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Risk assessment of heavy metals in honey consumed in Zhejiang province, southeastern China

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ABSTRACT

The levels of copper (Cu), zinc (Zn), cadmium (Cd), lead (Pb), arsenic (As), and mercury (Hg) in eight types of honey collected in China were determined. The average concentrations of the six heavy metals in the honey samples were 46.18, 1329.5, 1.34, 33.98, 13.44, and 1.65 μg kg^{-1}, respectively. All these values were below the maximum allowable contaminant levels in foods (GB2762-2005) and honey (GB14963-2011) in China. The hazard quotients of individual heavy metals and the hazard index of all six heavy metals were far below one, indicating no chronic-toxic risk from these metals for the inhabitants of Zhejiang under the current consumption rates of honey. However, the carcinogenic risk of As for the female inhabitants in Zhejiang exceeded the acceptable level of 10^{-4}. Therefore, As is the most concerning heavy metal in honey.

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1. Introduction

Honey has antioxidant, antibacterial, immunity-enhancing, and other physiological activities. Honey is recognized as a food with nutritional properties and a natural product with valuable therapeutic applications (Bilandžić et al., 2011). However, the presence of metals in honey may threaten the health of human consumers. These metals may come from external sources such as industrial smelter pollution, factory emissions, non-ferrous metallurgy, leaded petrol from busy highways, incorrect procedures during honey processing and conservation phases, as well as agrochemicals such as cadmium-containing fertilizers, organic mercury and arsenic-based pesticides still used in some countries (Pisani et al., 2008; Wang et al., 2010).

These metals can lead to quality impairment of human life when they accumulate to a toxic concentration level, which can be classified as essential and potentially toxic (Munoz-Olivas and Camara, 2001). Essential elements are safe and adequate for the body within a specific range of intake. Beyond this range, toxic effects are observed. Potentially toxic elements can be very toxic even at low concentrations. Dietary intake is considered to be the major route of human exposure to toxic elements, although air inhalation is also another dominant route, particularly in some developing countries. Guidelines for the intake of heavy metals by humans have been provided by several agencies and organizations, such as the Institute of Medicine of the National Academies, US Environmental Protection Agency (US EPA), as well as the Joint Food and Agriculture Organization/World Health Organization Expert Committee on Food Additives (JECFA). These guidelines list the safe intake levels that based on available scientific evidence (COT-Committee on Toxicity, 2004).

In recent years, the concentrations of different metals in honey have been determined in some European countries, such as Croatia (Bilandžić et al., 2011), France (Devillers et al., 2002), Italy (Pisani et al., 2008), Poland (Przybyłowski and Wilczyńska, 2001), Slovenia (Golob et al., 2005), and Turkey (Citak et al., 2012; Silici et al., 2008; Tuzen et al., 2007; Tuzen and Soylak, 2005). Most of these studies focused on the investigation of heavy metals in honey as a very important indicator of environmental pollution. China is a major producer and exporter of honey. Heavy metal pollution has become serious in China with the development of the mining, smelting, and metal treatment industries over the last few decades. Heavy metal pollution affects the production and quality of crops, as well as the qualities of the atmosphere and water bodies, thereby threatening the health and life of animals and human beings via the food chain. For the safe consumption of honey, the presence of heavy metals in honey and the associated health risks need to be evaluated. A few researchers have determined the metal contents of honey (Wang et al., 2011), but no risk assessment of heavy metals has been reported in China. The present study aimed to determine the concentrations of copper (Cu), zinc (Zn), cadmium (Cd), lead (Pb), arsenic (As), and mercury (Hg) in honey collected in China, as well as to examine the potential human health risks to honey consumers in Zhejiang Province, China.

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2. Materials and methods

2.1. Materials and reagents

A total of 48 samples of seven types of unifloral (acacia, linden, citrus, litchi, loquat, jujube, and yellow box) and multifloral honeys were provided by Tonglu Product Quality and Measurement Monitoring Center, Zhejiang province.

Analytical-reagent-grade chemicals were used in this study. Nitric acid (HNO_3) and hydrogen peroxide (H_2O_2) were of supra-pure quality (E. Merck, Darmstadt, Germany). High-purity water (18.2 $\Omega\,cm^{-1}$ resistivity) from a Milli-Q system (Millipore, Milford, MA, USA) was used throughout the study. Prior to use, polytetrafluoroethylene (PTFE) vessels and glassware were cleaned by soaking in diluted HNO_3 (10%, v/v) overnight and subsequent rinsing with high-purity water before drying. Cu, Zn, Cd, Pb, As, and Hg standard stock solutions were purchased from the National Standard Substance Research Center (NSSRC), China. The standard solutions used for the calibration procedures were prepared by diluting the stock solution with 1% (v/v) HNO_3.

2.2. Sample preparation

Samples (0.5 g) were digested with 3 mL of HNO_3 (65%, v/v) and 4 mL of H_2O_2 (30%, v/v) in PTFE vessels placed in a Multiwave 3000 microwave closed system (Anton Paar, Germany). A blank digest was similarly prepared. The digestion program began at a power of 500 W ramped for 1 min and held for 4 min. The second step began at a power of 1000 W ramped for 5 min and held for 5 min. The third step began at a power of 1400 W ramped for 5 min and held for 10 min. The digested samples were diluted to a final volume of 50 mL with high-purity water. For total inorganic As determination, a 5 mL aliquot of digested sample was injected to a 10 mL sample tube. To this tube, 0.5 mL of concentrated hydrochloric acid (HCl; 37%, v/v), 1 mL of 5% (w/v) thiourea, and 1 mL of 1% (w/v) ascorbic acid were added and the mixture was diluted to 10 mL. A parallel sample was prepared for each digested sample.

2.3. Determination of heavy metals

The Cu and Zn concentrations were determined using a flame atomic absorption spectrophotometry (FAAS) system (Varian 240FS, Agilent, USA). The Cd and Pb concentrations were determined using a graphite furnace atomic absorption spectrometry (GF-AAS) system equipped with an AAnalyst 800 atomic absorption spectrometer (PerkinElmer, USA). For graphite furnace measurements, argon was used as the inert gas. Pyrolytic-coated graphite tubes with a platform were used. The matrix modifier used to determine both metals was a mixture of 0.5% (w/v) ammonium dihydrogen phosphate ($H_2PO_4NH_4$) and 1% (v/v) HNO_3; 5 μL of matrix modifier was added if necessary. Most of the matrix was removed before the atomization step and less interference occurred during atomization. The As and Hg concentrations were determined by an AFS-970 dual-channel atomic fluorescence spectrometer (Haiguang Instrument Comp., Beijing, China). HCl (5%, v/v) was used as the carrier liquid, and 2% (w/v) potassium borohydride ($NaBH_4$) in 0.5% (w/v) sodium hydroxide (NaOH) was used as the reducing agent. The instrumental settings and optimal programs of FAAS, GF-AAS, and hydride generation-atomic fluorescence spectrometry (HG-AFS) are summarized in Table 1. The instrument was calibrated and standardized with different working standards. After ensuring that the instrument was properly calibrated and the results of the standards were within the confidence limit, the concentrations individual metals in each sample were measured. Triplicate analyses were performed for each sample. Blank and drift standards were run after 20 determinations to maintain instrument calibration. All heavy metal concentrations were determined on a natural weight basis and expressed in microgram per kilogram.

2.4. Quality assurance and control

Appropriate quality assurance procedures and precautions were carried out to ensure the reliability of the results. The standard reference material, chicken powder (GBW10018), was obtained from the NSSRC and digested with the samples to validate the analytical procedures. To calculate the recovery, we processed 10 honey samples that had been spiked with known amounts of Cu, Zn, Cd, Pb, As, and Hg analytical standards. The relative standard deviation (RSD) was calculated from repeated determination ($n = 7$). The method detection limit (MDL) was defined as the concentration of each element corresponding to three times the standard deviation of the digestion blanks ($n = 11$).

Table 2 summarizes the quality assurance and control results. The quality of data was checked by the recovery rate analysis of honey samples spiked with Cu, Zn, Cd, Pb, As, and Hg. High accuracy was found, with metal recovery rates of 97.42–104.54%. The RSDs were always less than 2%, suggesting high precision. Using FAAS, the MDL values were found to be 19.0 $\mu g\,kg^{-1}$ for Cu and 41.0 $\mu g\,kg^{-1}$ for Zn. Using GF-AAS, the MDL values were found to be 0.8 $\mu g\,kg^{-1}$ for Cd and 3.0 $\mu g\,kg^{-1}$ for Pb. Using HG-AFS, the MDL values were found to be 0.15 $\mu g\,kg^{-1}$ for Hg and 1.8 $\mu g\,kg^{-1}$ for As.

2.5. Health risk assessment

2.5.1. Deterministic estimation of health risks

In this study, the human health risks posed by chronic exposure to the heavy metals were assessed. The hazard quotient (HQ) was calculated to estimate the chronic-toxic risks posed by individual metals via honey consumption. The average daily exposure to heavy metals and HQs were estimated by the following equations (US EPA, 1992, 1999):

$$HQ = \frac{ADD}{RfD} \quad (1)$$

$$ADD = \frac{C \times IR}{BW} \quad (2)$$

where ADD is the average daily metal intake ($\mu g\,kg^{-1}\,day^{-1}$), RfD is the daily intake reference dose ($\mu g\,kg^{-1}\,day^{-1}$) suggested by the US EPA or derived from the provisional tolerable weekly intake (PTWI) given by the WHO, C is the mean heavy metal concentration in honey ($\mu g\,kg^{-1}$), IR is the honey consumption rate (kg person^{-1} day^{-1}), and BW is the average body weight (kg).

A questionnaire-based survey on daily honey intake, in reference to the dietary recall method and food frequency method that have been generally adopted by the food consumption survey, was conducted in Zhejiang province, China in May 2012. A total of 621 adult, gender-balanced participants were randomly recruited from 11 cities of Zhejiang. They were asked to recall the quantity and frequency of their honey consumption. For higher accuracy, the participants were also asked to show the spoon they usually used.

The hazard index (HI) was used to estimate total chronic-toxic risks of multiple metals on the assumption of dose additivity (US EPA, 1999):

$$HI = HQ_1 + HQ_2 + \ldots + HQ_n \quad (3)$$

Table 1
Instrumental analytical conditions for Cu, Zn, Cd, Pb, As, and Hg determination in honey samples.

Heavy metals	Analytical technique	Instrumental analytical conditions				
	FAAS	Wavelength (nm)	Lamp current (mA)	Slit width (nm)	Acetylene flow (L min^{-1})	Air flow (L min^{-1})
Cu		324.8	3.0	0.5	2.0	13.5
Zn		213.9	5.0	1.0	2.0	13.5

	GF-AAS	Wavelength (nm)	Lamp current (mA)	Slit width (nm)	Sample volume (μL)	Heating program temperature (°C) (ramp, time (s) hold time (s))			
						Drying	Ashing	Atomization	Cleaning
Cd		228.8	4.0	0.5	15	120 (5, 30)	350 (10, 20)	1750 (0, 5)	2400 (1, 5)
Pb		283.3	10.0	0.5	15	120 (5, 30)	450 (10, 20)	1800 (0, 5)	2400 (1, 5)

	HG-AAS	Negative high voltage (V)	Lamp current (mA)	Atomizer height (mm)	Carrier gas flow (mL min^{-1})	Shielding gas flow (mL min^{-1})
As		300	50	8	500	900
Hg		280	30	8	400	900

Note: FAAS, flame atomic absorption spectrophotometry; GF-AAS, graphite furnace atomic absorption spectrometry; HG-AAS, hydride generation-atomic fluorescence spectrometry.

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