text{Cs}$  and  $^{40}\text{K}$  are low in Iran [11], Israel [12], Venezuela [13] and Jordan [14].

Table 1. Activity concentration in radionuclides (in Bq. L<sup>-1</sup>)

Province	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>137</sup> Cs	<sup>40</sup> K
	2.7±0.1	0.4±0.1	0.7±0.1	15.1±1.1
Rize	5.5±0.4	0.8±0.1	N.D.	38.5±1.5
	4.1±0.3	1.6±0.2	N.D.	40.9±1.3
	3.5±0.3	0.1±0.1	0.4±0.1	18.3±0.5
Trabzon	1.5±0.1	0.3±0.1	0.4±0.1	25.9±1.3
	3.7±0.2	2.9±0.2	0.5±0.1	34.0±1.2
	3.5±0.2	1.5±0.1	0.5±0.1	30.1±1.1
	2.6±0.1	1.6±0.1	1.0±0.1	42.8±1.6
Giresun	2.7±0.2	1.5±0.2	0.7±0.1	36.7±1.7
	2.9±0.4	1.2±0.2	0.4±0.1	35.2±1.6
	2.5±0.5	0.8±0.2	0.5±0.1	27.2±1.3
	2.6±0.5	0.6±0.1	0.5±0.1	30.6±1.6
Mean	3.2	1.1	0.5	31.3

N.D: Not Detectable

The annual effective dose to an individual due to intake of radionuclides from the cow milk is estimated using the following equation [15]:

$$D_{\text{ing}} = C_R * I_F * E_D \quad (2)$$

where  $D_{\text{ing}}$  is the annual effective dose to an individual due to ingestion of radionuclides (sievert per year),  $C_R$  is the activity concentration of radionuclides in the ingested milk (becquerel per liter),  $I_F$  represents the annual intake of milk (litres per year). Assuming that every adult drinks 24 L.year<sup>-1</sup>, the annual effective doses caused by intake of the <sup>226</sup>Ra, <sup>232</sup>Th, <sup>137</sup>Cs and <sup>40</sup>K isotopes were calculated [16].  $E$  is the ingested dose conversion factor for the radionuclide (Sv Bq<sup>-1</sup>). The dose conversion factors used for the calculation were taken from the WHO publication [17] and were equal to  $2.8 \times 10^{-4}$ ,  $2.3 \times 10^{-4}$  and  $1.3 \times 10^{-5}$  mSv Bq<sup>-1</sup> for <sup>226</sup>Ra, <sup>232</sup>Th and <sup>137</sup>Cs, respectively. And for <sup>40</sup>K, intake was calculated using the conversion factor of  $6.2 \times 10^{-6}$  mSv Bq<sup>-1</sup> [18]. The calculated effective doses are given in Table 2.

The milk contribution to the total mean annual effective dose equivalent from the estimated effective doses was  $21.2 \mu\text{Sv year}^{-1}$  for  $^{226}\text{Ra}$ ,  $6.1 \mu\text{Sv year}^{-1}$  for  $^{232}\text{Th}$ ,  $0.1 \mu\text{Sv year}^{-1}$  for  $^{137}\text{Cs}$  and  $4.7 \mu\text{Sv year}^{-1}$  for  $^{40}\text{K}$  in Table 2. The annual effective dose from all measured radionuclides was much below the UNSCEAR [1] recommended reference level of  $200\text{-}800 \mu\text{Sv.y}^{-1}$  for all milk samples.

Table 2. Calculated total indicative doses from radionuclides (in  $\mu\text{Sv y}^{-1}$ )

Province	$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{137}\text{Cs}$	$^{40}\text{K}$
	18.1	2.2	0.2	2.2
Rize	37.0	4.4	N. D.	5.7
	27.6	8.8	N. D.	6.1
	23.5	0.6	0.1	2.7
Trabzon	10.1	1.7	0.1	3.9
	24.9	16.0	0.2	5.1
	23.5	8.3	0.2	4.5
	17.5	8.8	0.3	6.4
Giresun	18.1	8.3	0.2	5.5
	19.5	6.6	0.1	5.2
	16.8	4.4	0.2	4.0
	17.5	3.3	0.2	4.6
Mean	21.2	6.1	0.1	4.7

N.D:Not Detectable

$^{226}\text{Ra}$  is a highly radiotoxic radionuclide. When radium is absorbed into body, its metallic behavior is similar to that of calcium and an appreciable fraction is deposited in the bone, the remaining fraction being distributed almost uniformly in the soft tissues [19]. When people are exposed to very high levels of radium for a long time, it may result in cancer of the bone and the nasal cavity in human beings. From the table 2, it can also be seen that  $^{226}\text{Ra}$  contribution cumulative effective dose and annual effective dose are also higher than the other three radionuclides of  $^{232}\text{Th}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$ . Cesium is an important radionuclide for public health because of its presence in fallout and its relatively long life. Although the major source of Caesium body burden is from eating

foods which may contain radiocaesium or natural radiopotassium absorbed daily into different organs [20], the effect of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in milk cannot be neglected. There is a remarkable similarity between the distribution of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in the muscles of human beings, since  $^{40}\text{K}$  and  $^{137}\text{Cs}$  are both 90–100% absorbed from the diet into the tissues of the body [21]. This is because the two radionuclides have chemical similarity and are similarly metabolized by the same biological system [22]. Also, milk is considered to be a sensitive indicator of fall out contamination; this is because  $^{137}\text{Cs}$  is easily transferred to milk through the grass-cow-milk pathway [14].

#### 4. Conclusions

In this study, the radioactivity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in cow milk consumed in Eastern Black Sea region of Turkey were investigated. The measured values were found to be within the worldwide ranges as reported in the literature. Also, the calculated annual effective dose from all measured radionuclides was much below the UNSCEAR recommended reference level of 200-800  $\mu\text{Sv}\cdot\text{y}^{-1}$  for all samples. The activity concentrations of the  $^{137}\text{Cs}$  are generally detected in milk samples. This situation can be attributed to the effects of the  $^{137}\text{Cs}$  concentration that contaminated the Eastern Black Sea Region after the Chernobyl or nuclear weapons tests. The data obtained in this study may provide a baseline for setting standards for the quality of cow milk in the future estimations of the impact of radioactive pollution in this region since they might be consumed by the general public.

#### REFERENCES

- [1] UNSCEAR Sources and effects of ionizing radiation Report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly (New York: United Nations) (2000).
- [2] O. A. Osibote, J. B. Olomo, P. Tchokossa and F. A. Balogun, Radioactivity in milk consumed in Nigeria 10 years after Chernobyl reactor accident. *Nuclear Instruments and Methods in Physics A*, **422**, 778 (1999).
- [3] R. E. Black, S. M. Williams, I. E. Jones and A. Goulding, Children who avoid drinking cow milk have low dietary calcium intakes and poor bone health. *The American Journal of Clinical Nutrition*, **76**, 675 (2002).

- [4] B. Lonnerdal, Nutritional and physiologic significance of human milk proteins,” *The American Journal of Clinical Nutrition* **77**, 1537 (2003).
- [5] S. Murphy, K. T. Khaw, H. May and J. E. Compston, Milk consumption and bone mineral density in middle aged and elderly women. *BMJ*, **308**, 939 (1994)..
- [6] D. P. H. Laxen and R. M. Harrison, Cleaning methods for polyethene containers prior to the determination of trace metals in freshwater samples. *Analytical Chemistry*, **53**, 345 (1981).
- [7] Y. Tiryaki and C. Akbay, Consumers’ fluid milk consumption behaviors in Turkey: an application of multinomial logit model. *QualQuant*, **44**, 87 (2010).
- [8] U. Cevik, N. Damla, B. Koz and S. Kaya, Radiological characterization around Afsin Elbistan coal-fired power plant in Turkey,” *Energy Fuels*, **22**, 428 (2008).
- [9] N. Damla, U. Cevik, A. I. Kobya, A. Celik, R. Van Grieken and Y. Kobya, Radiological characterization of gas concrete used in building materials in Turkey. *Journal of Hazardous Materials*, **168**, 681 (2009).
- [8] N. Damla, U. Cevik, A. I. Kobya, B. Ataksor and U. Isik, “Assessment of environmental radioactivity for Batman, Turkey,” *Environmental Monitoring and Assessment*, Vol. 160, 2010, pp. 401-412.
- [9] T. Hosseini, A. A. Fathivand, H. Barati and M. Karimi, Assessment of radionuclides in imported foodstuffs in Iran. *Iranian Journal of Radiation Research*, **4**, 149 (2006).
- [10] N. Lavi, G. Golob and Z. B. Alfassi, Monitoring and surveillance of radio-cesium in cultivated soils and foodstuff samples in Israel 18 years after the Chernobyl disaster. *Radiation Measurements*, **41**, 78 (2006).
- [11] J. J. LaBecque, P. A. Rosales and O. Carias, The preliminary results of the measurements of environmental levels of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in Venezuela. *Nuclear Instruments and Methods in Physics A*, **312**, 217 (1992).
- [12] Q. Ababneh Zaid, A. M. Alyassin, K. M. Aljarrah and A. M. Ababneh, Measurement of natural and artificial radioactivity in powdered milk consumed in Jordan and estimates of the corresponding annual effective dose. *Radiation Protection Dosimetry*, **138**, 278 (2009).
- [13] N. M. Alam, M. I. Chowdhury, M. Kamal, S. Ghose, M. N. Islam and M. Anwaruddin, Radiological assessment of drinking water of the Chittagong region of Bangladesh. *Radiation Protection Dosimetry*, **82**, 207 (1999).
- [14] R. Yayar, Consumer characteristics influencing milk consumption preference. The Turkey case. *Economic Theory and Applications*, Vol. XIX, No. 7(572), pp. 25–42 (2012).
- [15] WHO (2006) Guidelines for drinking-water quality (First addendum to third edition, Vol. 1, p. 595).Recommendations. Geneva, Switzerland
- [16] D. Delacroix, J. P. Guerre, P. Leblanc and C. Hickman, Radionuclide and radiation protection data handbook. *Radiation Protection Dosimetry*, **98**, 1 (2002).
- [17] M. E. Wrenn, P. W. Durbin, B. Howard, J. Lipsztein, J. Rundo and E. T. Still, Metabolism of Ingested U and Ra. *Health Physics*, **48**, 601 (1985).

- [18] M. Fujita, J. Iwamoto and M. Kondo, Comparative metabolism of cesium and potassium in mammals-interspecies correlation between body weight and equilibrium level. *Health Physics*, 12, 1237 (1996).
- [19] International Commission Radiological Protection (1991) Recommendations of the International Commission on Radiological Protection. ICRP Publication 60 (Oxford: Pergamon Press)
- [20] P. F. Gustafson and J. E. Miller, The significance of  $^{137}\text{Cs}$  in man and his diet. *Health Physics*, 16, 167 (1969).
- [21] B. J. Howard, N. A. Beresford and K. Hove, Transfer of radiocaesium to ruminants in natural and semi natural ecosystems and appropriate countermeasures. *Health Physics*, 61, 715 (1991).
- [22] A. F. Nisbet and R. F. M. Woodman, Soil-to-plant transfer factor for radiocaesium and radiostrontium in agricultural systems. *Health Physics*, 78, 279(2000).

