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Radiostrontium Contamination of Milk in the Republic of Croatia

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INTRODUCTION

The dominant route for the introduction of artificial radionuclides into the environment until the nuclear accident in Chernobyl, Ukraine, on 26 April 1986 has been the radioactive fallout resulting from the atmospheric nuclear weapon tests. Atmospheric nuclear explosions have been conducted since 1945 and were specially intensive in 1960s, i.e., before a nuclear moratorium became effective. However, similar, but smaller tests were performed by the Chinese and French also 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 the anthropotropic radionuclides present in global fallout, ^{137}Cs and ^{90}Sr have been regarded as the fission products of a major potential hazard to living beings due to the unique combination of their relatively long half-lifes, and their chemical and metabolic properties resembling those of the potassium and calcium respectively. Milk, containing both potassium and calcium 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. Consequently, in the program of monitoring of radioactive contamination of human environment in Croatia (1, 2, 3, 4) investigations of radiostrontium and radiocaesium in dairy milk take significant part. In this paper are summarized the results of long-term systematic measurements of radiostrontium activities in milk.

MATERIAL AND METHODS

Fallout samples were collected monthly at the location of the Institute for Medical Research and Occupational Health (45° 50' 07.3" N, 15° 58' 58.7" E). The funnels used for fallout collection had 1 m^2 area. The precipitation amount was measured by Hellman pluviometer.

Milk samples from Zagreb dairy, 1 L every day, were obtained commercially.

In 1960s ^{90}Sr was determined by the conventional radiochemical analysis with fuming nitric acid separation. From 1970 to present, ^{90}Sr was determined by extraction with tributyl phosphate, except in the year of the Chernobyl nuclear accident, when fuming nitric acid was also used.

After the radiochemical treatment of fallout and milk samples, the radioactivity of ^{90}Sr was determined by beta counting its decay product, ^{90}Y, in a low-level anti-coincidence shielded GM counter. Counting time depended on ^{90}Sr activity concentration in samples, but was never less than 60,000 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 programmes.

RESULTS AND DISCUSSION

The highest recorded ^{90}Sr activity deposited in Croatia, being 1055.6 Bqm^{-2}, was recorded in Zagreb in 1963. Ever since that time ^{90}Sr fallout activities exponentially decreased, until the nuclear accident in Chernobyl that again caused a minor increase of ^{90}Sr activity in fallout (Figure 1). However, due to the prevailing meteorological conditions at the time after the accident that influenced the formation and spreading direction of Chernobyl plumes, in Croatia the ^{90}Sr peak was recorded only in fallout collected in Zagreb. 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 meteorological conditions the refractory components of the Chernobyl debris (e.g., ^{90}Sr) were deposited closer to the accident location than the more volatile constituents (e.g., radiocaesium) (5, 6). 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 contamination of environment, which was

especially true for the Adriatic region (7, 8). Consequently, the nuclear accident at Chernobyl did not cause any major increase in ^{90}Sr activity in environmental samples in Croatia, contrary to radioactive isotopes of caesium. It should be noted that the increased ^{90}Sr fallout activities were recorded mainly in May 1986, leading to the surface deposit on annual level of 211.3 Bqm^{-2} in Zagreb area (9). In 1987, ^{90}Sr fallout activities dropped to only 12.7 Bqm^{-2}, i.e., on pre-Chernobyl values.

The peak value of ^{90}Sr in milk (about 1420 Bqm^{-3}) was recorded in 1964 (Figure 1), after the most intensive nuclear weapon tests. As in the case of fallout ^{90}Sr levels were exponentially decreasing from 1964 to 1986, when the Chernobyl accident again caused ^{90}Sr activity peak, the annual surface deposit in 1986 being 431 Bqm^{-3}. It should be noted that the radiostrontium activities in milk in Croatia are log-normally distributed, reflecting the exponential decrease of activity.

By regression analysis it can be demonstrated that the ^{90}Sr fallout activity affects milk activity, the coefficient of correlation being 0.90. Therefore, from fallout data ^{90}Sr activities in milk can be modeled as:

$$A_{milk}(t) = 1.24 \times A_{fallout}(t) + 235.3 \qquad /1/$$

where:
$A_{milk}(t)$ is the ^{90}Sr activity in milk in Bqm^{-3} and
$A_{fallout}(t)$ is the ^{90}Sr fallout activity in Bqm^{-2}.

After the Chernobyl accident, the ^{90}Sr activities in milk very quickly decreased to pre-Chernobyl values, the activity in 1987 being 166 Bqm^{-3}. For comparison, in 1985 was recorded activity of 159 Bqm^{-3}. The lowest ^{90}Sr milk activity was recorded in 1997, being 76 Bqm^{-3}, while in 1998 it was 84 Bqm^{-3}. Such variations in activity of environmental samples can be attributed to the number of environmental parameters that naturally fluctuate.

^{90}Sr transfer from fallout to milk

To assess the ^{90}Sr transfer from fallout to milk was applied the mathematical model for food recommended by UNSCEAR (10), which was also previously tested on milk samples collected in Croatia (11). The function has the following form:

$$A_k(t) = b_1 U_k(i) + b_2 U_k(i-1) + b_3 \sum_{m=1}^{4} e^{-\mu m} U_k(i-m) \qquad /2/$$

where:
$A_k(t)$ is the activity concentration of radionuclide *k* in food (milk), the unit for $A_k(t)$ being Bqkg^{-1},
$U_k(i)$ is the fallout deposition rate of radionuclide *k* in a year *i* (Bqm^{-2}y^{-1}),
$\Sigma e^{-\mu m}$... is the cumulative fallout deposit for radionuclide *k* as the result of deposition in previous years (Bqm^{-2}y^{-1}),
μ^{-1} is the effective mean life of available ^{90}Sr in milk,
b_1,b_2, b_3 are the factors which can be derived from reported data by regression analysis. The unit is Bqykg^{-1}/(Bqm^{-2}).

The equation /2/ assumes the chain model for the transfer of radionuclides between environmental compartments (C_0 ...C_5), linking the input to the atmosphere to the dose in man:

Input (C_0) → Atmosphere (C_1) → Earth's surface (C_2) → Diet (C_3) → Tissue (C_4) → Dose (C_5)

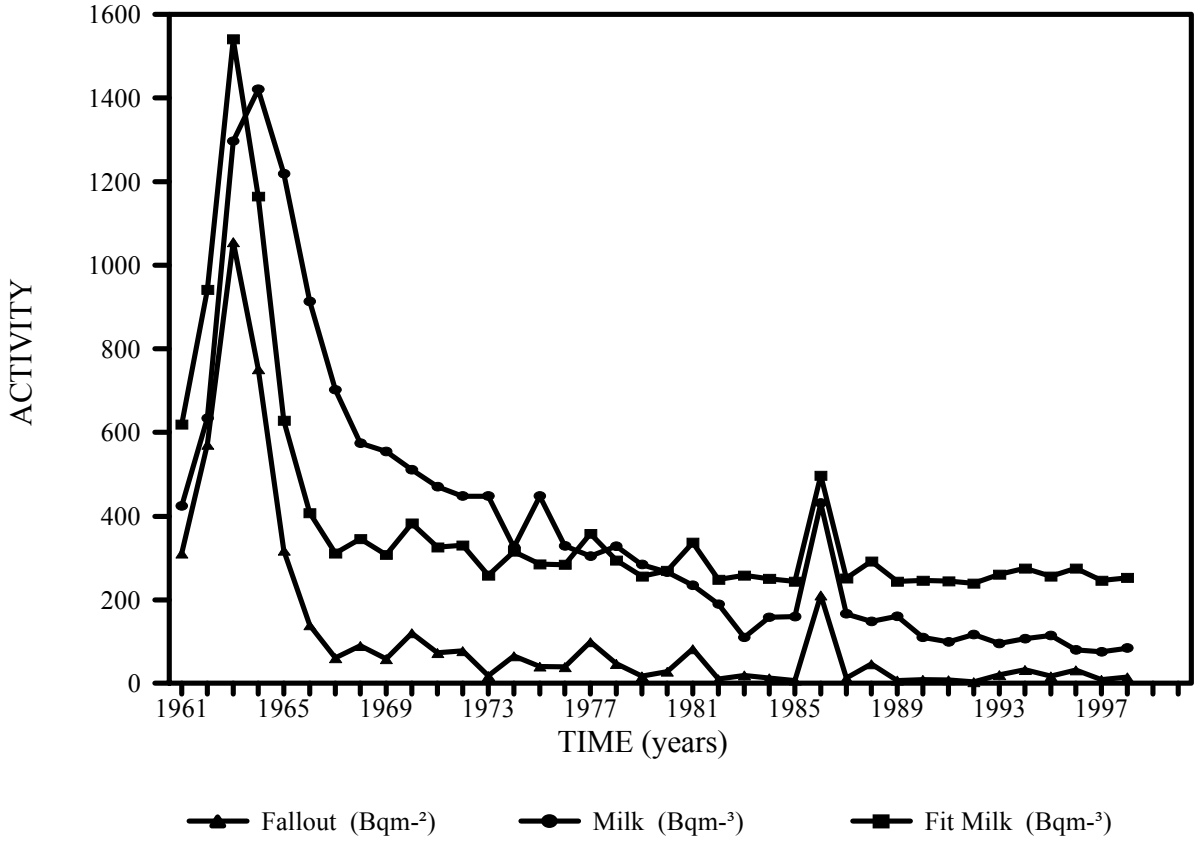

Figure 1. ^{90}Sr fallout and milk activities and modeled milk activities using equation /1/.

The physical meaning of terms in model /2/ is as follows: the first term (*rate factor*) describes direct deposition and transfer, the second term (*lag factor*) describes contamination through fallout from previous year, and the third term (*land factor*) reflects the contamination from fallout deposition accumulated from all preceding years, the exponential describing the combined physical decay and any other decrease in availability of considered radionuclide due to various other processes (like penetration in deeper soil layers, dilution etc.).

Regression analysis gives the following values: $b_1 = 1.00$, $b_2 = 0.09$, $b_3 = 0.97$ Bqykg^{-1}/(Bqm^{-2}) and $\mu = 0.32$ yr^{-1}, the coefficient of correlation between actual data and data predicted by model being 0.91. The effective mean life of ^{90}Sr in milk defined as $1/\mu$ is 3.12 years. The ^{90}Sr activity concentrations in milk and the fit obtained by equation /2/ are shown in Figure 2.

The transfer between successive steps in the pathway chains is described by transfer coefficients, which relate infinite time integrals of activity concentration in the relevant compartments. Thus, P_{23}, the transfer coefficient from fallout deposition (compartment 2) to diet (compartment 3) is given by the following equation:

$$P_{23} = \frac{\int_0^4 A_k(t)dt}{\int_0^4 \vartheta_k(t)dt} \qquad /3/$$

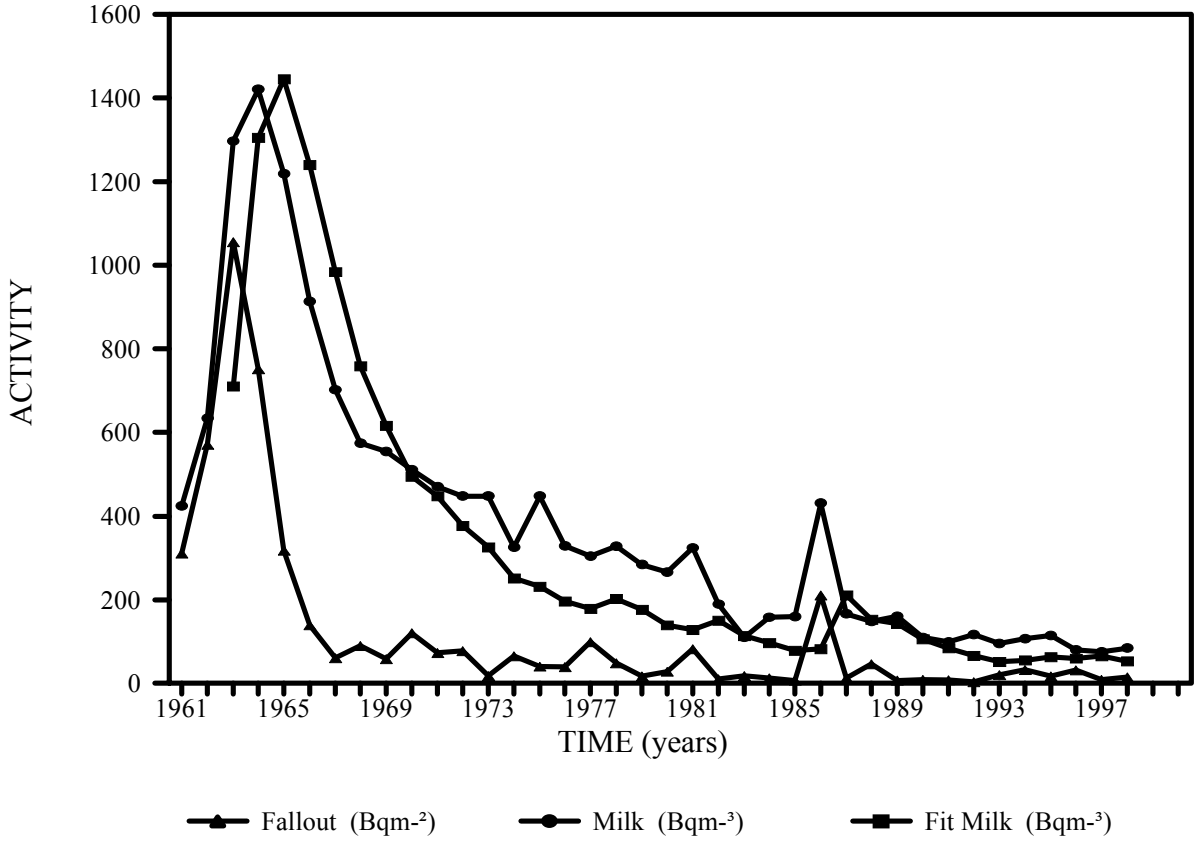

Figure 2. ^{90}Sr fallout and milk activities and modelled milk activities using equation /2/.

where:
$A_k(t)$ is the activity concentration of radionuclide *k* in food, i.e., milk (Bqkg^{-1}) and
$U_k(t)$ is the fallout deposition rate of radionuclide (Bqm^{-2}y^{-1}).

As for values of $A_k(t)$ and $U_k(t)$ assessed on the yearly basis the integration can be replaced by summation, the combination of equations /2/ and /3/ leads to:

$$P_{23} = b_1 + b_2 + b_3 \frac{e^{-\mu}}{1 - e^{-\mu}} \qquad /4/$$

P_{23} for ^{90}Sr in milk was assessed to be 2.72 × 10^{-3} Bqykg^{-1}/(Bqm^{-2}). That means that with each Becquerel deposited by fallout on an area of 1 m^2 of soil, the activity concentration of 1 m^3 milk increases by 2.72 Bq. For comparison, the transfer coefficient P_{23} for total diet was estimated to be 4 × 10^{-3} Bqykg^{-1}/(Bqm^{-2}) (10).

Radiostrontium doses incurred by milk consumption

In the Republic of Croatia, consumption of milk and diary products being approximately 100 L per year per person (12, 13) can potentially lead to significant radiation doses. The equivalent dose incurred due to milk consumption over certain period, depends on the activity of radionuclides that are present in milk and on the quantity of milk consumed. The dose can be expressed as:

$$H = C \int \sum_k D_{cf}(k) A_k(t) dt \qquad /5/$$

where:

H is the total 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* and
A_k the mean activity concentration of radionuclide *k* in milk ($Bqkg^{-1}$).

$D_{cf}(k)$, i.e., the equivalent dose per unit input, which converts the ingested activity to the equivalent dose (for age greater than 17 years) for ^{90}Sr is 2.8×10^{-8} $SvBq^{-1}$ (14).

For equal time increments of one month, integral from equation /5/ can be replaced by the respective sums of average monthly activity concentrations. Neglecting other isotopes apart from ^{90}Sr, the total dose due to that radionuclide in 1998 was approximately 0.2 μSv. For comparison, in 1994 the total dose due to ^{134}Cs and ^{137}Cs was about the same, i.e., 0.3 μSv, 94.4% being due to ^{137}Cs (15). The same year, the annual dose received by ^{90}Sr intake via food was estimated to be about 3 μSv (16).

Assuming similar radiostrontium milk activity concentrations in other Croatian regions, the collective effective dose for Croatian population (4.8 million inhabitants) due to ^{90}Sr ingestion by milk consumption in 1998 can be estimated to be 1.1 manSv, while the annual collective dose in the year of Chernobyl was 5.5 manSv.

CONCLUSIONS

The ^{90}Sr activity in milk decreases exponentially ever since 1960s. Peak values were recorded in 1964, following the most intensive nuclear weapon tests. Consequently, the ^{90}Sr activities in milk in Croatia are log-normally distributed, reflecting that exponential decrease.

Doses due to radiostrontium from milk consumption are small, in spite of relatively large consumption of milk by the Croatian population, the collective effective dose for Croatian population in 1998 being 1.1 Sv.

It should be noted that although radiocaesium levels in the environment in the year of Chernobyl accident were much higher than those of radiostrontium, since ^{90}Sr transfer from soil to the food chain is considerably more efficient, than for ^{137}Cs, in the following years the doses that Croatian population received from ^{137}Cs and ^{90}Sr were approximately the same.

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