Long term investigation of ^{137}Cs in chicken meat and eggs from northwest Croatia

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Abstract

This paper presents the results of long-term investigations of ^{137}Cs activity concentrations in chicken meat and eggs from northwest Croatia for the period 1987-2018. The research has been done as a part of monitoring program of radioactive contamination in Croatia. The highest activity concentrations in both of these foodstuffs were measured in 1987 and have been decreasing exponentially ever since. The Fukushima-Daiichi accident in 2011 did not cause any increase of ^{137}Cs activity concentrations. The ecological half-life for ^{137}Cs was estimated to be 8.0 and 8.4 years for chicken meat and eggs respectively. The correlation between ^{137}Cs in fallout and chicken meat as well as between ^{137}Cs in fallout and eggs is very good, the respective correlation coefficients being 0.79 and 0.72, indicating that fallout was the main source of ^{137}Cs contamination in both foodstuffs. The estimated effective doses received by adult members of the Croatian population due to the intake of radiocaesium by chicken meat and egg consumption for the overall observed period are very small, 2.0 and 0.6 μSv respectively. Therefore, chicken meat and chicken egg consumption was not a critical pathway for the transfer of radiocaesium to humans.

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Introduction

Nuclear tests conducted in the atmosphere and releases of radioactive material from nuclear facilities are the main causes of man-made radioactive contamination of the human environment. Once released into the atmosphere, long-range atmospheric transport processes can cause a widespread distribution of such radioactive matter, although it may, like in the case of the Chernobyl nuclear accident, originate in a single point of 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 plants and animals. In both cases, it causes a health hazard to the population through direct irradiation and internal contamination following the consumption of contaminated foodstuffs.

Among man-made radioactive nuclides, those of radiocaesium and radiostrontium, particularly ^{137}Cs and ^{90}Sr, are regarded as a great potential hazard to living beings. These fission products have unique combinations of relatively long half-lives (30.14 and 29.12 years, respectively) and chemical and metabolic properties resembling those of potassium and calcium, respectively. Once they enter the body of a mammal, radiocaesium and radiostrontium are excreted through milk, faces and urine. Meat and milk, containing both potassium and calcium are therefore recognized as sensitive indicators for the presence of fission products in the environment. Consequently, investigations of these two fission products take significant part in an extended monitoring program of radioactive contamination of human environment in Croatia. [1, 2, 3, 4] Regular investigations of radioactive contamination of poultry and eggs started in 1987, i.e., one year after the Chernobyl accident.

Regarding animal products, the lessons learned from Chernobyl's aftermath are that the detailed knowledge of decreasing behavior of radiocaesium activity concentrations in meat of animals bound for human consumption and effective decontamination measures are of the utmost importance for estimations and reductions of total ingestion doses. [5, 6, 7]

As these two poultry products are quite affordable and have high nutritional value, they are quite popular among consumers. In Croatia, the annual average for chicken meat consumption is about 22.8 kg [8] and for eggs about 9 kg, i.e., about 160 eggs per person. [9]

We present here the results of long-term investigations of ^{137}Cs activity concentrations in chicken meat and eggs from northwest Croatia for the period 1987-2018. In both feedstuffs activity concentrations steadily decreased through observed period.

In radioecology, decrease of activity concentrations is usually parametrized either by effective half-life or ecological half-life. Effective half-life (synonyms: observed half-life, effective ecological half-life, effective environmental half-life) is time during which the initial amount of activity concentration of observed radionuclide is reduced to half by an assumed exponential reduction which includes effects of environmental and ecological processes as well as physical decay. Ecological half-life ($T_{1/2,e}$) (synonyms: real ecological half-life, environmental half-life) is that part of the effective half-life attributed to ecological and environmental processes, but without any decrease to physical decay. It should be mentioned that ecological half-life is sometimes conceived as ecological residence time which is mathematically related to ecological half-life according to relation $T_{1/2,e}/\ln(2)$. Residence time is therefore measure of how much time particular radionuclide spends in observed ecological compartment. In radiobiology is important the biological half-life in an organism, which is that part of the effective half-life attributed to biological excretion and does not include the effect of physical decay. More detailed discussion is available elsewhere. [10] It should be

mentioned, however, that above definitions, although used in most of scientific literature dealing with radioecology, are not universally accepted.

Material and methods

Sampling and samples preparation

According to the national monitoring program prescribed by national authority for radiological and nuclear safety, approximately 5 kg of chicken meat (i.e., 3-5 chickens) and 60 eggs are yearly, usually in late spring or early summer, commercially obtained. All samples were obtained the same day on the civil market in the city of Zagreb from individual farmers from the nearby villages surrounding Zagreb, who claimed that the chickens were kept during the day outdoors. The data on the origin of chicken feed is not available. However, traditionally individual farmers produce most of the livestock and poultry feed by themselves.

Meat samples were cut into small pieces in order to obtain one composite sample which was dried in an oven. Dried meat was then ashed in a muffle furnace at 450 °C for 24 h.
The eggs were cracked and eggshells removed. Scrambled eggs were placed in the Pyrex beaker and heated to 100 °C in a fumehood, until charred well. The carbonaceous material was dried in an oven and ashed in a muffle furnace at 450 °C for 24 h.
Dry and wet fallout (rain water) was collected daily using funnels of 1 m^2 collection area. Fallout was collected in Zagreb, on the site of the Institute for Medical Research and Occupational Health which is good representation for the wider area of the city of Zagreb. On rainy days, the amount of precipitation that contained the fallout was measured by a Hellman

pluviometer. In days without precipitation, funnels were rinsed by 1 L of distilled water. Daily samples were merged into cumulative samples which were analyzed twice a year. Prior to gamma-spectrometric measurements, bi-annual samples were evaporated to a 1 L volume.

Measurement of activity concentrations

Fallout samples were measured in Marinelli beakers of a 1 L volume.

Ash resulting from chicken meat and eggs was placed in plastic cylindrical containers of 0.2 L volume and placed directly on the detector.

Activity concentrations of gamma-emitting radionuclides in the samples were analyzed from their gamma ray spectra by a gammaspectrometrical method accredited by the Croatian Accreditation Agency in 2010. [11]

In the period 1986-2003, gamma-ray spectrometry systems based on a low-level ORTEC Ge(Li) detector (FWHM 1.87 keV at 1.33 MeV ^{60}Co and relative efficiency of 15.4% at 1.33 MeV) and ORTEC HPGe detector (FWHM 1.75 keV at 1.33 MeV ^{60}Co and relative efficacy of 21% at 1.33 MeV) coupled to a computerized data acquisition system were used to determine radiocaesium and ^{40}K levels in the samples from their gamma-ray spectra.

Since 2003, for gamma spectrometry was used a low-level high-purity ORTEC HPGe detector (relative efficiency of 74.2% with FWHM resolution of 2.24 keV at 1.33 MeV).

To reduce background radiation, the detectors were shielded with 10 cm thick lead lined with 2 mm of cadmium and 2 mm of copper. The counting time for gammaspectrometric measurements depended on the sample activity, typically being 86,400 seconds.

The certified calibration standards, i.e., mixed gamma water standard sources were obtained from the Czech Metrological Institute covering energies 40-2000 keV. The efficiency calibration has been performed for all used geometries (cylindrical plastic containers and

Marinelli beakers) and matrices, i.e. ash matrix (for ashed meat and eggs) and water matrix (fallout). Quality assurance and intercalibration measurements were performed through intercalibration programs organized by the International Atomic Energy Agency (IAEA) and Joint Research Centre (JRC), which also included the regular performance of blanks (empty cylindrical containers and Marinelli beakers), background and quality control measurements.[12]

Results and discussion

^{137}Cs and ^{134}Cs activity concentrations

The data for ^{137}Cs activity concentrations in fallout, chicken meat and chicken eggs are presented in Table 1.

Table 1 about here

Following the Chernobyl accident, the activity concentrations of ^{137}Cs in chicken meat exponentially decreased from 3.30±1.21 in 1987 to only 0.04±0.01 Bqkg^{-1} in 2018 (Fig. 1). In chicken eggs, ^{137}Cs activity concentrations also exponentially decreased from 1.30±0.33 in 1987 to only 0.03±0.01 Bqkg^{-1} in 2018. Unfortunately, data for ^{137}Cs activity concentrations in 1986 are not available, but could be estimated from fallout data. Also, for chicken meat data for 1991-1994 period and year for 2000 are not available while for chicken eggs data for year 2000 are not available.

The observed ^{137}Cs activity concentrations both in chicken meat and eggs were in good correlation with ^{137}Cs activity concentrations deposited on the ground by fallout, the respective coefficients of correlation being 0.79 and 0.72, which confirms that fallout was a primary source of ^{137}Cs both in chicken meat and in chicken eggs. Using a simple linear regression model between fallout and chicken data for ^{137}Cs activity, concentrations were estimated at 14.8 Bqkg^{-1} and 5.7 Bqkg^{-1} in chicken meat and eggs, respectively (Figures 1 and 2). These values are consistent with the ^{137}Cs activity concentrations measured in 1986 in Serbia (4.72 Bqkg^{-1}) and Montenegro (23 - 61 Bqkg^{-1}). [13]

The surface deposit of ^{137}Cs activity delivered on ground by fallout after the Chernobyl accident also exponentially decreased; from 6410 Bqm^{-2} in 1986 to only 1.6 Bqm^{-2} in 2018. [2, 3, 4]

Figure 1 about here

Figure 2 about here

A few years after the Chernobyl accident, i.e. 1986-1988, the observed decrease of ^{137}Cs activity concentrations both in chicken meat and chicken eggs was quite rapid compared to the period that followed.

Similar bi-modal behavior regarding decrease of ^{137}Cs in chicken meat and eggs, parametrized by ecological half-life of ^{137}Cs was observed in some other foodstuffs in Croatia [13, 14] as well elsewhere. [15] The possible reasons for such bi-modal behaviour of ^{137}Cs half-lives in chicken and eggs could be direct contamination of chicken feedstuffs following the short-term fallout after the Chernobyl accident. In subsequent years, the direct contamination caused by Chernobyl fallout was not present since the radioactive material originating damaged reactor

did not reach stratosphere and the tropospheric residence time of radionuclides is much smaller compared to stratospheric. [16]

Pröhl et al. regarding bi-modal pattern of radiocaesium activity concentrations decrease in pasture vegetation growing on undisturbed soils argued that this behaviour seems to be specific for vegetation growing on peat and mineral soils low in clay minerals. [15]

Mück observed significant differences in ecological half-lives of ^{137}Cs in various foodstuffs (cereals, potatoes, lettuce, fruits, nuts, milk etc.) reflecting rapid steeper decline of activity concentrations in first year or two after the Chernobyl accident and much slower decline in subsequent years. [17] In the case of fruit Mück argued that it is not clear why those differences occur while for other foodstuffs as the possible reasons were mentioned direct contamination, and various uptake / removal processes characteristic for particular foodstuff.

It is reasonable to assume that the biological half-life of ^{137}Cs, in chickens remains the same regardless the extent of internal contamination and probably does not significantly affect the ecological half-life, especially because the latter one is much longer than the lifespan of chickens produced for human consumption.

Regarding ^{134}Cs, it was detected (being above the decision threshold of radiation measurements), but the activity concentrations were under the detection limit.

In addition to fission radionuclides, the gammaspectrometrical method allowed us to measure activity concentrations of naturally occurring ^{40}K. For the overall observed period, ^{40}K activity concentrations were measured to be 85.3±14.2 $Bqkg^{-1}$ in chicken meat and 52.2±6.1 $Bqkg^{-1}$ in chicken eggs. The maximum ^{40}K activity concentration in chicken meat was 115.0±2.0 $Bqkg^{-1}$ and minimum value was 54.4±0.6 $Bqkg^{-1}$. For eggs, the maximum and minimum ^{40}K activity concentration were 63.0±0.6 $Bqkg^{-1}$ and 37.9±0.2 $Bqkg^{-1}$respectively.

While ^{137}Cs activity concentrations in chicken meat and eggs were in excellent correlation, the coefficient of correlation being r=0.99, but no correlation was found for ^{40}K.

Ecological half-life of ^{137}Cs in chicken meat and eggs

A first order kinetic equation was used to parameterize time changes in the ^{137}Cs activity concentrations in chicken meat and egg samples and the measured data were fitted using the natural exponential function

$$A(t) = A(0)\, e^{-kt} \qquad /1/$$

where:

A (t) is the time-dependent activity concentration of ^{137}Cs in sample (Bqkg^{-1}),

A (0) initial activity concentration of ^{137}Cs in sample (Bqkg^{-1}) and

ln(2)/k effective half-life of ^{137}Cs in sample (years).

For the 1986-1988 period, by fitting the measured data to exponential curves /1/, the estimated ecological half-life of ^{137}Cs was found to be 0.43 and 0.47 years for chicken meat and eggs, respectively. However, for the 1988-2018 period, the effective half-life of ^{137}Cs was found to be 8.0 years for chicken meat (r=0.88) and 8.45 years in the eggs (r=0.85). However, in order to find the real ecological half-life as part of the effective half-life attributed to ecological processes only, the observed constants k from the equation /1/ should be corrected for the radioactive decay. Therefore, the formula can be written as:

$$k = \lambda + k_R \qquad /2/$$

where $\ln(2)/\lambda = 30.14$ y is the physical half-life of ^{137}Cs [18] and $T_{1/2,e} = \ln(2)/k_R$ is the ecological half-life for ^{137}Cs. From equation /2/, the real ecological half-life for ^{137}Cs in chicken meat and chicken eggs was found to be 10.8 and 11.8 years, respectively.

It should be mentioned that the radiocaesium uptake through feeding and the time of slaughtering could have impact on radiocaesium metabolism in chickens and transfer coefficient to eggs and therefore affect ecological half-life of ^{137}Cs in chicken meat and eggs. However, data on the origin of chicken feed is not available and these issues could not be addressed.

Regarding ^{134}Cs, in 1987, its activity concentration was found to be 1.20 ± 0.55 Bqkg^{-1} in chicken meat and 0.44 ± 0.17 Bqkg^{-1} in eggs. After 1990 it was under the detection limit but above the decision threshold quantifying the physical effect, which allows for a conclusion that ^{134}Cs was present in the samples.

In 1987, the $^{134}Cs/^{137}Cs$ activity ratio was 0.36 in chicken meat and 0.29 in chicken eggs, which is in both cases considerably lower compared to the theoretical value of 0.55. More precisely, the estimated amount of radiocaesium released after the reactor explosion at Chernobyl was ~85 PBq of ^{137}Cs and ~47 PBq of ^{134}Cs. [19] Therefore, the initial value for the ^{134}Cs:^{137}Cs activity ratio in radioactive material released in May 1986 from the damaged Chernobyl reactor was about 0.55.

The Fukushima-Daiichi nuclear accident (March 2011) did not cause an increase of ^{137}Cs activity concentrations both in chicken meat and chicken eggs compared to previous years.

Radioecological sensitivity

^{137}Cs fallout data along with activity concentrations in chicken meat and eggs allowed for an estimation of radioecological sensitivity, (R_s) as another important radioecological parameter. It is defined as the infinite integral of activity concentrations of a particular radionuclide in a given environmental sample to the integrated deposition. R_s is sometimes also called the transfer coefficient from fallout to sample and in the case of food samples it is equivalent to UNSCEAR's transfer coefficient P_{23}. [20] Mathematically, R_s (i.e., P_{23}) is defined as follows:

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

where:

A(t) is the activity concentration of a given radionuclide (Bqkg^{-1}) in a sample and

$\dot{U}$ (t) the fallout deposition rate of this radionuclide (Bqm^{-2}y^{-1}).

As for values of A(t) and $\dot{U}$ (t) assessed on a yearly basis, the integration can be replaced by summation, the value of P_{23} for ^{137}Cs in chicken meat for the 1987-2018 period can be easily calculated to be 4.06×10^{-3} Bqykg^{-1}/(Bqm^{-2}). For the same period, the P_{23} for ^{137}Cs in chicken eggs was 2.19×10^{-3} Bqykg^{-1}/(Bqm^{-2}). That means that with each Becquerel of ^{137}Cs deposited by fallout on an area of one square meter of land activity, the concentration of one ton of chicken meat increases approximately by 4.06 Bq, while the activity concentration of the same mass of chicken eggs i.e., one ton, increases approximately by 2.19 Bq.

To put the obtained values into perspective, the ^{137}Cs transfer coefficient P_{23} for a total diet was estimated to be approximately 1.2×10^{-2} Bqykg^{-1}/(Bqm^{-2}) for the 1962 - 1979 period in

Denmark [20], 2.1×10^{-2} Bqykg^{-1}/(Bqm^{-2}) for the 1987-2005 period in beef in Croatia [14] and 8.6×10^{-3} Bqykg^{-1}/(Bqm^{-2}) in honey for the 1986-1995 period in Croatia. [21]

Effective dose

The effective dose incurred due to the intake of a certain radionuclide over a specific time period by the consumption of contaminated food depends on the activity concentration in food and the consumed quantity. The dose can be expressed as:

$$E = C\sum_{m} D_m^{cf} A_m \qquad /4/$$

where:

E is the effective dose in Sv,

C is the total annual per capita consumption in Bqkg^{-1},

D_m^{cf} is the dose conversion factor for radionuclide *m* i.e., effective dose per unit intake, which converts the ingested activity to an effective dose and

A_m is the mean annual activity concentration of radionuclide *m* in sample (Bqkg^{-1}).

In the calculations, an annual consumption of 22.8 kg of chicken meat [8] and 9 kg of chicken eggs (about 160 pieces) [9] was assumed for the critical age class (>17 y). The dose conversion factor per unit intake via ingestion for adult members of the public for ^{137}Cs was 1.3×10^{-8} SvBq^{-1}. [18] The estimation of annual effective doses received by adult members of the Croatian population due to intake of ^{137}Cs by consumption of chicken meat and chicken eggs showed quite small doses in 1987 being 0.94 and 0.17 μSv respectively decreasing to 0.011 and 0.004 μSv in 2018. For the overall observed period, i.e, 1987-2018, the estimated effective doses received by adult members of the Croatian population due to the intake of radiocaesium by chicken meat and egg consumption are very small, 2.0 and 0.6 μSv

respectively. According to these estimations, the consumption of either chicken meat or chicken eggs is apparently not a critical pathway for the transfer of ^{137}Cs to humans.

Conclusion

The exponential decrease of ^{137}Cs cesium activity concentrations has been observed in both chicken meat and chicken eggs in the post-Chernobyl period. Since ^{137}Cs activity concentrations in chicken meat and eggs were in good correlation with its activity concentrations in fallout, this enables the development of simple mathematical models for the quick prediction of contamination in cases of nuclear accidents.

The transfer of ^{137}Cs from fallout to chicken meat and eggs, numerically represented as UNSCEAR's transfer coefficient P_{23}, has been found to be quite similar as for other foodstuffs.

Generally, a few years after the Chernobyl nuclear accident, the activity concentrations of ^{137}Cs in chicken meat and eggs were quite low, while ^{134}Cs activity concentrations after 1989 were below the detection limit of the instruments. Consequently, doses to the general population incurred by ^{137}Cs in chicken meat and eggs were quite small. It can therefore be concluded that, in Croatia, chicken meat and egg consumption was not a critical pathway for the transfer of radiocaesium from fallout to humans.

Monitoring for compliance with the accumulated maximum radioactive level in terms of total ^{134}Cs and ^{137}Cs activity concentration of 600 $Bqkg^{-1}$ for all food products except milk as stipulated in Council Regulation (EC) No 733/2008 of 15 July 2008 [22] implies a binary approach in addressing the hazard of the presence of radioactivity in foodstuffs intended for human consumption (below or above the prescribed level). However, advancements in

instrumentation that allow the lowering of detection limits of gammaspectrometrical instruments have brought about the possibility to obtain validated data on activity concentrations in analyzed foodstuffs and therefore the levels of radiation exposure. This allows one to observe and analyses trends of activity concentrations in various foodstuffs and implement appropriate protective measures if necessary.

Conflict of interest statement

The authors declare that there are no conflicts of interest.

References

[1]. Popović, V. (Ed.) Environmental Radioactivity in Yugoslavia, Annual Reports 1965–1977. Federal Committee for Labour, Health and Social Welfare: Belgrade, 1966–1978. In Croatian.

[2]. Bauman, A.; Cesar, D.; Franić, Z.; Kovač, J.; Lokobauer, N.; Marović, G.; Maračić, M.; Novaković, M. Results of Environmental Radioactivity Measurements in the Republic of Croatia, Annual Reports 1978–1991. Institute for Medical Research and Occupational Health: Zagreb, 1979-1992. In Croatian.

[3]. Kovač, J.; Cesar, D.; Franić, Z.; Lokobauer, N.; Marović, G.; Maračić, M. Results of environmental radioactivity measurements in the Republic of Croatia, Annual Reports 1992–1997. Institute for Medical Research and Occupational Health: Zagreb, 1993–1998. In Croatian.

[4]. Marović, G.; Franić, Z.; Kovač, J.; Lokobauer, N.; Maračić, M. Results of environmental radioactivity measurements in the Republic of Croatia, Annual Reports 1998–2005. Institute for Medical Research and Occupational Health. Zagreb: 1999–2006. In Croatian.

[5]. Petäjä, E.; Rantavaara, A.; Paakkola, O.; Puolanne, E. Reduction of radioactive caesium in meat and fish by soaking. J. Environ. Radioact. **1992**, *16*(3), 273-285.

[6]. Franić, Z.; Senčar, J.; Bauman, A. Speeding up meat decontamination by freezing Health Phys. **1993**, *65*(2), 216-217.

[7]. Ratnikov, A.N.; Vasiliev, A.V., Alexakhin, R.M.; Krasnova E.G.; Pasternak, A.D.; Howard B.J.; Hove, K.; Strand, P. The use of hexacyanoferrates in different forms to reduce radiocaesium contamination of animal products in Russia. Sci. Total Environ. **1998**, *223*(2–3), 167-176.

[8]. Kuskunović, M. Sabljak: Cekin chicken and Vindon turkey are the only ones that have a GMO free certificate in Croatia]. Agrobiz [Internet]. **2017**. Available from: https://www.agrobiz.hr/agrovijesti/sabljak-cekin-piletina-i-vindon-puretina-jedine-imaju-gmo-free-certifikat-u-hrvatskoj-7331

[9]. Senčić, Đ.; Samac, D. Nutritional value of eggs in human nutrition. Meso, **2017**, *1*(19); 68-72. In Croatian.

[10]. Dahlgaard H. (Ed.) Nordic Radioecology: The Transfer of Radionuclides through Nordic Ecosystems to man. Studies in environmental science 62. Elsevier:Amsterdam – Lausanne – New York,- Oxford – Shannon - Tokyo, 1994.

[11]. Accreditation Certificate No. 1288. Croatian Accreditation Agency. [Internet]. **2018**. Available from: http://www.akreditacija.hr/akreditacija/files/read.php?%20re=16244__akredFile&wr=1288

[12]. Petrinec, B.; Franic, Z.**;** Bituh, T.; Babić D. Quality assurance in gamma-ray spectrometry of seabed sediments. Arh Hig Rada Toksikol. **2011**, *62*(4), 17-23.

[13]. Bauman A. (Ed.) Environmental radioactivity in Yugoslavia – Data for year 1986. Federal Committee for Labour, Health and Social Welfare: Beograd, 1989. In Croatian.

[14]. Franic, Z.; Marovic, G.; Mestrovic, J. Radiocaesium contamination of beef in Croatia after the Chernobyl accident. Food and Chemical Toxicology. **2009**, *46*(6), 2096-2102.

[15]. Pröhl, G.; Ehlken, S.; Fiedler, I; Kirchner, G.; Klemt, E; Zibold, G. Ecological half-lives of ^{90}Sr and ^{137}Cs in terrestrial and aquatic ecosystems. Journal of Environmental Radioactivity . **2006**, *91*, 41-72.

[16]. Baskaran, M. (Editor) Handbook of Environmental Isotope Geochemistry. Springer: Heidelberg Dordrecht London New York, 2012.

[17]. Mück, K. Long-term effective decrease of cesium concentration in foodstuffs after nuclear fallout. Health Physics. **1997**; *72*, 659–673.

[18]. International Atomic Energy Agency (IAEA). Radiation protection and safety of radiation sources: International Basic Safety Standards. IAEA Safety Standards Series No. GSR Part 3. International Atomic Energy Agency: Vienna, 2014.

[19]. International Atomic Energy Agency (IAEA). Environmental consequences of the Chernobyl accident and their remediation: twenty years of experience. International Atomic Energy Agency: Vienna, 2006.

[20]. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). Ionizing Radiation: Sources and Biological Effects. United Nations: New York, 1982.

[21]. Franić, Z.; Branica, G. Long-term investigations of ^{134}Cs and ^{137}Cs Activity Concentrations in Honey from Croatia. Bulletin of Environmental Contamination and Toxicology. **2019**, *102*(4), 462-467.

[22]. Official Journal of the European Union. Council Regulation (EC) No 733/2008 of 15 July 2008 on the conditions governing imports of agricultural products originating in third

countries following the accident at the Chernobyl nuclear power station. European Council: 2008.

Table 1. ^{137}Cs and ^{134}Cs activity concentrations in fallout, chicken meat and chicken eggs (fresh weight). Activity concentrations are reported as ±2σ measurement uncertainties.

Year	Fallout	Chicken meat			Chicken eggs		
	Activity concentration ($Bqkg^{-1}$)						
	Bqm^{-2}	^{137}Cs	^{134}Cs		^{137}Cs	^{134}Cs	
	^{137}Cs						
1986	6410.0						
1987	1098.9	3.30 ± 1.21	1.20 ± 0.55		1.30 ± 0.33	0.44 ± 0.17	
1988	716.0	0.58 ± 0.38	0.50 ± 0.32		0.29 ± 0.07	0.24 ± 0.12	
1989	54.3	0.60 ± 0.39	0.27 ± 0.24		0.29 ± 0.07	0.24 ± 0.12	
1990	17.7	0.44 ± 0.33			0.27 ± 0.07		
1991	57.1				0.23 ± 0.06		
1992	31.0				0.09 ± 0.02		
1993	18.5				0.32 ± 0.08		
1994	10.4				0.32 ± 0.08		
1995	8.4	0.26 ± 0.06			0.26 ± 0.11		
1996	4.5	0.13 ± 0.02			0.10 ± 0.01		
1997	3.2	0.09 ± 0.01			0.05 ± 0.01		
1998	4.9	0.12 ± 0.02			0.09 ± 0.01		
1999	3.9	0.08 ± 0.01			0.12 ± 0.08		
2000	2.9						
2001	2.7	0.17 ± 0.02			0.06 ± 0.01		
2002	2.2	0.19 ± 0.02			0.03 ± 0.00		
2003	1.9	0.08 ± 0.01			0.06 ± 0.01		
2004	2.1	0.05 ± 0.01			0.06 ± 0.01		
2005	2.8	0.10 ± 0.01			0.08 ± 0.01		
2006	3.4	0.10 ± 0.01			0.04 ± 0.01		
2007	2.0	0.10 ± 0.01			0.07 ± 0.01		
2008	1.5	0.05 ± 0.03			0.06 ± 0.01		
2009	1.0	0.07 ± 0.01			0.04 ± 0.01		
2010	1.7	0.09 ± 0.01			0.02 ± 0.01		
2011	2.0	0.02 ± 0.00			0.03 ± 0.01		
2012	1.2	0.03 ± 0.01			0.06 ± 0.01		
2013	0.7	0.05 ± 0.01			0.04 ± 0.01		
2014	1.3	0.06 ± 0.01			0.03 ± 0.01		
2015	0.4	0.04 ± 0.02			0.03 ± 0.01		
2016	0.8	0.03 ± 0.01			0.02 ± 0.01		
2017	1.1	0.05 ± 0.01			0.04 ± 0.01		
2018	1.6	0.04 ± 0.01			0.03 ± 0.01		

Figure 1. ^{137}Cs activity concentrations in chicken meat (fresh weight) for the 1986-2018 period. The data for 1986 was estimated from fallout measurements.

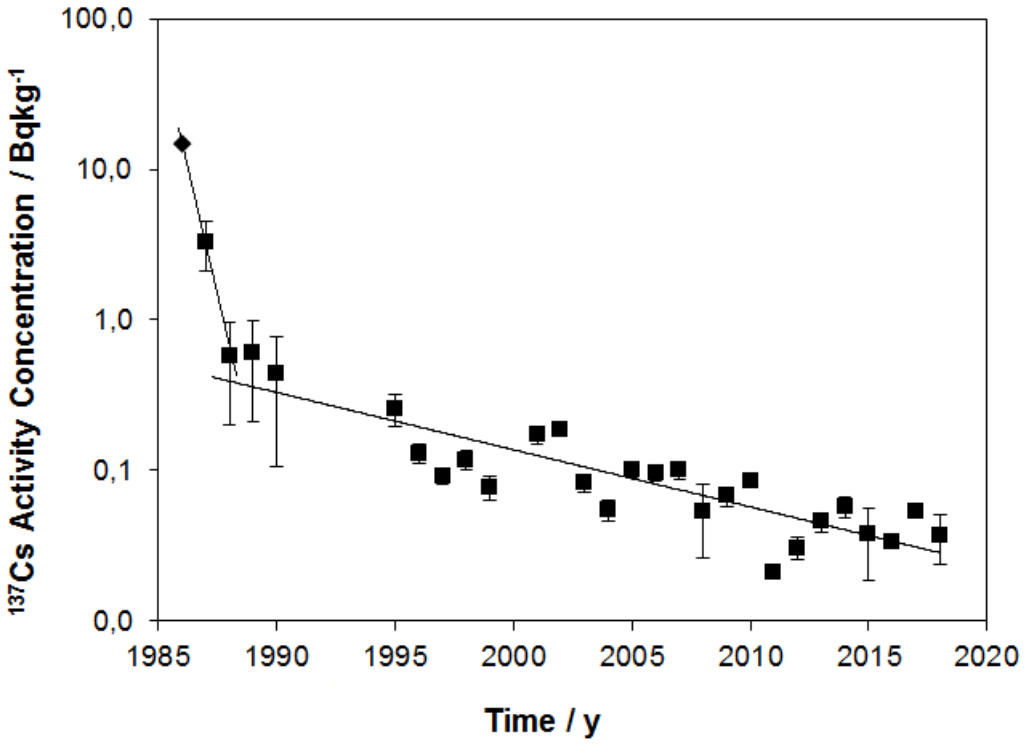

Figure 2. ^{137}Cs activity concentrations in chicken eggs (fresh weight) for the 1986-2018 period. The data for 1986 was estimated from fallout measurements.