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Long-Term Investigation (1968–2023) of ^{137}Cs in Apples

Branko Petrinec [1,2], Tomislav Bituh [1,*], Zdenko Franić [1], Branimir Zauner [1] and Dinko Babić [1]

1 Division of Radiation Protection, Institute for Medical Research and Occupational Health, Ksaverska cesta 2, HR-10000 Zagreb, Croatia; petrinec@imi.hr (B.P.)
2 Faculty of Dental Medicine and Health, Josip Juraj Strossmayer University of Osijek, Crkvena 21, HR-31000 Osijek, Croatia
* Correspondence: tbituh@imi.hr; Tel.: +385-(1)4682500

Abstract: Due to the consequences of nuclear and/or radiological accidents in the past (Chernobyl, Fukushima, etc.), and potential future events of that kind, the constant monitoring of environmental radioactivity is important. There are different pathways of the transfer of radionuclides from environment to humans (ingestion, inhalation and external). Food ingestion greatly contributes to the total effective dose; hence, it is of great importance to investigate exposure to radionuclides through food. This paper presents the results of a long-term investigation of ^{137}Cs activity concentration in apples in northwestern Croatia for the period 1968–2023. The highest ^{137}Cs activity concentration in apples was measured in 1986, decreasing exponentially ever since. The Fukushima-Daiichi accident in 2011 did not cause a significant increase in ^{137}Cs activity concentration, although the presence of the consequent fallout was detected via the appearance of ^{134}Cs in some parts of the environment. The observed residence time for ^{137}Cs in apples was estimated to be 4.5 and 3.9 years for the pre-Chernobyl and post-Chernobyl periods, respectively. The correlation between ^{137}Cs in fallout and apples is very good, the correlation coefficients being 0.99, which indicates that fallout is the main source of contamination. The estimated effective dose received by adult members of the Croatian public due to intake of radiocaesium from apples over the overall observed period is 6.4 µSv. Therefore, the consumption of apples was not a critical pathway for the transfer of radiocaesium to humans.

Keywords: apples; ecological half-life; residence time; ionizing radiation; monitoring of radioactivity; ^{137}Cs; effective dose

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1. Introduction

The monitoring of radioactivity in the environment is again becoming increasingly important due to recent global events that may pose a nuclear threat. Because of that, the long-term monitoring programs are of great importance. By analyzing data before the nuclear tests in the 1960s and those before, during and after large nuclear accidents (Chernobyl and Fukushima), one can infer how radioactivity levels in foodstuffs fluctuate.

Global pollution by anthropogenic radioactive matter is mainly caused by nuclear tests conducted in the atmosphere in the early 1960s and the releases of radioactive material from nuclear facilities. 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 Chernobyl and Fukushima-Daiichi nuclear accidents, originate in a (effectively) single point on the surface of earth [1,2]. This fallout can affect humans directly or indirectly (through food chain). Depending on whether radionuclides in the fallout have a short or long half-life, they may have adverse health effects on the population through direct irradiation and/or internal exposure after the ingestion of contaminated foodstuffs.

Two long-lived radioisotopes that have hazardous effects on humans are ^{137}Cs and ^{90}Sr. These radioactive elements are found mostly in the surface layers of soil, as their migration to lower layers is limited and depends on many chemical and physical parameters of a soil system. The low mobility of these radioactive elements in soil retains them in the root zone. However, the absorption can also occur through the stomata of the plants [3]. Plants assimilate radioactive and other non-radioactive substances necessary for their growth. This way, isotopes may propagate into animal tissues and finally, as part of foodstuff, into humans [4,5].

As fruits are sources of many essential nutrients, they are an important component of the human diet and are, therefore, included in the annual program of the radioactivity monitoring of the environment in Croatia. The regular investigations of the radioactive contamination of apples (*Malus domestica* Borkh.), as the most popular fruit among consumers, started in 1968. In Croatia, 15 kg/person of apples are consumed annually [6], which is below the world average. Some other orchard fruits, like grapes, plums, peaches, sweet cherries, raspberries, blackcurrant and strawberries, were investigated sporadically, especially after the Chernobyl accident. However, because of the lack of systematic data, these fruits are not addressed in this study.

Among anthropogenic radionuclides, those of the radiocaesium and radiostrontium families, particularly, ^{137}Cs and ^{90}Sr, are regarded as great potential hazards to living beings. Namely, 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 being excreted through milk, feces and urine [7]. It should be noted that, due to the lack of long-term data on ^{90}Sr, only ^{137}Cs was analyzed in this investigation.

Monte et al. [8] explain the three main pathways of the uptake of ^{137}Cs in apples:

- Deposition on soil $\rightarrow$ vertical migration in soil $\rightarrow$ root uptake $\rightarrow$ migration to the fruit and other components of plant.
- Deposition on exposed surfaces of plant $\rightarrow$ translocation to the interior of plant $\rightarrow$ migration to fruit.
- Deposition on exposed surfaces of fruit.

Of course, the relative importance of each of these migration pathways depends on the stage of the plant development and on the season during which contamination occurs. For instance, the direct deposition of a radionuclide on the external surface of a fruit is obviously negligible if the contamination takes place before the growth of the fruit itself. On the other hand, the efficiency of radionuclide translocation to the interior of a plant depends on seasonal conditions according to the state of development of the leaves [8].

According to statistical data, annual apple production in the Republic of Croatia in the period 2013–2017 ranged from 44.7 tons in 2016 to 128.2 tons in 2013 [6]. However, the consumption of apples per capita is relatively small and amounts to a modest 15 kg, compared to 50 kg, which is the average in the European Union [9].

The aim of this study is to present a long-term investigation of ^{137}Cs activity concentration in apples in northwest Croatia for the period 1968–2023 and compare the data to fallout sampled in the city of Zagreb during that same period.

2. Materials and Methods

According to the national monitoring program regulated by the Ministry of Interior [10], apples were commercially obtained in the open market after harvest season. 10 kg of apples were dried at 105 °C for 2–3 days, and then ashed in a muffle furnace at 400 °C for 24 h. It is important for the ashing temperature not to be higher than 400 °C due to the loss of ^{137}Cs in the samples. The samples were packed in 200 mL plastic cylindrical containers and analyzed by gamma-ray spectrometry.

Dry and wet (rainwater) fallout was collected daily using funnels of 1 m^2 collection area, positioned at 1.5 m above the ground. In days with rain and/or snow, the amount of precipitation that contained the fallout was measured by the Hellman rain gauge. In days without precipitation, funnels were rinsed by 1 L of distilled water in order to collect dry fallout. Daily samples were merged into monthly (pre-Chernobyl period) and quarterly (from the year of 1983) samples. After evaporation to a volume of 1 L, the samples were packed into Marinelli beakers (Nuclear Technology Services, Inc., Roswell, GA 30076, USA) for gamma-ray spectrometry analysis. The dry and wet deposition samples were sampled in the city of Zagreb (N45.833889; E15.978333) and represent the northwest of Croatia.

In the period 1968–2003, gamma-ray spectrometry systems based on a low-level ORTEC Ge(Li) detector (FWHM of 1.87 keV and relative efficiency of 15.4%, all at 1.33 MeV ^{60}Co) and an ORTEC HPGe detector (FWHM of 1.75 keV and relative efficiency of 21%, all at at 1.33 MeV ^{60}Co) coupled with a computerized data acquisition system were used. Since 2003, a low-level ORTEC HPGe detector (relative efficiency of 74.2% with FWHM of 2.24 keV, all at 1.33 MeV ^{60}Co) has been used. All of the gamma-ray spectrometry systems have been appropriately validated (trueness, precision/repeatability, trueness, limit of detection, matrix variation and measurement uncertainty) according to ISO 17025 requirements in order to assure an appropriate measurement quality [11].

To reduce background radiation, the detectors were shielded using a 10 cm thick lead container lined with 2 mm of Cd and 2 mm of Cu to avoid the X-rays of Pb shielding. The counting time for gamma-ray spectrometric measurements depended on the sample activity, typically being 250,000 s. ^{137}Cs activity concentration was calculated by analyzing 661 keV energy peak (85.01% probability). Corrections for self-attenuation and coincidence summing effects were applied. Certified calibration standards were obtained from the Czech Metrological Institute, covering energies in the range 40–2000 keV. Quality assurance was performed through proficiency testing organized by the International Atomic Energy Agency (IAEA) and Joint Research Centre (JRC), which also included regular checks of the background radiation and quality control measurements [12].

2.1. Effective Ecological Half-Life of ^{137}Cs in Apples

The effective half-life ($T_{1/2,eff}$) (synonyms: observed half-life, effective ecological half-life and effective environmental half-life) is a time over which the initial amount of the activity concentration of a studied radionuclide is reduced (usually following an exponential decrease) to the half of its initial value, which includes not only radioactive decay but also the effects of environmental and ecological processes. Residence time T_m is the measure of how long a particular radionuclide spends in a given ecological compartment (in this case, apples). It is related to $T_{1/2,eff}$ by the following equation:

$$T_m = 1/k = T_{1/2,eff}/ln(2). \quad (1)$$

However, the above definitions, although widely used in the literature on radioecology, are not accepted universally (for a more detailed discussion, see Dahlgaard, [13]).

Also, it should be noted that T_m (and, therefore, $T_{1/2,eff}$ as well) depends on the period for which it is estimated. From Figure 1, it is evident that after the Chernobyl accident, the ^{137}Cs activity concentration decreased quite rapidly until 1999.

2.2. Radioecological Sensitivity

Another important radioecological parameter, radioecological sensitivity (R_s), can be calculated from data on ^{137}Cs fallout and those on ^{137}Cs activity concentration in apples. It is defined as the ratio of the infinite integral of the activity concentration of a particular radionuclide in a given environmental sample and 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 [14] transfer coefficient from deposition to diet (P_{23}), defined as follows:

$$P_{23} = \frac{\int_0^\infty C(t)dt}{\int_0^\infty \dot{U}(t)dt} \quad (2)$$

where:

$C(t)$ is the concentration of a given radionuclide (Bq kg^{-1} or Bq L^{-1}) in a food sample and

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

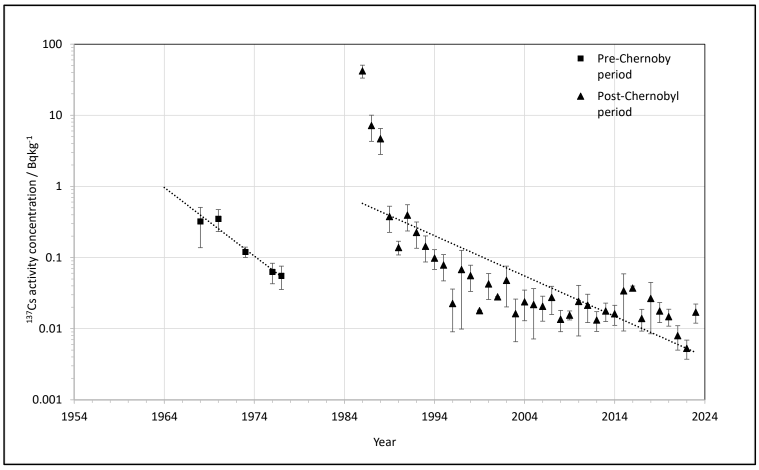

Figure 1. ^{137}Cs activity concentration in apples (Bq kg^{-1}) for the pre- and post-Chernobyl periods. The dotted line represents the exponential trendline for two periods. Error bars represent standard deviations of results for samples collected within a given year. (The data for the years 1987–1995, 1998 and 2000 are approximated).

2.3. Effective Dose

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

$$E = C \sum D_m A_m \quad (3)$$

where

E is the effective dose in Sv,

C is the total annual *per capita* ingestion of a radionuclide m by food consumption in Bq,

D_m is the dose conversion factor for a radionuclide m, i.e., the effective dose per unit intake, which converts the ingested activity to effective dose, and

A_m is the mean annual activity concentration of a radionuclide m in apples (Bq kg^{-1}).

3. Results and Discussion

3.1. ^{137}Cs Activity Concentration in Apples

The data for ^{137}Cs activity concentration in fallout and apples are presented in Table 1. The activity concentration of ^{137}Cs in apples decreased exponentially after the Chernobyl accident. The activity of ^{137}Cs per unit area due to deposition by fallout after the Chernobyl accident also decreased exponentially, from 6410 Bqm^{-2} in 1986 to only 0.1 Bq m^{-2} in 2022 [1,15,16]. The data on activity concentration in apples for 1987–1995, 1998 and 2000 are missing. In these years, the annual program for environmental monitoring in ex-Yugoslavia and, later, in the Republic of Croatia included other types of fruit; hence, there are no data on apples. If we a use simple linear relation, the ^{137}Cs activity concentrations in apples and fallout are correlated, the correlation coefficient being $r = 0.99$ ($P(t) < 0.05$). By using this, we can estimate their values in the years for which experimental data are missing. For the period 1986–2023, the activity concentration of ^{137}Cs in apples can be calculated using fallout data modelled as follows:

$$A_{apples}(t) = 0.0065 \times A_{fallout}(t) + 0.0169 \quad (4)$$

where

$A_{apples}(t)$ is the activity concentration of ^{137}Cs in apples (mBq kg^{-1}), and

$A_{fallout}(t)$ is the ^{137}Cs activity in fallout per unit area (Bq m^{-2}).

Table 1. ^{137}Cs activity concentration in fallout and apples from northwest Croatia.

Year	^{137}Cs in Fallout * [Bq m^{-2}]	Activity Concentration of ^{137}Cs in Apples A_{apples} [Bq kg^{-1}]	σ [Bq kg^{-1}]
1986	6410.0	42	9
1987	1098.9	** 7.2	2.9
1988	716.0	** 4.7	1.9
1989	54.3	** 0.4	0.2
1990	17.7	** 0.13	0.06
1991	57.1	** 0.4	0.2
1992	31.0	** 0.2	0.1
1993	18.5	** 0.13	0.06
1994	10.4	0.10	0.03
1995	8.4	** 0.07	0.03
1996	4.5	0.02	0.01
1997	3.2	0.07	0.06
1998	4.9	** 0.05	0.02
1999	3.9	0.018	0.001
2000	2.9	** 0.03	0.02
2001	2.7	0.028	0.001
2002	2.2	0.05	0.3
2003	1.9	0.02	0.01
2004	2.1	0.02	0.01
2005	2.8	0.02	0.01
2006	3.4	0.02	0.01
2007	2.0	0.02	0.01
2008	1.5	0.013	0.005
2009	1.0	0.015	0.002
2010	1.7	0.024	0.02
2011	2.0	0.02	0.01
2012	1.2	0.014	0.004
2013	0.7	0.02	0.01
2014	1.3	0.01	0.01
2015	0.4	0.034	0.002
2016	0.8	0.037	0.005
2017	1.1	0.014	0.02
2018	0.5	0.03	0.01
2019	0.6	0.018	0.004
2020	0.2	0.015	0.004
2021	0.1	0.008	0.003
2022	0.1	0.005	0.002
2023	0.6	0.017	0.005

Note: The data for activity concentration in apples are taken from the annual reports of the monitoring of environmental radioactivity in Yugoslavia and the Republic of Croatia from 1986–1989 and 1992–2023, respectively [17–22]. Other data is from unpublished results of measurements done by the Division of Radiation Protection of the Institute for Medical Research and Occupational Health. * Fallout activity data taken from [1,15,17–22]). ** Values calculated from fallout data using the model described in the text.

If we use the data from Table 1 and calculate the correlation adding the approximated data to the model, the new correlation coefficient is r = 0.99 ($P(t)$ < 0.05). Although these are rough predictions of activity concentration of ^{137}Cs in apples, the results prove that the linear model is adequate, and it can be used for further analysis.

3.2. Effective Ecological Half-Life of ^{137}Cs in Apples

The activity concentration of ^{137}Cs in apples for the pre- and post-Chernobyl period is shown in Figure 1. The error bars represent the standard deviations of activity concentrations for samples collected in a given year. The dotted line represents the exponential trendline for two periods. The data for the pre-Chernobyl period are taken from reports on environmental monitoring in the northern part of ex-Yugoslavia [18] and refer to northwestern Croatia. This ^{137}Cs originates in nuclear weapons tests in the 1960s.

After the mentioned intensive period of atmospheric nuclear weapons tests, the activity concentration of ^{137}Cs in apples decreased exponentially (Figure 1).

$$A_{apple}(t) = A_{apple}(0) \times e^{-kt} \quad (5)$$

where

$A_{apple}(t)$ is the ^{137}Cs activity concentrations in apples (in Bq kg^{-1}),

$A_{apple}(0)$ is the ^{137}Cs activity concentrations in apples in year in which measurements started, and

k is the exponential-decay constant.

The parameter k can be used to estimate two similar and yet conceptually different radioecological parameters: the effective half-life and the mean residence time of ^{137}Cs in apples. Both of them generally describe a decrease of an activity concentration in a given matrix.

The estimates of T_m and $T_{1/2,eff}$ from our data are presented in Table 2.

Table 2. Residence time, effective ecological half-lives and activity concentrations of ^{137}Cs in apples.

	Observed Period 1968–1976
k (years^{-1})	0.2219
Residence time = 1/k (years)	4.51
Effective (observed) ecological half-life $T_{1/2,eff}$ (years)	3.12
	Observed period 1986–1999
k (years^{-1})	0.5706
Residence time = 1/k	1.75
Effective (observed) ecological half-life $T_{1/2,eff}$ (years)	1.22
	Observed period 1986–2023
k (years^{-1})	0.257
Residence time = 1/k	3.89
Effective (observed) ecological half-life $T_{1/2,eff}$ (years)	2.70
Activity concentration (Bq kg^{-1})	**Year 1968**
	0.3 ± 0.2
	Year 1977
	0.055 ± 0.004
	Year 1986
	42 ± 9
	Year 2023
	0.017 ± 0.005

Namely, the presence of ^{137}Cs in fallout in the pre-Chernobyl period was a consequence of intense nuclear weapons tests in which the fission products reached the stratosphere. However, after the Chernobyl accident, radiocaesium from the damaged reactor core became part of the airborne plume, which remained confined to the troposphere. In the troposphere, dry and wet fallout remove any contaminant on the time scale of weeks to months, while in the stratosphere, the mean residence time for contaminants is much longer due to thermal stratification and separation from the troposphere by the tropopause. In comparison, the mean residence time for radiocaesium in the stratosphere has been estimated to be 2.5–5 years in the last few decades [23].

Our calculated results for apples are consistent with the values reported by Mück [24] on the basis of Austrian fruit sampled after the Chernobyl accident, where the estimated effective half-life for the period of 1987 to 1993 was found to be 507 d (1.38 y). In comparison, the effective half-life for the same period in Croatian apples is 602 d (1.65 y).

It should be noted that ^{137}Cs activity concentrations reported in 1986, immediately after Chernobyl accident, for some other fruits were quite similar to those in apples: 67 Bq kg^{-1} in raspberries, 51 Bq kg^{-1} in cherries and 37 Bq kg^{-1} in strawberries [17].

The Fukushima-Daiichi nuclear accident (March 2011) did not cause a significant increase in ^{137}Cs activity concentration (0.02 $\pm$ 0.01 Bq kg^{-1} in 2011 and 0.014 $\pm$ 0.004 Bq kg^{-1} in 2012) [21]. It should be noted that during the months following the Fukushima-Daiichi accident, ^{134}Cs was detected in some other parts of Croatia [25–27].

3.3. Radioecological Sensitivity

Since we assessed the values of $A(t)$ and $U(t)$ on a yearly basis, the integration can be replaced by a summation, which results in the value of P_{23} for the ^{137}Cs in apples for the 1986–2023 period being 0.006732 Bq y kg^{-1}/Bq m^{-2}. This means that 1 Bq of ^{137}Cs deposited by fallout over an area of one square meter increases the activity concentration of 1 kg of apples i by approximately 0.006732 Bq.

To put the obtained values into perspective, the ^{137}Cs transfer coefficient P_{23} for the total diet was estimated to be approximately 0.012 Bq y kg^{-1}/Bqm^{-2} for the 1962–1979 period in Denmark [14], 0.021 Bq y kg^{-1}/Bq m^{-2} in beef for the 1987–2005 period in Croatia [6], 0.0086 Bq y kg^{-1}/Bqm^{-2} in honey for the 1986–1995 period in Croatia [28] and 0.00334 Bq y kg^{-1}/Bqm^{-2} for chicken meat in 1987–2017 in Croatia [1].

3.4. Effective Dose

To estimate the effective dose E due to the intake of ^{137}Cs, an annual consumption of 15 kg of apples was assumed for the critical age (>17 y) [6]. The dose conversion factor per unit intake via ingestion for adult members of the public, for ^{137}Cs, is 1.3×10^{8} Sv Bq^{-1} [29].

The estimation of the annual effective doses received by adult members of the Croatian population due to the intake of ^{137}Cs by the consumption of apples showed quite small doses, ranging from 8.19 μSv in 1986, i.e., immediately after the Chernobyl accident, to only 1.56 nSv in 2021. In comparison, in 1968, i.e., in the year when measurements started, the estimated dose was 63 nSv. For the overall post-Chernobyl period (1986–2023), the estimated effective dose was 6.4 μSv, which is about 0.005% of the average yearly dose from the ingestion of foodstuffs by an adult Croatian member of the public [17–22]. Therefore, it can be concluded that in Croatia, the consumption of apples has not been a critical pathway for the transfer of ^{137}Cs to humans.

It should be noted, however, that after the Chernobyl accident, consumers were strongly advised to thoroughly wash and clean fruits and vegetables before consumption in order to remove radionuclides deposited by direct atmospheric deposition.

4. Conclusions

To our knowledge, this is the first report on the long-term monitoring of ^{137}Cs in apples. In the post-Chernobyl period, an exponential decrease in ^{137}Cs activity concentration has been observed in apples, similar to other foodstuffs that were exposed to the post-Chernobyl fallout. The residence times for ^{137}Cs were found to be 4.5 and 3.9 years for the pre-Chernobyl and post-Chernobyl periods, respectively.

Since the observed ^{137}Cs activity concentration in apples is in a good correlation with its activity concentration in the fallout, this enables the development of simple mathematical models as useful tools for a quick prediction of contamination in case of a nuclear accident.

The transfer of ^{137}Cs from fallout to apples, numerically represented as UNSCEAR's transfer coefficient P_{23}, has been found to be quite similar to that in other foodstuffs.

Generally, a few years after the Chernobyl nuclear accident, the activity concentration of ^{137}Cs in apples was quite low. Consequently, the doses to general population received by ^{137}Cs from apples were small. It can be concluded that in Croatia, the consumption of apples was not a critical pathway for the transfer of radiocaesium from fallout to humans.

Monitoring for compliance with the accumulated maximum radioactive levels in terms of total ^{137}Cs activity concentration of 600 Bq kg^{-1} for all food products except milk, as stipulated in European Commission Regulation 1158/2020 from 5th of August 2020 [30], implies a sort of binary approach to addressing the hazard of the presence of radioactivity in foodstuffs intended for human consumption (below or above the prescribed level). However, much lower detection limits of gamma-ray spectrometry instruments lead to a possibility to obtain validated data in analyzed foodstuffs and, therefore, the levels of radiation exposure. This allows to observe and analyze the trends of concentrations of radionuclides in foodstuffs and implement appropriate protective measures if necessary.

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Data Availability Statement: Data used in this investigation are publicly available within the annual reports for environmental monitoring of Republic of Croatia (1968–2023) and are deposited in the National and University Library in Zagreb, Croatia.

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Conflicts of Interest: The authors declare that there are no conflicts of interest.

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