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RADIOCAESIUM ACTIVITIES IN WHEAT IN CROATIA

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Results of systematic, long-term measurements of ^{137}Cs activities in wheat are summarized. ^{137}Cs fallout activity affected wheat activity the coefficient of correlation being 0.91. A model used by UNSCEAR was used to describe ^{137}Cs transfer from fallout deposition to wheat. The transfer coefficient was calculated to be 6.1×10^{-3} Bq yr kg^{-1}/(Bq m^{-2}). The dose incurred by wheat consumption was estimated for the Croatian population, the annual collective equivalent dose being approximately 550 man Sv for the 1965–1992 period. After the Chernobyl nuclear accident the ^{134}Cs:^{137}Cs activity ratio in wheat was ~ 0.5, and did not differ from that for other environmental samples.

Key terms:
^{134}Cs, ^{137}Cs, fallout, gammaspectrometric analysis, radioactive contamination

Nuclear tests conducted in the atmosphere and release of radioactive material from nuclear facilities cause radioactive contamination of the human environment. The fallout resulting from atmospheric dispersion of both short-lived and long-lived radionuclides not only directly affects humans, but also enters the food chain through plants and animals, causing a potential health hazard to the population consuming contaminated foods.

Among anthropotropic radioactive nuclides ^{137}Cs has been regarded as the fission product of great potential hazard to living beings because of the unique combination of its relatively long (30 years) half-life, and its structural as well as metabolic properties resembling those of potassium. Considering the long half-life of ^{137}Cs, dose assessment requires understanding of its behaviour in the environment. Therefore, research on the fate of ^{137}Cs has largely focussed on determining its activities in fallout, soil and along human exposure pathways (1). Generally, dose prediction models for humans depend crucially on the transfer coefficient values quantifying the movement of radionuclides through food chains.

By way of contaminated soil radioactivity is transferred to various food products, wheat (*Triticum vulgare*) being among the most important. Consumed daily energy per person in Zagreb for the 1975–1983 period was 9284 ± 243 J, wheat and various wheat products accounting for more than 30% (2). Therefore, in that period consumption of wheat and wheat products, like bread, pastry and flour was approximately 100 kg per year per person. To a limited extent, wheat is also used for making beer and industrial alcohol. Therefore, wheat as the predominant foodstuff in dietary habits in Croatia, is possibly a major source of radioactive contamination for the Croatian population that can lead to significant radiation doses.

MATERIAL AND METHODS

Fallout samples were collected monthly in the city of Zagreb. Wheat samples, if available, were obtained commercially on the markets in the cities of Zagreb, Osijek, Gospić and Zadar. Wheat samples were dried in the oven and ashed at 450 °C.

A gamma-ray spectrometry system based on a low-level ORTEC Ge(Li) detector (FWHM 1.82 keV at 1.33 MeV) coupled to a computerized data acquisition system (4096-channel pulse height analyzer 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 appropriate volume which were placed directly on the detector. Counting time depended on sample activity, but was never less than 60,000 seconds.

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.

RESULTS AND DISCUSSION

^{137}Cs *fallout activities*

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. Therefore, activity of

most environmental samples could be expected to be in correlation with fallout activity (i.e. surface deposit in Bq m^{-2}).

Severe radioactive fallout from highly radioactive air plumes that originated from the damaged Chernobyl nuclear reactor was spread and transported over Europe (3, 4). The radionuclides attributed to the Chernobyl accident were soon detected in Croatia, but only a few of those (caesium radioisotopes ^{137}Cs and ^{134}Cs) were responsible for most of the radioactivity (5). The fallout ^{137}Cs activities after the Chernobyl nuclear accident were much higher than in the mid-1960s, after a period of intensive atmospheric nuclear weapon tests. The baseline level of ^{137}Cs fallout activity in the pre-Chernobyl 1985 for the Zagreb area was 2 Bq m^{-3} leading to a surface deposit of 1.5 Bq m^{-2}.

Effective residence times of ^{137}Cs in fallout for the pre-Chernobyl and post-Chernobyl periods were estimated to be 3.7 and 0.7 years (6), reflecting the mechanism of release 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 years (7). As a consequence of the Chernobyl accident by explosions and subsequent fire of a graphite moderator, radioactive material was dispersed only to the troposphere where the mean residence time is a few months (7).

Contrary to ^{137}Cs the nuclear accident at Chernobyl did not cause any significant increase in ^{90}Sr activity in environmental samples. 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 refractory components of the Chernobyl debris (e.g. ^{90}Sr) were deposited closer to the accident location than the more volatile constituents (i.e. radiocaesium) (8, 9). 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, which was especially true of the Adriatic region (4, 9, 10).

Therefore, the ^{90}Sr from Chernobyl did not impose any additional threat to the Croatian population by way of the food chain, including wheat.

^{137}Cs transfer from fallout to wheat

Following the nuclear moratorium on atmospheric weapon tests in 1963 (7), ^{137}Cs activity in wheat exponentially decreased with time, with constant 0.31 yr^{-1} for the 1965–1985 period and 0.64 yr^{-1} for the 1986–1992 period. The mean residence times for the respective periods were 3.24 and 1.56 years. Transient increases in the activity (like in 1970), which were also observed elsewhere (11) can be explained both by Chinese nuclear tests as well as by a variety of naturally fluctuating physical environmental factors.

^{137}Cs activity in wheat is in good correlation with fallout activity, r = 0.91 with P(t) < 0.001 for 26 degrees of freedom. Thus, wheat activity can be modelled as:

$$A_w(t) = 0.0058\ A_f(t) + 0.9804 \qquad /1/$$

where:
$A_w(t)$ is ^{137}Cs activity in wheat (Bq kg^{-1}) and
$A_f(t)$ is ^{137}Cs deposited fallout activity (Bq m^{-2}).

To assess the ^{137}Cs transfer from fallout to wheat, several functions were tested, but the best fit was obtained by applying the function recommended by UNSCEAR for food (7):

$$A_k(t) = b_1\dot{U}_k(i) + b_2\dot{U}_k(i-1) + b_3 \sum_{m=1}^{\infty} e^{-\mu m}\, \dot{U}_k(i-m) \qquad /2/$$

where:
$A_k(t)$ is the activity concentration of radionuclide k in food (wheat), the unit for $A_k(t)$ being Bq kg^{-1},
$\dot{U}_k(i)$ is the fallout deposition rate of radionuclide k in year i (Bq m^{-2} yr^{-1}),
$\Sigma e^{-\mu m}$... is the cumulative fallout deposit for radionuclide k as the result of deposition in previous years (Bq m^{-2} yr^{-1}),
μ^{-1} is the effective mean life of accumulated deposition of ^{137}Cs and,
b_1, b_2, b_3 are the factors which can be derived from reported data by regression analysis. The unit is Bq yr kg^{-1}/(Bq m^{-2}).

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

Input → Atmosphere → Earth's surface → Diet → Tissue → Dose

0 1 2 3 4 5

The physical meaning of terms in model /2/ is as follows: the first term (rate factor) describes direct deposition and transfer, the second term (soil lag factor) describes contamination through fallout from previous year, and the third term (ultimate soil 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 to wheat of caesium in soil.

Regression analysis gives the following values:

$b_1 = 6.0 \times 10^{-3}$, $b_2 = 9.0 \times 10^{-5}$, $b_3 = 2.3 \times 10^{-3}$ Bq yr kg^{-1}/(Bq m^{-2}) and $\mu = 25.0\ yr^{-1}$, the coefficient of correlation being 0.93. Thus, the effective mean

half-life of the ^{137}Cs accumulated deposition in soil is $\ln(2)/\mu = 0.03$ years. As expected, $b_1 > b_3 > b_2$ since most of ^{137}Cs wheat contamination comes from direct deposition, contamination through fallout from previous year being of least importance. By function minimization of experimental data (5, 12) to exponential function it can be shown that the half-depth of radiocaesium penetration in uncultivated (i.e. undisturbed soils) is less than five centimetres, as observed elsewhere (13). Therefore most of caesium activity is found in the first few centimetres of soil. In cultivated soils (by ploughing) caesium penetrates into deeper layers. Nevertheless, *Triticum vulgare* having very deep roots, up to two metres, (14) has never been severely affected by caesium accumulated in soils.

The average (four sampling sites) ^{137}Cs activity in wheat and the fit obtained by equation /2/ are shown in the Figure. Relative error never exceeded 30%.

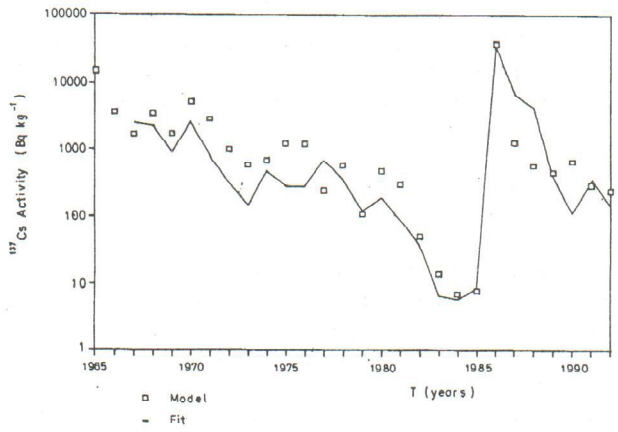

Figure. *Measured and modelled ^{137}Cs activities in wheat*

The environmental transfer of radioactivity between successive steps in the pathway chains is described by transfer coefficients, which relate infinite time integrals of activity in the relevant compartments. Thus, the transfer coefficient from fallout deposition (compartment 2) to diet (compartment 3) is given by equation (7):

$$P_{23} = \frac{\int_0^\infty A_k(t)dt}{\int_0^\infty \dot{U}_k(t)dt} \qquad /3/$$

where:

$A_k(t)$ is the activity of radionuclide k in food, i.e., wheat (Bq kg^{-1}) and
$U_k(t)$ is the fallout deposition rate of radionuclide k (Bq m^{-2} yr^{-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 ^{137}Cs in wheat was assessed to be 6.1×10^{-3} Bq yr kg^{-1}/(Bq m^{-2}). That means that with each Becquerel deposited on an area of one square metre of soil by fallout, the activity of one ton of wheat increases by 6.1 Bq. For comparison, the ^{137}Cs transfer coefficient P_{23} for total diet was estimated to be approximately 1.2×10^{-2} Bq yr kg^{-1}/(Bq m^{-2}) for the 1962–1979 period in New York (reference location for the northern hemisphere) and 8.0×10^{-3} Bq yr kg^{-1}/(Bq m^{-2}) for the 1963–1973 period in Argentina (the southern hemisphere) (7).

^{134}Cs wheat activities

The presence of ^{134}Cs (half-life of 2.06 years) was first detected in the environment in Croatia in May 1986. It could be immediately attributed to the Chernobyl nuclear accident, because being »shielded radionuclide« 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 owing to long irradiation times for nuclear fuel (typically three years residence time in the reactor core). This permits the build-up 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 which is over in milliseconds.

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) (15). This was more than 3% of radiocaesium that had been released to the environment by all nuclear weapon tests conducted in the atmosphere. Thus, the initial value for the ^{134}Cs:^{137}Cs activity ratio in May 1986 was 0.51. These two isotopes, ^{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.

As the half-life of ^{137}Cs compared to the half-life of ^{134}Cs is much longer (30.14 years), 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:

$$R(t) = \frac{1.9 \times 10^{16}}{3.7 \times 10^{16}} \times e^{\ln(2) \times t \times \left(\frac{1}{t_1} - \frac{1}{t_2}\right)} \qquad /5/$$

where:
t is the 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 wheat was 0.52 ± 0.10 and in 1987 0.42 ± 0.06.

The same ratio, decreasing according to equation /5/ was found in most environmental samples (9, 16, 17), the only exception being mushrooms (18). In mushrooms, (generally having shallow mycelium) the excess ^{137}Cs from the pre-Chernobyl fallout, affected the ^{134}Cs:^{137}Cs concentration ratios. As ^{134}Cs penetrated into deeper layers of soil, the observed ^{134}Cs:^{137}Cs concentration ratios approached the values predicted theoretically (18).

In 1988 and on, contamination of wheat by the fallout ^{134}Cs and ^{137}Cs that originated from the Chernobyl nuclear accident was detectable only at a very low level (5).

Dose incurred by wheat consumption

Per day intake of ^{137}Cs by wheat (and wheat products) in 1965 was estimated to be 4.4 Bq d^{-1}. In the year of the Chernobyl accident it increased to maximum 10.4 Bq d^{-1}, sharply decreasing ever since, to only 0.10 Bq d^{-1} in 1992.

Per caput dose incurred due to wheat consumption depends on the average activity of a radionuclide and on the quantity consumed. The dose can be expressed as:

$$H = C \sum_k D_{cf}(k) A_k \qquad /6/$$

where:
H is the mean annual per caput equivalent dose (Sv yr^{-1}),
C is total annual per caput consumption of wheat (100 kg yr^{-1}),
$D_{cf}(k)$ is dose conversion factor for radionuclide k i.e. equivalent dose per unit input, which converts the ingested activity to equivalent dose;
$D_{cf}(^{137}Cs) = 1.4 \times 10^{-8}$ Sv Bq^{-1};
$D_{cf}(^{134}Cs) = 2.0 \times 10^{-8}$ Sv Bq^{-1} (19) and
A_k is mean specific activity of radionuclide k in wheat (Bq kg^{-1}).

For the Zagreb area, the annual per caput equivalent dose, H, due to ^{137}Cs and ^{134}Cs ingestion by wheat consumption, estimated for 1992 when the ^{137}Cs and ^{134}Cs activities in wheat were ≈ 0.24 Bq kg^{-1} and ≤0.02 Bq kg^{-1} respectively, was 0.38 μSv; 0.34 μSv was due to ^{137}Cs and ≤0.04 μSv to ^{134}Cs.

To assess the incurred dose due to ^{137}Cs for a member of the Croatian population in the 1965–1992 period, the integrated activity of wheat (83.8 Bq kg^{-1} yr) should be multiplied by the total annual per caput consumption (100 kg yr^{-1}) and dose conversion factor which leads to the effective dose of 0.12 mSv. Assuming this value to be representative for the entire Croatian population (4.7×10^6 inhabitants), the collective effective dose, E_C, for this period was 550 man Sv. In 1986 (the year of the Chernobyl accident) the per caput effective dose was 0.05 mSv leading to the collective effective dose of 250 man Sv, which is 45% of the dose for the entire 1965–1992 period.

A similar result can be obtained using the estimated transfer coefficient P_{23} for ^{137}Cs transfer from fallout to wheat. Dose is given by relation:

$$H = P_{23} F_A (T)\, C\, D_{cf} (^{137}Cs) \qquad /7/$$

where:

P_{23} is the transfer coefficient for the 1965–1992 period: 6.1×10^{-3} Bq yr kg^{-1}/(Bq m^{-2}),

$F_A(T)$ is total ^{137}Cs activity delivered by fallout to the soil for the 1965–1992 period, i.e. 9.79 kBq m^{-2},

C is total annual per caput consumption of wheat in Croatia (kg yr^{-1}) and

$D_{cf}(^{137}Cs)$ is dose conversion factor for ^{137}Cs.

From equation /7/ the per caput equivalent dose is 0.084 mSv for a period of 27 years and the collective effective dose is 390 man Sv. Direct contamination of wheat grains is the most probable explanation for 30% lesser values than those obtained in the previous case.

CONCLUDING REMARKS

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

The ^{137}Cs transfer coefficient for wheat was of the same order of magnitude (10^{-3}) as for the other foodstuffs. Doses due to radiocaesium from wheat consumption are small in spite of large consumption of wheat by the Croatian population.

As the distribution of radiocaesium in wheat grain shows the activity to be highest in bran (20, 21), by the appropriate grinding technology flour activities can be reduced. The ^{137}Cs activity of white flour (T-500) is about 30%, and that in brown flour 50% of that in wheat (20). Therefore, in the case of major wheat contamination by radiocaesium, through consumption of white flour, the popu-

lation would receive much lesser doses. In addition, wheat products like macaroni (pasta) can be successfully decontaminated, up to 90% (21), by cooking in salty water. This is due to ion exchange between caesium ions in macaroni and sodium ions in the surrounding water. The same procedure can be used to decontaminate meat structurally contaminated by radiocaesium (22, 23).

Since low-grade wheat and by-products of the flour-milling (mainly bran), brewing and distilling industries are used as feed for livestock, in case of major wheat contamination by radiocaesium, this should be avoided.

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Sažetak

AKTIVNOSTI RADIOCEZIJA U PŠENICI U HRVATSKOJ

Prikazani su rezultati dugogodišnjih sustavnih mjerenja aktivnosti ^{137}Cs u pšenici. Aktivnost ^{137}Cs u radioaktivnim oborinama utječe na aktivnost pšenice, uz koeficijent korelacije 0,91. Transfer ^{137}Cs od radioaktivnih oborina do pšenice najbolje opisuje model UNSCEAR-a. Transferni koeficijent procijenjen je na 6.1 × 10^{-3} Bq god kg^{-1}/(Bq m^{-2}). Kolektivna ekvivalentna doza uslijed konzumacije pšenice za hrvatsku populaciju procijenjena je na oko 550 čovjek Sv za period od 1965. do 1992. godine. Poslije nuklearne nesreće u Černobilju omjer aktivnosti ^{134}Cs:^{137}Cs u pšenici bio je ≈ 0,5 i nije se razlikovao od vrijednosti omjera u ostalim uzorcima iz okoliša.

Ključne riječi:
^{134}Cs, ^{137}Cs, gamaspektrometrijska analiza, radioaktivna kontaminacija, radioaktivne oborine

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