Radioprotection 2017, 52(3), 171–176

DOI: 10.1051/radiopro/2017016

Available online at:
www.radioprotection.org

ARTICLE

Doses from Cs-137 and Sr-90 to Czech population due to milk consumption

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Received: 3 June 2016 / Accepted: 3 May 2017

Abstract – Activity concentrations of ^{137}Cs and ^{90}Sr in milk have been monitored in the Czech Republic on a long-term basis. The annual geometric means of ^{137}Cs activity concentrations in milk range from 12 Bq.L^{-1} in 1986 to 0.02 Bq.L^{-1} in 2014. The annual geometric means of ^{90}Sr activity concentrations range from 0.69 Bq.L^{-1} in 1965 to 0.02 Bq.L^{-1} in 2014. The values of effective and environmental half-lives in milk were calculated for both radionuclides from the annual geometric means and their temporal trends. Doses from both radionuclides due to milk ingestion were estimated.

Keywords: ^{137}Cs caesium / ^{90}Sr strontium / milk / dose

1 Introduction

The content of radionuclides in milk has been a subject of long-term monitoring in the Czech Republic. The most important artificial radionuclides in the food chain are ^{137}Cs and ^{90}Sr. Both have long physical half-life, both are chemical analogues of biogenic elements and both are transferred to milk. Caesium-137 with a half-life of 30.1 years, is an analogue of potassium depositing in muscular tissue and its behaviour is similar to potassium in plants and animals. Strontium has similar half-life, 28.8 years and behaves very much like calcium. A large portion of strontium will accumulate in bones and like calcium it transfers to milk. Since the Sr uptake by the human body from milk is an important pathway for radiostrontium incorporation, milk is a good indicator of ^{90}Sr content in human diet.

^{137}Cs and ^{90}Sr in the environment of the Czech Republic came from two sources. The first source has been relatively homogeneous surface contamination caused by the nuclear weapon tests especially in the 1960s. It used to reach (UNSCEAR, 1982) 5 kBq.m^{-2} for ^{137}Cs and 3.23 kBq.m^{-2} for ^{90}Sr on the north hemisphere between 40°–50° N. The second source was surface contamination caused by the fallout after the Chernobyl nuclear power plant accident. It spread inhomogeneously on the Czech Republic territory due to the variedly intensive rainfalls during the passage of the contaminated air over the territory. After the Chernobyl accident maximal surface activities of ^{137}Cs in some areas reached up to 100 kBq.m^{-2}; the average value was estimated (IHE, 1986; UNSCEAR, 1993; De Cort *et al.*, 1998) to be between 4 and 7 kBq.m^{-2}. Concerning ^{90}Sr, the activity increment in Central Europe caused by the Chernobyl accident was several times lower (Irlweck and Khademi, 1996; De Cort *et al.*, 1998) than that of ^{137}Cs.

As a result of the Chernobyl accident in 1986 the activity concentrations of ^{137}Cs in milk in many countries increased markedly by two to three orders from less than a tenth of Bq.L^{-1} to units up to tens of Bq.L^{-1}. The activity concentrations decreased relatively quickly during approximately a decade back to the pre-Chernobyl value. The decrease in the following years was only very slow (Grabowski *et al.*, 2002; Varga *et al.*, 2006; STUK, 2008; JRC, 2009). The development of ^{137}Cs activity concentrations in milk in the Czech Republic in the first years after the Chernobyl accident is described in several publications (Drábová *et al.*, 1988, 1990; Kameník *et al.*, 2009). The comprehensive results of milk and powdered milk monitoring till 2014 are presented in this paper.

The following values of ^{90}Sr activity concentrations in milk were detected in several countries neighbouring the Czech Republic: Poland – 0.3 Bq.L^{-1} in 1974 and 0.2 Bq.L^{-1} in 1975; East Germany – between 1974 and 1976 the values were in the interval (0.2–0.1) Bq.L^{-1}; West Germany – the values (UNSCEAR, 1982) between 1975 and 1980 were in the interval (0.3–0.1) Bq.L^{-1}. In Austria 0.078 Bq.L^{-1} was found (Mück *et al.*, 1990) in 1997. Already in the period of the nuclear weapon tests the content of ^{90}Sr in the Czech Republic environment had been monitored (Březík *et al.*, 1980; Radioaktivita, 1990).

2 Materials and methods

2.1 Sampling of milk

Samples of raw, processed – pasteurised, sterilised and powdered milk come in the Radiation monitoring network

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(RMN) from the retail network or producers (mostly dairies) for ^{137}Cs and ^{90}Sr determination. The aim is to achieve the most widespread and homogeneous monitoring of the Czech Republic territory and consequently to detect representative activities, which allow to estimate the population doses. Since the Chernobyl accident till the present time the sampling has been optimized as activities of radionuclides kept decreasing. Several tens of liquid and powdered milk samples from virtually homogeneously spread sites are collected in the Czech Republic per year.

Altogether 8107 samples of milk were collected within RMN since the Chernobyl accident till the end of 2014 for determination of ^{137}Cs. Out of the number there were 5350 liquid milk samples and 2757 powdered milk samples. Besides the National Radiation Protection Institute, the State Veterinary Institute has been participating in the milk sampling and ^{137}Cs evaluation since 2004. The results are collected in the common database RMN.

For the period 1965 – spring 1986 annual geometric means of ^{90}Sr activity are included in this study. Data on ^{90}Sr activity concentrations in milk from dairy (Praha – Radlice) since 1965 till 1995 are available. Samples for ^{90}Sr determination were collected systematically only in that dairy till 1986, afterwards the sampling network was enlarged markedly. Results from seven dairies in Northern Moravia and several others in Prague and its vicinity are used in this paper. Some of the dairies have been closed and the activity concentration of ^{90}Sr in liquid milk decreased, therefore the number of surveyed dairies was reduced. The dairy sampling was conducted in Prague till 2002, followed by liquid milk sampling from retail shops until now. In Northern Moravia one dairy: Ostrava – Martinov, has been surveyed since 1988–till present, with quarterly sampling. Three hundred and sixty four data on liquid milk have been obtained since 1986.

Since activity concentrations of ^{90}Sr in liquid milk were decreasing, determinations of its activity concentrations in powdered milk were initiated. The samples have been collected from one dairy since 1999 and from retail network in seven regions of the Czech Republic since 2006, currently 2 times a year. Altogether 173 powdered milk samples have been analysed since 1999.

2.2 Caesium-137 detection with gamma-ray spectrometry

Caesium-137 is detected using gamma-ray spectrometry, either with or without the sample pre-treatment. High purity germanium detectors of various efficiencies ranging from 10 to 150% were used. The efficiency calibration with gel calibration standards produced in the Czech Metrology Institute was used. In case of the difference between the measured sample density and calibration standard, correction to self-absorption was performed.

Activity of ^{137}Cs in powdered milk samples, and, less often, in liquid milk samples, was measured without previous treatment in identical geometry in cylindrical containers of 0.2 L placed around or on the detector. In samples of liquid milk with low activity concentration, ^{137}Cs was separated before the counting with composite inorganic ion exchanger NiFC–PAN, produced (Šebesta *et al.*, 1994) at the Czech Technical University and after drying measured in Petri dishes of 55 mm diameter. To calculate statistical characteristics, ^{137}Cs activity of powdered milk (in Bq.kg^{-1}) was converted into activity concentrations of liquid milk (in Bq.L^{-1}) by dividing by the following factors: 11.2 L.kg^{-1} (skimmed milk with lower than 0.5% fat on dry basis), 9.8 L.kg^{-1} (semi skimmed milk with 14% fat) and 8.2 or 8.4 L.kg^{-1} (whole milk with 26 and 28% fat) (Kněz, 1974). In case there was no fat indication on the given sample of powdered milk, the conversion factor for semi skimmed milk was used.

2.3 Radiochemical determination of ^{90}Sr

Activity of ^{90}Sr in liquid and powdered milk has been monitored exploiting a radiochemical method based on the U.S. Department of Energy, Environmental Measurements Laboratory method Sr-03-RC (HASL 300, 2015). Contrary to the original method strontium was not separated from calcium. A milk sample was ashed and the ash dissolved in hydrochloric acid. First, strontium and calcium were concentrated by oxalate precipitation. Radium, lead and barium were removed with barium chromate precipitation. Traces of other fission products were scavenged with iron hydroxide. After the ^{90}Sr + ^{90}Y equilibrium has been attained, yttrium was precipitated as the hydroxide and converted to the oxalate for counting on a low-background gas-flow proportional beta counter. The source was repeatedly measured, typically for a week to follow the decrease in ^{90}Y activity. Activity of ^{90}Sr was calculated from the measured data and the time of ^{90}Y ingrowth. Strontium yield was determined with ^{85}Sr tracer by counting in a gamma well detector. The separation yield of Y was determined by complexometric titration. Efficiency of the proportional counter for ^{90}Y was determined by counting ^{90}Y separated from a standard solution the same way as in sample analysis.

Various types of low-background alpha-beta instruments with proportional detectors were being used in the whole period. All the chemicals used in the analyses were of analytical grade.

When liquid milk was used, samples from 2 to 3.5 L were taken and divided into two parallel sub-samples. Tracer ^{85}Sr was added directly to the milk. When powdered milk was used, typically samples of 0.4 kg were taken for the analysis and tracer ^{85}Sr was added to the ash. For powdered milk the value of activity in the powdered milk was recalculated for the activity concentration in native state using factor of 9.8 L.kg^{-1}.

2.4 Statistical data processing

When processing the results, statistical characteristics of the individual data sets for ^{137}Cs and ^{90}Sr activities in milk were calculated from the individual values from all the sampling locations. Arithmetic mean (AM), geometric mean (GM) and geometric standard deviation (GSD) were estimated using programme R and a set of statistical programmes (Lee, 2009; R Development Core Team, 2011) NADA, which allow to evaluate data sets containing lower than minimum significant activity (MSA) values (Currie, 1968) and create their model substitutes. The calculations were based on the assumption of a log-normal distribution of the detected activities, while the MSA values were substituted with the model values, to comply with the assumption of the data log-normality as much as possible. The log-normality data compliance was verified by projection in quantile plots.

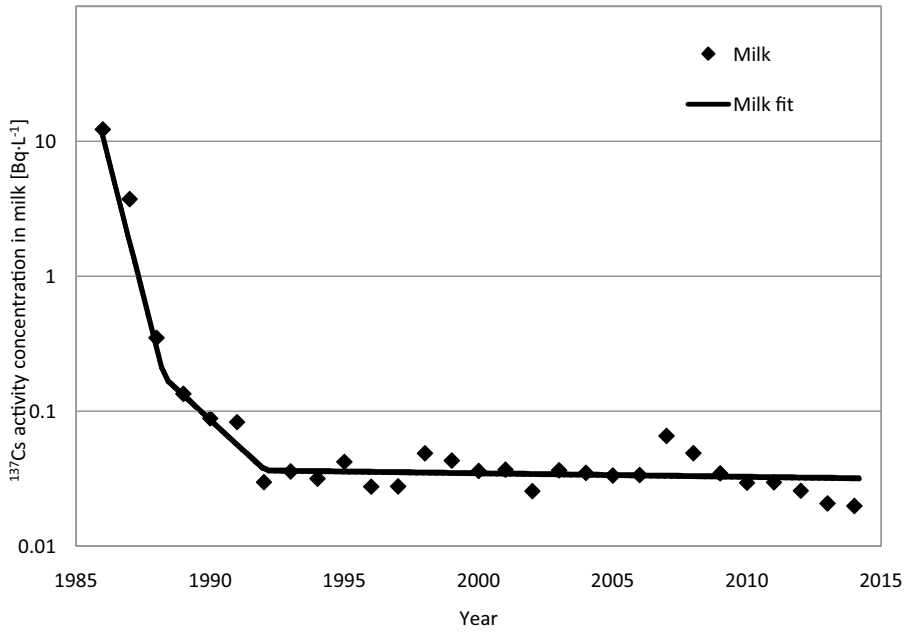

Fig. 1. The annual geometric means of ^{137}Cs activity concentrations in milk in the whole Czech Republic.

2.5 Calculation of half-lives and doses

Generating a best fit of time curve from radionuclide activity values in the matrix enables determination of effective and environmental half-lives of a given radionuclide in the given matrix. Effective half-life T_{EF} characterizes a decrease in content caused by all processes in progress in a natural ecosystem in the given period and it is determined by equation (1):

$$T_{EF} = \frac{\ln 2}{\lambda_{EF}}, \quad (1)$$

in which λ_{EF} means effective rate of loss. Environmental half-life T_{EN} characterizes a decrease in a radionuclide activity caused by all processes being in progress in a natural ecosystem except for radioactive decay. Environmental half-life T_{EN} can be expressed by equation (2):

$$T_{EN} = \frac{(T_{EN} \times T_P)}{T_P - T_{EF}}, \quad (2)$$

in which T_P is physical half-life of the given radionuclide, while it is always valid, that $T_P > T_{EF}$.

Logarithmic plots of all the geometric means were created for each radionuclide. The activity time courses of both radionuclides show different characters in various periods. The whole observed intervals were thus divided into segments characterized by straight data development as presented in the logarithmic plot. The location of the break points for ^{137}Cs was determined by iteration method (Muggeo, 2003). The fits and resulting equations in the individual sections were obtained by piecewise linear continuous regression implemented in the package "segmented" of the programme R (Muggeo, 2008).

For ^{90}Sr, at first the whole monitored interval was also analysed with the help of the above mentioned iteration method. It suggested the break points in the years 1974 and 1991. As especially the second suggested break did not correspond to the events important from the point of view of the radionuclide release into the environment, a different way of evaluation was chosen. The new break points at 1974 and 1986 were determined. Linear regression in Excel application was used to obtain the best fits for the chosen segments. Values of λ_{EF} (actually a slope of the best fit lines) and their deviations were calculated for the individual segments. According to equation (1) effective and according to equation (2) environmental half-lives were calculated for both radionuclides in the given periods.

Annual committed effective dose, E, was estimated using equation (3):

$$E = GM(c) \times CR \times h_{ing}, \quad (3)$$

in which $GM(c)$ means geometric mean of activity concentration, CR consumption rate of raw milk (in equivalent of milk products) and h_{ing} dose coefficient for intake by ingestion.

3 Results and discussion

3.1 Caesium-137

Since 1986 the number of annually analysed samples was gradually reduced from the original 3046 to 66 in 2014. In the same time interval the number of model (below MSA) values increased from 5% to 50%. As for the individual values of ^{137}Cs activity concentrations the maximum value in 1986 in one of the sampling locations 560 Bq.L^{-1} was found. In 2014 no activity concentration exceeded 0.3 Bq.L^{-1}.

Annual GMs of ^{137}Cs activity concentrations in milk are shown in Figure 1. Time course of ^{137}Cs activity concentrations in milk starts with the GM of 12 Bq.L^{-1} in 1986. Rapid decrease in the first years after the Chernobyl accident was

Table 1. Regression equation of the curves and the values of the effective (T_{EF}) and environmental (T_{EN}) half-lives of ^{137}Cs in milk in the Czech Republic between 1986 and 2014 calculated from them.

^{137}Cs in milk – whole CR			
Years	1986–1988	1989–1992	1992–2014
Break [year]	0	2.6 ± 0.1	6.4 ± 0.3
Equation	$y = 19.3e^{-1.8x}$	$y = 0.53e^{-0.42x}$	$y = 0.038e^{-0.0061x}$
λ_{EF} [year^{-1}]	1.80 ± 0.30	0.42 ± 0.05	0.006 ± 0.003
T_{EF} [year]	0.4	1.7	n/a
T_{EN} [year]	0.4	1.8	n/a

approximately from 1992 followed with almost constant development of activity concentrations on the level of hundredths of Bq.L^{-1} and the minimum value is 0.02 Bq.L^{-1} in 2014. The GSDs for annual GMs for the whole interval 1986–2014 were about from 2 to 4.

The values of ^{137}Cs activity concentrations were used to generate a best fit curve starting at the day of the Chernobyl accident (26 April 1986). Using programme R, the first break point on the curve to (2.6 ± 0.1) y and the second break point (6.4 ± 0.3) y after the accident was determined. As the first and the second segments are very short, with a small number of the annual GMs, individual measured activities and model values to find the fits for all three segments were used.

The best fit equations of the curves and effective (T_{EF}) and environmental (T_{EN}) half-lives calculated from them are presented in Table 1. The activity concentrations were decreasing from 1986 to 1988 with the T_{EF} = 0.4 y and T_{EN} = 0.4 y (λ_{EF} = [1.80 ± 0.30] y^{-1}). Between 1989 and 1992 the decrease slowed and both half-lives increased to T_{EF} = 1.7 y and T_{EN} = 1.8 y (λ_{EF} = [0.42 ± 0.05] y^{-1}). Since 1992 the level of ^{137}Cs activity concentration in milk has been stable. The effective half-life has not been calculated because it does not comply with the condition $T_P > T_{EF}$. Estimation of the coefficient λ_{EF} is in this case loaded with high uncertainty (approximately 50%, λ_{EF} = [0.006 ± 0.003] y^{-1}) caused by its low level and a relatively short observation interval.

For comparison, in Austria the environmental half-life values for ^{137}Cs in milk were found to be 4.5 y – 15 y in the period 1988–2007 and 4.3 y – 29.9 y for the shorter period 1993–2007. Both half-lives markedly depend (Lettner *et al.*, 2009) on the altitude where cows live. According to other sources the reduction of ^{137}Cs content in dairy milk in four variedly contaminated areas in Finland was slower than in the Czech Republic and different in unevenly contaminated areas. The environmental half-life values there were estimated (Kostiainen, 2005) to be 1.8 y – 3.2 y in the period 1987–1989 and 4.7 y – 8.7 y for the period 1990–2003. For milk sampled in 1987–1993 from ten farms in central Sweden the effective half-life was in the interval 0.8 y – 2 y (Gunnel *et al.*, 1995). Only a rough comparison of the values found by us and the values published is possible because individual papers differ in the survey period length and the number of break points on the best fit curves.

3.2 Strontium-90

Since 1965 altogether 537 milk samples were analysed in the whole Czech Republic. Out of that 19% were lower than the MSA. The number of samples increased after the Chernobyl accident in 1986, in the period 1986–1996 approximately 40 milk samples were analysed annually. No value was under MSA in this period. Since 1997 the number of samples has been optimised and gradually reduced to 14 samples per year in 2014. Since 2008 some single and later most values have been under MSA, altogether 64% of values in 2014.

Calculated annual geometric means of ^{90}Sr activity concentrations in milk fitted with a curve are presented in Figure 2. The highest activity concentration of ^{90}Sr in milk comes from 1965 and equals to 0.69 Bq.L^{-1}, in the following years first a sharp and from 1971 a slow decrease were observed. It is evident from the plot (Fig. 2) that ^{90}Sr activity concentration in milk promptly decreased after the atmospheric nuclear test moratorium (UNSCEAR, 1982) in 1963 (France signed in 1974, China did not sign). The decrease was caused by lower release of ^{90}Sr into the environment and its consequent transfer to the dairy milk cows and their milk. In 1986 due to the Chernobyl accident ^{90}Sr was further released into the environment, which resulted in an abrupt increase in GM values from 0.06 Bq.L^{-1} to 0.10 Bq.L^{-1} in the Czech Republic. As a consequence, the number of sampling sites and milk samplings were extended. Calculated GMs in the following years were thus loaded with higher uncertainty due to larger dispersion of the detected values. From the new maximum in 1986 the activity concentrations were decreasing and after approximately 10 years they reached the values observed before the Chernobyl accident, approximately 0.06 Bq.L^{-1}. Such a time course corresponds well to the activity concentrations found in the neighbouring states, mentioned earlier in the first chapter.

For ^{90}Sr, programme R suggested the break points in (9.3 ± 0.9) y and in (26.7 ± 3.3) y from the beginning in 1965, hence in the years 1974 and 1991. Comparing the suggestion of the programme with the course of events controlling ^{90}Sr content in the environment, it was found that the programme could not work appropriately with the abrupt, yet small increase in 1986 due to the Chernobyl accident. Therefore, the years 1974 (connection with the atmospheric nuclear test moratorium) and 1986 (the Chernobyl accident) were selected as the break points for the curves and further fits were conducted manually *via* linear regression in Microsoft Excel. The resulting regression equations of the curves and T_{EF} and T_{EN} calculated based on them are presented in Table 2.

3.3 Estimation of doses

The consumption rate of milk and milk products in terms of milk equivalent in Czech Republic is known (ČSÚ, 2012, 2014) for years 1965–2014, with average about 218 L. (year.person)$^{-1}$, minimal value 179.3 L. (year.person)$^{-1}$ and maximal value 252.0 L. (year.person)$^{-1}$.

The data for ^{137}Cs content in milk before Chernobyl accident do not exist and only rough estimation is possible based on UNSCEAR data (UNSCEAR, 1977). Maximal GM for ^{137}Cs activity concentration was determined in the year 1986. For adult population with dose coefficient (ICRP Publication 119, 2012) 1.3.10^{-8} Sv.Bq^{-1} annual committed effective dose 26 μSv.year^{-1} was calculated using equation (3). Since then, annual doses decreased stepwise to about 0.065 μSv.year^{-1}.

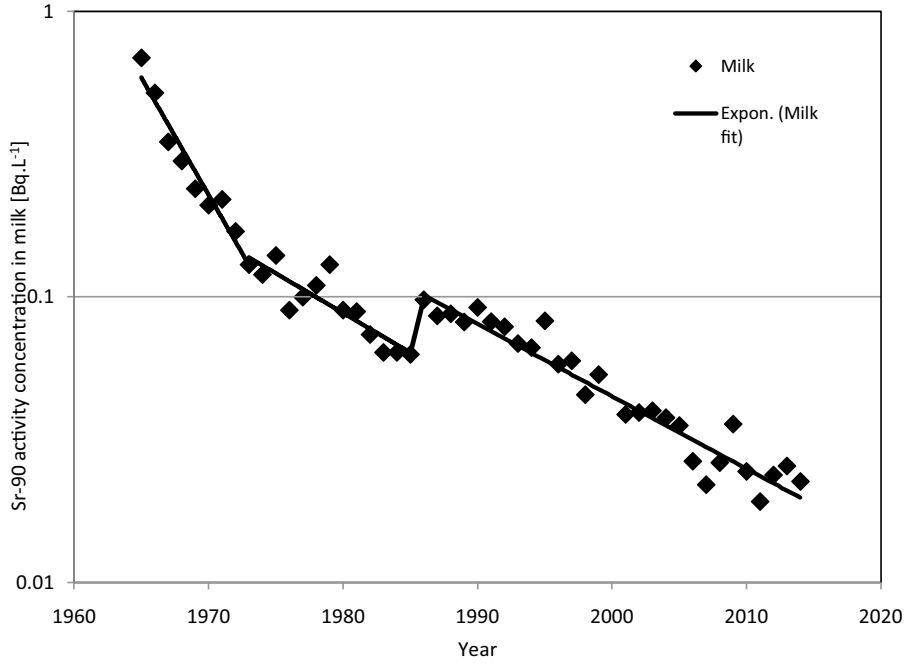

Fig. 2. The annual geometric means of ^{90}Sr activity concentrations in milk in the Czech Republic.

Table 2. Regression equation of the curves and the values of the effective (T_{EF}) and environmental (T_{EN}) half-lives of ^{90}Sr in milk between 1965–2013.

^{90}Sr in milk – whole CR			
Years	1965–1973	1974–1985	1986–2013
Point "0" in year	1965	1974	1986
Equation	$y = 0.59e^{-0.189x}$	$y = 0.14e^{-0.067x}$	$y = 0.10e^{-0.058x}$
λ_{EF} [year^{-1}]	0.19 ± 0.02	0.07 ± 0.01	0.06 ± 0.01
T_{EF} [year]	3.7	10.3	12.1
T_{EN} [year]	4.2	16.2	20.4

Due to maximal GM for ^{90}Sr activity concentration in milk, 0.69 Bq.L^{-1}, the highest annual dose from ^{90}Sr ingestion of milk was in the year 1965, namely 4.2 μSv.year^{-1} for adult population (dose coefficient (ICRP Publication 119, 2012) 2.8.10^{-8} Sv.Bq^{-1}). At present it is less than 0.24 μSv.year^{-1}.

As the contamination of Czech territory after the Chernobyl accident was not homogeneous and there are also groups of people with higher consumption rate of milk and milk products, individual doses could also be higher up to one order of magnitude. Doses to small children were also higher. Apart of this, it has to be born in mind that after the Chernobyl accident, dominant contributors to ingestion doses from milk were radioisotopes of iodine and also ^{134}Cs contributed significantly.

4 Conclusions

The activity concentration mean of ^{137}Cs in milk in the Czech Republic decreased from 12 Bq.L^{-1} below 0.5 Bq.L^{-1} during three years after the Chernobyl accident and since then it has remained on very low level. Since 1990 GM of activity concentrations of ^{137}Cs in milk appear under the level of 0.1 Bq.L^{-1} and since 1992 the content of ^{137}Cs in the forage components and hence in milk is stable except for the decrease caused by radioactive decay.

Activity concentrations of ^{90}Sr in milk have dropped from initial tenths Bq.L^{-1} to hardly detectable values during 1965–2014. Environmental and effective half-lives were calculated for the periods 1965–1973, 1974–1985 and 1986–2014, which are essential from the point of view of ^{90}Sr contamination. All T_{EF} and T_{EN} are lower than the physical half-life of ^{90}Sr, so the radionuclide disappears from milk faster than due to its physical decay. The effective and environmental half-lives are very close in the second and the third monitoring periods – the possible explanation is a low share of ^{90}Sr contained in the environment in the Czech Republic due to the Chernobyl accident, compared to the whole amount of ^{90}Sr.

The tests of nuclear weapon can be considered the largest source of man-made radionuclides in the environment in the Czech Republic.

The annual doses from ingestion of ^{137}Cs and ^{90}Sr are small. However, knowledge of the time course of activity of long-lived radionuclides in milk is important from the view of possible increases following irregular releases of radionuclides into environment.

Acknowledgement. This work was supported by Project of Ministry of Interior of the Czech Republic MV-25972-39/ OBVV- 2010. We acknowledge the State Office for Nuclear Safety for the possibility to exploit the data obtained within the frame of the Radiation Monitoring Network.

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Cite this article as: Bartusková M, Škrkal J, Schlesingerová E, Bečková V, Malátová I. 2017. Doses from Cs-137 and Sr-90 to Czech population due to milk consumption. *Radioprotection* 52(3): 171–176