RADIOCAESIUM ACTIVITY CONCENTRATIONS IN POTATOES IN CROATIA AFTER THE CHERNOBYL ACCIDENT

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ABSTRACT

In the paper are summarized the results of systematic investigations of ^{137}Cs and ^{134}Cs activity concentrations in potatoes (Solanum tuberosum) for the post-Chernobyl period in the Republic of Croatia. Potatoes are very important foodstuff in Croatia, the average annual consumption being about 40 kg per person.

Due to a comparatively high contribution of the ingestion doses to the total dose received by population after the exposure to nuclear fallout, a reliable prognosis of the expected ingestion doses is of utmost importance. The ingestion dose strongly depends on the consumption of various types of foodstuffs, and related activity concentrations of respective radionuclides in those foodstuffs, which themselves usually depend upon the transfer from fallout. In addition, a reliable prediction of the expected ingestion dose received by consumption of a particular foodstuff requires the detailed knowledge of decreasing behaviour of activity concentrations in the environment and respective foodstuffs.

The correlation between ^{137}Cs activity concentrations in fallout and potatoes, has been found to be very good, the correlation coefficient being $r^2=0.88$ with $P(t) < 0.001$ for 17 degrees of freedom. As the radiocaesium levels in potatoes decreased exponentially, the mean residence time of ^{137}Cs in potatoes was estimated by fitting the measured activity concentrations to the exponential curve. The mean residence time was found to be 6.3 ± 0.8 years, the standard deviation being estimated by the Monte Carlo simulations.

The initial observed ^{134}Cs:^{137}Cs activity ratio in potatoes has been found to be quite variable, but slightly lesser than theoretically predicted value of 0.5, calculated by applying the known inventory of these radionuclides in the Chernobyl reactor to the equation for the differential radioactive decay. This can be explained by presence of the pre-Chernobyl ^{137}Cs in soil that originated from nuclear fallout. As in other environmental samples, ^{134}Cs relatively quickly disappeared from potatoes and its activity concentrations were in 1990 under the detection limit of the instrument.

The annual effective doses received by ^{134}Cs and ^{137}Cs intake due to consumption of potatoes estimated for an adult member of Croatian population were found to be very small, as the per caput dose for the entire 1986 - 2004 period was calculated to be about 2.5 μSv, ^{134}Cs accounting approximately for 1/3 of the entire dose.

Consequently, it can be argued that after the Chernobyl accident consumption of potatoes was not the critical pathway for human intake of radiocaesium from the environment in Croatia.

Key words: *potato; concentration factor;* ^{137}Cs*;* ^{134}Cs*; Chernobyl accident; dose*

INTRODUCTION

Atmospheric nuclear tests and releases of radioactive material from nuclear facilities are the main causes of the man-made radioactive contamination of human environment. Once released to the atmosphere, long-range atmospheric transport processes can cause a widespread distribution of such radioactive matter, although it may, like in the case of Chernobyl accident, originate in a single point on the Earth's surface. The resulting fallout, consisting of short and long-lived radionuclides, eventually affects humans, either directly or indirectly by entering the food chain through consumption of plants and animals. In both cases it causes a health hazard to the population through the direct irradiation and consumption of contaminated foodstuffs.

Among man-made radioactive nuclides, those of the radiocaesium, particularly ^{137}Cs, are regarded as a great potential hazard to living beings. Namely, this fission product has unique combination of relatively long half-lives (30.14 years) and chemical and metabolic properties resembling those of the potassium, which is principal constituent of cell tissue.

Investigations of the distribution and fate of natural, nuclear weapons produced and reactor-released radionuclides in foodstuffs, have been conducted as a part of an extended and still ongoing monitoring programme of radioactive contamination of human environment in Croatia. (1, 2, 3, 4). However, the regular investigations of radioactive contamination of potatoes (Solanum tuberosum) have started in the year of the Chernobyl accident, i.e. in 1986.

The annual production of potatoes in Croatia is about 550,000 tonnes annually (5). Approximately 1/3 of this quantity is being consumed in Croatia. Since the population of Croatia is about 4.5 million of inhabitants, the consumption rate of potatoes is approximately 40 kg per person annually. Therefore it can be regarded as very important foodstuff in Croatia which can have, if contaminated, severe radiological consequences for the entire population.

MATERIALS AND METHODS

Potatoes have been obtained commercially on civil markets in the cities of Zagreb (45° 50' N, 16° 00' E), Osijek (45° 30' N, 18° 40' E) and Zadar (44° 06' N, 15° 15' E). From each site were obtained several kilograms of potatoes that were cut into small pieces in order to obtain the composite sample. Samples were dried in an oven and then ashed in a muffle furnace at 450 ^{o}C for 24 h.

Fallout samples were collected monthly in the city of Zagreb at the location of the Institute for Medical Research and Occupational Health (45° 50' 07.3" N, 15° 58' 58.7" E). The funnels, which were used for rainwater collection, had a 1 m^2 catchment area. Precipitation height was measured by Hellman pluviometer. Rainwater was evaporated to volume of 1 L in order to enrich the ^{137}Cs activity concentration.

A gamma-ray spectrometry system based on a low-level ORTEC Ge(Li) detector (FWHM 1.87 keV at 1.33 MeV ^{60}Co and relative efficacy of 15.4% at 1.33 MeV), coupled to a computerized data acquisition system, was used to determine radiocaesium levels in the samples from their gamma-ray spectra. Ash from the samples was measured in cylindrical plastic containers of appropriate volume, which were placed directly on the detector. Fallout samples were measured in Marinelli beakers. Counting times, which depend on radiocaesium activity concentrations in samples, ranged from 10,000 to 250,000 seconds and typically were 80,000 s.

Quality assurance and intercalibration measurements were performed through participation in an International Atomic Energy Agency (IAEA) and World Health Organization (WHO) international intercalibration programmes, which also include the regular performance of blank, background and quality control measurements.

Radiocaesium activity concentrations in samples in this paper are reported as averages of three sampling locations, implicitly implying similar characteristics and microclimate conditions, of soils in which potatoes have been grown, that is not necessarily true. However, typical relative error was about 25 %.

RESULTS AND DISCUSSION

^{137}Cs and ^{134}C activity concentrations in potatoes

The radioactive fallout resulting from large-scale nuclear weapon tests in the atmosphere conducted in the 1960s, followed by similar, but smaller scale tests by the Chinese and French in the 1970s and afterwards, was the dominant route for the introduction of artificial radionuclides in the environment until the nuclear accident at Chernobyl, in former USSR, on 26 April 1986. Fortunately, due to the prevailing meteorological conditions at the time after the accident that influenced the formation and spreading direction of Chernobyl plumes, Croatia was only on the North-Western region partially affected by the edge of one of the plumes (6).

The estimated amount of radiocaesium 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 (7). Thus, the initial value for the ^{134}Cs:^{137}Cs activity ratio in May 1986 was 0.51.

The highest observed ^{137}Cs activity concentrations in fallout were recorded in May 1986, resulting in 6200 Bqm^{-2} for the surface deposit by fallout in the year 1986 (8). The radioactive material introduced to the atmosphere by Chernobyl accident was by global dispersion processes distributed throughout the troposphere, causing the increased radiocaesium activity concentrations in the environment in years to come. However, ^{137}Cs showed a significant exponential decrease over time because of natural removal as well as radioactive decay. Also, no new releases of ^{137}Cs occurred after the Chernobyl reactor accident either from nuclear facilities or nuclear weapons testing. Therefore, in 2004 total ^{137}Cs surface deposit by fallout was only 2.1 Bqm^{-2}.

The highest ^{137}Cs activity concentration in potatoes, being 1.100 ± 0.650 Bq kg^{-1} was recorded in 1986 while in 2001 was recorded minimal value of only 0.033 ± 0.011 Bq kg^{-1}, the ^{137}Cs activity concentration in 2004 being 0.038 ± 0.005 Bq kg^{-1}. The increase of ^{137}Cs activity concentration in year 1999 could be explained by penetration of caesium to deeper soil layers as root uptake by plants of radionuclides as well as other stable elements depends upon their vertical distribution. It should be noted that these activity concentrations are comparable to the values observed elsewhere (9, 10).

When discussing ^{134}Cs activity concentrations, it should be noted that ^{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 value of 0.5, found at samples on the accident site, has not been altered as this ratio has been found in most of the environmental samples in Croatia (11, 12, 13). However, the initial observed ^{134}Cs:^{137}Cs activity ratio in potatoes has been found to be quite variable, but slightly lesser than 0.5. This can be explained by presence of the pre-Chernobyl ^{137}Cs in soil that originated from nuclear bomb fallout. Consequently, ^{134}Cs:^{137}Cs activity concentration ratio found in potatoes resembled that found in fallout.

In 1990 and afterwards, contamination of potatoes by the fallout ^{134}Cs that originated from the Chernobyl nuclear accident was detectable only at a very low level.

^{137}Cs activity concentrations in potatoes are in good correlation with fallout activity when related by a simple linear relation, with coefficient of correlation r = 0.88 with P(t) < 0.001 for 17 degrees of freedom. Thus, for the observed period, i.e. from 1986 - 2004, ^{137}Cs activity concentrations in potatoes can be from fallout data modelled as:

$$A_{pot}(t) = 0.00015 \times A_{fall}(t) + 0.14576 \qquad /1/$$

where:

$A_{pot}(t)$ *is ^{137}Cs activity concentration in potatoes (Bq kg^{-1}) and*

$A_{fall}(t)$ *^{137}Cs fallout activity (Bq m^{-2}).*

By plotting the observed (measured) ^{137}Cs activity concentrations data in potatoes and data modelled by the equation /1/ against the time scale, it is clear that the fit overestimates the real data ever since year 1995 (Figure 1).

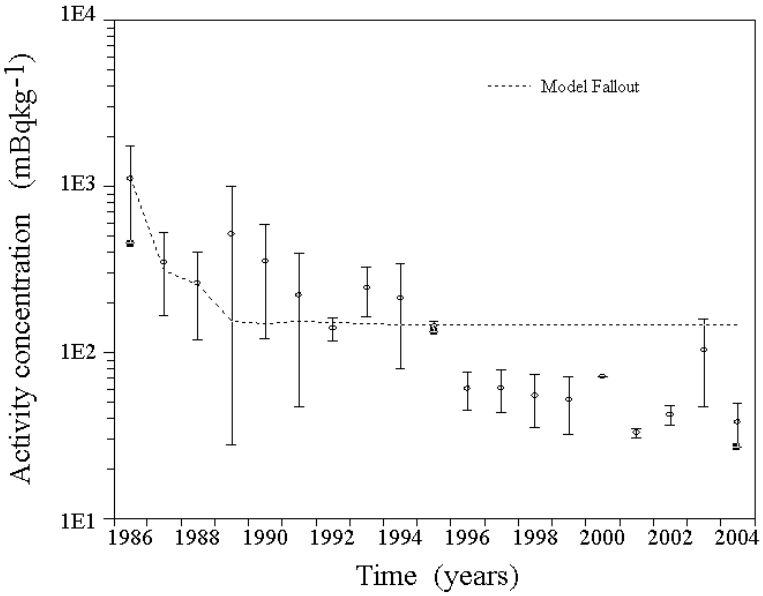

Figure 1
Observed and modelled ^{137}Cs activity concentrations in potatoes as a function of time

It should be noted that equation /1/ showing mere functional dependence, is just simple tool for rough prediction of ^{137}Cs activity concentration data in potatoes from fallout data, having no deeper physical meaning.

Mean residence time of ^{137}Cs in potatoes

A reliable prediction of the expected ingestion dose received by consumption of a particular foodstuff requires the detailed knowledge of decreasing behaviour of activity concentrations in the environment and respective foodstuffs. To evaluate the decrease activity concentrations and to assess the effective mean residence time (sometimes also called ecological time) of ^{137}Cs in potatoes the observed data were fitted to the exponential function:

$$A_{pot}(t) = A_{pot}(0)\, e^{-kt} \qquad /2/$$

where:

$A_{pot}(t)$ is time-dependant activity concentration of ^{137}Cs in potatoes ($Bqkg^{-1}$),

$A_{pot}(0)$ initial activity concentration of ^{137}Cs in potatoes ($Bqkg^{-1}$) and

$1/k=T_M$ effective (observed) mean residence time of ^{137}Cs in potatoes (years).

By fitting the data for ^{137}Cs activity concentration in potatoes to the curve /2/ ($r = 0.84$ with $P(t) < 0.001$ for 17 degrees of freedom) the observed mean residence time for ^{137}Cs in potatoes, for the overall observed post-

Chernobyl period (1986 – 2004) was estimated to be 6.3 years. This value is just slightly lesser effective (observed) mean residence time of 6.7 ∀ 1.3 reported (effective half-life is 1688 ∀ 338 days) for potatoes in Austria (9), and similar to mean residence time for the other foodstuffs (9).

The fit is shown on Figure 2.

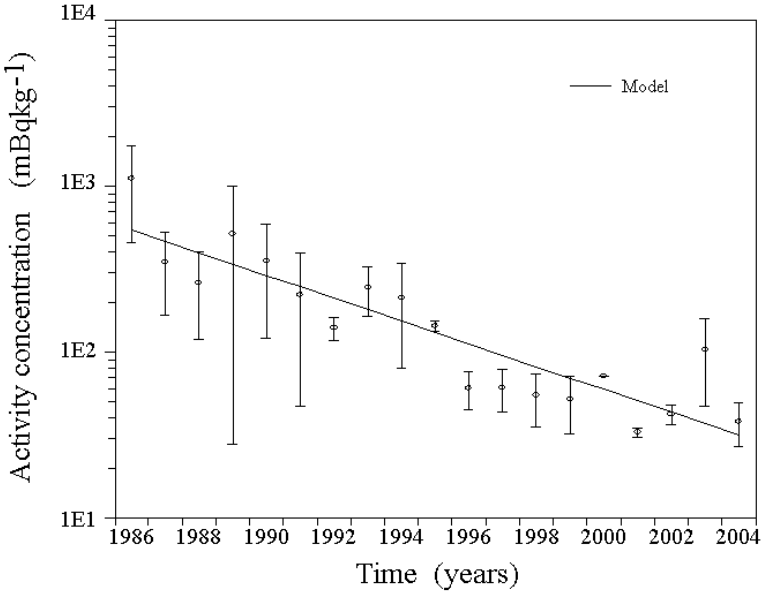

Figure 2

Observed and modelled ^{137}Cs activity concentrations in potatoes as a function of time

However, to find the real residence time, T_R, the observed constant k should be corrected for the radioactive decay. Therefore constant k from the equation /2/ can be written as:

$$k = 8 + k_R \qquad /3/$$

where ln(2)/8 = 30.14 y is the physical half-life of ^{137}Cs.

From equation (3) real mean residence time for ^{137}Cs in potatoes $T_R = 1/k_R$, was found to be 7.4 y, which is one year more compared to the effective mean residence time T_M.

In order to obtain the standard deviation of T_M, Monte Carlo simulations were performed. To be on a conservative side, as well as to simplify calculations, the uniform distribution has been assumed over the A ± 2Φ value of ^{137}Cs activity concentrations in potatoes for respective years, although normal would be more realistic. For each year the random value was generated over the interval [A - 2Φ, A + 2Φ] and then from such set of data was estimated the $1/k_o$ value by fitting to the equation (2). The process has been repeated 100 times and 100 values for $1/k = T_M$ were obtained. From

such set of data the mean value and standard deviation for T_M were calculated to be 6.3 ∀ 1.2 years.

Dosimetry

Data on activity concentrations of ^{137}Cs and ^{134}Cs in potatoes allow for the estimate of the doses incurred by their consumption. Dose conversion factors, i.e. effective dose per unit intake via ingestion for the member of public older than 17 years are 1.3×10^{-8} $SvBq^{-1}$ and 1.9×10^{-8} $SvBq^{-1}$ for ^{137}Cs and ^{134}Cs respectively (15). As the ratio of dose conversion factors for ^{137}Cs and ^{134}Cs is ≈ 0.7, it implies that ingestion of ^{134}Cs contributes about 30% more to the dose, compared to ingestion of the same activity concentration of ^{137}Cs.

Collective effective dose incurred due to food consumption over certain time period, depends on the activity of a radionuclide and on the quantity of food that is consumed. The dose can be expressed as:

$$E = C \ \square \ D^k_{cf} \ A^k(t) \qquad /4/$$

where:

- E *is the effective dose in Sv,*
- C *total annual per caput consumption of food,*
- D^k_{cf} *dose conversion factor for radionuclide k, i.e. effective dose per unit input, which converts the ingested activity to effective dose and*
- A^k *mean yearly specific activity of radionuclide k in food ($Bqkg^{-1}$).*

Based on the statistical data for annual consumption rate for potatoes of 40 kg, the estimated total effective dose due to ^{137}Cs and ^{134}Cs ingestion by potato consumption for the Croatian population and overall observed period, i.e. 1986-2004 is 13.1 person-Sv. Out of this, 9.5 person-Sv could be attributed to ^{137}Cs and 3.6 person-Sv to ^{134}Cs and 4.2 person-Sv refers to the year 1986 (2.6 person-Sv and 1.6 person-Sv for ^{137}Cs and ^{134}Cs respectively).

The annual effective collective doses are shown on Figure 3.

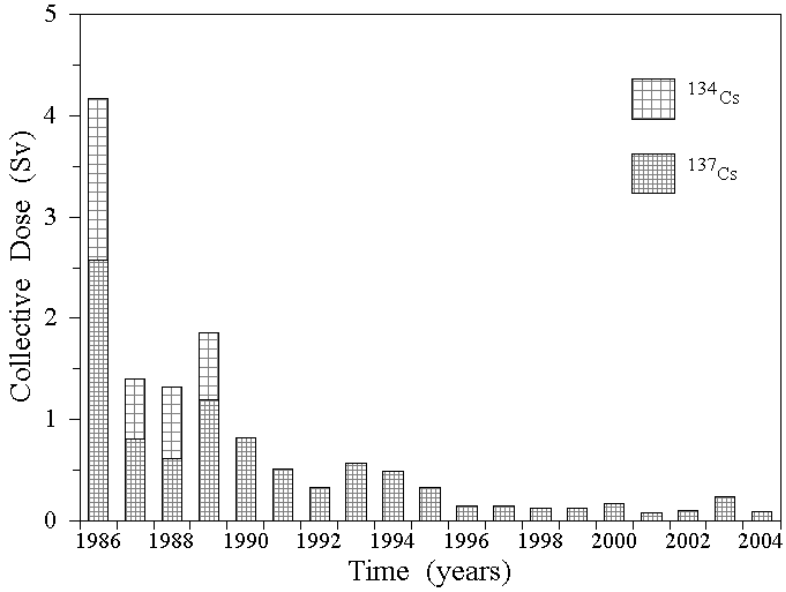

Figure 3

Estimated annual effective doses received by ^{134}Cs and ^{137}Cs intake due to consumption of potatoes for an adult member of Croatian population.

It should be noted that these doses are rather small since for the overall 1986-2004 period per caput dose is . 2.9 :Sv. Consequently, it can be argued that after the Chernobyl accident consumption of potatoes was not the critical pathway for human intake of radiocaesium from the environment in Croatia.

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