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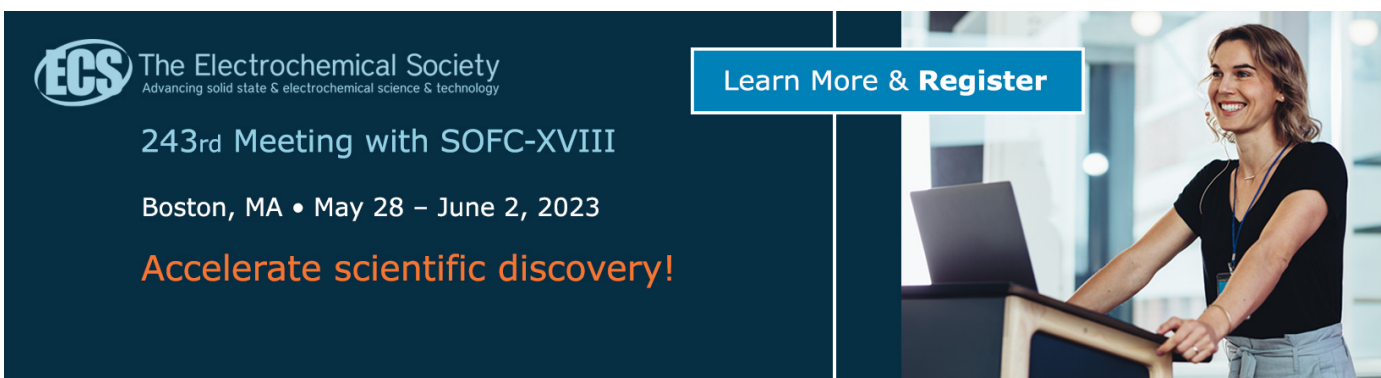
## The radioactivity distribution and radiation hazard in honey samples from Italian large retailers

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# The radioactivity distribution and radiation hazard in honey samples from Italian large retailers

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**Abstract.** The natural (<sup>40</sup>K) and anthropogenic (<sup>137</sup>Cs) radioactivity concentration in twenty-five honey samples of five different typologies (*acacia*, *chestnut*, *wildflower*, *linden* and *honeydew*), coming from Italian large retailers, was investigated by High Purity Germanium (HPGe) gamma spectrometry.

The study was carried out in order to estimate the background levels of detected radionuclides in various honey samples, as well as to assess the dose levels due to their ingestion, taking into account the average yearly direct consumption in Italy and assuming this requirement satisfied from a single kind of honey.

Experimental results were compared with the total natural radioactivity value (external + internal) for humans, in order to evaluate any possible radiological health risk.

## 1. Introduction

Honey is one of the most complex foods produced naturally [1]. It is made from the nectar collected by bee workers from various flowers and spread in the surrounding areas where the bees are raised [2].

From a general point of view, its chemical composition and predominant simple sugars content [3], even considering the expected variability related to its botanical and geographical origin [4], make honey a precious energy source, with valuable nutritional and healing properties. As a matter of fact, it contains a mixture of different carbohydrates, high sugars, proteins, aminoacids, vitamins and minerals [5], and is also used as an ingredient in foodstuffs because of its flavor, color and sweetness [6]. Honey has, in addition, healing and antibacterial properties [7,8].

As a foodstuff used also for beneficial purposes, it must be free of any of unpleasant contents [9,10]. In particular, although, as well established, radionuclides can pose a health risk for humans, their presence in honey has not been the subject of a systematic investigation to date.

As apiculture is popular in Italy, i.e. every year the country produces approximately 20000 tons of honey [11], authors decided to investigate the levels of radionuclides in honey samples.

With this aim in mind, twenty-five honey samples, of five different typologies (*acacia*, *chestnut*, *wildflower*, *linden* and *honeydew*), coming from Italian large retailers, were analyzed in the present



study, as far as natural ( $^{40}\text{K}$ ) and anthropogenic ( $^{137}\text{Cs}$ ) radioactivity concentration is concerned. In particular, as far as natural specific activity is concerned, our attention was focused only on  $^{40}\text{K}$  since natural radioactivity in food mainly comes from this radioisotope, uranium and thorium daughter products being usually present in traces [12].

The assessment of the dose levels due to honey consumption, together with a comparison with the total natural radioactivity value (external + internal) for humans ( $2.4 \text{ mSv y}^{-1}$ ), were also performed in order to evaluate any possible radiological health risks for the population.

The main advantages of the presented research methodology are due to the possibility of evaluating:

- any possible anthropogenic radioactivity contamination of the investigated samples;
- the content of the most significant natural radioisotope in food ( $^{40}\text{K}$ );
- if the honey consumption is safe from the radiological point of view and if it does not adversely affect the human health.

## 2. Materials and Methods

### 2.1 Samples description

Investigated samples were divided into 5 groups (G#, # = 1, 2, 3, 4, 5), based on their typology, according to what reported in Table 1.

Group ID	Typology	Number of samples
G1	<i>Acacia</i>	5
G2	<i>Chestnut</i>	5
G3	<i>Wildflower</i>	5
G4	<i>Linden</i>	5
G5	<i>Honeydew</i>	5

**Table 1.** Investigated groups, together with their identification code (ID), typology, and number of samples for each group.

*Acacia* honey is obtained from the *robinia pseudoacacia*, a plant belonging to the *mimosaceae* family. It is from its flowers, when pollinated by bees, that this type of honey is obtained, with a vaguely floral scent and sweet taste. *Acacia* is then a monofloral honey, very light yellow in color, transparent and with a liquid consistency. It has a very low crystallization rate and can remain in the liquid state for a very long time, thanks to its high concentration of fructose compared to glucose. By virtue of its organoleptic and gustatory characteristics, as well as its excellent solubility, *acacia* honey is excellent as a sweetener for drinks (tea, herbal teas, milk, yogurt, coffee, vegetable milk, etc.). It does not excessively alter the taste and aroma of the products; on the contrary, it generally exudes its main characteristics [13].

*Chestnut* honey is a monofloral one, produced by bees that live near chestnut trees and use their flowers practically exclusively. That of the chestnut tree (*Castanea*) is a botanical genus that belongs to the *Fagaceae* family.

It is dark amber or brown in color, vaguely translucent and with a fluid but very viscous consistency. Second only to *acacia* honey, *chestnut* one has a low crystallization rate and can remain liquid for a long time. Its taste is sweet with a strong bitter aftertaste; aroma and flavor are decisive, with hints typical of the aromas left by the botanical species of origin. In Italy, *chestnut* honey is among the most used to accompany cheeses with an intense flavor [14].

*Wildflower* honey is produced by the natural mixing of the nectar collected by bees inside the hive. There is, therefore, no preminent floral type, but is possible to find flavors and characteristics of various types of flowers.

Depending on the color it can be divided into two types:

- *wildflower* honey with a light color, almost transparent: its flavor, very delicate and sweet, makes it pleasant for everyone and suitable for multiple uses and recipes;

- *wildflower* honey with a slightly darker and amber color: it has a stronger taste and can vaguely recall honeydew. It is typical of mountain areas, where there are numerous chestnut trees and berries.

At nutritional level, *wildflower* honey is more complete than monofloral ones, presenting all the main characteristics of honey: it is very good for children, pregnant women and the elderly being an important energy food for those who are growing or for those who need to reinvigorate their strength; it also promotes bone development and is capable of improving cognitive functions [15].

*Linden* honey is obtained from the flowers of the plant of the same name, that belongs to the *Tiliaceae* family and can be found both in its “wild” version (*Tilia Cordata*) and as an ornamental tree (*Tilia Platyphyllos* and *Tilia americana*). *Linden* honey is presented in a rather light, amber color, with reflections tending to yellow-green in its purest varieties. In addition to the normal anti-inflammatory, anti-bacterial and anti-oxidant action common to other types of honey, *linden* honey has a real anti-spasmodic effect, acting as a calming of the nervous system, and is often used to combat anxiety and the resulting disorders [16].

*Honeydew* honey originates from a sugary substance secreted by some small insects on tree sap, such as spruce, pine, oak, lime and maple. This type of honey is much less sweet than the others and has a bitter aftertaste. Its color ranges from dark brown to almost black; the dark color is due to the richness of antioxidants, especially polyphenols. This type of honey has the greatest number of mineral salts and trace elements. It contains many vitamins and numerous essential amino acids, it has a viscous consistency, but it never crystallizes completely. Due to its strong antibacterial properties, it is particularly recommended for relieving diseases involving the respiratory system. It is an excellent source of energy for athletes and can be used to supplement a lack of minerals and trace minerals [17].

## 2.2 Radioactivity measurements

For High Purity Germanium (HPGe) gamma spectrometry evaluations, investigated samples were directly inserted in Marinelli containers of 1 L capacity, without any preparation [18], and counted for 70000 s.

The 661.66 keV and 1460.8 keV  $\gamma$ -ray lines of  $^{137}\text{Cs}$  and  $^{40}\text{K}$ , respectively, were used to quantify their specific activity.

Two Ortec HPGe detectors and integrated digital electronics were employed for the analysis. The first is a negative biased detector (GMX), with full width at half maximum (FWHM) of 1.94 keV, peak to Compton ratio of 65:1 and relative efficiency of 37.5 % at 1.33 MeV ( $^{60}\text{Co}$ ). GMX is cooled by the Ortec recycler condensing liquid nitrogen cooling Mobius system [19]. The second one is a positive biased detector (GEM), with FWHM of 1.85 keV, peak to Compton ratio of 64:1 and relative efficiency of 40 % at 1.33 MeV ( $^{60}\text{Co}$ ). GEM is cooled by liquid nitrogen [20].

The Gamma Vision (Ortec) software was used for data acquisition and analysis [21,22]. The specific activity (Bq kg $^{-1}$ ) of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  was calculated as follows [23]:

$$C = \frac{N_E}{\varepsilon_E t \gamma_d M} \quad (1)$$

where  $N_E$ ,  $\varepsilon_E$  and  $\gamma_d$  account for the net area, efficiency and yield of a photopeak at energy  $E$ , respectively,  $M$  is the mass sample (kg) and  $t$  is the live time (s).

The data uncertainty (coverage factor  $k=2$ ) takes into account the contribution of the counting, calibration source, efficiency calibration, background subtraction,  $\gamma$ -branching ratio and internal absorption estimation [24].

The reliability of the experimental results was certified by the Italian Accreditation Body (ACCREDIA) on the basis of the quality controls performed according to the UNI 11665:2017 [25].

## 2.3 Radiological health risk assessment

The assessment of the radiological health risk was performed as the effective dose for honey samples ingestion [26]:

$$(2)$$

$$D_{ing} (Sv y^{-1}) = h_{ing,K-40} \times J_{ing,K-40}$$

where  $h_{ing, K-40}$  indicates the  $^{40}K$  conversion coefficient of effective dose, estimated [27] equal to  $4.2 \times 10^{-8} Sv Bq^{-1}$  for the age category between 1 and 5 years old,  $2.1 \times 10^{-8} Sv Bq^{-1}$  between 5 and 10 years old,  $1.3 \times 10^{-8} Sv Bq^{-1}$  between 10 and 15 years old,  $7.6 \times 10^{-9} Sv Bq^{-1}$  between 15 and 17 years old, and  $6.2 \times 10^{-9} Sv Bq^{-1}$  for adults, respectively, and  $J_{ing, K-40}$  accounts for the intake of  $^{40}K$  ( $Bq year^{-1}$ ) and it is obtained by multiplying the yearly honey consumption (kg) for the experimentally measured activity concentration of the investigated radionuclide ( $Bq kg^{-1}$ ) [28]. The assessment of the dose levels due to honey consumption is a critical point in order to evaluate if it is safe from the radiological point of view and does not adversely affect human health.

### 3. Results and Discussion

#### 3.1 Radionuclides concentration

The specific activities of  $^{40}K$  and  $^{137}Cs$  for all the analyzed honey samples are reported in Table 2. For each group, the mean values are also reported, together with the standard deviation.

Group ID	Specific activity	
	$^{40}K$ ( $Bq kg^{-1}$ )	$^{137}Cs$ ( $Bq kg^{-1}$ )
G1	$5 \pm 1$	$< 0.15$
	$15 \pm 2$	$< 0.17$
	$7 \pm 3$	$< 0.19$
	$18 \pm 3$	$< 0.18$
	$34 \pm 5$	$< 0.16$
Mean value $\pm$ standard deviation	$16 \pm 3$	$< 0.17$
G2	$66 \pm 10$	$< 0.19$
	$108 \pm 12$	$< 0.21$
	$71 \pm 10$	$< 0.17$
	$84 \pm 10$	$< 0.18$
	$74 \pm 10$	$< 0.17$
Mean value $\pm$ standard deviation	$81 \pm 10$	$< 0.18$
G3	$40 \pm 8$	$< 0.13$
	$56 \pm 10$	$< 0.12$
	$30 \pm 6$	$< 0.12$
	$47 \pm 7$	$< 0.14$
	$51 \pm 9$	$< 0.16$
Mean value $\pm$ standard deviation	$56 \pm 8$	$< 0.13$
G4	$43 \pm 7$	$< 0.19$
	$65 \pm 9$	$< 0.18$
	$15 \pm 5$	$< 0.13$
	$43 \pm 7$	$< 0.14$
	$39 \pm 7$	$< 0.17$
Mean value $\pm$ standard deviation	$41 \pm 7$	$< 0.16$
G5	$114 \pm 19$	$< 0.22$
	$109 \pm 17$	$< 0.19$
	$116 \pm 20$	$< 0.23$
	$106 \pm 16$	$< 0.21$
	$111 \pm 14$	$< 0.17$

Mean value $\pm$ standard deviation	$111 \pm 17$	$< 0.20$
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**Table 2.** Specific activities (Bq kg<sup>-1</sup>) of <sup>40</sup>K and <sup>137</sup>Cs in the analyzed honey samples.

The variation of the <sup>40</sup>K content from honey to honey is mainly attributed to the differences in botanical structure, as well as in the mineral composition of the soil in which the plants are cultivated. Other factors responsible for the observed variation are preferential absorbability of the plant and climatic conditions [29].

The <sup>137</sup>Cs activity concentration was in particular quantified in order to investigate about any possible artificial contamination. In all cases, it was found to be lower than the minimum detectable activity, giving evidence of the absence of any residual contamination from anthropogenic radioactivity.

All these experimental results are in good agreement, being of the same order of magnitude and also very similar, with those reported in the database of the “Italian Institute for the Environmental Protection and Research” (ISPRA) [30].

It should be noted that the specific activity of <sup>40</sup>K, measured for each sample, indicates the amount of radioactivity, and not the radiological hazard to individuals. In order to evaluate it, additional factors have to be taken into account, as reported in the following.

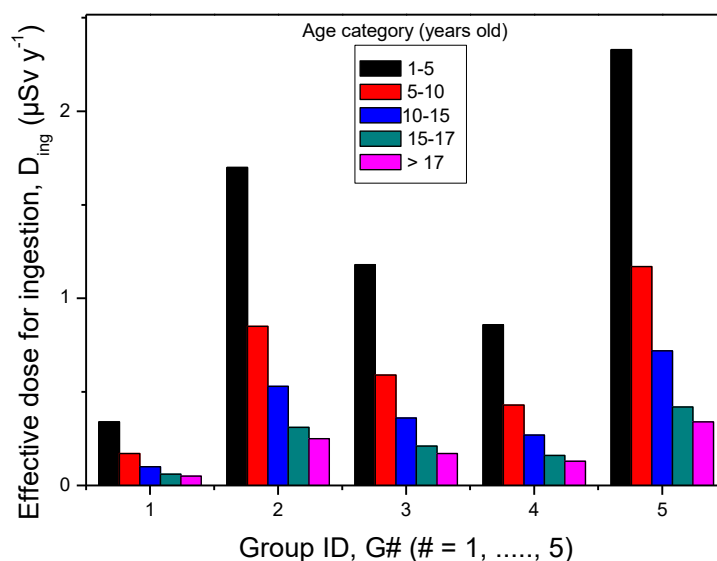
### 3.2 Potential health hazards resulting from honey consumption

In addition to the environmental concern regarding honey element composition, the analysis of the quality of honey is of great relevance, given the increasing global trends in total honey production and the fact that the European Union is the world’s largest consumer of this foodstuff.

With regard to national and supranational regulations related to honey, the Directive 2001/110/EC refers to some properties related to its composition, without however making any mention of the radionuclide content [31].

The regulation EC 733/2008, which sets the maximum levels for certain contaminants in foodstuffs, does not address apiary products [32].

Figure 1 reports the annual effective dose due to the ingestion of the investigated honey samples, as calculated by the Equation (2). Obtained results take into account the average yearly honey consumption per person in Italy (500 g) [33] under the a priori hypothesis that this need can be satisfied by a single variety of honey [34].



**Figure 1.** The calculated annual effective dose for honey ingestion.

The consumption of this foodstuff for infants (lower than 1 year old) is not recommended by the World Health Organization [35], for this reason this age group was not considered in the analysis.

The annual effective dose due to the honey ingestion was found to be:

- 0.34  $\mu\text{Sv y}^{-1}$ , 0.17  $\mu\text{Sv y}^{-1}$ , 0.10  $\mu\text{Sv y}^{-1}$ , 0.06  $\mu\text{Sv y}^{-1}$  and 0.05  $\mu\text{Sv y}^{-1}$  for the age category between 1 and 5 years old, between 5 and 10 years old, between 10 and 15 years old, between 15 and 17 years old and higher than 17 years old (adults), respectively, for the acacia honey consumption;
- 1.70  $\mu\text{Sv y}^{-1}$ , 0.85  $\mu\text{Sv y}^{-1}$ , 0.53  $\mu\text{Sv y}^{-1}$ , 0.31  $\mu\text{Sv y}^{-1}$  and 0.25  $\mu\text{Sv y}^{-1}$  for the age category between 1 and 5 years old, between 5 and 10 years old, between 10 and 15 years old, between 15 and 17 years old and higher than 17 years old (adults), respectively, for the chestnut honey consumption;
- 1.18  $\mu\text{Sv y}^{-1}$ , 0.59  $\mu\text{Sv y}^{-1}$ , 0.36  $\mu\text{Sv y}^{-1}$ , 0.21  $\mu\text{Sv y}^{-1}$  and 0.17  $\mu\text{Sv y}^{-1}$  for the age category between 1 and 5 years old, between 5 and 10 years old, between 10 and 15 years old, between 15 and 17 years old and higher than 17 years old (adults), respectively, for the wildflower honey consumption;
- 0.86  $\mu\text{Sv y}^{-1}$ , 0.43  $\mu\text{Sv y}^{-1}$ , 0.27  $\mu\text{Sv y}^{-1}$ , 0.16  $\mu\text{Sv y}^{-1}$  and 0.13  $\mu\text{Sv y}^{-1}$  for the age category between 1 and 5 years old, between 5 and 10 years old, between 10 and 15 years old, between 15 and 17 years old and higher than 17 years old (adults), respectively, for the linden honey consumption;
- 2.33  $\mu\text{Sv y}^{-1}$ , 1.17  $\mu\text{Sv y}^{-1}$ , 0.72  $\mu\text{Sv y}^{-1}$ , 0.42  $\mu\text{Sv y}^{-1}$  and 0.34  $\mu\text{Sv y}^{-1}$  for the age category between 1 and 5 years old, between 5 and 10 years old, between 10 and 15 years old, between 15 and 17 years old and higher than 17 years old (adults), respectively, for the honeydew honey consumption;

Worth of note, the aforementioned doses are about three order of magnitude lower than the total (external + internal) natural radioactivity value for humans, equal to 2.4  $\text{mSv y}^{-1}$  [36]. Then, the calculated effective doses reasonably appear to be too low to induce significant health hazards [37].

#### 4. Conclusions

In the present article, the activity concentration of natural and anthropogenic radionuclides ( $^{40}\text{K}$  and  $^{137}\text{Cs}$ , respectively) was measured for honey samples of five different typologies, collected from Italian large retailers.

Samples were analyzed using HPGe gamma spectrometry and results indicate that there is a variation of the  $^{40}\text{K}$  content in the investigated samples, mainly due to the differences in botanical structure, as well as in the mineral composition of the soil in which the plants are cultivated. The  $^{137}\text{Cs}$  activity concentration was found to be lower than the minimum detectable activity in all cases, thus excluding a contamination due to anthropic radionuclides.

In order to evaluate the radiological hazard effects for the population, the effective dose for honey ingestion was assessed. The calculated values were compared with the total natural radioactivity value for humans, resulting about three orders of magnitude lower. It is therefore possible to conclude that radionuclide intoxication resulting in human adverse effects is not a concern.

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