



ESTIMATION OF CHANGE DYNAMICS OF MILK CONTAMINATION WITH ^{90}Sr AND ^{137}Cs IN LITHUANIA IN 1965–2003

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Abstract. The paper describes the trends of concentrations of ^{90}Sr and ^{137}Cs in milk in Lithuania in 1965–2003. Sampling of milk was performed in five regions of the biggest cities of Lithuania, and since 1976 – also in the region of possible impact of Ignalina Nuclear Power Plant (NPP). Data of production and consumption of milk are taken from Lithuanian statistical yearbooks. Activity concentrations of radionuclides were measured at Radiation Protection Centre. Daily diet products are connected with different components of the environment – soil, air and water. For this reason food contamination reflects pollution of the environment. This paper describes estimation of trends of activity concentrations of ^{90}Sr and ^{137}Cs in one of the food products – in milk. Comparison applies to two periods – the so-called before Chernobyl period (1965–1985) and that after the accident in Chernobyl NPP up to now (1986–2003). Contamination of milk was analysed using correlation tests. Though the trend of average annual activity concentrations is complicated, during the two periods plenty of high enough correlation ratio values were calculated (0,63–0,74). Regression curves using 3–6 degree of polynoms show the decrease of ^{90}Sr and ^{137}Cs activity concentrations during the periods analysed. Factors of influence on the value of an average annual effective dose for an inhabitant of Lithuania based on activity concentrations of ^{90}Sr and ^{137}Cs in milk were evaluated. Estimation shows that accuracy of calculating an average annual effective dose due to ^{90}Sr and ^{137}Cs in milk does not exceed 60 %.

Keywords: ionizing radiation, dose, radiological measurements.

1. Introduction

Man-made radionuclides appeared in the environment after the first nuclear bomb tests in the atmosphere, later – during operation of nuclear installations and accidents at nuclear power plants. Most dangerous from the view of the ecology are long-lived radionuclides. Food products are connected with the environment by a food chain which is shorter for vegetables and longer for meat, milk or fish. All food products are connected in different ways with the components of the environment: air, water, soil and plants growing in the soil. The way of behavior of two of long-lived radionuclides – ^{90}Sr (strontium) and ^{137}Cs – (cesium) is the same as their stable isotopes calcium and potassium that are in the body of man.

There exist at least three most important sources of contamination of the environment with ^{90}Sr and ^{137}Cs . The first one is connected with tests of nuclear bombs.

The first tests of nuclear bombs were performed in Japan in 1945 [1]. According to the data of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 1993) during the period before 1962, when a triangular moratorium was signed by the Soviet Union, USA and United Kingdom for the restriction of nuclear tests in the atmosphere, already 360 nuclear bomb tests were performed. During these tests releases to the environment were up to 0,76 EBq ($0,76 \times 10^{18}$ Bq) of ^{90}Sr and 1,14 EBq of ^{137}Cs . Even after the agreement was signed France and China continued tests and 63 more tests were performed that released to the atmosphere 0,09 EBq of ^{90}Sr and 0,14 EBq of ^{137}Cs [2]. Most of these releases deposited in the North hemisphere, close to the 40° latitude, less part of releases deposited close to the North Pole and direction to the equator. Only one third of all the releases deposited in the South hemisphere.

The next source of contamination of the environment with ^{90}Sr and ^{137}Cs is releases from nuclear installations and nuclear reprocessing plants. Low-level radioactive waste was released from Sellafield nuclear fuel reprocessing facility to the Irish Sea. During the period 1950–1990 about $3\text{--}4 \times 10^{16}$ Bq of ^{137}Cs was released from this installation [1].

One more source of contamination of the environment are accidents in nuclear power plants and nuclear reprocessing plants. Paper [1] states that releases during accidents are smaller than from nuclear bomb tests. Releases of ^{137}Cs during the accident at Chernobyl NPP were approximately about 100 PBq ($1 \text{ P} = 10^{15}$) or 20–40 % of the whole content of ^{137}Cs which was in the active zone of the reactor [3], and approximately about 8 PBq of ^{90}Sr . One third of all these releases deposited in the European part of the territory of the Soviet Union – approximately 30 PBq of ^{137}Cs and approximately 90 % of ^{90}Sr (7 PBq) [2]. Paper [4] states that the density of precipitation of ^{137}Cs in Lithuania was less than $18,5 \text{ kBq/m}^2$. ^{137}Cs activity concentrations in the soils of Lithuania were measured in 1992 dividing the territory into sectors of $16 \cdot 16 \text{ km}$ [4]. Sampling was performed up to a depth of 5 cm. Three cesium “spots” were estimated where ^{137}Cs activity concentrations were the highest – in the South and West part of Lithuania and the Curonian Spit. The highest activity concentration in the soil was measured in 1992 in the samples from the Curonian Spit – approximately $19\,700 \text{ Bq/m}^2$. Activity concentrations were measured at the same time in the samples of the two other “spots”. The highest activity concentration of ^{137}Cs was measured – 1600 Bq/m^2 [4].

Paper [5] presents the investigation data on soil contamination with ^{137}Cs in Lithuania during the period 1974–2000. An average activity concentration of ^{137}Cs in soils of Lithuania before the accident at Chernobyl NPP was $(6,8 \pm 1,8) \text{ Bq/kg}$, after the accident an average amount of ^{137}Cs is $6,7\text{--}28,5 \text{ Bq/kg}$ [5].

One more source of contamination of the Lithuanian territory with ^{137}Cs is forest fire and resuspension from the soil at highly contaminated regions of Belarussia and the Ukraine [6]. ^{137}Cs activity concentration in the atmosphere of Lithuania after forest fire in Belarussia increased significantly. Activity concentration of ^{137}Cs in the atmosphere was up to $300 \mu\text{Bq/m}^3$ during 30 August – 1 September 1992, up to $250 \mu\text{Bq/m}^3$ in 9 April 1996 and up to $200 \mu\text{Bq/m}^3$ at 6–9 September 2002 [6].

Measurements of activity concentrations of ^{90}Sr and ^{137}Cs in fresh food samples in Lithuania were started in 1965 when the network of environmental radiological monitoring was established. Sampling was started in 5 regions, each area was in the surroundings of the biggest cities of the country. The coordinates of sampling points are identified as an area name – Kaunas, Klaipėda, Panevėžys, Šiauliai and Vilnius. The monitoring network was the same during the period from the start until the accident at Chernobyl NPP. After the accident at

Chernobyl NPP the monitoring network, especially for the measurement of ^{131}I , was expanded [7].

This paper analyses only data from the environmental radiological monitoring network and does not include statistical analysis of additional measurements of milk after the accident at Chernobyl NPP and a special surveillance of regions where the radioactive cloud passed. The results of milk radiological measurements at Radiation Protection Centre starting with 1965 are analysed. Milk is one of food products using which for daily products ^{137}Cs and ^{90}Sr concentrations can easily reach the human body. According to models described in UNSCEAR approximately 32 % of ^{90}Sr intake is due to milk products. In the case of ^{137}Cs the intake is approximately 25 % [2]. Measurements were performed for raw fat-free milk. In cases when a sample contains fat on the top of milk, it was taken out from the sample analysed.

The aim of this paper is to analyse contamination of milk in Lithuania with ^{90}Sr and ^{137}Cs during the period 1965–2003 and to evaluate factors that may influence the value of an average annual effective dose for an inhabitant of Lithuania.

2. Methods used

Starting with 1965 activity concentrations of ^{137}Cs and ^{90}Sr were measured in samples from the regions of Vilnius, Kaunas, Klaipėda, Šiauliai and Panevėžys. Since 1976 monitoring was established in the region of Ignalina NPP, within a radius of 30 km around the Plant. The results of measurements are available in the vicinity of Ignalina NPP: Ignalina municipality since 1976, Zarasai municipality since 1979 and Utena municipality since 1992. An average value of samples analysed per year is 4, during some years – up to 8, for example, during 1972 in Vilnius region. Sampling was performed from milk containers in farms, milk factories, from places where it is possible to obtain mixed milk samples that may represent a region. The sample volume was 1–6 liters.

Procedures for measuring ^{137}Cs and ^{90}Sr in milk were also different during the period analysed. The first procedures that were adopted in 1965 for radiochemical analysis of ^{137}Cs and ^{90}Sr were in use until 1996. Before 1996 for a beta counting of ^{137}Cs and ^{90}Sr a proportional counter type UMF radiometer with SBT-13 type detector was used, since 1996 – a liquid scintillation counter.

Radiochemical separation of ^{137}Cs during the period analysed in this paper was performed according to the procedure described in paper [8]. Using this procedure ashed sample is dissolved in diluted nitric acid, separation of ^{137}Cs from the solution is carried out using Ni ferrocianate salt and precipitation process of cesium to $\text{Cs}_3\text{Sb}_2\text{J}_9$. Chemical yield is determined by gravimetric using stable cesium tracer that is added to a sample. The efficiency of counting is determined using

certified reference material – radioactive ^{37}Cs solution. The limit of detection is 10 mBq/sample, total uncertainty, when $k = 2$, is 10–15 % (is indicated in the description of the procedure).

Radiochemical separation of strontium from the ash of a sample is performed using extraction of ^{90}Sr by an organic solvent and counting of beta particles by beta radiometer type UMF. The limit of detection is 20 mBq/sample. The total uncertainty, when $k = 2$, is 15–30 %.

Since 1996 the activity concentration of ^{90}Sr and its daughter ^{90}Y that is in the equilibrium with ^{90}Sr is determined by counting in liquid scintillation counter Tri-Carb 2770 TR/SL a Cherenkov radiation. According to the procedure an ashed sample is dissolved in concentrated nitric acid, after the sample is evaporated to dryness and when again dissolution using diluted chloral acid is used. Extraction of yttrium is performed using HDEHP, after this step precipitate is dissolved in 1M HNO_3 acid and Cherenkov counting is performed. A scintillation counting vial used is plastic, the volume of the vial is 20 ml, counting is performed using 0,0–200,0 keV energy window, the time of counting is 120–200 min. The efficiency of counting is determined using a standard reference solution – a radioactive solution of strontium/yttrium made in Amersham, Great Britain. Counting efficiency was 0,55, background value – 2,30 counts per minute. Chemical yield in milk samples was high (0,4–0,87) and was determined by adding stable yttrium to the initial sample before dissolution of ash in diluted chloral acid. The total uncertainty, when $k = 2$, is 10–30 %, it depends on the activity measured. In case when the value of activity concentration is close to the detection limit, the total uncertainty becomes higher (up to 90 %).

The measurement results were summarized using Ms Excel files. Separate files were prepared for ^{137}Cs and ^{90}Sr according to the regions of milk sampling. Calculation of an annual average value was performed using at least 10 separate values of measurement results.

3. Results

Statistical evaluation of the measuring results was performed for three periods. The first period was selected for the whole monitoring period when milk measurements were started (1965–2003). Two more periods were selected – before the accident at Chernobyl NPP (1965–1985) and the period 1982–2003.

Estimation of trends in the activity concentration of ^{137}Cs during the period 1965–2003 was performed using 552 data, for ^{90}Sr – 559 data. The quantity of data in the other two periods was approximately 300 in each. Figs 1 and 2 show histograms in which the number of samples according to activity concentrations of ^{137}Cs and ^{90}Sr is set. The histograms demonstrate a lot of values with low-activity concentrations, and only during the period 1965–1985 some higher-activity concentration

values were found to be even ten times higher.

Contamination of milk with ^{137}Cs and ^{90}Sr was analysed using correlation testing. Correlation ratio evaluates the strength of statistical ratio (any ratio, not only linear as it is in the case of correlation coefficient) and is expressed by number 0 and 1. This paper analyses only a ratio between the activity concentration of ^{90}Sr and ^{137}Cs and the time period. Though the trend of average annual activity concentrations is complicated, during two periods (1965–2003 and 1965–1985) plenty of high enough correlation ratio values were calculated (0,63–0,74). Regression curves using 3–6 degree of polynoms show decrease of ^{90}Sr and ^{137}Cs activity concentrations in milk during the periods analysed. Negative values of the correlation coefficient are not very high (from –0,34 to –0,44), but they are reliable enough from the point of view of statistics (99 %).

Exemption from this evaluation is the period after the accident at Chernobyl NPP. During this period the activity concentration of ^{137}Cs in milk becomes higher (for ^{90}Sr such an expression is not evident) but an average annual value is less than during the year 1965. The variation coefficient increases significantly only for ^{137}Cs activity concentration in milk (from 1,5 to 2,8). The variation coefficient for ^{90}Sr activity concentration decreases slowly from 1,5 to 1. Regression curves in Figs 1 and 2 show these trends.

An annual effective dose due to ^{137}Cs and ^{90}Sr in milk is calculated using activity concentrations of radionuclides analysed, annual consumption of milk (including milk products) and dose coefficients [9]. Average of milk consumption per year was estimated using statistic data.

Statistic data for consumption of milk per capita were listed in the statistics yearbooks of Lithuania only for the period 1996–2002 [10, 11] (Fig 3, Table). For the period since 1965, data were found in other statistics yearbooks of Lithuania [12–14].

An average annual value of milk produced during the period 1965–2002 was (2466±147) thousand tons (standard deviation from an average value is approximately 6 %) (Fig 4). Annual milk consumption per capita was postulated the same as during the period 1996–2002. This assumption is based on the fact that decrease of population was slight during the period 1965–2003. An average annual value of population since 1965 is (3437,9±78,3) thousand people (standard deviation from an average value is 2,36 %). No substantial changes were estimated analysing milk consumption for the country's internal use since 1996. An average annual value of milk consumption for the Lithuanian inhabitants is (1079±115) thousand tons. Standard deviation is less than 11 %. Production of milk per year in the period since 1996 was the same as during the period 1965–2003 – (1803±86) thousand tons. So, a conclusion can be drawn that an average annual consumption of milk (including milk products) during the period 1965–2003 is (1079±115) thousand tons.

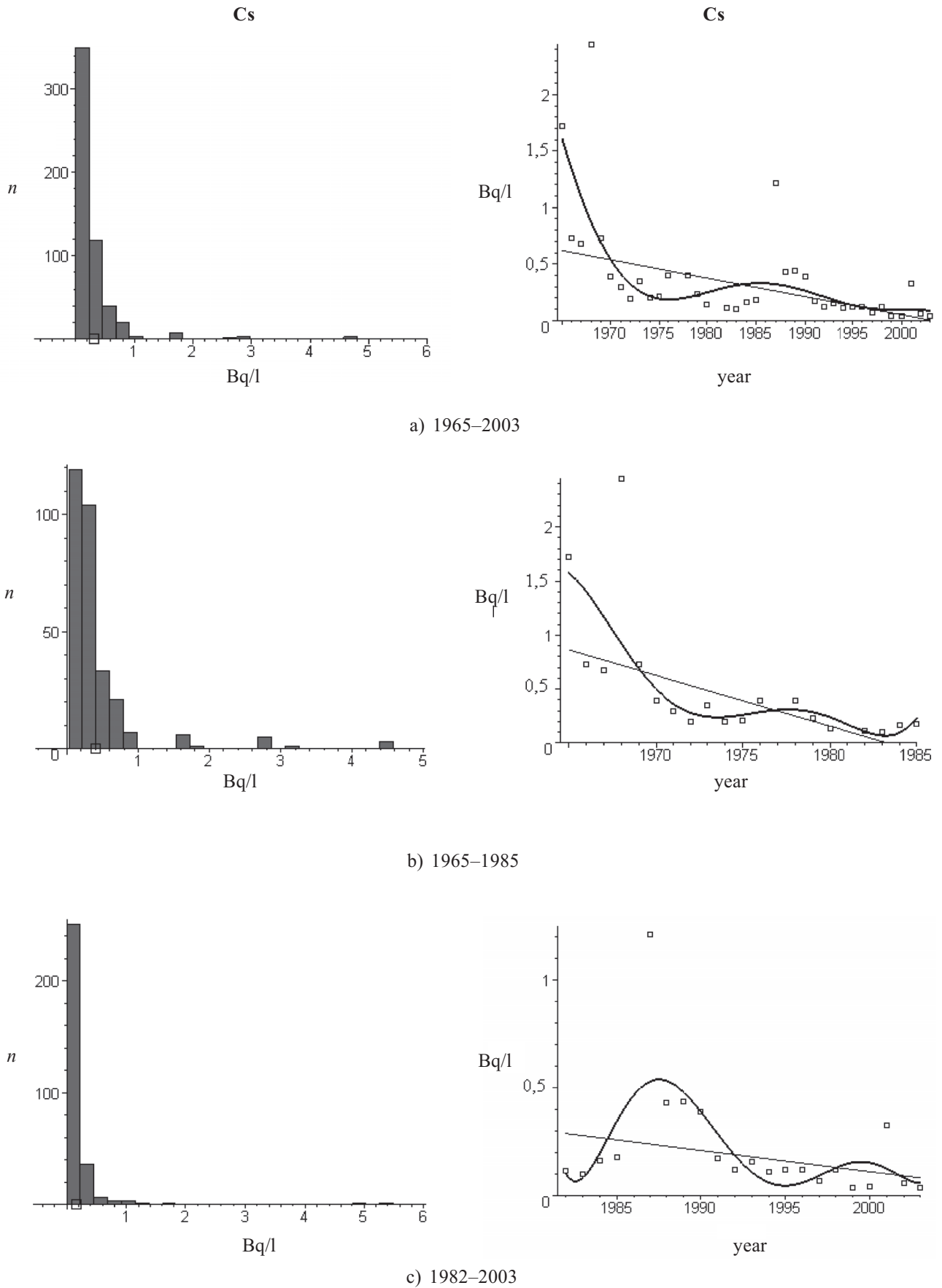


Fig 1. ¹³⁷Cs activity concentrations in milk: a) 1965–2003, b) 1965–1985, c) 1982–2003. Left histogram: *n* – quantity of samples, right symbol – □ – average annual value of activity concentration in samples analysed

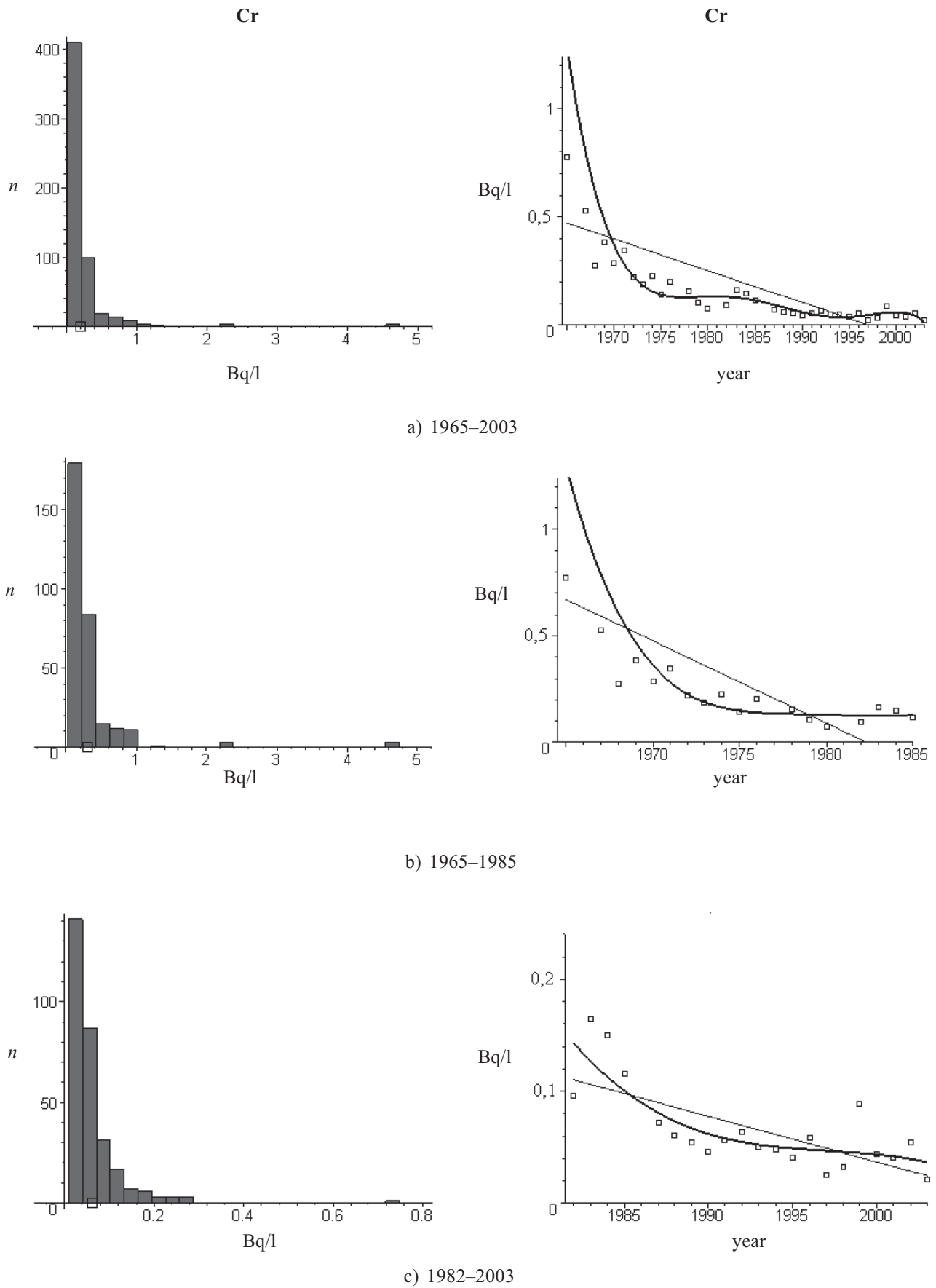


Fig 2. ⁹⁰Sr activity concentrations in milk: a) 1965–2003, b) 1965–1985, c) 1982–2003. Left histogram: *n* – quantity of samples, right symbol – □ – average annual value of activity concentration in samples analysed

The data in the Table show that milk imported to Lithuania is insignificant comparing with annual consumption and is not higher than 5 % of the total milk produced in the country. Import of milk products increased during the period 2000–2001 but it was less than 6 % to compare with the total milk. So, activity concentration of ^{137}Cs and ^{90}Sr in milk does not affect an annual effective dose.

Analysis of statistic data shows that an average

annual milk consumption per capita is 313,85 liters. Uncertainty of this evaluation is not less than 25 %. Average uncertainty of radiological measurements is approximately 30 %. So, uncertainty of estimation of an average annual effective dose for an adult caused by ^{137}Cs and ^{90}Sr in milk is not less than 60 %. The trends of dose are the same as the regression curves in Figs 1 and 2.

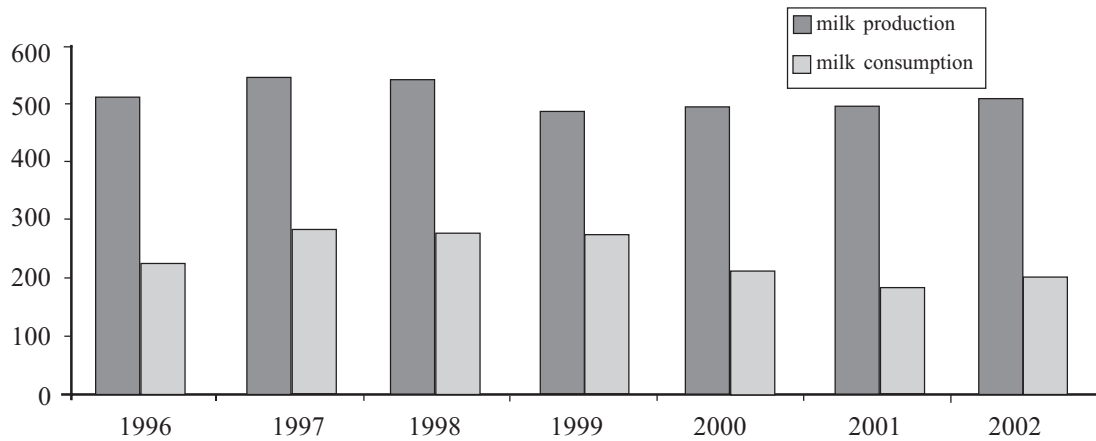


Fig 3. Annual milk consumption per capita in Lithuania during the period 1997–2002, kg

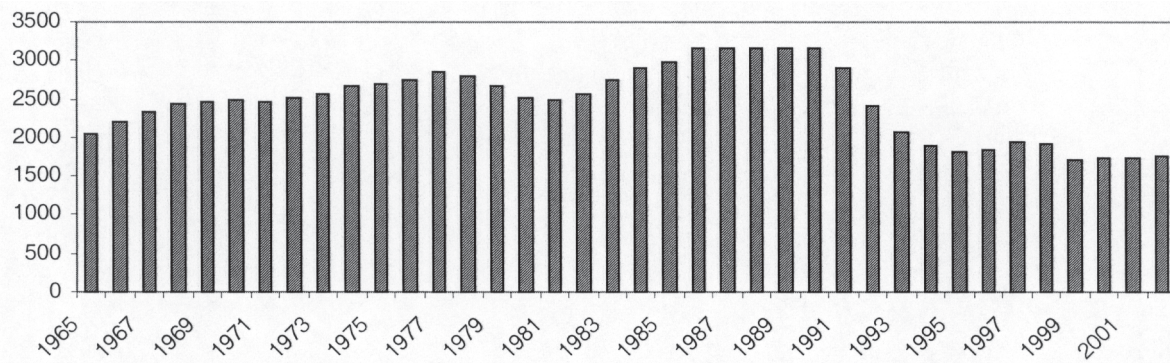


Fig 4. Annual milk production in Lithuania during the period 1965–2002, in thousand tons

Data on milk production and consumption for the country's internal use, import and export during the period 1997–2002 in Lithuania, in thousand tons

Year	Production	Consumption for country's internal use	Import	Export
1997	1949,7	988,20	100,2	965,6
1998	1929,9	913,00	54,8	1168,8
1999	1714,2	945,70	43,4	715,8
2000	1724,7	1199,50	170,5	700,2
2001	1729,8	1214,90	134,1	668
2002	1770,9	1211,00	61,4	615,4
Average value	1803	1079	94	806
Standard deviation from average value	108	144	50	215
Interval of confidence, $k = 2$	86	115	40	172

4. Conclusions

1. Trends in the concentrations of ^{90}Sr and ^{137}Cs in milk from 1965 to 2003 were estimated. Though the trend of average annual activity concentrations is complicated, during two periods – 1965–2003 and 1965–1985 – plenty of high enough correlation ratio values were calculated (0,63–0,74);

2. Regression curves using 3–6 degree of polynoms show decrease of ^{90}Sr and ^{137}Cs activity concentrations during the periods analysed;

3. During the period after the accident at Chernobyl NPP the activity concentration of ^{137}Cs in milk becomes higher (for ^{90}Sr such an expression is not evident), but an average annual value is less than during the year 1965. The variation coefficient increases significantly only for ^{137}Cs activity concentration in milk (from 1,5 to 2,8). The variation coefficient for ^{90}Sr activity concentration decreases slowly from 1,5 to 1;

4. Uncertainty of estimation of an average annual effective dose for an adult caused by ^{137}Cs and ^{90}Sr in milk is not less than 60 %.

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PIENO UŽTARŠOS ^{90}Sr IR ^{137}Cs KAITOS DINAMIKOS 1965–2003 m. LIETUVOJE ĮVERTINIMAS

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S a n t r a u k a

Straipsnyje analizuojama ^{90}Sr ir ^{137}Cs tūrinio aktyvumo piene kaita nuo 1965 m. iki 2003 m. Pieno ėminiai imti penkiuose didžiausių Lietuvos miestų regionuose, o nuo 1976 m. – ir Ignalinos atominės elektrinės regione. Pieno gamybos ir suvartojimo duomenys paimti iš Lietuvos statistikos metraščių, o radionuklidų tūrinis aktyvumas piene matuotas Radiacinės saugos centre. Žmogaus raciono produktai tiesiogiai ar netiesiogiai yra susiję su aplinkos komponentais – dirva, oru ir vandeniu. Todėl maisto produktų tarša atspindi žmogaus aplinkos taršos mastus. Šiame straipsnyje įvertinta vieno iš maisto produktų – pieno užterštumo ^{90}Sr ir ^{137}Cs kitimas laikui bėgant. Lyginta užtarša per du periodus – vadinamąjį „ikičernobylinį“ (1965–1985 m.) ir per laikotarpį nuo avarijos Černobylio atominėje elektrinėje iki šių dienų (1986–2003 m.). Pieno užterštumo tendencijos nagrinėtos atliekant koreliacinę analizę. Nors metinių tūrinio aktyvumo vidurkių kitimo eiga gana sudėtinga, tačiau apskaičiuotos 1965–2003 ir 1965–1985 m. laikotarpiu pakankamai didelės jų koreliacinių santykių vertės (0,63–0,74). Pagal regresijos kreives su 3–6 laipsnio daugianariais nustatytas ^{90}Sr ir ^{137}Cs tūrinio aktyvumo piene mažėjimas per visą tiriamąjį laikotarpį. Įvertinti veiksniai, turintys įtakos metinės efektinės dozės vidutiniam Lietuvos gyventojui kaitai dėl piene esančių ^{90}Sr ir ^{137}Cs jonizuojančiosios spinduliuotės. Nustatyta, kad ^{90}Sr ir ^{137}Cs piene jonizuojančiosios spinduliuotės sukeltos vidutinės metinės efektinės dozės įvertinimo tikslumas neviršija 60 %.

Raktažodžiai: jonizuojančioji spinduliuotė, dozė, radiologiniai tyrimai.

АНАЛИЗ ДИНАМИКИ ИЗМЕНЕНИЙ ЗАГРЯЗНЕНИЯ МОЛОКА РАДИОНУКЛИДАМИ ^{90}Sr И ^{137}Cs В ЛИТВЕ В ПЕРИОД 1965–2003 гг.**Р. Ладигене, Д. Буткус, Й. Клейза****Резюме**

Представлены данные анализа загрязнения молока радионуклидами ^{90}Sr и ^{137}Cs в Литве в период 1965–2003 гг. Образцы молока отобраны в пяти регионах страны, а после 1976 г. и в регионе Игналинской атомной электростанции. Статистические данные о производстве и потреблении молока в Литве взяты из годовых отчетов Департамента статистики Литвы. Исследования молока производились в Центре радиационной защиты.

Динамика загрязнения молока радионуклидами ^{90}Sr и ^{137}Cs в Литве проанализирована за период до аварии на Чернобыльской АЭС и в период 1986–2003 гг. Для анализа применялся метод корреляции. Кривые регрессий показывают постоянное снижение концентраций радионуклидов ^{90}Sr и ^{137}Cs в молоке. Установлены факторы, оказывающие влияние на среднюю годовую эффективную дозу, приходящуюся на жителя Литвы из-за ионизирующего излучения находящихся в молоке ^{90}Sr и ^{137}Cs .

Ключевые слова: ионизирующее излучение, доза, радиологические измерения.

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