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Analytical Methods

Radioactivity in honey of the central Italy

Maria Assunta Meli [a], Donatella Desideri [a,*], Carla Roselli [a], Laura Feduzi [a], Claudio Benedetti [b]

[a] Department of Biomolecular Sciences, Urbino University "Carlo Bo", P.zza Rinascimento 6, 61029 Urbino, Italy
[b] Health Agency Urbino (ASUR), Via Gramsci 29, 61029 Urbino, 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 Bq kg^{-1} with a range of 7.28–101 Bq kg^{-1}. The mean of ^{210}Po activity resulted 0.40 ± 0.46 Bq kg^{-1} with a range of 0.03–1.98 Bq kg^{-1}. The mean of ^{238}U activity resulted 0.020 ± 0.010 Bq kg^{-1}. ^{226}Ra and ^{228}Ra resulted always <0.34 and <0.57 Bq kg^{-1} respectively, ^{235}U, ^{228}Th and ^{232}Th were always <0.007 Bq kg^{-1}. ^{137}Cs resulted <0.10 Bq kg^{-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.

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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, Protano, & Riccobono, 2008; Tuzen, Silici, Mendil, & Soylak, 2007).

Honey provides energy, with valuable nutritional, healing and prophylactic properties attributed to chemical composition and predominant simple sugars content (Belitz, Grosch, & Schieberle, 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, Cavalli, Mevoli, & Sharma, 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 & 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, Desideri, Roselli, Benedetti, & Feduzi, 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, Fraschetti, Gubiani, & Masci, 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

* Corresponding author.
E-mail address: donatella.desideri@uniurb.it (D. Desideri).

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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 & Holm, 2011). It is also known that natural levels of ^{210}Pb and ^{210}Po in the environment can locally be increased by anthropogenic activities like phosphate ore processing, coal-fired power stations, coal mining, metal smelting, etc. which produce enhanced levels of ^{210}Pb and ^{210}Po.

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, Kyštym 1957 and Chernobyl 1986 (De Cort et al., 1998; Mitrovic, Vitorovic, Vitorovic, Pantelic, & Adamovic, 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, Roselli, et al., 2014).

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 and ^{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 DirCouncil dirCouncil directive 2001/110/EC relating to honey (EC, 2001).

2.2. Analytical methods

2.2.1. 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, Meli, Feduzi, & Roselli, 2006; Desideri, Meli, et al., 2014).

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 8 K 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.

2.2.2. 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, Meli, et al., 2014). 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 intercom-

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