RADIOCAESIUM ACTIVITY CONCENTRATIONS IN MILK IN THE REPUBLIC OF CROATIA AND DOSE ASSESSMENT

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Abstract. Results of systematic measurements of radiocaesium activities in milk after the Chernobyl nuclear accident are summarized. ^{137}Cs fallout activity affects milk activity, the coefficient of correlation being 0.89. The ^{137}Cs activities in milk in Croatia are log-normally distributed, reflecting the exponential decrease of activity. After the Chernobyl nuclear accident the ^{134}Cs: ^{137}Cs activity ratio in milk was $\approx$0.5, and did not differ from that for other environmental samples. The dose due to radiocaesium ingestion by milk consumption was estimated for the Croatian population, the annual collective equivalent dose being approximately 205 manSv in 1986 and 1.5 manSv in 1994.

Key words: Chernobyl accident, dose assessment, milk, radiocaesium

1. Introduction

The dominant route for the introduction of artificial radionuclides in the environment until the nuclear accident at Chernobyl, Ukraine, on 26 April 1986 (UNSCEAR, 1988) has been radioactive fallout resulting from atmospheric nuclear weapon tests. Atmospheric nuclear explosions that have been conducted since 1945 were specially intensive in the 1960s, followed by similar, but smaller scale tests performed by the Chinese and French in the 1970s and afterwards. Therefore, radioactivity of most environmental samples, including those that enter human food chain, could be expected to be in correlation with fallout activity (i.e. surface deposit in Bqm^{-2}).

Among anthropotropic radioactive nuclides ^{137}Cs and ^{90}Sr have been regarded as the fission products of potential hazard to living beings due to the unique combination of their relatively long half-lives, and their chemical and metabolic properties resembling those of the sodium and potassium respectively. Milk, containing both ^{137}Cs and ^{90}Sr is therefore the sensitive indicator for presence of fission products in the environment. In addition, milk as the very important foodstuff in dietary habits, is potentially a major source of radioactive contamination by ingestion. Therefore, in the program of monitoring of radioactive contamination of human environment in Croatia (Bauman *et al*., 1986–1995; Popović, 1964–1978) investigations of radiocaesium in milk take significant part.

The nuclear accident at Chernobyl did not cause any significant increase in ^{90}Sr activity in environmental samples in Croatia, contrary to radioactive isotopes of

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caesium, (mainly ^{137}Cs), commonly referred as radiocaesium. Unlike the debris from the atmospheric testing of nuclear weapons, the radionuclides that originated from the Chernobyl accident were not released directly into the upper atmosphere. As the result of the release mechanism and the prevailing meteorological conditions at the time, the less volatile components of the Chernobyl debris (e.g. ^{90}Sr) were deposited closer to the accident location than the more volatile constituents (i.e. radiocaesium) (UNSCEAR, 1988; Aarkrog, 1988). Thus, ^{90}Sr was not subjected to the global dispersion processes, being deposited to the Earth's surface within a period of a few days to a few weeks after the accident. In addition, the late spring and early summer of 1986 in Croatia were rather dry, leading to relatively low direct radioactive deposition, which was especially true for the Adriatic region (Franić and Bauman, 1993). Therefore, as the consequence of the Chernobyl accident unlike ^{137}Cs, ^{90}Sr did not impose any additional threat to the Croatian population via the food chain, including milk.

Considering the long half-life of ^{137}Cs (30 yr), dose assessment requires understanding of its behaviour in the environment. Therefore, research on the fate of ^{137}Cs has been largely focused on determining its activity concentrations in fallout, soil and along human exposure pathways.

2. Material and Methods

Fallout samples were collected quarterly (four times per year) in the city of Zagreb. Milk samples from Zagreb dairy, 1 L every day, were obtained commercially. An aliquot (10 L) of cumulative monthly sample of liquid milk was placed in the Pyrex beaker and heated to 100 °C in a fumehood, until well charred. The carbonaceous material was dried in an oven and ashed at 450 °C.

A gamma-ray spectrometry system based on a low-level Ge(Li) detector (efficiency 18.7% at 1.33 MeV ^{60}Co; resolution (FWHM) 1.82 keV at 1.33 MeV ^{60}Co) coupled to a computerized data acquisition system (4096-channel pulse height analyser and personal computer) was used to determine radiocaesium levels in the samples from their gamma-ray spectra. Samples were measured in cylindrical plastic containers of 110 cm^3 volume that were placed directly on the detector. Counting time depended on sample activity concentration, but was never less than 60000 seconds.

The efficiency calibration was carried out using sources provided by the International Atomic Energy Agency (IAEA) and World Health Organization (WHO). Intercalibration was performed also on samples provided by the IAEA and WHO as part of the international intercalibration programme.

3. Results and Discussion

3.1. ^{137}Cs FALLOUT ACTIVITY CONCENTRATIONS

Radioactive fallout from highly radioactive air plumes that originated from the damaged Chernobyl nuclear reactor was spread and transported over Europe (UNSCEAR, 1988; UNEP, 1991) and radionuclides attributed to the Chernobyl accident were soon detected in Croatia. As in the rest of Europe, only a few of the radionuclides (radiocaesium, i.e. caesium radioisotopes ^{137}Cs and ^{134}Cs) were responsible for most of the radioactivity (IAEA, 1986). The ^{137}Cs fallout activities after the Chernobyl nuclear accident were much higher than in mid 1960s, after a period of intensive atmospheric nuclear weapon tests. The baseline level of ^{137}Cs wet fallout activity concentration in pre-Chernobyl 1985 for the Zagreb area was 2 Bqm^{-3} leading to a surface deposit of 1.5 Bqm^{-2}.

However, in May 1986, in the Zagreb area the ^{137}Cs surface deposit was 6200 Bqm^{-2}, while in 1994 it decreased to pre-Chernobyl values, being only 1.3 Bqm^{-2}. Once deposited to ground, radiocaesium very slowly penetrates to deeper layers in undisturbed (unplowed) soils. The half-depth of radiocaesium penetration, depending on the soil type, in undisturbed soils is on average less than five centimetres. (Korun *et al.*, 1990; Franić and Lokobauer 1994). Therefore, besides the common spring activity peak in fallout, which can be attributed to meteorological phenomena, radiocaesium is every spring by grass vegetation by root uptake carried to the soil surface (UNSCEAR, 1988). As a consequence, grass vegetation is usually more active in springtime, compared to other seasons. However, it should be noted that the foiler deposition of radiocaesium, especially in the immediate period after the Chernobyl accident has been the important pathway to the grazing animals and therefore milk as well.

Effective residence times of ^{137}Cs in fallout in the Zagreb area for the pre-Chernobyl and post-Chernobyl periods were estimated to be 3.7 and 0.7 yr respectively (Franić, 1992a), reflecting the release mechanism to the atmosphere. By atmospheric nuclear weapon tests ^{137}Cs was released in the stratosphere where the mean residence time of radioactive fallout is more than two yr (UNSCEAR, 1982). As a consequence of the Chernobyl accident by explosions and subsequent burning fire of a graphite moderator, radioactive material was dispersed only to the troposphere where the mean residence time is a few months (UNSCEAR, 1982).

3.2. ^{137}Cs ACTIVITY CONCENTRATIONS IN MILK

After the Chernobyl accident, ^{137}Cs activity concentrations in milk for 1986–1994 period dropped exponentially with time from 46000 Bqm^{-3} in May 1986 to 270 Bqm^{-3} in December 1994 (Bauman *et al.*, 1986–1995). From that exponential drop, the lognormal distribution of ^{137}Cs activity concentrations in milk is to be expected. However, on the cumulative probability plot, bimodal behaviour is

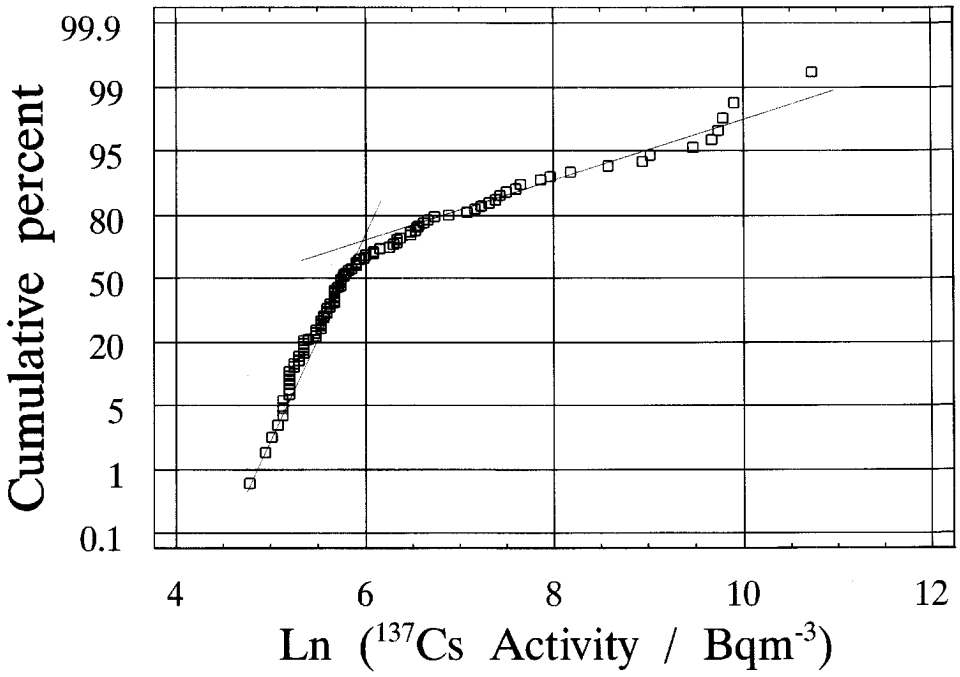

Figure 1. Cumulative probability plot of ^{137}Cs activity concentrations in milk.

exhibited. Exponential decrease was initially very fast, compared with the rather slow decrease rate few years after the accident (Figure 1).

For comparison, activity concentration of naturally occurring ^{40}K in milk in overall period ranged from 49000 to 58000 Bqm^{-3}. In international intercomparison performed by World Health Organization, International Reference Center for Radioactivity (IRC/WHO) in 1992, in which participated 48 laboratories worldwide, mean ^{40}K activity concentration in a fresh milk sample was 52.3 Bq/L (WHO, 1994).

The ^{137}Cs milk activity concentrations for the periods May 1986 – June 1988 and July 1988 – December 1994 are well fitted to lognormal distribution with geometric mean 4653 and 309 Bqm^{-3} respectively and geometric standard deviations of 3 and 2 Bqm^{-3}. In both cases, the hypothesis that the parent distribution is lognormal cannot be rejected, even at a significance level of 0.15, according to Kolmogorov-Smirnov test.

The retention curve for radiocaesium in mammals (e.g. in humans) has at least two exponential terms (ICRP, 1978; Voigt *et al.*, 1989). Although the biological elimination half-life is as short as several months, due to continuous exposure to fallout (pseudo-equilibrium), radiocaesium is routinely found in cow's milk even one decade after the Chernobyl accident. Therefore, the different mean residence times of radiocaesium in milk reflected in bimodality in Figure 1, are related

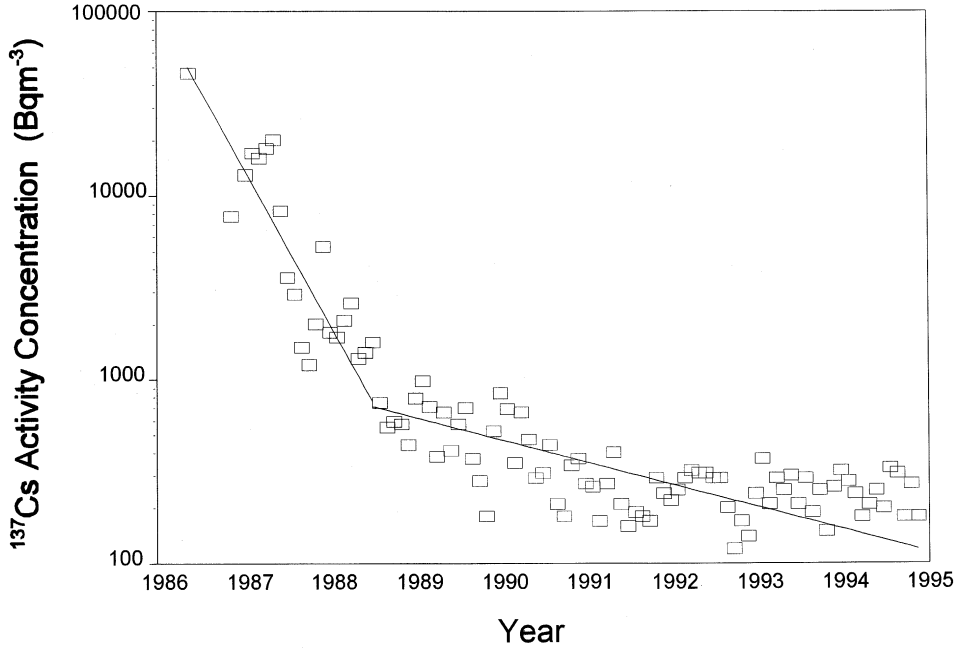

Figure 2. Measured and fitted ^{137}Cs activity concentrations in milk for 1985–1994 period.

not only with two terms in the radiocaesium retention curve, but also with other factors like pseudo-equilibrium achieved, seasonal activity variations, weathering processes etc.

By plotting ^{137}Cs milk activity data against time scale and fitting to the exponential curve

$$A_m(t) = A_m(0)e^{-kt} \tag{1}$$

where $A_m(t)$ and $A_m(0)$ are ^{137}Cs activity concentrations in milk (Bqm^{-3}) at time t and zero respectively, the decay constant k was determined.

For period May 1986 – June 1988 k is 1.92 yr^{-1} and for period July 1988 – December 1994 k is 0.28 yr^{-1}. Rough estimates of mean residence time of ^{137}Cs in milk, being reciprocal value of k is therefore 0.5 and 3.6 yr for respective periods (Figure 2). Therefore, when multiplied by ln(2), half residence times for respective periods are 0.36 yr (130 d) and 2.5 yr.

For comparison, the ^{137}Cs biological half-life modelled using data from controlled feeding experiment was 1 to 2 days for fast and 10 to 20 days for slow component (Voigt *et al.*, 1989). However, for the milk of cows fed under field condition, ^{137}Cs half-time for slow component was for the immediate period after the Chernobyl accident up to 175 days (CEC, 1992). Pseudo-equilibrium achieved after that time affects the mean residence time. In Croatia, cows often graze outdoors, therefore being constantly exposed to residual radiocaesium. In addition, transient

increases in the ^{137}Cs milk activity (Figure 2) can be explained by a variety of environmental physical factors that naturally fluctuate.

^{137}Cs activity in milk is in good correlation with fallout activity, r = 0.89, with P(t) $<$0.001 for 32 degrees of freedom. Thus, milk activity can be modelled as

$$A_m(t) = 6.97A_f(t) + 1008.00 \tag{2}$$

where

$A_m(t)$ = ^{137}Cs activity concentration in milk (Bqm^{-3});
$A_f(t)$ = ^{137}Cs fallout activity deposited on earth (Bqm^{-2}).

It should be noted that in spite of very good correlation, between fallout and milk ^{137}Cs activities, milk activity strongly depends also on the ^{137}Cs activity of soil, and therefore on the activity of animal feed. The other factors affecting the radiocaesium activity concentrations in animal feed are foiler deposition (external deposition on vegetation) and internal vegetation uptake of ^{137}Cs from soil at a plant root zone.

3.3. ^{134}Cs MILK ACTIVITY CONCENTRATIONS

In May 1986, the presence of ^{134}Cs (half-life of 2.06 yr) was for the first time detected in the environment in Croatia. It could be immediately attributed to the Chernobyl nuclear accident, because it is not produced in the nuclear weapons explosions. This means that the nuclide that would produce ^{134}Cs by β decay following production in the fission process (^{134}Xe) is, itself, stable. Therefore, the mass 134 fission product decay chain stops with ^{134}Xe and ^{134}Cs is not formed. ^{134}Cs is found, however, in reactor fission product inventories due to long irradiation times for nuclear fuel (typically three years residence time in the reactor core). This permits the buildup of the stable end-product nuclide ^{133}Cs in the core and the corresponding neutron capture by the ^{133}Cs results in the ingrowth of significant quantities of radioactive ^{134}Cs. This does not occur in the weapons blast that is over in milliseconds. Therefore, the sudden presence of ^{134}Cs in the environment clearly indicates the nuclear Chernobyl accident.

The amount of caesium released after the reactor explosion at Chernobyl was 3.7×10^{16} Bq of ^{137}Cs (13% of total reactor inventory) and 1.9×10^{16} Bq of ^{134}Cs (10% of total reactor inventory) (IAEA, 1986). Thus, the initial value for the ^{134}Cs:^{137}Cs activity ratio in May 1986 was 0.51.

The environmental pathways and consequent impact of some chemical elements sometimes depend on chemical or isotope form, (tritium being the most obvious example). However, ^{134}Cs and ^{137}Cs being the most conservative in behaviour, have undergone no selective removal in transit between the accident site at Chernobyl and Croatia as their activity ratio has not been altered.

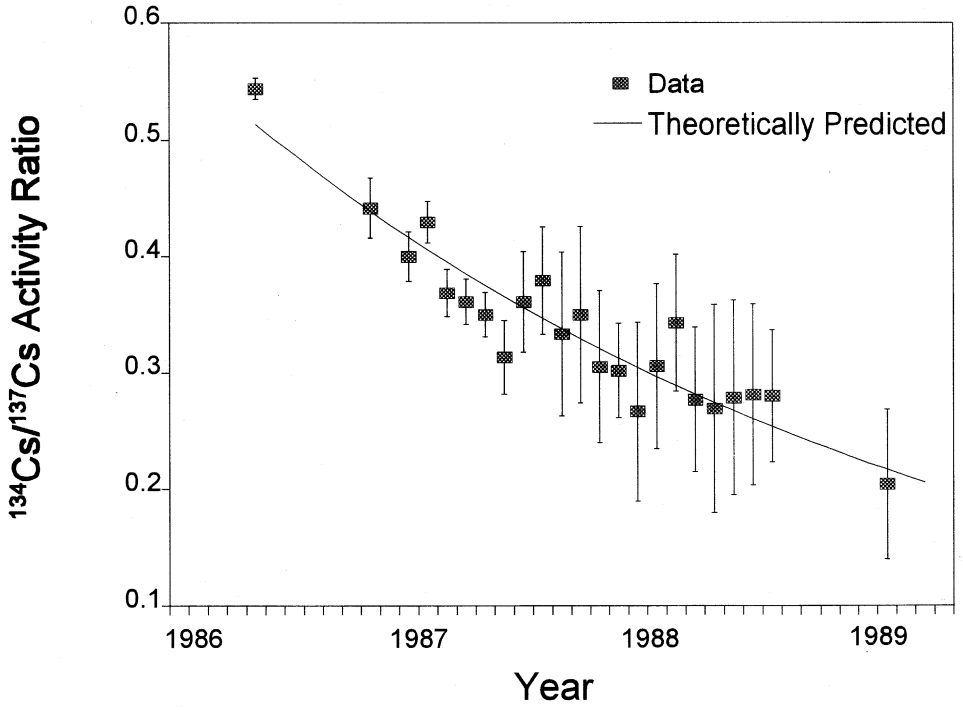

Figure 3. ^{134}Cs: ^{137}Cs activity ratio in milk samples.

As the half-life of ^{137}Cs compared to the half-life of ^{134}Cs is much longer (30.14 yr), the ^{134}Cs:^{137}Cs activity ratio *R(t)* decreased, due to differential radioactive decay, from the initial value of 0.5 according to relation

$$R(t) = \frac{1.9\times10^{16}}{3.7\times10^{16}}\times e^{\ln(2)\times t\times\left(\frac{1}{t_1}-\frac{1}{t_2}\right)} \tag{3}$$

where

t is time elapsed after the Chernobyl accident;
t_1 and t_2 are physical half-lives for ^{137}Cs and ^{134}Cs.

Thus in 1986 the ^{134}Cs:^{137}Cs activity ratio in milk was 0.50±0.10 and in 1987 0.42±0.06. On Figure 3. are shown observed ^{134}Cs: ^{137}Cs activity ratios in milk and theoretically predicted curve according to Equation (3).

The same ^{134}Cs: ^{137}Cs ratio, decreasing according to Equation (3) was found in most of the environmental samples (Franić *et al.*, 1989; Franić *et al.*, 1991), the only exception being mushrooms (Franić *et al.*, 1992b). In the mushrooms, (generally having shallow mycelium) the excess ^{137}Cs the pre-Chernobyl fallout, affected the ^{134}Cs: ^{137}Cs activity concentration ratios.

In 1988 and afterwards, contamination of milk by the fallout ^{134}Cs and ^{137}Cs that originated from the Chernobyl nuclear accident was detectable only at a very low level (UNSCEAR, 1988).

3.4. RADIOCAESIUM DOSES INCURRED BY MILK CONSUMPTION

In the Republic of Croatia, consumption of milk and dairy products being approximately 100 L per year per person (CEDCZ, 1987; CEDCZ, 1993), can lead to significant radiation doses. The equivalent dose incurred due to milk consumption over certain period, depends on the activity of a radionuclide and on the quantity of milk consumed. The dose can be expressed as

$$H = C \int \sum_k D_{cf}(k) A_k(t) dt \tag{4}$$

where

H = the equivalent dose in Sv;
C = total annual *per caput* consumption of milk (100 Ly^{-1});
$D_{cf}(k)$ = the dose conversion factor for radionuclide *k*;
A_k = the mean activity concentration of radionuclide *k* in milk (BqL^{-1}).

$D_{cf}(k)$, i.e. the equivalent dose per unit input, which converts the ingested activity to the equivalent dose (for age greater than 17 yr) is 1.3×10^{-8} SvBq^{-1} and 1.9×10^{-8} SvBq^{-1} for ^{137}Cs and ^{134}Cs respectively (IAEA, 1996).

For equal time increments of one month, integral from Equation (4) can be replaced by the respective sums of average monthly activity concentrations. Neglecting other radiocaesium isotopes apart from ^{134}Cs and ^{137}Cs, the total dose due to those two radionuclides in 1986 was 43.7 μSv, 58.8% being due to ^{137}Cs and 41.2% due to ^{134}Cs. However, in 1994 the total dose was only 0.3 μSv, 94.4% being due to ^{137}Cs (Figure 4)

Assuming similar radiocaesium milk activity concentrations in other Croatian regions, the collective equivalent dose for Croatian population (4.7 million inhabitants) decreased from 205.4 manSv in 1986 to only 1.5 manSv in 1994.

4. Conclusions

The ^{137}Cs activities in milk in Croatia are log-normally distributed, reflecting the exponential decrease of activity. However, due to initially very fast and subsequent slower reduction of milk activity concentration after the Chernobyl nuclear accident, cumulative probability plot of ^{137}Cs activities exhibit bimodal behaviour.

Generally, a few years after the Chernobyl nuclear accident the activities of fission radionuclides, ^{137}Cs and ^{134}Cs in milk in Croatia were relatively low, amounting to less than 1% of the naturally occurring ^{40}K activity.

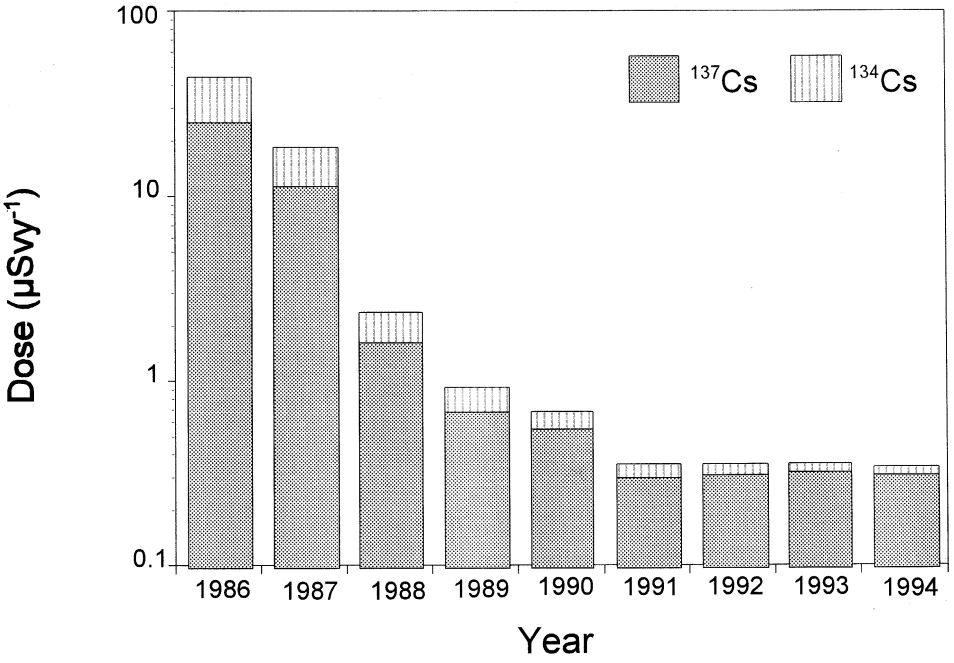

Figure 4. The equivalent dose for an adult person, incurred by ingestion of ^{137}Cs and ^{134}Cs by consumption of 100 Ly^{-1} of cow milk.

Doses due to radiocaesium from milk consumption are small in spite of large consumption of milk by the Croatian population, collective equivalent dose for Croatian population in 1994 being 1.5 manSv. For comparison, background collective equivalent dose for early 1980s was ≈0.2 manSv.

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