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Radioactivity in honey of the central Italy

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Abstract

Natural radionuclides and ^{137}Cs in twenty seven honeys produced in a region of the Central Italy were determined by alpha (^{235}U, ^{238}U, ^{210}Po, ^{232}Th and ^{228}Th) and gamma spectrometry (^{137}Cs, ^{40}K, ^{226}Ra, and ^{228}Ra). The study was carried out in order to estimate the background levels of natural (^{40}K, ^{238}U and ^{232}Th and their progeny) and artificial radionuclides (^{137}Cs) in various honey samples, as well as to compile a data base for radioactivity levels in that region. ^{40}K showed a mean activity of 28.1±23.0 $Bqkg^{-1}$ with a range of 7.28-101$Bqkg^{-1}$. The mean of ^{210}Po activity resulted 0.40 ± 0.46 $Bqkg^{-1}$ with a range of 0.03-1.98 $Bqkg^{-1}$. The mean of ^{238}U activity resulted 0.020 ± 0.010 $Bqkg^{-1}$. ^{226}Ra and ^{228}Ra resulted always <0.34 and < 0.57 $Bqkg^{-1}$ respectively, ^{235}U, ^{228}Th and ^{232}Th were always <0.007 $Bqkg^{-1}$.^{137}Cs resulted <0.10 $Bqkg^{-1}$ in all samples. The committed effective doses due to ^{210}Po from ingestion of honey for infants, children and adults account for 0.002-5.13 % of the natural radiation exposure in Italy. The honeys produced in Central Italy were of good quality in relation to the studied parameters, confirming the general image of a genuine and healthy food associated to this traditional products.

Keywords: Italian honeys; natural and artificial radionuclides; alpha and gamma spectrometry

1. Introduction

Honey, a product of the elaboration of flower nectar or honeydew by bees, is one of the most complex foods produced naturally (Pisani et al., 2008, Tuzen et al., 2007).

Honey provides energy, with valuable nutritional, healing and prophylactic properties attributed to chemical composition and predominant simple sugars content (Belitz et al., 2004). The general features and elemental composition of honey depend upon its botanical and geographical origin. Honey contains mixture of different carbohydrates, including fructose, glucose, maltose, sucrose, high sugars, proteins, amino acids, vitamins and minerals (Buldini et al., 2001). The contribution of minerals, that depends upon the soil type, is relatively low and normally accounts for 0.1–0.2% of nectar honeys (Pohl, 2009).

Honey is also used as an ingredient or preservative in foodstuffs because of its flavor, color and sweetness. This foodstuff has healing properties where the moisturizing action of honey around a wound facilitates healing process and high viscosity of honey inhibits infections to penetrate into the body. The antibacterial properties are due to its low acidity and low-level hydrogen peroxide release (Akbari et al., 2012).

Honey may be useful as biomonitor for collecting information regarding the environment within the bees' forage area (a surface of more than 7 km^2). Honey bees come into a contact with different parts of the surroundings and are exposed to potential pollutants when they forage for nectar, pollen, honeydew, or other exudates within such a territory (Bratu and Georgescu, 2005). In this manner, contaminants in air, water, and soil reach the honey and change its composition and quality. As a food stuff used also for healing purposes, honey should contain only small amounts of pollutants as heavy metals and radionuclides (Meli et al, 2015). The content of radionuclides in honey is of interest in terms of quality and potential adverse human health risks.

Taking into account that apiculture is popular in Italy, in fact the number of hives in Italy exceeds 1,000,000 and yearly the country produces approximately 20,000 tons of honey (Benvenuti et al., 2009), it was decided to determine the levels in honeys of natural and artificial radionuclides.

In our daily lives, we are each exposed to various types of naturally occurring ionizing radiation which is commonly referred to as background radiation. Naturally occurring background radiation comes from a number of source that include terrestrial radiation, cosmic radiation, inhaled radionuclide and internal radionuclide. Any radioactivity present on air or more importantly in the ground and soil may transfer into food grown on it. It happens, however, that some naturally occurring radioactive elements find their way into our body. The most important radionuclide that gives the largest part of the dose to an average person from ingestion are ^{40}K, a primordial radionuclide, and ^{210}Po, a radionuclide of ^{238}U radioactive family.

^{40}K is a natural radioisotope present in soil and as the element K, an essential plant nutrient, enters in the plant roots via ion channels or specific transporters. The percentage made up by the natural radionuclide ^{40}K is 0.0117% K is generally abundant in the food (Sugiyama et al., 2009).

Among the alpha emitters ^{210}Po is estimated to contribute about 7% of the effective dose equivalent to man from ingested natural internal radiation (UNSCEAR, 1988). This radionuclide and his grandfather ^{210}Pb belong to ^{238}U series. Their presence in the terrestrial environment arises from ^{222}Rn which, once produced, may remain in soil interstitial air spaces, decay in ^{210}Pb and ^{210}Po within the mineral matrix of soil or be released to the atmosphere. ^{210}Pb and ^{210}Po return to the earth's surface via both wet and dry deposition. Atmospheric fallout of these decay products result in the contamination of plants and the top layer of soil. Most of the natural radioactivity content in wild leafy plants is ^{210}Po as the result of the direct deposition of ^{222}Rn daughters from atmospheric precipitation and their presence in all terrestrial foodstuffs is

inevitable (Brown et al., 2011, Persson et Holm, 2011). There are other factors from anthropogenic activities contributing to increase the levels of ^{210}Pb and ^{210}Po in herbal medicinal plants: atmospheric deposition from industrial emissions, from town wastes, phosphate ore processing, coal-fired power stations, coal mining, metal smelting, etc.

About 18% of the average internal dose of the population is due to ingestion of ^{210}Po along with its precursor ^{210}Pb. ^{210}Po, in fact, causes considerable radiation risk even at minimal intake due to its high linear energy transfer (LET). The ^{210}Po toxicity is comparable to ^{239}Pu and about 5 times greater than ^{226}Ra (NRC, 1988).

The environment is also contaminated by the presence of the artificial and biologically significant radionuclides as^{131}I, ^{134}Cs, ^{137}Cs; their presence is due to atmospheric nuclear weapon testing (1945-1963) and to a series of nuclear accidents, Windscale 1957, Kyštym1957 and Chernobyl, 1986 (De Cort et al., 1998; Mitrovic et al., 2009), or as consequences of natural disasters (Fukushima 2011). The most important long-lived radionuclide is ^{137}Cs with long half-life (30.17 years). Its chemical similarity to potassium means that it is rapidly adsorbed by the bloodstream and can be distributed in all cell of the body particularly in all soft tissues in animals including muscle. Its activity concentration in samples of human food decreases with time after deposition according to its biological, ecological and physical half life. Recently, there has been a growing concern about the effect of low level radioactivity on human health (Desideri et al., 2014a).

Taking into account that safety of the honey is of great importance, the aim of this study was the determination of natural (^{40}K, ^{238}U and ^{232}Th and their progeny) and artificial radionuclides (^{137}Cs) to 1) carry out a radiological characterization of various honey samples, 2) investigate possible environmental contamination as well as 3) compile a data base for radioactivity levels in that region.

^{235}U, ^{238}U, ^{210}P, ^{228}Th and ^{232}Th were determined by alpha spectrometry; gamma spectrometry was used to measure ^{137}Cs, ^{40}K, ^{226}Ra, ^{228}Ra.

2. Materials and methods

2.1. Samples

Twenty-seven samples of honey (25 wildflower, 1 honeydew and 1 acacia) were analyzed. The samples were produced and collected in 2013 from individual beekeepers in Central-Eastern Italy (Marche region), in an area near S. Marino, with small-scale mixed farming and scarce big industries. Further, the urban centers are small and the main town, Urbino, is an historical city. Each sample, furnished by the local Health Agency (ASUR), was accompanied by a sheet in which the type of honey and provenance were indicated. Table 1 shows the botanical origin of the honey samples and the collection area (four different areas of sampling). The same table shows the content of sugars, the degree of humidity, the pH, the free acidity, the combined and the total acidity determined previously by authors (Meli et al., 2015); these parameters were in good agreement to those reported by the Council Directive 2001/110/EC relating to honey (EC, 2001).

2.2. Analytical methods

Gamma spectrometry. It is possible to determine simultaneously many radionuclides by a direct γ-spectrometry of the sample without any specific pre-treatment of this. Nevertheless it is impossible to determine directly all the radionuclides of interest because some of them, as ^{210}Po or ^{232}Th, are not gamma emitters. Particular care must be taken to ensure that the overall analytical procedure does not give erroneous results. The principal cause of error is that some

procedures automatically assume secular equilibrium for all members of the series. Some matrixes can result from complex chemical processes causing breaks in the radioactive equilibria. In these cases, if the daughters are long lived radionuclides, it is impossible to restore these equilibria in laboratory. For example, in the ^{238}U family the equilibrium breaks between ^{234}Pa and ^{234}U, ^{234}U and ^{230}Th, ^{230}Th and ^{226}Ra, ^{226}Ra and ^{222}Rn, ^{226}Ra and ^{210}Pb can be found. For ^{226}Ra and ^{222}Rn the equilibrium can be restored in laboratory. So it is possible to determine ^{238}U by its daughter gamma emitters ^{234}Th, ^{234m}Pa and ^{234}Pa, and ^{226}Ra by ^{214}Pb and ^{214}Bi, but it is impossible to determine by this way the activity of ^{234}U and ^{230}Th activities (Desideri et al., 2006; Desideri et al., 2014b).

In this study the dried sample was packed in 500 ml plastic container, sealed for about thirty days prior to the measurement to ensure that equilibrium had been established between ^{226}Ra and its short-living decay products.

All the measurements were performed with a reverse-electrode coaxial Ge detector (REGe), with resolution of 1.8 keV for the 1332 keV ^{60}Co photopeak, for 4096 chambers (Canberra, USA). Peak detection efficiencies were automatically calculated through a computer system interfaced to an 8K multichannel analyser; the energy and efficiency calibration was performed by means of gamma–ray reference standards of mixed radionuclides. The ^{226}Ra activity was determined by taking the mean activity of four separate photopeaks of its daughter nuclides (^{214}Pb at 295.22 and 351.99 keV, and ^{214}Bi at 609.32 keV and 1120.28 keV). The ^{228}Ra of the samples was determined by measuring the intensities of the 338.3, 911.1 and 969.11 of ^{228}Ac. The ^{40}K and ^{137}Cs were directly measured from the 1460.8 keV and 661.66 peak energies, respectively.

Alpha spectrometry. This radiometric technique consists in measurements of the sources of the radionuclides after their separation (by extraction chromatography, precipitation,

electrodeposition etc.) from the solution arising from the complete dissolution of the sample (Desideri et al., 2014b). This technique requires lengthy preparation and source counting; however, it does have the advantage of being inexpensive, highly sensitive and specific, while providing complete information on concentration and isotopic ratios of ^{238}U, ^{235}U,^{234}U, ^{232}Th, ^{230}Th and ^{228}Th. The radioanalytical method accuracy was regularly checked through participation in intercomparison exercises organized by the International Atomic Energy Agency (IAEA).

The method consists in two steps: a) source preparation and b) source counting.

a) Source preparation

Uranium and thorium were separated by Extraction Chromatography using, as extractant, the diamyl, amylphosphonate (DAAP); this is a selective extractant in nitric medium for tetravalent and hexavalent actinides (Th, U, Pu, Np). DAAP forms nitrate complexes with the actinide elements The formation of these complexes is driven by the concentration of nitrate in the sample solution. Therefore, the uptake of the actinides increases with increasing nitric acid concentration: all have strong retention above 5M nitric acid. In 4-6 M HCl, uranium, but not thorium, was retained. The very different behaviour of the two elements in the 4-6M HCl range allows the selective elution of thorium from the resin after both thorium and uranium have been loaded. It is necessary to reduce the effect of phosphate anion occurring quite commonly in a variety of biological and environmental samples. The anion phosphate readily forms complexes tetravalent with actinides that are not extracted by the DAAP. Fortunately the addition of aluminium to the sample matrix can significantly reduce this issue. Added aluminium can effectively tie up the phosphate preventing its interference with actinide uptake by the resin (Thakkar, 2000, Roselli et al., 2015).

Taking these considerations into account, the radiochemical method employed in the study is as follows:

10 g of honey, after addition of a known activity of ^{236}U and ^{229}Th as the yield internal standards, were 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 was filtered. The solution was passed through a chromatographic column (UTEVA Resin, Eichrom Technologies) constituted by DAAP supported onto a inert polymer (acrylic ester) and conditioned with 5M HNO_3; 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 was electroplated from ammonium sulphate solution at pH 4.

^{210}Po emits only alpha particles at 5.407 MeV. 10 g of honey, after addition of a known activity of ^{209}Po as the yield internal standard, were dissolved in 150 ml of 1M HCl; the solution was stirred (30') and, finally, was filtered. Polonium was deposited at 85-90°C and pH 1.5-2.0, continuously for 4 h, on a silver disk, placed in a syringe and immersed in the solution arising from the sample dissolution (200 mL of 1 M HCl) and containing 10 ml of 20% hydroxylamine hydrochloride and 10 ml of 25% sodium citrate. No preliminary separation was required and essentially quantitative recoveries were calculated by using a standard ^{209}Po tracer.

b) *Source counting*

The measurements of the polonium, uranium and thorium isotopes alpha emitters were carried out using an α-spectrometer equipped with a semiconductor silicon detector of surface barrier type (300 mm^2 active surface, resolution 20 keV, 31.7±3.1%. of counting efficiency, and

190 2 x 10^{-6} s^{-1} of the background in the interested energy region) (Canberra Industries, Inc., 800
191 Research Parkway, Meriden, CT 06450) and connected to a computerized multichannel analyzer.
192 The mean chemical yield resulted 67.9±18.4, 77.8±16.4 and 88.1±9.2 %, for uranium,
193 thorium and polonium respectively. The minimum detectable activity concentration (MDC) was
194 found to be 0.007 $Bqkg^{-1}$for uranium, thorium and polonium.
195
196 *2.3. Statistical analyses*
197 Data were grouped according to the collection area and, for every group, the
198 concentration of every sample with the relevant uncertainty was reported; uncertainties of all
199 measurements were calculated taking into account statistical fluctuations of the peaks and of the
200 backgrounds, and efficiency calibration; the median, the arithmetical mean with relevant
201 standard deviation, the minimum and maximum values are reported. Statistical analysis
202 (Student's *t*-test) were carried out after logarithmic transformation of the individual data.
203 Concentration differences between samples collected in different areas were calculated;
204 significance was accepted at $P < 0.05$.
205
206 *2.4. ^{210}Po annual intake and committed effective dose calculation*
207 The ^{210}Po annual intake was calculated by the product of the ^{210}Po concentration and the
208 ingestion rate; for the consumption rate, two different values of honey daily ingestion were taken
209 into account: a minimum of 5g (a teaspoon) and a maximum of 25 g.
210 The annual committed effective dose for an individual, as a result of ^{210}Po intake, was
211 calculated using the following formula:
212
213 $$D_{Po\text{-}210} = Q \ x \ C_{Po210} \ x \ I$$
214

where $D_{Po\text{-}210}$ is the annual committed effective dose for ^{210}Po (μSv y^{-1}), $C_{Po\text{-}210}$ is the ^{210}Po concentration (Bq kg^{-1}), I is the ingestion rate (kg y^{-1}), and Q is the conversion factor; in this paper, the annual committed effective dose was calculated using the conversion factors 8.8, 2.6, 1.2 μSv Bq^{-1} for infants (< 1 year), children (10 years) and adults respectively recommended by UNSCEAR (2000).

3. Results

3.1. Radionuclides concentration

Labeling of honey must be supported by analysis that confirms its provenance and safety. The EU is the world's largest consumer of honey (Vanhanen et al., 2011) and the need of assuring low levels of radionuclides in honey is very high given the constantly rising global trends in honey production.

Table 2 shows the ^{210}Po, ^{238}U and ^{40}K activity concentration ($Bqkg^{-1}$), the minimum and maximum, the median, the arithmetical mean and the relevant standard deviation. For the concentrations below the detection limit, MDC was considered.

^{235}U, ^{232}Th, ^{228}Th always resulted < 0.007 $Bqkg^{-1}$; ^{214}Pb and ^{214}Bi (^{226}Ra) were always < 0.33 and 0.34 $Bqkg^{-1}$ respectively and ^{228}Ac (^{228}Ra) < 0.57 $Bqkg^{-1}$.

In the ^{238}U series, ^{210}Po and ^{238}U range from 0.03 to 1.98 $Bqkg^{-1}$ (mean value 0.40 ± 0.46 $Bqkg^{-1}$) and from <0.007 to 0.043 $Bqkg^{-1}$ (mean value 0.020 ± 0.010 $Bqkg^{-1}$) respectively; ^{210}Po was detectable in the 100% and ^{238}U in the 85% of the samples.

Figure 1 shows, for ^{238}U and ^{210}Po, the comparison between the their mean content calculated for every area. A significant difference ($P < 0.01$) was found for ^{210}Po in samples from Candigliano Valley and those from other three areas. As far as ^{238}U activity concentration, no statistically significant differences were found for the different areas.

^{40}K ranges from 7.28 to 101 $Bqkg^{-1}$ (mean value 28.1 ± 23.0 $Bqkg^{-1}$) and it was detectable in the 100% of the samples.

The variation of natural radionuclides 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 a variation in natural radionuclides content are preferential absorbability of the plant, use of fertilizers, irrigation water and climatic conditions.

^{137}Cs was always below the MDC (0.10 $Bqkg^{-1}$); it means that residual contamination from anthropogenic radioactivity is not present in these areas.

The determined values were generally comparable to those reported by other authors (Table 3) for others Italian or European regions. However, it is difficult to draw conclusions by extrapolating data from different botanical types of honey, this feature may substantially affect the chemical composition. Furthermore, different methods of sample solubilization and different analytical techniques may also affect the results.

3.2. Potential health hazards resulting from honey consumption

In addition to the environmental concern regarding honey element composition, quality control of honey is also important given the increasing global trends in total honey production and the fact that the European Union is the world's largest consumer of honey. There are no specific regulation about the presence of radionuclides in honey. The Directive 2001/110/EC of the European Union Commission relating to honey includes some general and specific properties of honey composition but no guidelines about the content of radionuclides are given (European Commission, 2001).

The regulation EC 733/2008, which set the maximum levels for certain contaminants in foodstuffs, does not address apiary products (EC, 2008); the limits set for ^{137}Cs were 370 $Bqkg^{-1}$

for milk, its derivatives and products for children, and 600 Bq kg^{-1} for other foods. Consequently, now, there are not legal criteria with which to compare the results obtained by our studio. In any case, the contents of ^{137}Cs, being always < 0.10 Bq kg^{-1} in all honey samples tested, were always below the maximum levels indicated in Community legislation.

Table 4 shows a comparison between the ^{210}Po concentration determined in honey samples, that found by the authors in the previous study for diet (Desideri et al., 2014b) and those reported by UNSCEAR (2000) for European diet (except Italy) and for the reference values.

In this study the contribution to the radiation dose due to ^{210}Po ingestion, the most radiotoxic among the radionuclides considered in this study, is calculated for infants (< 1 year), children (1 year < age < 10 years) and adults (>10 years) considering two different values of honey daily ingestion: 5 g (a teaspoon) and 25 g.

Table 5 shows the^{10}Po committed effective dose (μSv y^{-1}) calculated for individuals of the three groups of population, the minimum and maximum, the median, the arithmetical mean and the relevant standard deviation. The relative uncertainty on the committed effective dose ranged from 20 to 25%. The minimum and maximum values of dose (0.07 and 31.8 μSv y^{-1}) due to ^{210}Po from 5 g daily honey ingestion account for 0.002-1.03% of the natural radiation exposure in Italy (3.1 mSv y^{-1}); the minimum and maximum values of dose (0.33-159μSv y^{-1}) due to ^{210}Po from 25 g daily honey ingestion account for 0.01- 5.13%. The maximum dose obtained (159 μSvy^{-1}) should not be a cause for concern as the scenario selected for this study was extremely conservative: in fact, this scenario was based on: 1) daily consumption of 25 g of honey, which is a large amount for infant 2) the maximal activity concentration in honeys analyzed.

Table 5 shows, for the committed effective dose due to ^{210}Po from honey ingestion, the comparison between the arithmetical mean ($\mu Sv\ y^{-1}$) calculated for every area. A significant difference ($P < 0.01$) was found between the doses from Candigliano Valley and those from other three areas due to the significant different concentrations.

4. Conclusion

Natural radionuclides and ^{137}Cs were determined by alpha and gamma spectrometry in 27 kinds of honey produced in central Italy. The quality control is important given the increasing global trends in total honey production and the fact the European Union is the world's largest consumer of honey.

The radionuclides were found to be present in honey in various proportions depending on the area foraged by bees from flower type visited for the collection of nectar, the quality of water in the vicinity of the hive.

The honeys produced in Central Italy were of good quality in relation to the standard parameters, in fact, the level of the radionuclides taken into account is far below those assumed as safe in food products by the international commissions of radiological protection; therefore it is possible to conclude that radionuclide intoxication resulting in human adverse effects is not a concern.

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Table 1 List of honey samples, location of production, kind and botanical origin and chemical composition (Meli et al., 2015)

Sample code	Kind	Botanical origin	Sampling area	% humidity	Sugars (Brix)	pH	Free acidity (meq/kg)	Combined acidity (meq/kg)	Total acidity (meq/kg)
1	wildflower	Alfalfa, Clover, Sulla	Urbino	17.7	80.6	4.00	28.1	4.3	32.4
2	wildflower	Alfalfa	Urbino	18.3	80.0	3.74	34.4	2.0	36.4
3	wildflower	Sulla, Grasses	Urbino	18.9	79.4	4.08	32.1	2.2	34.3
4	wildflower	Alfalfa, Woods, Sunflower	Urbino	18.4	80.2	3.98	29.9	3.5	33.4
5	wildflower	Alfalfa, Woods	Urbino	18.2	80.4	4.18	31.1	5.7	36.8
6	wildflower	Alfalfa, Woods, Sunflower, Coriander	Urbino	18.4	80.2	3.94	24.7	5.3	30.0
7	wildflower	Woods	Urbino	18.4	80.2	4.05	28.8	4.8	33.6
8	wildflower	Alfalfa, Sunflower, Bramble	Urbino	20.0	78.6	3.98	29.8	3.2	33.0
9	wildflower	Lime, Pastures	Urbino	18.8	79.8	4.30	31.3	5.1	36.4
10	wildflower	Alfalfa, Woods, Clover	Foglia Valley	17.6	80.7	3.84	19.3	1.2	20.5
11	wildflower	Alfalfa, Sulla, Woods	Foglia Valley	16.8	81.5	4.08	26.7	1.1	27.8
12	wildflower	Alfalfa, Woods	Foglia Valley	17.9	80.4	3.85	23.4	2.5	25.9
13	wildflower	Alfalfa, Chestnut, Woods	Foglia Valley	16.9	81.2	3.92	24.0	1.3	25.3
14	wildflower	Alfalfa, Woods	Foglia Valley	16.9	81.2	3.87	21.7	1.7	23.4
15	wildflower	Alfalfa, Woods, Clover,Orchard	Foglia Valley	16.1	82.0	3.78	22.0	1.9	23.9
16	wildflower	-	Metauro Valley	18.9	79.4	3.90	23.9	2.0	25.9
17	wildflower	Alfalfa, Robinia, Sulla	Metauro Valley	18.9	79.4	3.90	23.4	3.3	26.7
18	wildflower	Alfalfa, Cherry	Metauro Valley	18.3	80.0	3.99	27.5	3.3	30.8
19	wildflower	Alfalfa,Sunflower,Chestnut	Metauro Valley	18.8	79.8	5.40	25.6	3.0	25.3
20	wildflower	Robinia pseudoacacia, Alfalfa,Woods	Metauro Valley	18.0	80.4	3.74	25.0	2.0	23.4
21	wildflower	Meadows, Pastures, Woods	Metauro Valley	19.0	79.6	3.85	31.1	3.1	34.2
22	wildflower	Robinia pseudoacacia, Alfalfa,Sunflower	Metauro Valley	19.0	79.6	3.85	31.1	3.1	34.2
23	acacia	Robinia pseudoacacia	Candigliano Valley	18.0	80.6	3.87	16.2	2.3	18.5
24	honeydew	Woods, Orchard,Lime	Candigliano Valley	18.2	80.4	4.38	37.9	4.6	42.5
25	wildflower	Woods, Orchard	Candigliano Valley	15.4	83.0	4.98	30.0	4.5	34.2
26	wildflower	Alfalfa, Sunflower, Robinia, Lime	Candigliano Valley	18.2	80.4	3.81	35.7	4.3	40.0
27	wildflower	Alfalfa, Meadows,Woods	Candigliano Valley	18.0	80.4	4.87	37.9	4.7	42.6

Table 2 Activity concentration ($Bqkg^{-1}$) of ^{210}Po ,^{238}U (alpha spectrometry) and ^{40}K (gamma spectrometry)

Sample code	^{210}Po	^{238}U	^{40}K
1	0.25±0.050	0.011±0.005	19.1±0.91
2	0.03±0.008	0.024±0.011	23.9±1.22
3	0.25±0.050	0.024±0.011	29.9±1.29
4	0.36±0.070	0.033±0.015	25.6±1.10
5	0.72±0.140	0.012±0.005	38.9±1.60
6	0.23±0.050	0.043±0.019	19.7±0.98
7	0.62±0.120	<0.007	31.1±1.29
8	0.27±0.050	0.013±0.006	25.8±1.20
9	0.71±0.140	0.027±0.012	49.6±1.90
10	0.08±0.020	<0.007	7.28±0.76
11	0.54±0.110	0.039±0.018	24.2±1.04
12	0.24±0.050	0.035±0.016	11.6±0.87
13	0.26±0.050	0.017±0.008	17.2±0.94
14	0.09±0.020	0.019±0.009	10.0±0.73
15	0.06±0.015	0.020±0.009	28.2±1.41
16	0.32±0.060	0.014±0.006	18.3±0.99
17	0.12±0.020	0.013±0.006	11.7±0.94
18	0.36±0.070	0.016±0.007	25.3±1.72
19	0.16±0.030	0.011±0.005	9.04±0.84
20	0.14±0.030	<0.007	9.86±0.63
21	0.14±0.030	0.022±0.010	20.6±1.16
22	0.22±0.040	<0.007	22.6±1.04
23	0.09±0.020	0.023±0.011	8.39±0.74
24	0.87±0.170	0.026±0.012	65.6±2.33
25	1.63±0.330	0.030±0.014	83.2±3.03
26	0.14±0.030	0.021±0.009	20.8±0.95
27	1.98±0.400	0.020±0.009	101±3.56
Median	0.25	0.020	21.7
Mean	0.40	0.020	28.1
Standard Deviation	0.46	0.010	23.0
Minimum	0.03	<0.007	7.28
Maximum	1.98	0.043	101

Table 3 Comparison of the some radionuclide activity concentration ($Bqkg^{-1}$) of honey produced in different regions of Italia and Europe

	^{137}Cs	^{40}K	^{238}U	^{235}U	^{210}Po	^{232}Th	^{226}Ra
Marche, Italy (this paper)	<0.10	7.28-100.7	<0.007-0.043	<0.007	0.03-1.98	<0.007	<0.34
Yuguslavia (Esposito et al., 2002) (	<0.02-<1.7	<8.0-87	<1.5-<38			<0.20-<2.2	<0.18-<2.8
Poland (Borawska et al., 2013)	0.11-16.39	5.51-98.89					
West Serbia (Djuric et al., 1996	2.4±0.45	27.1±3.2	2.3±1.5	0.11±0.09	-	0.26±0.04	
Croazia (Barisic et al., 1999)	0.44±0.37	28.3±15.9					

Table 4 Comparison between the ^{210}Po mean concentration ($mBqkg^{-1}$) found for honey, that determined by authors (Desideri et al., 2014b) in the previous study $mBqkg^{-1}_{ww}$) for diet and that reported by UNSCEAR (2000) as reference value and for European diet (except Italy)

Food	By authors	By Reference value	UNSCEAR European diet
Milk products	20	15	2-220
Meat products	90	60	37-67,000
Grain products	50	60	20-1,900
Leafy vegetables	130	100	4-7,400
Roots and fruits	20	40	12-5,200
Fish products	5,760	2.000	50-120,000
Water and beverages	30	5	0.1-7,600
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Table 5 ^{210}Po committed effective dose (μSvy^{-1}) from honey (5 and 25 g daily consumption)

Area	Sample code	5 gd^{-1}			25 gd^{-1}		
		infant	children	adult	infant	children	adult
Urbino	1	4.02	1.19	0.55	20.1	5.93	2.74
	2	0.48	0.14	0.07	2.41	0.71	0.33
	3	4.02	1.19	0.55	20.1	5.93	2.74
	4	5.78	1.71	0.79	28.9	8.54	3.94
	5	11.6	3.42	1.58	57.8	17.1	7.88
	6	3.69	1.09	0.50	18.5	5.46	2.52
	7	9.96	2.94	1.36	49.8	14.7	6.79
	8	4.34	1.28	0.59	21.7	6.41	2.96
	9	11.4	3.37	1.55	57.0	16.8	7.77
Mean ±stand.dev.		6.14±3.91	1.81±1.15	0.84±0.53	30.7±19.5	9.07±5.17	4.19±2.66
Foglia Valley	10	1.28	0.38	0.18	6.42	1.90	0.88
	11	8.67	2.56	1.18	43.4	12.8	5.91
	12	3.85	1.14	0.53	19.3	5.69	2.63
	13	4.18	1.23	0.57	20.9	6.17	2.85
	14	1.45	0.43	0.20	7.23	2.14	0.99
	15	0.96	0.28	0.13	4.82	1.42	0.66
Mean ±stand.dev		3.40±2.93	1.00±0.86	0.46±0.40	17.0±14.6	5.02±4.32	2.32±2.00
Matauro Valley	16	5.14	1.52	0.70	25.7	7.59	3.50
	17	1.93	0.57	0.26	9.64	2.85	1.31
	18	5.78	1.71	0.79	28.9	8.54	3.94
	19	2.57	0.76	0.35	12.8	3.80	1.75
	20	2.25	0.66	0.31	11.2	3.32	1.53
	21	2.25	0.66	0.31	11.2	3.32	1.53
	22	3.53	1.04	0.48	17.7	5.22	2.41
Mean ±stand.dev		3.35±1.54	0.99±0.45	0.46±0.21	16.7±7.69	4.95±2.27	2.28±1.05
Candigliano Valley	23	1.45	0.43	0.20	7.23	2.14	0.99
	24	14.0	4.13	1.91	69.9	20.6	9.53
	25	26.2	7.73	3.57	131	38.7	17.8
	26	2.25	0.66	0.31	11.2	3.32	1.53
	27	31.8	9.40	4.34	159	47.0	21.7
Mean ±stand.dev		15.1±13.7	4.47±4.06	2.06±1.87	75.6±68.7	22.3±20.3	10.3±9.36
Total	Median	4.02	1.19	0.55	20.1	5.93	2.74
	Mean	6.47	1.91	0.88	32.4	9.56	4.41
	Stand. Dev.	7.43	2.19	1.01	37.1	11.0	5.06
	Minimum	0.48	0.14	0.07	2.41	0.71	0.33
	Maximum	31.8	9.40	4.34	159	47.0	21.7