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RADIOACTIVITY AND HEAVY METAL CONCENTRATIONS IN VARIOUS HONEY SAMPLES

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Abstract. In this study, the contents of natural radionuclide ^{40}K and fission product ^{137}Cs in floral honey samples from Rize and some other cities in Turkey, were investigated. The results indicated that 80% of honey samples were contaminated by ^{137}Cs , but in 50% of contaminated samples the content was lower than 10 Bq/kg. The found activities of ^{137}Cs in honey samples did not exceed the highest permitted level of the radionuclide for food and only slightly contributed to the natural radioactivity of honey. In addition, the contents of heavy metals like cadmium, chrome, iron, lead, zinc, copper, nickel, cobalt and manganese in honey samples were analysed using ICP-OES. Consequently, these results indicate that mean intake of heavy metals due to daily honey consumption is generally tolerable and fairly satisfactory.

Keywords: honey, radioactivity, heavy metal, high-purity germanium (HPGe) detector.

AIMS AND BACKGROUND

Radioactivity, found naturally in our living environment which is actually stemmed from certain industrial activities or human errors, has caused negative effects to human health in recent years. Especially, high levels of radioactivity results in very harmful consequences to human body¹. Humans are exposed to external radiation from cosmic rays and terrestrial radiation, while ingestion and inhalation of natural radioisotopes result in exposure to internal radiation. Naturally occurring radioisotopes are the main source of both external and internal radiation exposure in humans. Terrestrial radioisotopes enter the human body primarily by ingestion of foods, while inhalation of these isotopes is limited².

^{137}Cs is an artificial radionuclide and has a half-life of 30 years. It emits gamma and beta radiations and mainly contaminates leaves and flowers directly from the atmosphere through a fine layer of dust but also through absorption from soil. Organisms process it in a very similar way to potassium³.

Bee honey is a very important food and it can be a good source of major and trace elements needed by humans, but if they exceed safety levels, they can be toxic⁴. Also, it is used as a biomonitor to determine the quality in the environments

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which are polluted by heavy metals, radioactivity and pesticides⁵⁻⁷. Food safety is a major public concern worldwide. During the last decades, the increasing demand for food safety has stimulated research regarding the risk associated with consumption of foodstuff contaminated by pesticides, heavy metals and toxins⁸.

Heavy metals are one of the major contaminants of the food supplies and may be considered the most important problem to our environment⁹. This problem is becoming more serious all over the world, especially in developing countries. Heavy metals, in general, are not biodegradable, have long biological half-lives and have the potential for accumulation in various body organs, leading to undesired side effects¹⁰⁻¹³.

Keeping in view the potential toxicity, persistent nature and cumulative behaviour as well as the consumption of honey, it is necessary to test and analyse to ensure that the levels of these contaminants meet the agreed international requirements. Regular survey and monitoring programs of heavy metal contents in foodstuffs have been carried out for decades in most developed countries¹⁴⁻¹⁹.

In recent studies, heavy metals such as Cd, Cu, Zn, Ni, Pb, Cr, Mn, Fe and Se in honey samples were investigated from Central Anatolia using Plasma Optical Emission Spectrophotometry (ICP-OES) (Ref. 20). Also in Kahramanmaraş city of Turkey, heavy metal concentrations of honey samples had been studied previously²¹.

In Czech Republic the maximum activity of ¹³⁷Cs was found to be 39.16 Bq/kg in all types of honey and the mean activity concentration of ⁴⁰K in honey was found as 91.45 Bq/kg, the highest concentration in honeydew honey was 132.02 Bq/kg and the lowest average activity of radiopotassium in floral honey was 23.01 Bq/kg (Ref. 22).

In this study the main purpose was to measure the radiocaesium (¹³⁷Cs), radiopotassium (⁴⁰K) and heavy metals like nickel, chrome, lead, zinc, copper, iron, manganese, cadmium, cobalt in honey samples which are produced in different locations of Turkey.

EXPERIMENTAL

Honey sampling method. 20 samples of honey of each province were collected from Rize, Erzurum, Ankara and Artvin for radioactivity and heavy metal measurements. Samples were put in cylindrical plastic containers and hermetically sealed.

The radiation levels of samples were analysed using gamma spectrometry which was equipped with a 55% efficiency high-purity germanium (HPGe) detector and a multichannel analyser. Efficiency of the detector was determined with a ¹⁵²Eu source with known activity. ¹⁵²Eu sources have been widely used for calibration and efficiency determination because of their large range of energies (122, 244, 344, 411, 443, 779, 964, 1112 and 1408 keV) with emission probabilities of 3–29% (Refs 23 and 24).

Honey samples preparation. For heavy metal measurements, honey samples were dried in oven to evaporate any remaining water. The samples which had been converted to ash were dissolved in nitric acid (HNO₃). All the samples were centrifuged and then made up to volume with %1 HNO₃ to 25 ml (Ref. 19). Then the samples were analysed with Perkin Elmer Optima 7000 DV ICP-OES device.

RESULTS AND DISCUSSION

ACTIVITY CONCENTRATIONS OF RADIONUCLIDES

⁴⁰K and ¹³⁷Cs activity concentrations in analysed samples of honey are given in Table 1.

Table 1. ⁴⁰K and ¹³⁷Cs activities of honey samples

| Radionuclide | Range of activities (Bq/kg) | Mean±SEM (Bq/kg) |
|-------------------|-----------------------------|------------------|
| ⁴⁰ K | MSA – 168.95±6.77 | 68.48±4.07 |
| ¹³⁷ Cs | MSA – 105.09±0.98 | 18.2±0.45 |

MSA=0.09±0.002 for ⁴⁰K and MSA=0 for ¹³⁷Cs; SEM – standard error of the mean.

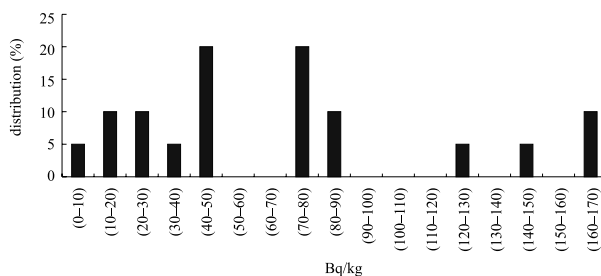


Fig. 1. Distribution of ⁴⁰K activity concentration in honey

The ⁴⁰K activity concentrations ranged between minimum significant activity (MSA) to 168.95 Bq/kg and it is higher than published results. The distribution of ⁴⁰K activity concentrations in honey samples is given in Fig. 1. The highest ⁴⁰K activity was 168.95±6.77 Bq/kg in all honey samples. In Czech Republic, the highest activity concentration ⁴⁰K was found as 132.02 Bq/kg in honey²². Natural radiopotassium (⁴⁰K) gets into the human organism via water and foodstuffs, where its specific activity ranges between 20 and 240 Bq/kg (Ref. 25). The average human (70 kg) comprises of 0.1–0.3 % of potassium, which represents an activity of 22–76 kBq of ⁴⁰K and yearly average dose from internal exposition of ionising radiation of 200 μSv. Therefore, the detected average activities of radiopotassium in honey (59.70 ± 3.94 Bq/kg), at a current consumption of honeybee honey, would present only a minor hazard to the inner irradiation of man.

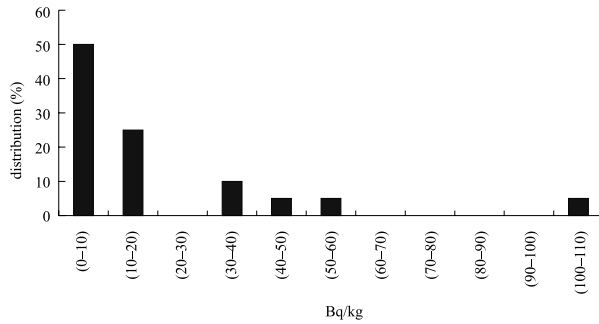


Fig. 2. Distribution of ¹³⁷Cs activity concentration in honey samples

The distribution of ¹³⁷Cs activity concentrations in honey samples is given in Fig. 2. The ¹³⁷Cs activity concentrations ranged from MSA to 105.09±0.98 Bq/kg in all honey samples. The average ¹³⁷Cs value of all honey samples was 18.20±0.45 Bq/kg. In Czech Republic, the highest activity concentration of ¹³⁷Cs was found as 39.16 Bq/kg in all types of honey²². Compared to the published data the average content of ¹³⁷Cs in honey was higher with results in the same type of honey in Czech Republic.

That isotope ¹³⁷Cs, being one of the anthropogenic radionuclides, is produced by several types of nuclear activity, including recent testing of nuclear weapons, accidents in nuclear facilities, reprocessing of used nuclear fuel and nuclear power reactors. After the Chernobyl event on 26 April 1986 the radioactive plume from the accident area reached Turkey on 5 May 1986, and substantially contaminated various regions including the Black Sea region. The main reason of high ¹³⁷Cs activities in the honey samples is that 80% of honey samples were taken from the Black Sea region of Turkey.

HEAVY METAL CONCENTRATIONS

Average heavy metal concentrations in honey samples are given with their standard deviations in Table 2.

Table 2. Metal concentrations of honey samples (µg/l)

| Metals | Mean±SEM | Min±SEM | Max±SEM |
|--------|--------------|--------------|--------------|
| Cr | 0.007±0.0004 | 0.004±0.0003 | 0.029±0.0003 |
| Mn | 0.603±0.0084 | 0.023±0.0000 | 3.898±0.0136 |
| Fe | 0.424±0.0026 | 0.160±0.0008 | 0.693±0.0024 |
| Cu | 0.064±0.0086 | 0.044±0.0004 | 0.116±0.0007 |
| Zn | 3.976±0.0416 | 0.771±0.0004 | 9.576±0.0985 |
| Cd | 0.011±0.0002 | 0.008±0.0003 | 0.016±0.0002 |
| Pb | 0.078±0.0036 | 0.036±0.0038 | 0.185±0.0065 |
| Co | 0.010±0.0004 | 0.009±0.0001 | 0.013±0.0005 |
| Ni | 0.041±0.0014 | 0.018±0.0016 | 0.065±0.0013 |

The average Cr concentrations were 0.007 µg/l and it is lower than previous studies. Reported studies showed that chrome values between 0.01 and 1.07 µg/g (Refs 20 and 21).

The average Mn concentrations found in that study were 0.603 µg/l. Some reported manganese values in the literature for honey were 0.32–1.70 µg/g and 0.02–1.56 µg/g (Refs 20 and 26). Our samples are at the same levels as these studies.

The highest Fe concentration was found 0.693 µg/l. According to the Codex alimentarius commission the maximum iron value must be found in honey is reported as 15 µg/g (Ref. 27). Our results are lower than this value.

The highest Cu concentration was found 0.116 µg/l in honey samples. Copper values in the literature have been reported as 0.51–1.30 and 0.01–0.80 µg/g (Refs 20, 26 and 28).

The highest Zn concentration was found 9.576 µg/l. Zinc values in literature have been reported as 2.2–11.0 µg/g and 0.15–5.39 µg/g (Refs 20 and 21).

The average Pb concentration in honey samples was found 0.078 µg/l. The values for Pb contents in our samples are at the same level as literature values (Refs 20, 28 and 29).

The average Cd concentration was found 0.11 µg/l in honey samples. Cadmium values in literature have been reported as 0.09–0.24 µg/g and 0.001–0.10 µg/g (Refs 20, 28, 30 and 31).

The Ni concentration was found between 0.018 and 0.065 µg/l. Nickel values in the literature have been reported as 0.03–1.44 µg/g and 0.2–0.8 µg/g, respectively (Refs 20 and 21).

CONCLUSIONS

Protection of human health is the first priority in scientific research into pollution in food categories. The observed ¹³⁷Cs activities proved that the Chernobyl radiocaesium is still present in nature and transported by bees from the environment into honey. However, the results of the present study indicate that ¹³⁷Cs and ⁴⁰K activities in addition to concentrations of Cr, Mn, Fe, Cu, Zn, Cd, Pb, Co, Ni obtained from the samples were far below the limits established by FAO/WHO, and present no risk to public health.

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