Long-term investigations of ^{134}Cs and ^{137}Cs Activity Concentrations in Honey from Croatia

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Introduction

Honey is among the top products at risk of food fraud. This is especially true for net importers of honey like the European Union (EU). Although the EU produces about 250,000 tonnes per year domestic production covers only around 60% of consumption and the EU is also a net importer of honey, which makes it the largest consumer of honey with an average per capita consumption of 0.9 kg (European Commission, 2017). In the EU, honey is consistently among the top 10 products with cases of food fraud. Counterfeit honey not only economically endangers beekeeping, but also poses a public health risk for consumers since it can be contaminated with various substances and drugs illegal in the food industry.

Therefore, in order to assure the health safety of honey and other bee products, it is of increasing importance to perform chemical analyses as well as radiological characterization. In addition, using radionuclides such as radiocaesium as radiotracers can be a useful tool in assuring the provenance of honey as well as other food products, easing their labelling with quality labels.

In addition to naturally occurring radionuclides, atmospheric nuclear weapons tests, resulting in global fallout and accidental or routine releases of radioactive material from various nuclear facilities are the main causes of man-made radioactive contamination that enters the environment. The main contributions of artificial radionuclides to the Croatian environment were global fallout from previous atmospheric nuclear weapons tests and fallout from nuclear accidents in the Chernobyl and Fukushima-Daiichi nuclear power plants.

Among man-made radionuclides, those of radiocaesium, especially ^{137}Cs and ^{134}Cs, as the most abundant volatile isotopes of caesium, are regarded as a particular hazard to organisms. This is the consequence of their relatively long physical half-lives (30.00 and 2.06 years, respectively) and the chemical and metabolic properties of these radionuclides, which closely resemble those of potassium.

In Croatia, ^{137}Cs and ^{134}Cs in honey were first investigated after the Chernobyl accident in 1986 as part of an extended and still ongoing monitoring programme of radioactive contamination in the environment performed by the Radiation Protection Unit of the Institute for Medical Research and Occupational Health (IMI) ever since 1959.

The aim of this paper was to present long-term, post-Chernobyl, investigations of radiocaesium activity concentrations in multifloral and sweet chestnut (*Castanea sativa Mill.*) honey from north-west Croatia and broaden the knowledge regarding radioecological characterizations of honey. Such "radioecological fingerprinting" can be used to trace the geographical origin of honey as well as to help combat honey adulteration. More precisely, it could be expected that in certain geographical areas, activity concentrations of radionuclides (natural and man-made) and other radioecological parameters lead to a distinctive radiological imprint providing valuable additional tools evidencing floral, vegetable, regional, territorial or topographical origin or specific quality criteria.

It should be noted that in the EU there is high demand, but limited availability, of sweet chestnut honey due to the relatively low presence of sweet chestnut trees in European forests, limited distribution area of such mixed forests and decrease or even disappearance of sweet chestnut trees from some mixed stands (Conedera et al, 2016). In Croatia, mixed sweet chestnut forests cover 135,000 ha, while forests that have a high presence of sweet chestnuts (70-90%) cover about 15,000 ha and are mostly situated in the Sisačko-moslavačka County.

These forests therefore provide an opportunity for intensive beekeeping and production of sweet chestnut honey of the highest quality. Sweet chestnut honey is very rich in pollen and can be declared to be unifloral only if it contains >85% of sweet chestnut pollen (Official Gazette, 2009).

Materials and methods

The total of 163 samples of multifloral and 12 samples of sweet chestnut honey were obtained in late spring and summer, i.e., at the end of beekeeping season, preferably from individual beekeepers. Data for years in which honey samples were not available for analysis were taken from literature (Barišić et al, 1994; Barišić et al, 2002).

Samples of multifloral honey were collected in the area of north-west Croatia, while samples of sweet chestnut honey were collected in Banovina region in Sisačko-moslavačka county.

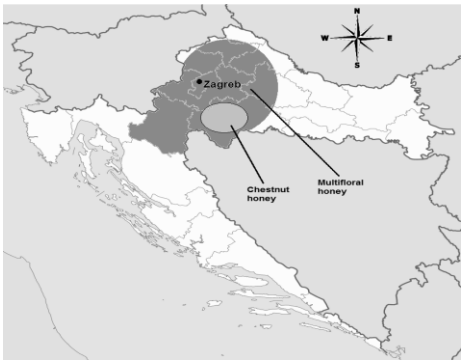

Fig 1. Approximate sample location areas of multifloral and sweet chestnut honey and fallout

As certified reference materials (CRM) for honey are not available for radioecological analyses, samples of certified organic honey obtained in Banovina region (*Banski med* from the village of Klinac) were used instead.

Fallout samples were collected in the city of Zagreb. Dry and wet fallout (rain water) is collected daily using funnels of 1 m^2 collection area. In rainy days the amount of precipitation that contained the fallout was measured by the Hellman pluviometer. In the days without precipitation, funnels were rinsed by 1 L of distilled water. Daily samples are merged into cumulative samples which were analysed quarterly. Prior to gamma-spectrometric measurements, samples were evaporated to 1 L volume.

^{137}Cs and ^{134}Cs levels in the samples from their gamma ray spectra have been analysed by gammaspectrometrical method accredited by Croatian Accreditation Agency in 2010.

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 for gamma spectrometry has been use a low-level high-purity ORTEC HPGe detector (relative efficacy of 74.2% with FWHM resolution of 2.24 keV at 1.33 MeV).

The counting time for radiocaesium measurements depended on the sample activity, was typically 80,000 s. Fallout samples were measured in Marinelli beakers of 1 L volume, while honey samples were placed in plastic cylindrical containers of 0.2 L volume.

Quality assurance and intercalibration measurements were performed through intercalibration programs organized by 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 (Petrinec et al, 2011).

Results and discussion

After the nuclear accident at the Chernobyl nuclear power plant in Ukraine on 26 April 1986, fallout from highly radioactive atmospheric plumes originating from the damaged nuclear reactor was spread and transported all over Europe causing contamination of the environment. In 1986, the total surface deposition of radiocaesium measured in the fallout collected in the city of Zagreb was 6,410 Bqm^{-2} and 2,812 Bqm^{-2} for ^{137}Cs and ^{134}Cs respectively (Bauman et al., 1987).

Consequently, soon after the Chernobyl accident in environmental samples and foodstuffs from the Republic of Croatia elevated levels of ^{137}Cs and, for the first time, of ^{134}Cs were detected. ^{134}Cs is not produced in significant amounts in nuclear explosions of fission weapons which occur in milliseconds. However, it is found in reactor inventories due to long irradiation times of reactor fuels where it is produced via neutron capture from nonradioactive ^{133}Cs, which is a common fission product. Therefore, the presence of this radionuclide in the environment clearly indicated that a nuclear accident occurred.

The highest ^{137}Cs and ^{134}Cs activity concentrations in honey, like in most of the other environmental samples and foodstuffs were recorded in May 1986 (Franic et al, 1991) decreasing exponentially afterwards. In the mid-1990s, activity concentrations in honey for both radionuclides were under the detection limit. However, ^{137}Cs activity concentrations were for the period 1996-2010 above the decision threshold quantifying the physical effect, which allows the conclusion that ^{137}Cs was present in samples, contrary to the ^{134}Cs activity concentrations that were below decision threshold of measurement. Both ^{137}Cs and ^{134}Cs activity concentrations after the Fukushima Daiichi accident were again detectable, but never exceeded 1.5 $Bqkg^{-1}$ for both radionuclides. For comparison, in Poland, ^{137}Cs activity concentrations in multifloral honey after the Fukushima Daiichi accident ranged from 0.24 to 10.57 $Bqkg^{-1}$ (Borawska et. al., 2013).

The activity concentrations of ^{137}Cs and ^{134}Cs in Croatian honey are shown in Table 1.

Table 1 about here

For comparison, ^{137}Cs activity concentrations in samples of honey collected in May-June 1986 in Italy ranged from 106.8 ± 115 $Bqkg^{-1}$ in the Friuli region to 18.3 ± 11 $Bqkg^{-1}$ in Lazio (Tonelli et al., 1990). Regarding honey samples harvested in Slovenia, the values of ^{137}Cs activity concentrations in Slovenian honeys in the period 1987-1995 varied between 8 and 51 $Bqkg^{-1}$ (Bogdanov, 2006).

Activity concentrations of naturally occurring ^{40}K in sweet chestnut honey ranged from 267.0 $Bqkg^{-1}$ in 2012 to 19.7 $Bqkg^{-1}$ in 2017. The reason for this variability is not clear and calls for further research. However, the lack of ^{40}K in honey would indicate an absence of pollen, since potassium, and therefore ^{40}K is the principal constituent of pollen as well as other plant tissues. The absence of pollen therefore raises suspicion in adulteration.

As the main mechanism of environmental contamination by radiocaesium is fallout, measured ^{137}Cs and ^{134}Cs activity concentrations in honey are correlated with fallout activity. When data for ^{137}Cs and ^{134}Cs fallout and honey activity concentrations from Table 1 are related to fallout by a simple linear equation, the coefficients of correlation are r = 0.92 and r = 0.99 for ^{137}Cs and ^{134}Cs respectively. Thus, from fallout data ^{137}Cs activity concentrations in honey can be modelled as:

$$A_h(t) = 0.005 \times A_{fall}(t) + 2.041 \qquad /1/$$

where:
$A_h(t)$ is the time-dependent activity concentration of ^{137}Cs in honey (Bqkg^{-1}) and
$A_{fall}(t)$ is the time-dependent activity concentration of ^{137}Cs in fallout (Bqm^{-2}).

Equation /1/ opens up the possibility to use activity concentrations of ^{137}Cs to authenticate the region of origin and production year of honey, similarly to authenticating wine vintages (Hubert et al. 2009). In addition, we can argue that good correlations lead to greater significance of radioecological sensitivity of honey as a useful tool that helps to compare sensitivities of various environmental samples to radioactive contamination.

Another useful radioecological parameter that can also be readily assessed from long term data on radiocaesium activity concentrations in honey is the ecological half-life. To study the ecological half-life of ^{137}Cs and ^{134}Cs in honey, the mean values of ^{137}Cs and ^{134}Cs from Table 1 were fitted to the exponential function:

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

where:
$A_h(t)$ is the time-dependent activity concentration of radiocaesium in honey (Bqkg^{-1}),
$A_h(0)$ the initial activity concentration of radiocaesium in honey (Bqkg^{-1}) and
$ln(2)/k=T_{1/2,eff}$ the effective (observed) ecological half-life of radiocaesium in honey (years).

For the 1986-1995 period, the observed effective ecological half-life for ^{137}Cs in honey was about 1.11 years. As ^{134}Cs activity concentrations in honey after 1991 were under the detection limit, and therefore not reported, its activity concentrations were analysed for the 1986-1991 period. The observed effective ecological half-life for ^{134}Cs in honey was found to be 0.67 years. The difference between respective ecological half-lives of ^{137}Cs and ^{134}Cs in honey is, in addition to variety of environmental parameters that naturally fluctuate, mainly influenced by different radioactive decay rates of ^{137}Cs and ^{134}Cs. To find the real ecological half-lives, T_R, the observed constant k from the equation /2/ should be corrected for the radioactive decay. Therefore, it can be written as:

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

where $ln(2)/\lambda$ is the physical half-life and $ln(2)/k_R = T_{1/2,e}$ is the ecological half-life and the physical half-lives for ^{137}Cs and ^{134}Cs are 30.0 and 2.06 years, respectively (IAEA, 2014).

From equation the /3/, the real ecological half-lives for ^{137}Cs and ^{134}Cs were found to be 1.16 and 1.01 years, respectively.

In 1986, the observed ^{134}Cs:^{137}Cs activity ratio in honey was 0.60, which reflected the theoretical ratio of 0.55 calculated by dividing the amount of radiocaesium released to air after the reactor explosion at Chernobyl i.e., ~85 PBq Bq of ^{137}Cs and ~47 PBq of ^{134}Cs as

reported by the IAEA (2006). Afterwards, this ratio decreased according to differential radioactive decay. The observed ^{134}Cs:^{137}Cs activity ratio in honey was similar to the ratio found in other environmental samples (Franić et al., 2008; Franic et al., 2009).

As the half-life of ^{137}Cs is about 15 times longer than that of ^{134}Cs, the ^{134}Cs:^{137}Cs activity ratio is decreasing due to differential radioactive decay according to the relationship:

$$R(t) = R(0) \times e^{ln(2) \times t \times (1/T_1 - 1/T_2)} \qquad /4/$$

where:

R(0) is the initial ^{134}Cs:^{137}Cs activity ratio in May 1986, i.e., 0.55,
t is the time elapsed after the Chernobyl accident and
T_1 and T_2 are the physical half-lives for ^{137}Cs and ^{134}Cs, respectively.

After the Fukushima Dai-ichi nuclear accident in March 2011, the initial ^{134}Cs:^{137}Cs ratio in honey was 1.08, which was consistent with the ^{134}Cs:^{137}Cs activity ratio reported to be about 0.9 – 1.0 at the site of accident (Masson et al., 2011). Therefore, it can be concluded that ^{137}Cs and ^{134}Cs have the same environmental fate as no discrimination between those two radionuclides has been observed during the transport from the place of their origin (Chernobyl or Fukushima) to the sampling site. However, it should be noted that in 2011, after the Fukushima Dai-ichi accident, ^{134}Cs:^{137}Cs activity concentration value of 0.9-1.0 could be observed only in samples in which pre-Fukushima, i.e., already existing ^{137}Cs from previous depositions was not present in larger quantities.

Radiocaesium fallout data along with activity concentrations in honey allow an estimation of radioecological sensitivity, (Rs) 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. Rs 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} (UNSCEAR, 1982). Mathematically, P_{23} is defined as follows:

$$P_{23} = \frac{\int_0^\infty A(t)dt}{\int_0^\infty \dot{U}(t)dt} \qquad /5/$$

where:

A(t) is the activity concentration of given radionuclide ($Bqkg^{-1}$) in food 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 honey for the 1986-1995 period can be easily calculated to be 8.6×10^{-3} $Bqykg^{-1}/(Bqm^{-2})$, while the P_{23} for ^{134}Cs from the 1986-1991 period is 1.1×10^{-2} $Bqykg^{-1}/(Bqm^{-2})$. That means that with each Becquerel of ^{137}Cs and ^{134}Cs deposited by fallout on an area of one square meter of land, the activity of one ton of honey increases approximately by 8.6 and 11.1 Bq of ^{137}Cs and ^{134}Cs respectively.

However, as in the year 1986, direct radiocaesium, i.e. both ^{137}Cs and ^{134}Cs, deposition was very high, it affects the overall results. Therefore, when 1986 is excluded from analysis, the P_{23} for ^{137}Cs and ^{134}Cs in honey for 1987 – 1995 and 1987-1991 respective periods were calculated to be 2.0×10^{-2} $Bqykg^{-1}/(Bqm^{-2})$ for ^{137}Cs and 1.9×10^{-2} $Bqykg^{-1}/(Bqm^{-2})$ for ^{134}Cs. It can therefore be clearly seen that, for honey, the radioecological sensitivities of those two radionuclides were almost equal.

To put the obtained values into perspective, the ^{137}Cs transfer coefficient P_{23} for total diet was estimated to be approximately 1.2×10^{-2} Bqykg^{-1}/(Bqm^{-2}) for the 1962 - 1979 period in Denmark (UNSCEAR, 1982) and 2.1×10^{-2} Bqykg^{-1}/(Bqm^{-2}) for the 1987 – 2005 period in beef in Croatia (Franic et al., 2008).

To estimate radiocaesium health risk related to honey consumption, a reliable knowledge of ingestion dose is of particular importance. This is especially true when the ingestion dose is a significant part of the total dose received by the population after nuclear fallout, as was demonstrated to be the case in Croatia (Lokobauer et al., 1998). The dose received by members of the general public by consumption of honey (or any other food) contaminated by a mixture of radionuclides can be expressed as:

$$E = C\sum_{m} D_m^{cf} A_m \qquad /6/$$

where:

E is the annual effective dose in Sv,

C is the total annual *per caput* consumption of food in kgy^{-1}

D_m^{cf} is the dose conversion factor for radionuclide *m*, i.e. effective dose per unit intake, for a member of adult population which converts the ingested activity to effective dose; 1.3×10^{-8} SvBq^{-1} and 1.9×10^{-8} SvBq^{-1} for ^{137}Cs and ^{134}Cs respectively (IAEA, 2014) and

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

The annual effective doses received by an adult member of the Croatian population due to radiocaesium intake, assuming an annual consumption of about 0.9 kgy^{-1} of honey per person, which is consistent with the honey consumption in EU (European Commission 2017), are estimated to be quite small, both after the Chernobyl and the Fukushima-Daiichi nuclear accidents. The effective dose after the Chernobyl accident was estimated at ~0.8 μSv in 1986, decreasing to 1.2 nSv in 1995. After the Fukushima-Daiichi nuclear accident, the per capita effective dose in 2011 was only 12 nSv (4.7 due to ^{137}Cs and 7.5 nSv due to ^{134}Cs). The total annual collective effective dose due to ^{137}Cs and ^{134}Cs intake for the Croatian population (4×10^{6} inhabitants) was estimated at 3.3 Sv in 1986 and ~6.0 Sv for the overall observed period.

Although the consumption of honey in Croatia is not a critical pathway for human intake of radiocaesium from the environment, radioecological characterizations of honey and other bee products add another level of confidence to present radioecological monitoring programmes due to the mobility of honey bees and their ability to integrate all exposure pathways. In addition, "radioecological fingerprinting" of honey on the European level could be a useful tool in assuring its traceability to a particular geographical origin thus combating honey adulteration and related health and other risks. As potassium and therefore ^{40}K as well, is a principal constituent of pollen, whose presence indicates that honey is non-adulterated, measurements of ^{40}K activity concentrations in honey could be used as triage method to check for honey adulteration. However, further research is needed in order to assure adequate statistics of ^{40}K activity concentrations in different honey types (floral varieties).

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Table 1.

Activity concentrations of ^{137}Cs and ^{134}Cs in honey and fallout

Data for 1986 and 1987 are from Franić et al., 1991, data for 1988 and 1989 from Bauman et al., 1987-1990, data for 1990-1991 from Barišić et al., 1994 and Barišić et al., 2002, data for 1992-1995 form Barišić et al., 2002 and data for 2011-2017 are unpublished data measured in Radiation Protection Unit of the Institute for Medical Research and Occupational Health.

Year	Activity concentration in multifloral honey Bqkg^{-1}		^{134}Cs:^{137}Cs Activity ratio		No. of samples	Activity concentration in sweet chestnut honey Bqkg^{-1}		No. of samples	Activity concentration in fallout Bqm^{-2}	
	^{137}Cs	^{134}Cs	Observed	Theoretical		^{137}Cs	^{134}Cs		^{137}Cs	^{134}Cs
1986	32.20 ± 20.57	26.40 ± 3.61	0.60	0.55	5				6410.0	2812.0
1987	18.10 ± 4.24	6.00 ± 2.44	0.33	0.40	7				1098.9	381.9
1988	9.03 ± 3.02	2.80 ± 1.67	0.31	0.30	6				716.0	163.0
1989	6.05 ± 2.45	1.65 ± 1.28	0.28	0.22	9				54.3	13.1
1990	4.00 ± 2.40	0.50 ± 0.30	0.13	0.16	12				17.7	4.0
1991	1.90 ± 1.10	0.10 ± 0.10	0.05	0.12	16				57.1	6.6
1992	0.70 ± 0.30				11				31.0	
1993	0.50 ± 0.20				17				18.5	
1994	0.30 ± 0.20				20				10.4	
1995	0.10 ± 0.10				10				8.4	
2011	0.36 ± 0.08	0.39 ± 0.03	1.07	1.00	2				2.0	0.2
2012	0.44 ± 0.06	0.30 ± 0.08	0.69	0.73	3				1.2	<0.1
2013	0.67 ± 0.23	0.43 ± 0.27	0.64	0.53	6				0.7	
2014	0.91 ± 0.51	0.46 ± 0.25	0.47	0.39	14	3.06 ± 0.20	<0.52±0.39	3	1.3	
2015	0.96 ± 0.17	0.48 ± 0.01	0.45	0.29	18	0.83 ± 0.08	---	3	0.4	
2016	0.36 ± 0.03			0.21	3	0.36 ± 0.01	---	3	0.8	
2017	0.82 ± 0.25				4	0.64 ± 0.01	---	3	1.1	