NUKLEONIKA 2020;65(3):193–198
doi: 10.2478/nuka-2020-0030

ORIGINAL PAPER

Long-term investigation of ^{137}Cs and ^{134}Cs in drinking water in the city of Zagreb, Croatia

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Abstract. This paper presents the results of long-term investigations of ^{137}Cs and ^{134}Cs activity concentrations in drinking water in the city of Zagreb for the period 1987–2018. The highest activity concentrations of both radionuclides were measured in 1987, decreasing exponentially ever since, while ^{134}Cs in several subsequent years fell under the detection limit. After the Fukushima Daiichi accident in 2011, the presence of ^{134}Cs in drinking water was detected again. The environmental residence time for ^{137}Cs was estimated to be 8.1 years in drinking water and 5.7 years in fallout. The correlation between ^{137}Cs in fallout and in drinking water is very good, and this indicates that fallout is the main source of water contamination. The observed ^{134}Cs/^{137}Cs activity ratio in drinking water for the post-Chernobyl period was similar to the ratio found in other environmental samples. The estimation of annual effective doses received by the adult members of the Croatian population due to the intake of radiocaesium in drinking water showed quite small doses of 0.28 μSv in 1987 decreasing to 2.5 nSv in 2018, which indicated that drinking water was not a critical pathway for the transfer of radiocaesium to humans.

Keywords: Drinking water • Ecological half-life • Ionizing radiation • Monitoring of radioactivity • Radiocaesium

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Received: 4 January 2019
Accepted: 18 February 2020

Introduction

Owing to its daily consumption by living beings and the ability to transport pollutants, radiometric investigations of drinking water and the associated dose estimations are of considerable concern in environmental studies. More precisely, water is an early indicator of radioactive contamination. Drinking water and household water are potentially important pathways of various radionuclides to humans and animals, either directly or through their use in food preparation and processing. Water consumed by livestock and its use for irrigation purposes can also be a source of various radionuclides in foods.

Public water supplies are of the utmost importance for a progressive increase in urbanization, linking natural and urban water cycles. However, although they are clearly beneficial in distributing water of uniform quality through public water supplies, a large number of people become exposed to the risk of suffering adverse effects when the water is unsafe to drink.

The use of radiologically contaminated water as drinking water for humans and animals may result in high doses of ionizing radiation, particularly to the gastrointestinal tract. Regarding internal exposure from ingestion of water and the contamination of environmental materials, which are part of the immediate pathways leading to contamination of food, the most important radionuclides to be as-

sessed following a release of radionuclides from a uranium-fuelled reactor to the environment are ^{134}Cs, ^{137}Cs (^{137m}Ba), ^{131}I as well as other gamma emitters.

The official requirements for the protection of the health of the general public with regard to radioactive substances in water intended for human consumption were laid down in Council Directive 2013/51/Euratom [1]. This directive requires that each EU member state has to establish an appropriate monitoring programme for water intended for human consumption. This was implemented in Croatian legislative through the Water for Human Consumption Act and subsequent amendments [2].

The European and national legal frameworks set out indicator parameters related to the monitoring of radioactive substances and related provisions. However, in the Republic of Croatia, monitoring of fission products in drinking water started after the Chernobyl nuclear accident as part of an extended monitoring programme of radioactive contamination of the human environment in Croatia. The monitoring results, reported annually [3], provide a large database of gamma emitters in drinking water.

This article presents the results of a long-term investigation of the main radiocaesium isotopes, that is, ^{134}Cs and ^{137}Cs in drinking water in the city of Zagreb, the capital of Croatia. Unfortunately, data for 1986, that is, the year of Chernobyl accident, are not available as monitoring started only from 1987.

Material and methods

As the population of the Zagreb metropolitan area is more than 1 million people, that is, about one-fourth of the entire Croatian population, the monitoring of radioactive contamination in drinking water devotes special attention to the city of Zagreb. The main and basic source of drinking water for the city of Zagreb and the neighbouring city of Samobor is the groundwater from an alluvial granular aquifer, which is influenced by the Sava River. Therefore, tap water in the entire city of Zagreb has uniform properties, and the location of the Institute for Medical Research and Occupational Health (Zagreb) has been chosen by Croatian state authorities to be a representative place for its radiological characterization. Samples of drinking water supplied to a tap (valve) are collected daily, which amounts to 1 L. The collected water is combined in a quarterly sample of about 90 L, which is evaporated to 1 L volume.

Dry and wet fallout (rainwater) is collected daily using funnels of 1 m^2 collection area. On rainy days, the amount of precipitation that contained the fallout was measured by the Hellman pluviometer. On days without precipitation, funnels were rinsed by 1 L of distilled water. Daily samples are merged into cumulative samples that are analysed twice a year. Before gamma-spectrometric measurements, bi-annual samples were evaporated to a 1 L volume.

The most widespread analytical method available for detecting, measuring, and monitoring caesium in environmental samples, including drinking water and fallout, is gamma spectrometry [4].

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 efficacy 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 gamma spectrometry has been using a low-level high-purity ORTEC HPGe detector (relative efficacy of 74.2% with full width at half maximum (FWHM) resolution of 2.24 keV at 1.33 MeV). Both gamma-ray spectrometry systems have been appropriately validated to assure the quality of activity concentration measurements.

To reduce background radiation, the detectors are shielded with a 10 cm thick lead that is lined with 2 mm of cadmium and 2 mm of copper. The counting time for gamma spectrometric measurements depended on the sample activity, typically being 250 000 s.

Fallout and drinking water samples were measured in Marinelli beakers of 1 L volume that were placed directly onto the detector.

Gamma spectrometrical method was accredited in 2010 by the Croatian Accreditation Agency and therefore internationally recognized [5].

The detection limit of Council Directive 2013/51/Euratom [1] requires that the method of analysis used for ^{134}Cs and ^{137}Cs activity concentrations in water must, at a minimum, be capable of measuring activity concentrations with a limit of detection 0.5 Bq·L^{-1}, but the detection limit (which mainly depends upon sample counting time) of the accredited method described earlier was more than 10^3 times lower, being $\sim 10^{-4}$ Bq·L^{-1}.

Certified calibration standards, that is, mixed gamma water standard sources were obtained from the Czech Metrological Institute covering energies 40–2000 keV.

Quality assurance and intercalibration measurements were performed through intercalibration programmes organized by the International Atomic Energy Agency (IAEA) and Joint Research Centre (JRC), which also included the regular performance of blanks (empty cylindrical containers), background and quality control measurements [6].

Results and discussion

The activity concentrations of ^{137}Cs in drinking water exponentially decreased following the Chernobyl accident from 17.68 ± 6.61 Bq·m^{-3} in 1987 to only 0.27 ± 0.03 Bq·m^{-3} in 2018, which is shown in Fig. 1.

The measured data (excluding the years for which proper data are not available, i.e., years 1992 and 1993) were fitted to an exponential curve

$$A_{\text{water}}(t) = A_{\text{water}}(0) \times e^{-\lambda t} \quad (1)$$

where $A_{\text{water}}(t)$ is time-dependant ^{137}Cs activity concentrations in drinking water (in Bq·m^{-3}),

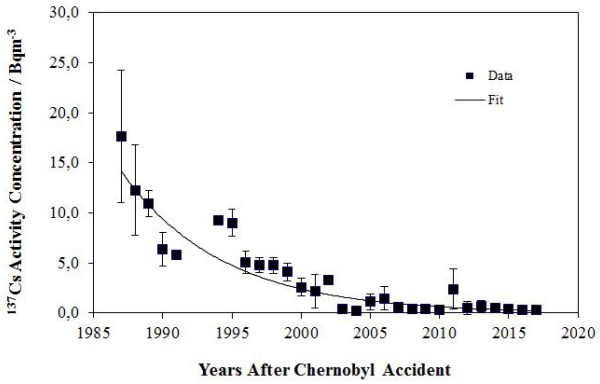

Fig. 1. ^{137}Cs activity concentrations in drinking water.

$A_{water}(0)$ = 17.68 Bq·m^{-3} is ^{137}Cs activity concentrations in drinking water in year 1987, and λ = 0.124 year^{-1} is parameter obtained from exponential function fitted to the observed data.

The coefficient of correlation between measured data and fit is r = 0.94. The value of the parameter λ obtained from exponential function (1) fitted to the observed data for estimation of residence time which is mathematically equal to $1/\lambda$. We have not used correction for radio decay constant of respective radionuclides; therefore, observed residence time estimated that way is environmental residence time. Environmental residence time for drinking water was found to be 8.1 years, which is slightly higher than the mean environmental residence time of this radionuclide in the fallout, that is, 5.7 years. For comparison, for the period 1988–1997, the environmental ^{137}Cs residence time in cistern waters in Croatia has been reported to be 6.9 ± 0.8 years [7].

The ^{137}Cs residence time in drinking water is connected with the characteristics of the aquifer from which that water originates. The ^{137}Cs residence time in aquifers (as well as in freshwater ecosystems) depends upon water residence time, effects of temporal variations in water demand, runoff, biological activity, geological characteristics of the aquifer, and so on. The water residence time, itself, depends among other factors upon inflow and outflow and the volume of water body (lake or aquifer). Some other affecting factors are the climate of the area, precipitation, drainage, evapotranspiration, and discharge. It should be noted that an estimated ^{137}Cs residence time in drinking water reflects the characteristics of the aquifer in the Zagreb area.

Regarding ^{134}Cs, after the Chernobyl accident, in 1987, its activity concentration in drinking water was found to be 6.28 ± 2.08. In several subsequent years, it was under the detection limit but above the decision threshold which indicated that it was present in the sample.

It should be noted that the derived concentrations for radioactivity in water intended for human consumption for adult man, calculated for a dose of 0.1 mSv, an annual intake of 730 L and using the dose coefficients laid down in Annex III, Table A of Directive 96/29/Euratom [8] that are in the cases of ^{134}Cs and ^{137}Cs identical to those of the International Atomic Energy Agency [9] are 11 000 Bq·m^{-3} for ^{137}Cs and 7200 Bq·m^{-3} for ^{134}Cs.

In 1987, the ^{134}Cs/^{137}Cs activity ratio was 0.36, which is close to the theoretical value of 0.40. More precisely, the estimated amount of caesium released after the reactor explosion at Chernobyl was ~85 PBq of ^{137}Cs and ~47 PBq of ^{134}Cs [10]. Therefore, the initial value for the ^{134}Cs : ^{137}Cs activity ratio in May 1986 was about 0.55. As the half-life of ^{137}Cs, being 30 years, is about 15 times longer than that of ^{134}Cs (2.04 years), the ^{134}Cs : ^{137}Cs activity ratio decreases according to differential radioactive decay. This study, as well as radioecological studies of some other environmental samples [11, 12], showed that this activity ratio has not been altered during the passage of the radioactive plumes from Chernobyl to other geographical areas in Europe.

The Fukushima Daiichi nuclear accident (March 2011) caused only a minor increase of ^{137}Cs activity concentrations in drinking water, from 0.33 ± 0.14 Bq·m^{-3} in 2010 to 2.40 ± 2.01 Bq·m^{-3} in 2011. Again, the presence of ^{134}Cs in drinking water was detected, but it was under the detection limit.

In addition to fission radionuclides, the gamma spectrometrical method allowed us to measure the activity concentrations of naturally occurring ^{40}K in drinking water. For the overall observed period, ^{40}K activity concentrations were measured to be 86.8 ± 34.7 Bq·m^{-3}. These values were consistent with activity concentrations of ^{40}K measured in the river Sava, the major river that flows through the city of Zagreb influencing its aquifer.

The observed ^{137}Cs activity concentrations in drinking water were correlated to the ^{137}Cs activity concentrations deposited by fallout on the ground using a simple linear equation:

$$A_{water}(t) = a + b \times A_{fallout}(t) \quad (2)$$

where $A_{water}(t)$ is time-dependant ^{137}Cs activity concentrations in drinking water (in Bq·m^{-3}), $A_{fallout}(t)$ is time-dependant ^{137}Cs activity concentrations of fallout deposited to ground (in Bq·m^{-2}), and a and b are parameters.

The respective values of parameters a and b estimated from Eq. (2) by linear regression are a = 2.67 Bq·m^{-3} and b = 0.014 m^{-1}.

^{137}Cs activity concentrations in drinking water are in good correlation with ^{137}Cs activity concentrations deposited by fallout on the ground, r = 0.75, which confirms that fallout is a primary source of radiocaesium in drinking water. The surface deposit of ^{137}Cs activity on the ground after the Chernobyl accident also exponentially decreased from 1098.9 Bq·m^{-2} in 1987 to 1.6 Bq·m^{-2} in 2018.

Radiocaesium fallout data along with activity concentrations in drinking water allow 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 (United Nations Scientific Committee on the Effects of Atomic Radiation) transfer

coefficient P_{23} [13]. Mathematically, R_s (i.e., P_{23}) is defined as follows:

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

where $A(t)$ is the activity concentration of given radionuclide (in Bq·kg^{-1} or Bq·L^{-1}) in sample and $\dot{U}(t)$ the fallout deposition rate of this radionuclide (in Bq·m^{-2}·year^{-1}).

As for values of $A(t)$ and $\dot{U}(t)$ assessed on a yearly basis, the integration can be replaced by summation and the value of P_{23} for ^{137}Cs in drinking water for 1987–2018 period can be easily calculated from Table 1, i.e., [(0.127 Bq·L^{-1})/(2061.939 Bq·m^{-2}·year^{-1})] = 6.16 × 10^{-5} Bq·year·L^{-1}/(Bq·m^{-2}). That means that with each becquerel of ^{137}Cs deposited by fallout on an area of 1 m^2 of land, the activity of 1 m^3 of drinking water increases approximately by 61.5 mBq of ^{137}Cs.

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

$$E = C\sum_m D_m^{cf} A_m \quad (4)$$

where E is the effective dose (in Sv), C is the total annual *per capita* intake of water (in L), D_m^{cf} is the dose conversion factor for radionuclide m, i.e., effective dose per unit intake, which converts the ingested activity to effective dose, and A_m is the mean annual specific activity of radionuclide m in water (in Bq·L^{-1}).

In the calculations, a daily consumption of 2 L of drinking water was assumed for the critical age class (>17 years) [14]. The dose conversion factor per unit intake via ingestion for adult members of the public is 1.3 × 10^{-8} Sv·Bq^{-1} and 1.9 × 10^{-8} Sv·Bq^{-1} for ^{137}Cs and ^{134}Cs, respectively [9].

Our estimation of annual effective doses received by the adult members of the Croatian population due to intake of radiocaesium in drinking water revealed quite small doses of 0.28 μSv in 1987

Table 1. ^{137}Cs and ^{134}Cs activity concentrations in fallout and drinking water and related effective doses to an adult person drinking 2 L of water per day

Year	Fallout [Bq·m^{-2}]	Drinking water				
		Activity concentration [Bq·m^{-3}]		Effective dose [nSv]		
	^{137}Cs	^{137}Cs	^{134}Cs	^{137}Cs	^{134}Cs	Total
1986	6410.0					
1987	1098.9	17.68 ± 6.61	6.28 ± 2.08	167.7 ± 62.7	110.3 ± 28.9	278.0 ± 69.0
1988	716.0	12.28 ± 4.48	<3.48 ± 1.19	116.5 ± 42.5	<48.2 ± 16.5	<164.7 ± 45.6
1989	54.3	10.93 ± 1.27	<2.18 ± 0.90	103.7 ± 12.0	<30.2 ± 12.6	<133.9 ± 17.4
1990	17.7	6.38 ± 1.66		60.5 ± 15.7		78.1 ± 19.0
1991	57.1	5.84 ± 0.46		55.4 ± 4.3		55.4 ± 4.3
1992	31.0	<12.24 ± 7.85		<116.1 ± 74.5		<116.1 ± 74.5
1993	18.5	<12.61 ± 1.62		<119.7 ± 15.4		<119.7 ± 15.4
1994	10.4	9.28 ± 0.44		88.1 ± 4.2		88.1 ± 4.2
1995	8.4	9.01 ± 1.31		85.5 ± 12.5		85.5 ± 12.5
1996	4.5	5.07 ± 1.14		48.1 ± 10.8		48.1 ± 10.8
1997	3.2	4.81 ± 0.74		45.6 ± 7.0		45.6 ± 7.0
1998	4.9	4.75 ± 0.78		45.1 ± 7.4		45.1 ± 7.4
1999	3.9	4.08 ± 0.85		38.7 ± 8.0		38.7 ± 8.0
2000	2.9	2.58 ± 0.86		24.5 ± 8.1		24.5 ± 8.1
2001	2.7	2.19 ± 1.67		20.7 ± 15.9		20.7 ± 15.9
2002	2.2	3.27 ± 0.40		31.1 ± 3.8		31.1 ± 3.8
2003	1.9	0.39 ± 0.28		3.7 ± 2.7		3.7 ± 2.7
2004	2.1	0.22 ± 0.03		2.1 ± 0.3		2.1 ± 0.3
2005	2.8	1.12 ± 0.81		10.6 ± 7.7		10.6 ± 7.7
2006	3.4	1.47 ± 1.19		14.0 ± 11.3		14.0 ± 11.3
2007	2.0	0.57 ± 0.28		5.4 ± 2.7		5.4 ± 2.7
2008	1.5	0.36 ± 0.18		3.4 ± 1.7		3.4 ± 1.7
2009	1.0	0.37 ± 0.13		3.5 ± 1.3		3.5 ± 1.3
2010	1.7	0.33 ± 0.14		3.1 ± 1.3		3.1 ± 1.3
2011	2.0	2.40 ± 2.01	<0.31 ± 0.11	22.7 ± 19.1	<4.3 ± 1.0	<27.0 ± 19.2
2012	1.2	0.50 ± 0.34		4.7 ± 3.2		4.7 ± 3.2
2013	0.7	0.68 ± 0.54		6.4 ± 5.1		6.4 ± 5.1
2014	1.3	0.46 ± 0.11		4.4 ± 1.1		4.4 ± 1.1
2015	0.4	0.38 ± 0.10		3.6 ± 0.9		3.6 ± 0.9
2016	0.8	0.28 ± 0.02		2.6 ± 0.2		2.6 ± 0.2
2017	1.1	0.29 ± 0.07		2.8 ± 0.7		2.8 ± 0.7
2018	1.6	0.27 ± 0.03		2.5 ± 0.3		2.5 ± 0.3

Activity concentrations are reported as ±2σ measurement uncertainties.

(0.17 ± 0.06 μSv due to ^{137}Cs and 0.11 ± 0.03 μSv due to ^{134}Cs), decreasing to only 2.5 ± 0.3 nSv in 2018. The overall dose in 1987–2018 period was estimated to be <1.4 μSv. Therefore, it can be concluded that drinking water is not a critical pathway for the transfer of radiocaesium to humans. In Table 1, ^{137}Cs and ^{134}Cs activity concentration in fallout and drinking water and related effective doses with estimated uncertainties are presented.

Conclusion

When monitoring for compliance with the indicative dose (ID), very high detection limit for gamma emitters in water, as stipulated in 2013/51/Euratom Council Directive [1], implies a sort of binary approach in addressing the hazard of the presence of radioactivity in water intended for human consumption (below or above ID). However, much smaller detection limits lead to validate data on activity concentrations in drinking water and therefore the levels of radiation exposure. This allows to observe and analyse trends and implement appropriate protective measures if necessary.

The activity concentrations of ^{137}Cs in drinking water in Zagreb exponentially decreased following the Chernobyl accident from 17.68 ± 6.61 Bq·m^{-3} in 1987 to 0.27 ± 0.03 Bq·m^{-3} in 2018. The Fukushima Daiichi accident did not cause a significant increase of radiocaesium in drinking water in Zagreb.

The observed mean residence time of ^{137}Cs in drinking water was found to be 8.1 years, which is a little bit longer than in fallout, that is, 5.7 years.

As the ^{137}Cs activity concentrations in drinking water are very well correlated with its activity concentrations in fallout, it is reasonable to assume that fallout is, either directly or indirectly, the main source of radioactive contamination of drinking water in Zagreb. However, radioecological sensitivity that relates ^{137}Cs activity concentrations in drinking water and fallout for the period 1987–2018 is quite small, being 6.15×10^{-5} Bq·year·L^{-1}/(Bq·m^{-2}).

The total effective dose incurred by ^{134}Cs and ^{137}Cs that a hypothetical adult person >17 years drinking 2 L of water per day would receive would have been about 0.28 μSv in 1987, decreasing to 2.5 nSv in 2018.

Generally, it can be concluded that the use of water from the public supply system in Zagreb as drinking water is not a critical pathway for the transfer of radiocaesium to humans after the Chernobyl accident. However, the analysis of radioactive contamination of drinking water is a valuable tool for the assessment of environmental radioactive contamination of fission radionuclides.

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