

# Evaluation of Cs-137 Content in Powdered Cow Milk from Four Regions of Poland

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## Abstract

This study presents the results of determinations of Cs-137 in full-fat cow milk powder originating from four regions of Poland. The assays performed demonstrated differences in the content of Cs-137 in milk powder depending on the region. The higher content of Cs-137 was found in powdered milk samples collected from Opole (3.50 Bq/dm<sup>3</sup>). The lowest content of Cs-137 was in milk samples from Września (0.98 Bq/dm<sup>3</sup>). In powdered milk samples from Łapy and Krotoszyn the amount of Cs-137 was 2.02 and 1.11 Bq/dm<sup>3</sup>, respectively. The average content of Cs-137 in full-fat cow milk powder from all examined regions was 1.90 Bq/dm<sup>3</sup> after reconstituting to liquid milk, and did not exceed the admissible level of Cs-137 in food.

**Keywords:** cesium 137, cow milk powder, regions

## Introduction

Increasing industrialization has brought about substantial pollution of the soil, water and air with various harmful substances, including heavy metals such as cadmium and lead, organochlorine compounds, polychlorinated biphenyls and others [1, 2]. These compounds have contributed to environmental changes and caused the occurrence of hazardous substances in food, especially in food for infants [3]. Food is the major gateway for hazardous substances into the human body. Milk is a sensitive indicator of many contaminants due to fallout of radionuclides or other contaminants because of the feeding of cows on grass, a plant with a high absorbency of various dangerous substances [4].

Exploitation of nuclear energy for both peaceful and military purposes poses the problem of radioactive contamination of the biosphere. A vast number of explosions of atomic and thermonuclear bombs have been conducted since 1945. Radioactive elements of nuclear explosions are absorbed by atmospheric dust and rainwater, then steadily

fall and penetrate the surface of the Earth. They are then gradually released and absorbed by plants [5].

Radiation may result in disturbances in data transfer between a cell and its environment as well as between cells inside a body. Radioactive activity may lead to cell apoptosis. It may also evoke changes in DNA, damage pyrimidine and purine bases, and form untypical links between them. This, in turn, is likely to result in an incapacity to produce essential proteins that may be subject to deformation and, consequently, to disorders of the immune system and susceptibility to infections. Exposure of an organism induces symptoms determined by the dose of radiation. Acute radiation sickness is linked with the occurrence of encephalic syndrome, marrow syndrome, etc. Each of the symptoms has the following phases: initial latent phase and a main phase leading to death. After recovery from radiation sickness, secondary symptoms usually emerge, mainly in the form of neoplastic changes [6].

Caesium is the major radionuclide generated during nuclear explosions. There are 35 known isotopes of caesium [7]. It has good solubility in water, which facilitates its penetration into plants and exposure to the subsequent links of the food chain [8-10].

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Cs-137 and Cs-134, as well as products of their degradation, emit  $\gamma$  radiation and  $\beta$  radiation absorbed in tissues. Radioactive isotopes of caesium in the human body disintegrate after 100 to 140 days. This compound is completely absorbable in the respiratory and alimentary organs. In humans, most of the absorbed radiocaesium is accumulated in muscles (60%), and the rest in organs of the gastrointestinal tract, in liver, lungs, kidneys, brain, blood. Other radionuclides, for example iodine, are accumulated in the thyroid or strontium in bones and teeth. Caesium also accumulates in the reproductive organs and joints. It may be a cause of miscarriages and genetic damage to fetuses [11].

A number of factors and phenomena of both physical, chemical, meteorological and biological origin determine the process of environmental "purification" of radiocaesium. The half-life of Cs-137 is ca. 30 years [7, 12].

Most of the radioactive caesium occurring in the environment originates from contamination resulting from the reactor breakdown in Chernobyl on April 26, 1986.

The occurrence of Cs-137 in food of both plant and animal origin has been confirmed by ample research [9, 13-15]. Investigations into the transformation of Cs-137 from soil to chicken meat and eggs [13] to milk and dairy products [15], and the transfer of Cs-137 and Sr-90 to horse milk and meat have also been conducted [10].

On January 13, 2008 the National Atomic Energy Agency announced that in Polish cow milk the average content of Cs-137 was 0.6 Bq/dm<sup>3</sup> and ranged from 0.2-1.2 Bq/dm<sup>3</sup>.

Taking into account the detrimental effects of radioactive compounds on human health, special attention should be paid to the level of radionuclides in food, especially in milk. Cow milk and dairy products play an important role in human nutrition.

The level of radiocontamination of milk is the basis for the evaluation of radioactive isotopes absorption by food [16]. Powdered cow milk is a good indicator representing radioactive contamination of foodstuffs by radioactive isotopes such as Cs-137 [17].

The objective of our study was to evaluate the content of Cs-137 in full-fat cow milk powder originating from four regions of Poland.

## Material and Methods

The experimental material was powdered cow milk originating from four regions of Poland (Fig. 1). The full fat milk powder subjected to analyses originated from dairy plants in Łapy, Krotoszyn, Opole and Września. Milk was purchased in retail shops or directly from producers in the years 1998-99. Cow milk powder from this region was most often used and available on the Olsztyn market.

The methodology was described by the Central Laboratory for Radiological Protection [18]. Samples of milk powder were weighed and dried at ca. 80°C for 12 h.

They were then carbonized on an electrical plate and incinerated in a muffle furnace at a temperature not exceeding 450°C until grey ash was obtained. The ash was transferred to a high beaker with a volume of 100 cm<sup>3</sup>, then 10 cm<sup>3</sup> of distilled water and 60 cm<sup>3</sup> of nitric acid (1:1) were added, and the beaker was heated until the grey ash was completely dissolved. The resultant solution was filtered through a hard filter into a cylinder that was then filled with water up to a volume of 150 cm<sup>3</sup>.

Preparation of ammonia phosphomolybdenate (AMP) bed: 7 cm<sup>3</sup> of ammonium nitrate solution (71.4 g of NH<sub>4</sub>NO<sub>3</sub> was dissolved in 143.5 cm<sup>3</sup> of concentrated HNO<sub>3</sub> with  $d=1.4$  g/cm<sup>3</sup> and filled to a volume of 1,000 cm<sup>3</sup>), 3 cm<sup>3</sup> of ammonium heptamolybdate (26.5 g of tetrahydrous salt (NH<sub>4</sub>)<sub>6</sub>Mo<sub>7</sub>O<sub>27</sub> · H<sub>2</sub>O and dissolved in 250 cm<sup>3</sup> of distilled H<sub>2</sub>O at a temperature of 70°C) and 1 cm<sup>3</sup> of ammonium dihydroorthophosphate solution (3.64 g of NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub> was dissolved in 500 cm<sup>3</sup> of distilled water) were weighed into a beaker with a volume of 50 cm<sup>3</sup>. The resultant precipitate was heated at 80°C for 20 minutes for crystallization. Next, it was cooled to room temperature and filtered through a radiochemical funnel with a filter made of hard blotting paper. The bed was rinsed with distilled water until the disappearance of acidic reaction according to litmus. Dissolved ash was vacuum-filtered through a radiochemical funnel with an AMP bed. The filtration rate was adjusted so that the filtration process lasted 20 minutes. The blotting paper filter was transferred to a measuring cuvette, coated with a 4% collodion in acetone and left to dry. Radioactivity was measured with the use of a kit for contamination control and measurements (ZAPKS-1 by Polon, Warsaw). As a standard, use was made of 250 mg of KCl fixed in a measuring cuvette. The activities of the KCl standard and Cs-137 standard were compared and the resultant conversion factor reached 1.2220. It was used in calculations of the activity of the examined samples.



Fig. 1. Powdered milk sampling locations.

$$A_p = \frac{N_0 \cdot k_{Cs}}{m \cdot s}$$

...where:

$$k_{Cs} = \frac{A_w}{N_w - N_T} \left[ \frac{Bq}{imp/min} \right]$$

$$N_0 = N_p - N_T \text{ [imp/min]}$$

$A_p$  - activity of Cs-137 in the research sample;  $A_w$  - activity of standard,  $k_{Cs}$  - calibration coefficient Cs isotopes;  $N_p$  - counting number of samples with background;  $N_T$  - counting number of background;  $N_w$  - counting number of standard with background;  $m$  - samples weight used for analyses;  $s$  - sorption efficiency ( $s = 1$ ).

### Results and Discussion

The obtained results of Cs-137 content in milk powder are collated in Table 1. The mean content of Cs-137 accounted for 1.90 Bq/dm<sup>3</sup> of milk prepared from full fat milk powder. The highest content of Cs-137 was observed in milk powder originating from the Dairy Plant in Opole – 3.5 Bq/dm<sup>3</sup> of milk, whereas the lowest was in milk produced in Września – 0.98 Bq/dm<sup>3</sup> of milk.

Content of Cs-137 in cow's milk from different years and countries shows Table 2.

Bierska et al. reported that the mean annual value of Cs-137 for Poland in 1982 was 555±41 mBq/dm<sup>3</sup> and in 1983 it had fallen to 434±43 mBq/dm<sup>3</sup> [19].

After the Chernobyl nuclear accident, the level of radioactive contamination increased. In powdered cow milk produced between May 9-15, 1986, in Mława was measured at 270-445 Bq/dm<sup>3</sup> after recounting to liquid milk [20].

The concentration of Cs-137 isotope in drinking milk has decreased in subsequent years. In 1986 in Poland, its level accounted for 5.2 Bq·dm<sup>-3</sup>, in 1987 for 4.2 Bq·dm<sup>-3</sup>, and in 1988 for as low as 1.8 Bq·dm<sup>-3</sup> [21]. In 2003 the level was 0.8 Bq/kg of Cs-137 [22].

Investigations carried out in Sweden in June 1988 demonstrated that the Cs level in cow milk had reached 300.0 Bq·kg<sup>-1</sup> milk, whereas in August of the same year it had dropped to 170 Bq·kg<sup>-1</sup>. In the period from August 1989 until 1990, the mean content of Cs-137 in milk was relatively high and reached 100 Bq·kg<sup>-1</sup>. In turn, in the years 1992-93, its level in cow milk was shown to account for as little as 2 to 21 Bq·kg<sup>-1</sup> [15].

Kostiainen reports that before the atomic reactor accident in Chernobyl, levels of Cs-137 in all regions of Finland were similar and ranged from 5 to 35 Bq/dm<sup>3</sup> of milk. After 1986, however, Cs levels in milk had increased to 60 Bq per liter of milk. What is more, higher concentrations were recorded in regions with a prevalence of peat soil and lower ones in areas with a predominance of loamy soil [23].

Table 1. Content of Cs-137 in cow milk powder originating from different regions of Poland (Bq/dm<sup>3</sup> of milk prepared from full fat milk powder).

Regions	Number of samples	Content of Cs-137	Standard Deviation	Average
Łąpy	6	<b>2.02</b>	0.24	<b>1.90</b>
Krotoszyn	6	<b>1.11</b>	0.33	
Opole	6	<b>3.50</b>	0.72	
Września	6	<b>0.98</b>	0.61	

Table 2. Content of Cs-137 in cow milk from different years and countries.

Countries	Year	Content of Cs-137	Denomination	References
Poland	1982	<b>555</b>	mBq/dm <sup>3</sup>	[19]
	1983	<b>434</b>	mBq/dm <sup>3</sup>	[19]
	1986	<b>270 - 445</b>	Bq/dm <sup>3</sup>	[20]
	1986	<b>5.2</b>	Bq/dm <sup>3</sup>	[21]
	1987	<b>4.2</b>	Bq/dm <sup>3</sup>	[21]
	1988	<b>1.8</b>	Bq/dm <sup>3</sup>	[21]
	2003	<b>0.8</b>	Bq/kg	[22]
Austria	1986	<b>942 - 3480</b>	Bq/kg	[4]
	1991	<b>12 - 50</b>	Bq/kg	[4]
Brazil	1998	<b>3.7</b>	Bq/kg	[24]
Sweden	1988 - June	<b>300</b>	Bq/kg	[15]
	1988 - August	<b>170</b>	Bq/kg	[15]
	1990	<b>100</b>	Bq/kg	[15]
	1992-93	<b>2 - 21</b>	Bq/kg	[15]
Finland	1985	<b>5 - 35</b>	Bq/dm <sup>3</sup>	[23]
	1986	<b>60</b>	Bq/dm <sup>3</sup>	[23]

Mück described a change in the content of caesium in full fat milk powder originating from large dairy plants in Austria. Results of seven-year experiments proved that in 1986 the content of caesium ranged from 942 to 3480 Bq/kg of milk powder, and seven years later it was from 12 to 50 Bq/kg of milk powder, depending on the dairy plant [4].

In 1998, Melquiades and Appoloni measured Cs-137 in Brazilian milk powder at a level of 3.7 Bq/kg [24].

The present study shows that in cow powder milk, the average content of Cs-137 is 1.9 Bq/dm<sup>3</sup> and is higher in comparison with content of Cs-137 in other kinds of milk. Investigations indicates that human breast milk contained 0.13±0.08 Bq/dm<sup>3</sup> and infant formulas (which are produced

with the use of powdered cow milk) contained  $1.39 \pm 0.26$  Bq/dm<sup>3</sup> of Cs-137 [3].

Appendix 1 to the government regulations of the April 27, 2004 stipulates the permissible levels of radioactive substances in contaminated food and contaminated water for human consumption. The level of caesium-134 and 137 in milk and dairy products reaches 1000 Bq/kg (for concentrated or dried products, this value is applicable to ready-to-eat preparations) [25].

### Conclusions

The study demonstrated that the content of Cs-137 in full fat milk powder originating from four regions of Poland was low and did not exceed the permissible levels of this radionuclide in milk as stipulated in the government regulations.

The contents of Cs-137 in full fat milk powder were demonstrated to differ depending on the region the samples were collected in. The highest concentrations of caesium were reported in milk powder originating from the region of Opole and the lowest were in milk powder originating from the region of Września. The differences of Cs-137 content in powdered milk depend on several factors, including region, type of soil, climatic characteristic, other environmental conditions, and the type of feed and age of the cow.

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