Published in final edited form as:
Food Control (110), April 2020, doi:10.1016/j.foodcont.2019.107001

# Assessment of radioactivity in commercially available honey in Italy

Maria Assunta Meli*, Donatella Desideri*§, Paolo Battisti°, Isabella Giardina°, Daniela Gorietti°, Carla Roselli*

**Department of Biomolecular Sciences, Urbino University "Carlo Bo", P.zza Rinascimento 6, 61029 Urbino, Italy*

°Integrated Laboratory of Radioactivity Measurement and Monitoring, Radiation Protection Institute, ENEA-CR, Casaccia, Via Anguillarese 301, 00123 Rome, Italy

§Corresponding author: Donatella Desideri, Biomolecular Sciences Department, University of Urbino Carlo Bo, P.zza Rinascimento 6, 61029 Urbino, Italy, Tel: ++39 0722 303308

E-mail address: donatella.desideri@uniurb.it

**Abstract**

Activity concentration of natural radionuclides and $^{137}Cs$ were estimated in 28 honeys purchased from large supermarket chains in Italy. Uranium, polonium and thorium were measured by alpha spectrometry, $^{40}K$ and $^{137}Cs$ by gamma spectrometry and $^{226}Ra$ by liquid scintillation. The concentrations were 50.6±46.3, 0.023±0.010, 0.019±0.017, 0.27±0.40 $Bqkg^{-1}$ for $^{40}K$, $^{238}U$, $^{234}U$ and $^{210}Po$ respectively. $^{235}U$, $^{228}Th$, $^{232}Th$ were consistently <0.007 $Bqkg^{-1}$ and $^{226}Ra$ <0.200 $Bqkg^{-1}$. $^{137}Cs$ was <2.1 $Bqkg^{-1}$ in 93% of the samples. The activity of radiocesium was found to be above the minimum detectable level in only two samples and did not exceed the highest permitted level for food contributing to the overall radioactivity only slightly. The effective doses attributable to $^{210}Po$ accounts for 0.0026-5.31% of global human exposure to natural radiation. The honeys that were tested were found to be of good quality with regard to the parameters under study, confirming the general image of honey as a genuine healthy product.

**Keywords:** honeys; natural and artificial radionuclides; ingestion dose

## 1.Introduction

For the general population ingestion of food or beverage is the main path in which radionuclides may be incorporated into the body, and thus the main source of internal dose. Any radioactivity present in the air, or more importantly in the ground and soil, can be transferred to the crops that are grown in that soil. Indeed, some naturally occurring radioactive elements find their way into our bodies. The most important radionuclides, accounting for the highest percentage of the radiation dose present in the body due to food intake, are the primordial radionuclides $^{40}$K, thorium and uranium (Meli et al., 2016).

$^{232}$Th and $^{228}$Th are present in the Earth's crust. They enter the human body through the food chain and inhalation of the suspended dust in the air accumulating in the lungs, liver and skeletal tissues and causing radiation damage as well as biochemical and morphological changes.

A high percentage of the internal radiation dose is attributable to $^{210}$Po, a naturally occurring radionuclide of the $^{238}$U series with a half-life of 138.4 days. According to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), $^{210}$Po is estimated to contribute about 8% of the effective dose equivalent to humans due to natural radioactivity (UNSCEAR, 2000). $^{210}$Po distribution in the environment depends to a large extent on the other radionuclides in the decay series, namely $^{226}$Ra and $^{210}$Pb. Part of $^{210}$Po is deposited on plants via the $^{222}$Rn-daughters, mainly $^{210}$Pb, through precipitation, caused by the outgassing of $^{222}$Rn from soils (= unsupported $^{210}$Po). The second source of $^{210}$Po is the decay of $^{226}$Ra in the soil. Its presence in all terrestrial foodstuffs is, therefore, unavoidable (Brown et al., 2011, Persson et Holm, 2011). Other anthropogenic activities that increase the levels of $^{210}$Pb and $^{210}$Po in plants include atmospheric deposition from industrial emissions, waste disposal, phosphate ore processing, coal-fired power plants, coal mining, metal smelting, etc.

Potassium, on the other hand, plays a key role in regulating many bodily functions such

as digestion and heart rate as well as the water content of cells. Potassium content in the body is therefore held constant by metabolic processes (homeostatic control), although gender and age related variations have been observed. Its distribution within the body is more or less uniform. $^{40}K$ is a single occurring natural radionuclide which has a very long half-life of $1.251\times10^{9}$ years. It makes up 0.012% of the total amount of potassium found in nature (Sugiyama et al., 2009) and its level in the body, like that of stable potassium is constant. In an average 70 kg human potassium content is 0.1-0.3% with a corresponding total activity ranging from 22 to 76 kBq of $^{40}K$ (Altekin et al., 2015).

Atmospheric nuclear weapon tests (1945-1963) and a series of nuclear accidents have contaminated the environment with artificial and biologically significant radionuclides such as $^{131}I$, $^{134}Cs$, $^{137}Cs$ and $^{90}Sr$. $^{137}Cs$ (characterized by a half life of 30.17 years) is among the most problematic of the short-to-medium-lifetime fission products because it spreads readily through the environment thanks to the high water solubility of Cs salts. It mainly contaminates leaves and flowers directly from the atmosphere but also through uptake from the soil. Organisms process caesium in a very similar way to potassium (Altekin et al., 2015). $^{90}Sr$ is even more problematic than $^{137}Cs$, but, due to the complexity of the methods requested for its analysis, much more analyses have been performed on $^{137}Cs$.

Honey, a nutritious product of global economic importance, is a naturally sweet substance produced by honeybees from the nectar of blossoms or from honeydew. The nectar or deposits are then modified and stored in honeycombs. Honeybees (Apis mellifera L.) are the main pollinating insects for numerous plants and fruit trees. Honey is one of the most complex food products, and its composition varies according to many factors including the botanical/floral origins of the plants from which the nectar is collected and the species of the bees. Honey can be considered a supersaturated solution composed of four main sugar (75-80%), fructose, glucose, maltose and sucrose (Poschl et al., 2011). It has a water content of less than 20% and contains

minerals and organic substances, including amino acids, enzymes, proteins, vitamins, organic acids, pigments and phenolics, (1%–2%) (Buldini et al., 2001).

Honey is a popular natural product, not only for its taste and nutritional value, but also for its health benefits. Its high contents of glucose and fructose make honey one of the most digestible foods (Belitz et al., 2004) and it is commonly used as an ingredient in foodstuffs because of its sweetness, color, flavor, caramelization, and viscosity. Honey also has preservation properties due to its sugar-content.

The European population is the world's largest consumer of honey (Vanhanen, 2011), and in light of the growing consumption of this natural food, it is of paramount importance to ensure its safety. Honey, in fact, can be contaminated by several toxic compounds, including radionuclides. Contamination can occur as a result of beekeeping practices or environmental factors (Bogdanov, 2006). Regarding environmental contamination, during their foraging flights, when honeybees collect nectar, pollen, plant resins, and water, they inadvertently come into contact with a wide array of inorganic and organic pollutants (heavy metals, radionuclides, pesticides etc.), which are often taken back to the colony. Hence, contaminants migrate through the chain air-water soil-plants-bees chain into honey and change its composition and quality. Of these contaminants, radionuclides are of particular interest because of their impact on the quality of honey as well as their potential adverse effects on human health. Hence, ensuring low concentrations of radionuclides in honey is imperative (Borawska et al., 2013), and commercially available honey must therefore be monitored closely to guarantee its quality, provenance and safety.

Hence, the aim of this study was to perform a radiological characterization of a range of commercially available honeys, purchased from major Italian supermarket chains in order to evaluate the risk posed by natural and artificial radioactivity present in these products. For this

purpose, gamma and alpha spectrometry and liquid scintillation counting were used to determine the concentrations of $^{137}Cs$ and the natural radionuclides, $^{40}K$, $^{238}U$ and $^{232}Th$ and their progeny.

## 2. Experimental setup

### *2.1. Samples*

The honeys used in the present study were purchased from major Italian supermarket chains. Twenty-eight samples were analyzed. These samples were classified into six groups according to their botanic origin: 9 wildflower, 6 acacia, 3 eucalyptus, 3 chesnut, 5 citrus, 1 linden and 1 forest.

Twenty-six of the tested honeys were produced in various European countries (Italy, Hungary, Serbia, Ukraine) and two respectively in Argentina and Uruguay.

Table 1 shows the sampling area, botanical origin and chemical composition of the honeys namely their sugar contents, degree of humidity, pH and free acidity, combined and total acidity. These fndings were obtained from a previous investigation (Meli et al., 2018) and are consistent with those reported by the Council Directive 2001/110/EC relating to honey (EC, 2001).

**Table 1** List of honey samples, sampling area, botanical origin and chemical composition (Meli et al., 2018)

| Sample code, N | Botanical origin | Sampling area | Humidity (%) | Sugar (Brix) | pH | Free Acidity (meq/kg) | Combined acidity (meq/kg) | Total acidity (meq/kg) |
|---|---|---|---|---|---|---|---|---|
| 1 | wildflower | Hungary, Ukraine | 17.7 | 80.2 | 3.9 | 23.6 | 3.3 | 26.8 |
| 2 | wildflower | Hungary, Italy | 18.2 | 79.8 | 4.0 | 27.4 | 1.7 | 29.1 |
| 3 | wildflower | Italy | 16.2 | 81.6 | 4.6 | 60.9 | 3.0 | 63.9 |
| 4 | wildflower | Italy | 19.0 | 79.0 | 4.1 | 33.4 | 4.0 | 39.5 |
| 5 | wildflower | Hungary, Italy | 18.2 | 79.5 | 4.1 | 28.4 | 1.6 | 29.9 |
| 6 | wildflower | Italy | 17.4 | 80.2 | 4.4 | 19.8 | 3.7 | 23.5 |
| 7 | wildflower | Italy | 18.6 | 79.1 | 5.1 | 17.2 | 1.7 | 18.8 |
| 8 | wildflower | Argentina | 18.7 | 79.0 | 4.2 | 32.9 | 2.2 | 35.0 |
| 9 | wildflower | Italy | 17.5 | 80.1 | 4.3 | 31.9 | 1.5 | 33.6 |
| 10 | acacia | Hungary | 18.0 | 80.0 | 4.2 | 10.6 | 2.1 | 12.7 |
| 11 | acacia | Hungary | 18.3 | 79.8 | 4.1 | 12.0 | 1.7 | 13.7 |
| 12 | acacia | Italy | 17.6 | 80.4 | 4.1 | 15.5 | 1.7 | 17.1 |
| 13 | acacia | Hungary | 17.9 | 79.8 | 4.2 | 13.0 | 1.2 | 14.2 |
| 14 | acacia | Italy-Lunigiana | 17.5 | 80.3 | 4.4 | 10.0 | 1.0 | 11.0 |
| 15 | acacia | Serbia | 17.2 | 80.5 | 4.5 | 9.6 | 1.0 | 10.6 |
| 16 | eucalyptus | Uruguay | 20.7 | 77.1 | 4.3 | 35.5 | 3.7 | 39.3 |
| 17 | eucalyptus | Italy | 17.7 | 80.3 | 4.3 | 27.3 | 2.9 | 30.2 |
| 18 | eucalyptus | Italy | 18.6 | 79.0 | 4.3 | 20.5 | 3.2 | 23.7 |
| 19 | chesnut | Italy -Calabria | 17.2 | 80.6 | 4.8 | 32.7 | 5.1 | 37.8 |
| 20 | chesnut | Italy | 17.5 | 80.3 | 5.5 | 14.5 | 2.7 | 17.2 |
| 21 | chesnut | Italy | 17.0 | 81.0 | 4.5 | 45.7 | 3.4 | 49.1 |
| 22 | citrus | Italy | 18.6 | 79.4 | 4.1 | 14.3 | 1.5 | 15.8 |
| 23 | citrus | Italy | 17.9 | 80.1 | 3.9 | 31.1 | 2.5 | 33.6 |
| 24 | citrus | Italy | 19.9 | 77.9 | 3.9 | 29.3 | 1.3 | 30.6 |
| 25 | citrus | Italy | 18.1 | 79.6 | 4.2 | 19.2 | 2.1 | 21.3 |
| 26 | citrus | Italy | 19.2 | 78.8 | 3.9 | 22.8 | 1.3 | 24.1 |
| 27 | linden | Italy | 17.9 | 80.0 | 4.9 | 17.3 | 4.0 | 21.3 |
| 28 | forest honey | Italy | 17.3 | 80.3 | 5.0 | 27.1 | 2.2 | 29.2 |
| Minimum | | | 16.2 | 77.1 | 3.9 | 9.6 | 1.0 | 10.6 |
| maximum | | | 20.7 | 81.6 | 5.5 | 60.9 | 5.1 | 63.9 |
| Mean | | | 18.1 | 79.8 | 4.3 | 24.4 | 2.4 | 26.9 |
| SD | | | 0.91 | 0.91 | 0.40 | 11.6 | 1.1 | 12.2 |

*2.2. Analytical methods*

Three different methods were used to measure the radioactive content of the honey samples: gamma spectrometry, alpha spectrometry and liquid scintillation.

*Gamma spectrometry.* This technique is able to determine many different radionuclides directly and simultaneously without any specific pre-treatment of the sample. However, some of radionuclides of interest in the present study, namely $^{210}$Po, $^{234}$U, $^{230}$Th or $^{232}$Th, are not gamma emitters or only poor ones and therefore it is difficult to measure them using gamma spectrometry.

$^{40}$K and $^{137}$Cs were assessed in honey by measuring all samples with a p-type coaxial HPGe detector by Ortec®, Model GEM-C5060P4 Profile C; it is characterized by an energy range of 3 – 3000 keV, a relative efficiency of 20% at 1.33 MeV and an energy resolution (FWHM) of 1.8 keV at 1.33 MeV, 850 eV at 122 keV and 725 eV at 5.9 keV. The detector was cooled to near liquid nitrogen temperature and it was placed in a shielded house built from low-background, 10 cm thick lead, lined with a sheet of copper (1.5 mm thick). The samples were counted for 150,000 s. Gamma spectra were analysed by Canberra®) Genie-2000 software. To determine the activity concentration of $^{40}$K and $^{137}$Cs the gamma-ray energies of 1460.75 and 661.62 keV were considered respectively. For full quantitative analysis, the system was calibrated in efficiency and energy using multiradionuclides aqueous standard solution certified by the Italian National Institute of Ionizing Radiations Metrology (ENEA INMRI). The performances of the measuring system are periodically checked through the participation to international intercomparison exercices (Procorad and IAEA-Almera). 100 g of each honey sample were dissolved in bidistilled water with the aid of slight heating and the solution was then placed into a 180 ml Marinelli beaker. Taking into account that the density of the analysed matrices after dilution was around 1 (similar to the calibration solution), no correction for self-absorption was applied. The background spectra were counted under the same conditions using a

container with bidistilled water.

*Alpha spectrometry.* This radiometric technique is applied to pure alpha emitting radionuclides and involves a complex and time consuming pre-treatment of the sample to make measurement of the investigated radionuclide possible. The method includes a complete dissolution of the sample with nitric acid followed by a separation of the radionuclide by extraction chromatography or precipitation, and an electrodeposition to prepare the source for the measurement, etc. This technique requires very long preparation and source counting, however, it makes possible to perform a specific determination of activity concentration with extremely low detection limits providing complete information on concentration and isotopic ratios of $^{238}U$, $^{235}U$,$^{234}U$, $^{232}Th$, $^{230}Th$ and $^{228}Th$ (Desideri et al., 2006).

Taking these considerations into account, the following radiochemical method was employed for uranium and thorium measurement: 10 g of honey, after the addition of a known activity of $^{236}U$ and $^{229}Th$ as the yield internal standards, was dissolved in 70 ml of 5M $HNO_3$, 25 ml of 1M $Al(NO_3)_3$ in 5 M $HNO_3$; the solution was stirred (30') and after 12 hours it was filtered. The solution was then passed through a chromatographic column (UTEVA Resin, Eichrom Technologies) constituted by dipentyl,pentylphosphonate (or diamyl, amylphosphonate), DAAP, supported on an inert polymer (acrylic ester) and conditioned with 5M $HNO_3$; DAAP is a selective extractant in nitric medium for tetravalent and hexavalent actinides (Th, U, Pu, Np) (Thakkar, 2000). Thorium was eluted by 5M HCl and then uranium was eluted by 0.02 M HCl. The two elution solutions were evaporated, dried and mineralised. Finally, the residues were dissolved in conc. $H_2SO_4$ and transferred into an electrolytic cell. Uranium and thorium were electroplated from ammonium sulphate solution at pH 4.

For the determination of $^{210}Po$, no preliminary chemical separation was required. The polonium source was, in fact, prepared by the spontaneous deposition of the element on silver plate after complete dissolution of the sample. The following radiochemical method was

177 employed for polonium determination: 10 g of each sample was spiked with a known activity of
178 $^{209}$Po as an internal standard yield and then treated several times with concentrated nitric acid
179 and hydrogen peroxide to aid the oxidization of the organic compounds. When the solution was
180 clear, the liquid was evaporated to dryness and the residue was dissolved in 80 ml of 1M
181 hydrochloric acid. After the addition of 100 mg of ascorbic acid to eliminate the ferric ion
182 interference and 10 ml of 25% sodium citrate, both $^{210}$Po and $^{209}$Po were plated at 85-90°C and
183 pH 1.5-2.0 continuously for 4 h onto a silver disk, placed in a syringe and immersed in the
184 solution (Meli et al., 2014).

185 For quality control, these analytical methods were tested using standard reference
186 materials. .

187 The measurements of the isotopes alpha emitters of polonium, uranium and thorium were
188 carried out using an α-spectrometer equipped with a semiconductor silicon detector of surface
189 barrier type (300 $mm^2$ active surface, resolution 20 keV, 31.7±3.1% of counting efficiency, and 2
190 x $10^{-6}$ $s^{-1}$ of the background in the region of interest (ROI)) (Canberra Industries, Inc., 800
191 Research Parkway, Meriden, CT 06450) and connected to a computerized multichannel analyzer.
192 Counting times ranged from 1000 to 3000 min to achieve good counting statistics.

193 For uranium, thorium and polonium percentage chemical recoveries resulted respectively
194 67.9±18.4, 77.8±16.4 and 88.1±9.2 % respectively, and the minimum detectable activity
195 concentration (MDA) for a 20 g sample was consistently around 7 $mBqkg^{-1}$. A blank sample was
196 prepared because reagents, containers and equipment could release impurities that could lead to
197 miscalculation

198 *Liquid Scintillation (LSC).* This method was employed for the measurement of $^{226}$Ra. It
199 makes use of the volatility of $^{222}$Rn and the secular equilibrium between $^{226}$Ra and $^{222}$Rn. In fact;
200 this radionuclide transfers completely and selectively from the aqueous solution containing $^{226}$Ra
201 to an overlying immiscible organic phase containing the scintillation cocktail. 25 g of each honey

sample was dissolved in water and evaporated at 200°C until a final volume of 10 ml was reached. This solution was then placed in a PTFE vial, and 10 ml of the scintillation cocktail Ultima Gold F was added. The closed vial was kept in dark for four weeks to achieve secular equilibrium between $^{226}$Ra and its daughter $^{222}$Rn. For each $^{226}$Ra decay, three alpha decays (of $^{222}$Rn, $^{218}$Po and $^{214}$Po) and two beta decays (of $^{214}$Pb and $^{214}$Bi) were considered. The samples were counted in the ultra low-level scintillation counter Quantulus 1220 from Perkin Elmer applying a special alpha/beta discrimination tool. By discrimination of the β-decays, the alpha decays of $^{222}$Rn, $^{218}$Po and $^{214}$Po were countable with an efficiency of around 240% and a low background could be achieved. In our measurements, a minimum detectable concentration of around 0.2 Bq/kg (not so good) was achieved due to the small sample amount (25g). Longer counting times (1000 minutes) and a low a-background could not overcome this limitation. In addition, for liquid scintillation as well as for alpha spectrometry, a blank sample was prepared because reagents, containers and equipment could release impurities that could lead to miscalculation (Desideri et al, 2019).

*2.3. Activity concentrations and MDA calculation*

The activity concentration ($Bqkg^{-1}$) of each radionuclide present in the sample was calculated from the formulas reported by IAEA (IAEA, 1989) and ISO (ISO 2018a; ISO 2018b). The total uncertainty of each measurement was calculated taking into account all the major sources of variability, particularly statistical fluctuations of counting both of the sample and of the blank (including background) and those associated to efficiency calibration and sample weighting. The minimum detectable activity (MDA) of the measurements system was done following the procedure shown in ISO 11929.

227 *2.4. Statistical analyses*

228 For each group of samples, the arithmetical mean with its standard deviation (SD) was
229 reported. The median, the minimum and maximum values are also reported. Statistical analyses
230 (Student's *t*-test) were carried out following the logarithmic transformation of the individual data
231 in order to evaluate the presence of significant difference in term of radioactivity content
232 between honeys of different botanical origin (wildflower, acacia, eucalyptus, chesnut, citrus,
233 linden, forest honey); significance was accepted at $P < 0.05$.

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236 *2.4. Radionuclide annual intake and committed effective dose calculation*

237 The annual intake of radionuclides was calculated as the product of the radionuclide
238 activity concentration and the annual consumption rate. The following dose calculations were
239 based on two different daily consumption rates: 5g (a teaspoon) and 25 g of honey. The annual
240 committed effective dose was calculated for each radionuclide using the following formula:

241

242 $$D = Q \ x \ C \ x \ I$$

243 where D is the annual committed effective dose for every radionuclide taken into account
244 ($\mu Sv\ y^{-1}$), C is the radionuclide concentration ($Bq\ kg^{-1}$), I is the annual consumption rate ($kg\ y^{-1}$),
245 and Q is the conversion factor for ingestion ($\mu Sv\ Bq^{-1}$) recommended by ICRP (2012) for infants
246 (1-2 year), children (7-10 years) and adults (>17).

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## 3. Results

### *3.1. Radionuclids concentrations*

Table 2 shows the measurement of alpha and gamma emitters for each sample. Each activity concentration value was affected by uncertainties of 10%, 15%, 25% and 25% for $^{40}K$ $^{210}Po$, $^{238}U$ and $^{234}U$ respectively.

For the $^{238}U$ series, $^{210}Po$ $^{238}U$ and $^{234}U$ were detectable in the all the samples; $^{210}Po$ ranges from 0.03 to 1.62 $Bqkg^{-1}$ (mean value 0.27 ± 0.40 $Bqkg^{-1}$); the mean activity concentration was 0.023 ± 0.010 $Bqkg^{-1}$ and 0.019 ± 0.017 $Bqkg^{-1}$ for $^{238}U$ and $^{234}U$ respectively.$^{235}U$, $^{232}Th$ and $^{228}Th$ activity concentrations were consistently below MDA (0.007 $Bqkg^{-1}$) such as $^{226}Ra$ (0.200 $Bqkg^{-1}$).

Table 2 also shows the $^{40}K$ activity concentration. $^{40}K$ was detectable (above MDA) in the 55% of the samples and this radionuclide was responsible for most of the activity in the honeys honey. This is due to the fact that potassium, an essential element for humans and widely distributed in the environment (Uwatse et al., 2015), can be easily transferred through the food chain. The $^{40}K$ activity concentration ranged from <22 to 180 $Bqkg^{-1}$ (mean value 50.6 ± 46.3 $Bqkg^{-1}$). For the mean calculation, when activity concentration was below the MDA, the MDA was considered.

Concentration differences between samples collected from honeys with different botanical origins were calculated and the results were showed in table 3.

Regarding the $^{210}Po$, a significant difference ($P < 0.05$) was found between the activity concentration of this radionuclide in forest honey and its concentration in all the different kinds of tested honey, excluding chesnut. No significant difference ($P > 0.05$) was found between $^{210}Po$ concentration in wildflower honey and that in all the different kinds of honey excluding forest honey.

**Table 2**

Activity concentration ($Bqkg^{-1}$) of $^{238}U$, $^{234}U$, $^{210}Po$, and $^{40}K$ in honey samples, minimum, maximum value, median, mean and relative standard deviation (SD)

| Code, N | Group | $^{238}U$ | $^{234}U$ | $^{210}Po$ | $^{40}K$ |
|---|---|---|---|---|---|
| 1 | wildflower | 0.042 | 0.031 | 0.07 | 180 |
| 2 | wildflower | 0.049 | 0.089 | 0.11 | <22.0 |
| 3 | wildflower | 0.017 | 0.013 | 0.26 | 105 |
| 4 | wildflower | 0.020 | 0.021 | 0.18 | 49.0 |
| 5 | wildflower | 0.024 | 0.011 | 0.08 | <22.0 |
| 6 | wildflower | 0.020 | 0.013 | 0.11 | <22.0 |
| 7 | wildflower | 0.019 | 0.011 | 0.91 | 65.2 |
| 8 | wildflower | 0.023 | 0.012 | 0.05 | 33.4 |
| 9 | wildflower | 0.028 | 0.017 | 0.33 | 38.7 |
| | Mean± SD | 0.027±0.011 | 0.024±0.025 | 0.23±0.27 | 59.7±52.5 |
| 10 | acacia | 0.024 | 0.013 | 0.03 | <22.0 |
| 11 | acacia | 0.017 | 0.007 | 0.05 | <22.0 |
| 12 | acacia | 0.029 | 0.025 | 0.03 | <22.0 |
| 13 | acacia | 0.015 | 0.013 | 0.05 | <22.0 |
| 14 | acacia | 0.017 | 0.012 | 0.03 | <22.0 |
| 15 | acacia | 0.007 | 0.007 | 0.06 | <22.0 |
| Mean± SD | | 0.018±0.008 | 0.013±0.008 | 0.04±0.01 | <22.0 |
| 16 | eucalyptus | 0.028 | 0.033 | 0.12 | 57.4 |
| 17 | eucalyptus | 0.014 | 0.010 | 0.23 | 40.5 |
| 18 | eucalyptus | 0.017 | 0.007 | 0.09 | 44.0 |
| | Mean± SD | 0.020±0.007 | 0.017±0.014 | 0.15±0.07 | 47.3±8.90 |
| 19 | chesnut | 0.041 | 0.028 | 0.52 | 151 |
| 20 | chesnut | 0.021 | 0.010 | 1.45 | 157 |
| 21 | chesnut | 0.015 | 0.019 | 0.32 | 77.1 |
| | Mean± SD | 0.026±0.014 | 0.019±0.009 | 0.76±0.60 | 128±44.5 |
| 22 | citrus | 0.017 | 0.012 | 0.04 | <22.0 |
| 23 | citrus | 0.029 | 0.015 | 0.30 | 31.8 |
| 24 | citrus | 0.044 | 0.051 | 0.10 | <22.0 |
| 25 | citrus | 0.015 | 0.013 | 0.04 | <22.0 |
| 26 | citrus | 0.025 | 0.028 | 0.08 | <22.0 |
| | Mean± SD | 0.026±0.012 | 0.024±0.017 | 0.11±0.11 | 24.0±4.38 |
| 27 | linden | 0.013 | 0.011 | 0.59 | 87.9 |
| 28 | forest honey | 0.015 | 0.011 | 1.62 | 104 |
| Total | Minimum | 0.007 | 0.007 | 0.03 | <22.0 |
| | Maximum | 0.049 | 0.089 | 1.62 | 180 |
| | Median | 0.020 | 0.013 | 0.10 | 26.9 |
| | Mean± SD | 0.023±0.010 | 0.019±0.017 | 0.27±0.40 | 50.6±46.3 |

**Table 3**.

Presence of significant (S) or not significant (NS) difference in term of radioactivity content between honeys with different botanical origin (significance was accepted at $P < 0.05$)

| $^{210}$Po | wildflower | acacia | eucalyptus | chesnut | citrus | linden | forest |
|---|---|---|---|---|---|---|---|
| **wildflower** | - | NS | NS | NS | NS | NS | S |
| **acacia** | NS | - | S | S | NS | S | S |
| **eucalyptus** | NS | S | - | NS | NS | S | S |
| **chesnut** | NS | S | NS | - | S | NS | NS |
| **citrus** | NS | NS | NS | S | - | S | S |
| **linden** | NS | S | S | NS | S | - | S |
| **forest** | S | S | S | NS | S | S | - |

| $^{40}$K | wild | acacia | eucalyptus | chesnut | citrus | linden | forest |
|---|---|---|---|---|---|---|---|
| **wildflower** | - | NS | NS | NS | NS | NS | NS |
| **acacia** | NS | - | S | S | NS | S | S |
| **eucalyptus** | NS | S | - | S | S | NS | S |
| **chesnut** | NS | S | S | - | S | NS | NS |
| **citrus** | NS | NS | S | S | - | S | S |
| **linden** | NS | S | NS | NS | S | - | NS |
| **forest** | NS | S | S | NS | S | NS | - |

No significant difference ($P > 0.05$) was found between $^{210}Po$ concentration in wildflower honey purchased from large supermarket chains ($0.23 \pm 0.27$) and the concentration in wildflower honey samples produced by local beekeepers in Central Italy ($0.40 \pm 0.46$ $Bqkg^{-1}$) (Meli et al., 2016).

Regarding the $^{40}K$, significant difference ($P < 0.05$) was found between the concentration of this radionuclide in acacia honey and that in eucalyptus, chesnut, linden and forest honey. Significant difference was found between its concentration in eucalyptus honey and that in acacia, chesnut, citrus and forest honey. No significant difference ($P > 0.05$) was found between $^{40}K$ activity concentration in wildflower honey and its concentration in all other kind of honey.

$^{137}Cs$ was consistently below the MDA (2.1 $Bqkg^{-1}$) except in samples 27 (linden) and 28 (forest) which showed an activity concentration of 5.14 and 4.00 $Bqkg^{-1}$respectively. This points to the presence of residual contamination from anthropogenic radioactivity in the analyzed samples.

Table 4 (part A) shows a comparison between the values found in this study with those reported by other authors for Italy and other European regions. Due to variations in concentrations of natural radioisotopes in the environment, honeys of different origins are expected to contain variable levels of radioactiviy. Such variations in the content of natural radionuclides can be attributed to the botanical types of the plants from which the nectar is collected, the preferential absorbability of those plants, variability of their soils, use of fertilizers, radioactivity content of the irrigation water and the climatic conditions (Borawska et al., 2013). Furthermore, different methods of sample solubilization and different analytical techniques may also affect the results. In any case the values that were measured in this study were comparable with those found by other researchers.

**Table 4** A) Comparison of the some radionuclide activity concentration (mBqkg$^{-1}$) of honey produced in different regions of Italia and Europe; B) Comparison between the $^{210}$Po,$^{238}$U, $^{226}$Ra mean concentration (mBqkg$^{-1}$) found for honey in the present study and that reported by UNSCEAR (2000) as reference value and as mean concentration in European diet (excluding Italy)

| **Part A** | **$^{137}$Cs** | **$^{40}$K** | **$^{238}$U** | **$^{235}$U** | **$^{210}$Po** | **$^{232}$Th** | **$^{226}$Ra** |
|---|---|---|---|---|---|---|---|
| Italy (this study) | <2100-5140 | <22000-180000 | 7-49 | <7 | 30-1620 | <7 | <200 |
| Marche, Italy (Meli et al., 2016) | <100 | 7280-100700 | <7-43 | <7 | 30-1980 | <7 | <340 |
| Poland (Borawska et al., 2013) | 110-16390 | 5510-98890 | | | | | |
| Czech Republic (Poschl et al., 2011) | <201-39200 | <2670-132000 | | | | | |
| Yuguslavia (Esposito et al., 2002) ( | <20.0-<1700 | <8000-87000 | <1500-<38000 | | | <200-<2200 | <180-<2800 |
| Croazia (Barisic et al., 1999) | 440±370 | 28300±15900 | | | | | |
| West Serbia (Djuric et al., 1996) | 2400±450 | 27100±3200 | 2300±1500 | 110±90 | - | 260±40 | |

| **Part B** | **$^{210}$Po** | **By UNSCEAR** | **$^{238}$U** | **By UNSCEAR** | **$^{226}$Ra** | **By UNSCEAR** |
|---|---|---|---|---|---|---|
| Food | Reference value | European diet | Reference value | European diet | Reference value | European diet |
| Milk products | 15 | 2-220 | 1 | 0.1-4.9 | 5 | <0.4-200 |
| Meat products | 60 | 37-67,000 | 2 | 1.6-5.6 | 15 | 2-220 |
| Grain products | 60 | 20-1,900 | 20 | 4.7-400 | 80 | 0.7-5200 |
| Leafy vegetables | 100 | 4-7,400 | 20 | 6-2200 | 50 | 2.2-1150 |
| Roots and fruits | 40 | 12-5,200 | 3 | 0.9-2900 | 30 | 5-9400 |
| Fish products | 2,000 | 50-120,000 | 30 | 2.5 | 100 | 8.5-7400 |
| Water and beverages | 5 | 0.1-7,600 | 1 | 0-1000 | 0.5 | 0-4000 |
| *Range* | *5-2,000* | | *1-30* | | *0.5-100* | |
| *Honey (this work)* | *270* | | *23* | | *<200* | |

342 *3.2. Potential health hazards resulting from honey consumption*

343 Quality control of honey is of great importance given the overall increasing global
344 consumption of honey, in particular, in the European Union which is, as already mentioned, the
345 world's largest consumer of honey. There is no specific regulation regarding the presence of
346 radionuclides in honey. The Directive 2001/110/EC of the European Union Commission relating
347 to honey includes some general and specific properties regarding its composition but no
348 guidelines are provided regarding the content of radionuclides are given (European Commission,
349 2001). The Council Regulations EC 733/2008 and EC 1048/2009, which set the maximum levels
350 for certain contaminants in foodstuffs, do not address apiary products (EC, 2008 and 2009). For
351 milk, its derivatives and products for children, the limits for $^{137}Cs$ were set at 370 $Bqkg^{-1}$ while
352 for other foods, the limits for $^{137}Cs$ were set at 600 Bq $kg^{-1}$. Consequently, at present, there are no
353 legal criteria with which to compare the results obtained in this study. The contents of $^{137}Cs$
354 found in this investigation were <2.1 $Bqkg^{-1}$ in 26 samples and 5.14 and 4.00 $Bqkg^{-1}$ respectively
355 in two samples (27, linden and 28, forest) and therefore were consistently below the limit set by
356 European Community Regulations (EC, 2008 and 2009) and the Guideline levels established by
357 WHO in 1995 and amended in 2010, (WHO, 1995 (2010)).

358 Table 4 (part B) shows a comparison between the $^{210}Po$,$^{238}U$ and $^{226}Ra$ mean
359 concentrations ($mBqkg^{-1}$) measured in honeys in this study and the corresponding values
360 reported by UNSCEAR (2000) which serve as reference values and as mean concentrations for
361 the European diet (excluding Italy). The results of the present investigation are comparable with
362 the reference values and with those found in the European diet indicated by UNSCEAR.

363 The final aim of our investigation of honey was to estimate the dose of radiation
364 attributable to the consumption of honey. The contribution to the radiation dose attributable to
365 the ingestion of natural radionuclides was calculated for three segments of the population: infants

366 (1-2 year), children (7-10 years) and adults (>17 years). In addition, two different values of daily
367 honey ingestion were considered: 5 g (a teaspoon) and 25 g.

368 Table 5 shows the $^{210}$Po committed effective dose (nSv $y^{-1}$) calculated for individuals
369 belonging to the three segments of the population, the minimum and maximum, the median, the
370 arithmetical mean and the relative standard deviation for every group of honey and for all
371 samples. Considering a daily honey ingestion of 5 g, the maximum contribution to the internal
372 dose (26000 nSv $y^{-1}$) by $^{210}$Po accounts for about 1.1% of the average annual effective dose to
373 which the world population (2.4 $mSvy^{-1}$) is exposed, whereas for a daily honey ingestion of 25 g,
374 that value rises to 5.3%.

375 Table 5 shows the $^{238}$U and $^{234}$U committed effective dose (nSv $y^{-1}$) calculated for
376 individuals belonging to the three above-mentioned segments of population, the minimum and
377 maximum, the median, the arithmetical mean and the relative standard deviation for every group
378 of honey and for all samples. For a daily honey ingestion of 5 g, the maximum contributions to
379 the internal dose from $^{238}$U (10.6 nSv $y^{-1}$) and $^{234}$U (20.8 $nSvy^{-1}$) correspond respectively to 4.4
380 $10^{-4}$% and 8.7 $10^{-4}$% of the natural radiation exposure in the world population, whereas, for a
381 daily honey ingestion of 25 g, these values rose to 53 ($^{238}$U) and to 104 nSv $y^{-1}$ ($^{234}$U)
382 respectively accounting for 2.2 $10^{-3}$% and 4.3 $10^{-3}$% of natural radiation exposure.

383 It is quite clear that the maximum dose obtained for $^{238}$U, $^{234}$U and $^{210}$Po should not be a
384 cause for concern even assuming the most conservative hypothesis (maximum dose value for a
385 daily consumption of 25 g of honey).

386 Table 6 shows the mean concentrations and the mean committed effective doses for all
387 radionuclides under study ($^{238}$U, $^{234}$U, $^{235}$U, $^{210}$Po, $^{232}$Th, $^{228}$Th, $^{226}$Ra, $^{40}$K and $^{137}$Cs) in infants,
388 children and adults assuming a daily honey ingestion of 5g and 25g.

389 **Table 5.** $^{210}$Po, $^{238}$U and $^{234}$U committed effective dose (mean and relative standard deviation
390 (SD) for every group of honey in nSvy$^{-1}$) from a daily honey ingestion of 5 and 25 g, calculated
391 for individuals belonging to three segments of the population (infants, children and adults)
392

| Group of honey | Radionuclide | 5 gd$^{-1}$ | | | 25 gd$^{-1}$ | | |
|---|---|---|---|---|---|---|---|
| | | infant | children | adult | infant | children | adult |
| wildflower | $^{210}$Po | 3700±4280 | 1090±1270 | 500±580 | 18500±21400 | 5460±6320 | 2520±2920 |
| acacia | | 660±210 | 190±60 | 0.09±0.03 | 3300±1050 | 970±310 | 450±140 |
| eucalyptus | | 2320±1170 | 609±340 | 320±161 | 1.600±5830 | 3430±1720 | 1580±0.800 |
| chesnut | | 12100±9550 | 3570±.820 | 1650±1300 | 60400±47700 | 17900±14100 | 8240±6510 |
| citrus | | 1770±1710 | 520±510 | 240±230 | 8807±8570 | 2620±2530 | 1210±1170 |
| linden | | 9340±140 | 2760±414 | 1207±181 | 46700±7005 | 13800±2070 | 6370±956 |
| forest honey | | 25700±3855 | 7580±1137 | 3500±525 | 128300±19245 | 37900±5685 | 17500±2625 |
| Total $^{210}$Po | Minimum | 470 | 140± | 60± | 2380 | 700 | 320 |
| | Maximum | 25700 | 7580± | 3500± | 128300 | 37900 | 17500 |
| | Mean±SD | 4320±6390 | 1280±1890 | 590±870 | 21600±32000 | 6380±9450 | 2940±4360 |
| | Median | 1580 | 470 | 216 | 7920 | 2340 | 1080 |
| wildflower | $^{238}$U | 5.810±2.41 | 3.29±1.37 | 2.18±0.90 | 29.04±12.05 | 16.46±6.83 | 10.89±4.52 |
| acacia | | 3.890±1.71 | 2.20±0.97 | 1.460±1.38 | 19.44±8.53 | 11.02±4.83 | 7.290±3.20 |
| eucalyptus | | 4.250±1.59 | 2.410±0.90 | 1.590±0.60 | 21.24±7.96 | 12.04±4.51 | 7.960±2.98 |
| chesnut | | 5.540±2.94 | 3.140±1.67 | 2.080±1.10 | 27.72±14.70 | 15.71±8.33 | 10.39±5.51 |
| citrus | | 5.620±2.50 | 3.180±1.42 | 2.110±0.94 | 28.08±12.50 | 15.91±6.93 | 10.53±4.69 |
| linden | | 2.808 | 1.591 | 1.053 | 14.04 | 7.96 | 5.265 |
| forest honey | | 3.240 | 1.836 | 1.215 | 106.2 | 9.18 | 6.075 |
| Total $^{238}$U | Minimum | 1.533 | 0.869 | 0.575 | 6.48 | 3.67 | 2.430 |
| | Maximum | 10.58 | 6.00 | 3.97 | 52.92 | 29.99 | 19.84 |
| | Mean±SD | 4.930±2.20 | 2.790±1.24 | 1.850±0.82 | 24.65±10.99 | 13.97±6.22 | 9.240±4.12 |
| | Median | 4.320 | 2.45 | 1.62 | 21.6 | 12.24 | 8.100 |
| wildflower | $^{234}$U | 5.668±5.882 | 3.226±3.348 | 2.136±2.217 | 28.34±29.41 | 16.13±6.742 | 10.68±11.09 |
| acacia | | 3.010±1.533 | 1.714±0.873 | 1.135±0.578 | 15.05±7.665 | 8.569±4.363 | 5.674±2.889 |
| eucalyptus | | 3.900±3.328 | 2.220±1.895 | 1.470±1.255 | 19.50±16.64 | 11.10±9.473 | 7.350±6.273 |
| chesnut | | 4.446±2.106 | 2.531±1.199 | 1.676±0.794 | 22.23±10.53 | 12.65±5.994 | 8.379±3.969 |
| citrus | | 5.569±3.864 | 3.170±2.199 | 2.099±1.456 | 27.85±19.32 | 15.85±11.00 | 10.49±7.282 |
| linden | | 2.574 | 1.465 | 0.970 | 12.87 | 7.326 | 4.851 |
| forest honey | | 2.574 | 1.465 | 0.970 | 12.87 | 7.326 | 4.851 |
| Total $^{234}$U | Minimum | 1.660 | 0.945 | 0.626 | 8.304 | 4.727 | 3.130 |
| | Maximum | 20.83 | 11.85 | 7.850 | 104.1 | 59.27 | 39.25 |
| | Mean±SD | 4.543±3.96 | 2.586±2.254 | 1.712±1.493 | 22.71±19.8 | 12.93±11.27 | 8.561±7.463 |
| | Median | 3.042 | 1.732 | 1.147 | 15.21 | 8.658 | 5.733 |

393

394 **Table 6.** Radionuclide mean concentration ($Bqkg^{-1}$), conversion factor for ingestion, Q, ($\mu Sv\ Bq^{-1}$) recommended by ICRP (2012) for three
395 segments of population (infants, children and adults), Mean Committed Effective Dose ($\mu Svy^{-1}$) by daily honey ingestion of 5 and 25 g and comparison
396 with the Committed Effective Dose from diet (UNSCEAR, 2000)
397
398

| **Radionuclide** | **Conversion** Infant | **Factor,** Children | **Q** Adult | **Mean** concentration | **Committed** by Infant | **effective** ingestion Children | **Dose** $5gd^{-1}$ Adult | **Committed** by Infant | **effective** ingestion Children | **Dose** $25gd^{-1}$ Adult | **Committed** from Infant | **Effective** European Children | **Dose** diet Adult |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $^{232}Th$ | 0.45 | 0.29 | 0.23 | <0.007 | <0.0057 | <0.0036 | <0.0029 | <0.028 | <0.0183 | <0.0145 | 0.26 | 0.32 | 0.38 |
| $^{228}Th$ | 0.37 | 0.15 | 0.072 | <0.007 | <0.0047 | <0.0019 | <0.0009 | <0.023 | <0.0095 | <0.0045 | 0.38 | 0.30 | 0.22 |
| $^{238}U$ | 0.12 | 0.068 | 0.045 | 0.023 | 0.0049 | 0.0028 | 0.0018 | 0.025 | 0.014 | 0.0092 | 0.23 | 0.26 | 0.25 |
| $^{235}U$ | 0.13 | 0.071 | 0.047 | <0.007 | <0.0017 | <0.0009 | <0.0006 | <0.0083 | <0.0045 | <0.0030 | 0.012 | 0.012 | 0.012 |
| $^{234}U$ | 0.13 | 0.074 | 0.049 | 0.019 | 0.0045 | 0.0026 | 0.0017 | 0.022 | 0.013 | 0.0086 | 0.25 | 0.28 | 0.28 |
| $^{226}Ra$ | 0.96 | 0.80 | 0.28 | <0.200 | <0.346 | <0.288 | <0.101 | <1.728 | <1.440 | <0.504 | <7.5 | <12 | <6.3 |
| $^{210}Po$ | 8.8 | 2.6 | 1.2 | 0.27 | 4.32 | 1.28 | 0.59 | 21.6 | 6.38 | 2.94 | 180 | 100 | 70 |
| $^{40}K$ | 0.042 | 0.013 | 0.006 | 50.6 | 3.88 | 1.20 | 0.55 | 19.4 | 6.00 | 2.77 | | | |
| $^{137}Cs$ | 0.012 | 0.010 | 0.013 | <2.1 | <0.046 | <0.038 | <0.050 | <0.230 | <0.192 | <0.249 | | | |

399
400

As regards $^{232}Th$, taking into account a concentration equal to the MDA value (0.007 $Bqkg^{-1}$), the committed effective dose ($\mu Sv\ y^{-1}$) calculated for individuals belonging to the three above mentioned segments of the population ranges from 0.0029 to 0.0057 $\mu Svy^{-1}$ and from 0.0145 to 0.028 $\mu Svy^{-1}$ for a daily honey ingestion of 5 g and 25 g respectively. These values account in any case for an absolutely negligible contribution to the average annual effective dose to which the world population is exposed. The same results were also obtained for $^{228}Th$, $^{235}U$ and $^{226}Ra$. The $^{40}K$ dose calculated in this study for comparison purposes taking into account a mean concentration of 50.6 $Bqkg^{-1}$ was also reported in table 6. Potassium is an essential element under homeostatic regulation in the body and it is not influenced by variations in environmental levels (Desideri et al., 2019). For $^{40}K$, the committed effective dose ($\mu Sv\ y^{-1}$) calculated for individuals belonging to the three segments of the population (infants, children and adults) ranged from 0.55 to 3.88 and from 2.77 to 19.4 $\mu Svy^{-1}$ for a daily honey ingestion of 5 g and 25 g respectively. These mean values account for 0.0245-0.169% and 0.120-0.844% respectively of the natural radiation exposure in the world. The committed effective dose trend of natural radionuclides was $^{210}Po \sim {}^{40}K > {}^{226}Ra >> {}^{238}U, \sim {}^{234}U \sim {}^{232}Th \sim {}^{228}Th > {}^{235}U$.

Table 6 also shows the comparison between the Committed Effective Dose of natural radionuclides due to a daily honey ingestion of 5 g or 25 g and that which is due to diet reported by UNSCEAR (2000) for individuals belonging to the three segments of the population under study. In particular, for $^{232}Th$ and $^{228}Th$, the dose values account for 0.8-10.8% and 0.46-6.0% respectively of the dose from diet reported by UNSCEAR; for $^{238}U$ and $^{234}U$, the dose values account for 0.7-10.9% and 0. 6-8.8% respectively; for $^{235}U$ the dose values account for 5-69%; for $^{226}Ra$ and $^{210}Po$, the dose values account for 1.6-23.0% and 0.84-12.0% respectively of the dose from diet reported by UNSCEAR (2000).

As far as $^{40}K$ is concerned, UNSCEAR (2000) indicates an annual effective dose from $^{40}K$ naturally present in our bodies of 165 and 185 μSv for adult and children respectively; this dose value contributes to about 60% of the total exposure due to food ingestion (290 μSv). Therefore, the detected average activities of $^{40}K$ in honey (50.6±46.3 $Bqkg^{-1}$), for the daily ingestion of 5 or 25g of this food, would appear to pose only a minor risk (0.55-19.4 μSv $y^{-1}$) to the internal radiation of the population.

For $^{137}Cs$, the only artificial radionuclide that was detected, taking into account the maximum concentration observed (5.14 Bq $kg^{-1}$), the committed effective dose (μSv $y^{-1}$) calculated for individuals belonging to the three segments of the population under study (table 6) ranged from 0.093 to 0.122 and from 0.470 to 0.609 $\mu Svy^{-1}$ for a daily honey ingestion of 5 g and 25 g respectively.

The results of the radiological characterization of the honey show that the levels of natural and artificial radioactivity were far below those assumed to be safe in food products by UNSCEAR for diet.

## 4. Conclusion

Natural radionuclides and $^{137}Cs$ were determined by alpha, gamma spectrometry and liquid scintillation in 28 kinds of honey widely consumed in Italy and purchased in major supermarket chains. Quality control is an important issue in regard to the rising global trends in total honey production and the fact that the European Union is the world's largest consumer of honey.

The radionuclides that were detected in honey showed a wide range of activity. These activity levels were influenced by the area foraged by bees, by the type of flowers where the

nectar was collected and by the quality of the water in the vicinity of the hive. The radioactivity found in the investigated honeys was chiefly attributable to $^{40}K$, as honey is rich in potassium.

The honeys consumed in Italy were found to be of good quality. In fact, the levels of the radionuclides that were measured were far below those assumed to be safe in food products by UNSCEAR. We can therefore conclude that honey currently consumed in Italy is an absolutely negligible source for internal contamination by radionuclides and thus a food of no concern in terms of radiation risk.

**Funding**

This study was supported by the University of Urbino Carlo Bo.

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