



Health risk assessment of eight heavy metals in nine varieties of edible vegetable oils consumed in China

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ARTICLE INFO

Article history:

Available online 22 September 2011

Keywords:

Edible oils
Heavy metals
Microwave digestion
Risk assessment
China

ABSTRACT

Eight heavy metals, namely Cu, Zn, Fe, Mn, Cd, Ni, Pb and As, in nine varieties of edible vegetable oils collected from China were determined by inductively coupled plasma atomic emission spectrometry (ICP-AES) and graphite furnace atomic absorption spectrometry (GF-AAS) after microwave digestion. The accuracy of procedure was confirmed by certified reference materials (GBW10018 and GBW08551). The relative standard deviations were found below 10%. The concentrations for copper, zinc, iron, manganese, nickel, lead and arsenic were observed in the range of 0.214–0.875, 0.742–2.56, 16.2–45.3, 0.113–0.556, 0.026–0.075, 0.009–0.018 and 0.009–0.019 $\mu\text{g g}^{-1}$, respectively. Cadmium was found to be 2.64–8.43 $\mu\text{g/kg}$. In general, iron content was higher than other metals in the investigated edible vegetable oils. Comparing with safety intake levels for these heavy metals recommended by Institute of Medicine of the National Academies (IOM), US Environmental Protection Agency (US EPA) and Joint FAO/WHO Expert Committee on Food Additives (JECFA), the dietary intakes of the eight heavy metals from weekly consumption of 175 g of edible vegetable oils or daily consumption 25 g of edible vegetable oils for a 70 kg individual should pose no risk to human health.

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1. Introduction

Vegetable oils are widely used in cooking and alimentary, cosmetic, pharmaceutical and chemical industries (Dugo et al., 2004). Vegetable oils are beneficial and popular due to their cholesterol-lowering effect. In contrast to animal fats, which are predominantly saturated and hence do not react readily with other chemicals, especially oxygen, unsaturated vegetable oils are more reactive (Mendil et al., 2009).

The quality of edible oils regarding their freshness, storability and toxicity can be evaluated by the determination of several trace metals. Levels of trace metals like Cu, Zn, Fe, Mn and Ni are known to increase the rate of oil oxidation while other elements such as As, Cd and Pb are very important on account of their toxicity and metabolic role (Anthemidis et al., 2005). The presence of metals in vegetable oils depends on several factors. They might come from the soil, environment, genotype of the plant, fertilisers and/or metal-containing pesticides, introduced during the production process or by contamination from the metal processing equipment (Zeiner et al., 2005; Jamali et al., 2008).

Heavy metals can be classified as potentially toxic (arsenic, cadmium, lead, etc.), probably essential (vanadium, cobalt) and essential (copper, zinc, iron, manganese, etc.). Toxic elements can

be very harmful even at low concentration when ingested over a long time period (Unak et al., 2007). The essential metals can also produce toxic effects when the metal intake is excessively elevated (Gopalani et al., 2007). It is necessary to assess the levels of heavy metals in edible vegetable oils and to report possible contamination that would represent a health hazard. A few researchers have made some progress on the determination of heavy metals in edible oils (Cindiric et al., 2007; Pehlivan et al., 2008; Mendil et al., 2009). Food consumption had been identified as the major pathway of human exposure to toxic metals, compared with other ways of exposure such as inhalation and dermal contact. US Environmental Protection Agency (US EPA), Joint FAO/WHO Expert Committee on Food Additives (JECFA) and Institute of Medicine of the National Academies (IOM) have provided guidelines on the intake of trace elements by humans. The IOM of the National Academies recommended the adequate intake (AI) and the tolerable upper intake level (UL) values for some essential elements (IOM, 2002, 2003; the JECFA recommended permissible tolerable weekly intakes (PTWIs) and acceptable daily intakes as guidelines for food additives and certain contaminants in foods (JECFA, 2003). Also, the US EPA provided reference dose (RfDo) values in $\mu\text{g/kg}$ body wt/day for some elements (US EPA, 2007a).

China is one of the largest producers and consumers of edible vegetable oils in the world. In the year of 2010, the total edible vegetable oil consumption in China reached 24.75 million tons. However, there has been no report, to our knowledge, on the heavy metal levels in

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edible vegetable oils in China. The main objective of this study were to determine the concentrations of copper, zinc iron, manganese, cadmium, nickel, lead and arsenic in edible vegetable oils consumed in China and estimate the potential human health risks from weekly consumption of 175 g of edible vegetable oils or daily consumption 25 g of edible vegetable oils for a 70 kg individual.

2. Materials and methods

2.1. Sample collection

Total 109 samples of edible vegetable oils (13 soybean oil, 12 corn oil, 14 peanut oil, 12 sesame oil, 11 rapeseed oil, 12 cottonseed oil, 12 olive oil, 11 blend oil and 12 sunflower oil samples) were purchased in Chinese supermarkets during 2009 and 2010. The most widely accepted and most frequently consumed several brands such as Kinlongyu, Fortune and Luhua were selected. The collected oil samples were packed in polyethylene bags and stored below -20°C until analysis.

2.2. Reagents and apparatus

All reagents were of analytical reagent grade unless otherwise stated. HNO_3 and H_2O_2 were of suprapure quality (E. Merck, Germany). Double deionised water (Milli-Q Millipore 18.2 $\text{M}\Omega\text{-cm}$ resistivity) was used for all dilutions. The element standard solutions were prepared by dilution of 1000 mg/l certified standard solutions (Nssrc, China).

XT-9912 model microwave system (Xintuo, China) equipped with advanced composite PTFE vessels was used for digestion of the samples. A Perkin Elmer Analyst 700 (CT, USA) atomic absorption spectrometer equipped with HGA graphite furnace (GF-AAS) was used to determined Cd, Ni, Pb and As. For graphite furnace measurements, argon was used as inert gas. Pyrolytic-coated graphite tubes (Perkin Elmer part No. B3 001264) with a platform were used. Samples were injected into the graphite furnace using Perkin Elmer AS-800 autosampler. Optima 2100 DV model ICP-AES (PE, USA) was used for simultaneous multielement detection of Cu, Zn, Fe and Mn. The operating conditions for GF-AAS for the Cd, Ni, Pb and As analysis were as reported in a previous experiment (Tuzen, 2009), the operating conditions of ICP-AES were reported in Table 1.

2.3. Microwave digestion

One gram samples were digested with 6 ml of concentrated HNO_3 (65%) and 2 ml of concentrated H_2O_2 (30%) in microwave digestion system for 32 min and finally diluted to 10 ml with 2% HNO_3 . All sample solutions were clear. A blank digest was carried out in the same way. Digestion conditions for microwave system were applied as 3 min for 500 W, 5 min for 800 W, 8 min for 1000 W, 10 min for 1300 W, vent: 8 min, respectively.

2.4. Quality assurance

Appropriate quality assurance procedures and precautions were carried out to ensure the reliability of the results. During the experiments, all glasswares and equipments were carefully cleaned starting with 2% HNO_3 and ending with repeated rinsing distilled deionised water to prevent contamination. Reagent blank determinations were used to correct the instrument readings. The accuracy and precision of the analytical method was calculated by analyzing the certified reference materials (GBW10018 and GBW08551). Detection limit is defined as the concentration corresponding to three times the standard deviation of ten blanks.

2.5. Statistical analysis

Statistical analysis of data was carried out using SPSS 17.0 statistical package program. Two-way ANOVA was employed to find the significant differences of heavy metal concentrations in nine varieties of edible vegetable oils with regard to oil types and oil sources (several brands such as Kinlongyu, Fortune, Luhua and Longda). The significance was set at 0.05.

Table 1
The operating conditions of ICP-AES.

RF power (W)	1200
Gas flow rate (ml/min)	
Auxiliary gas	0.6
Coolant gas	18
Nebuliser gas	0.8
Sample up take (ml/min)	1.5
Torch	Axial
Elements monitored (wavelength, nm)	Cu (327.4), Zn (206.2), Fe (238.2), Mn (257.6)

3. Results and discussion

Detection limit values of elements as milligram per liter were found to be 0.018 for Cu, 0.010 for Zn, 0.016 for Fe, 0.020 for Mn, 0.09 for Cd, 0.35 for Ni, 0.18 for Pb and 0.17 for As in this study. The recovery values were nearly quantitative ($\geq 95\%$) for microwave digestion method. The relative standard deviations were less than 10% for all investigated elements. In order to validate the method for accuracy and precision, certified reference materials, namely chicken (GBW10018) and porcine liver (GBW08551) were analyzed for corresponding elements. As shown in Table 2, the achieved results were in good agreement with certified values.

The results of two-way ANOVA, concentration ranges and averages with standard deviations of heavy metals in the analyzed nine edible vegetable oils were given in Table 3. Among all determined heavy metals, iron was found to be the dominant elemental ion followed by zinc and copper. The results of two-way ANOVA showed the oil types influenced significantly the levels of all heavy metals in the edible oils except nickel. However, sources (oil brands) and interaction of types $\times$ sources did not show any significant effect on the levels of all heavy metals.

Copper is essential for good health but very high intake can cause adverse health problems such as liver and kidney damage (Ikem and Egiebor, 2005). Copper deficiency leads to hypochromic anemia, leucopenia and osteoporosis in children (Kanumakala et al., 2002). Minimum and maximum values of copper in our samples were 0.021 and 0.265 $\mu\text{g g}^{-1}$ in corn oil and olive oil. In the literature copper levels in edible oil samples have been reported in the range of 0.02–0.33 $\mu\text{g g}^{-1}$ (Garrido et al., 1994), 12.71–50.5 $\mu\text{g g}^{-1}$ (Buldini et al., 1997), 21.0–31.0 $\mu\text{g g}^{-1}$ (Ajayi et al., 2006), 0.0184–0.2870 $\mu\text{g g}^{-1}$ (Pehlivan et al., 2008) and 0–130 ng g^{-1} (Llorent-Martínez et al., 2011a, b). Our copper values in the investigated oil samples were in agreement with those reported in the literatures (Zeiner et al., 2005; Cindiric et al., 2007; Mendil et al., 2009). According to national and international requirements, the approved contents of these metals in oils are: 1–1.5 $\mu\text{g g}^{-1}$ (Fe), 0.2 $\mu\text{g g}^{-1}$ (Ni), 0.1 $\mu\text{g g}^{-1}$ (Cu, Pb, As) and 0.05 $\mu\text{g g}^{-1}$ (Cd) (Kowalewska et al., 2005). Copper levels in peanut oil, rapeseed oil and olive oil were found to be higher than the recommended legal limits. The FAO/WHO has set a limit for heavy metals intakes based on body weight. For an average adult (60 kg body weight), the provisional tolerable daily intake (PTDI) for copper, zinc, iron and lead are 3 mg, 60 mg, 48 mg and 214 $\mu\text{g g}^{-1}$, respectively (FAO/WHO 1999).

The zinc content of the samples ranged from 0.742 to 2.56 $\mu\text{g g}^{-1}$, soybean oil had the lowest copper concentration whereas corn oil had the highest. Zinc is known to be involved in most metabolic pathways in humans and zinc deficiency can lead to loss of appetite, growth retardation, skin changes and immunological abnormalities. The zinc levels in the oil samples were lower than those in previous reports (Cindiric et al., 2007; Mendil et al., 2009), but the levels were higher than these literature values 0.04–0.70 $\mu\text{g g}^{-1}$ (Garrido et al., 1994) and 0.0484–0.2870 mg kg^{-1} (Pehlivan et al., 2008).

The minimum and maximum iron levels observed were 16.2 $\mu\text{g g}^{-1}$ in corn oil and 45.3 $\mu\text{g g}^{-1}$ in peanut oil. The reported iron values for oil samples were 0.22–220 $\mu\text{g g}^{-1}$ (Garrido et al., 1994), 52.0–291.0 $\mu\text{g g}^{-1}$ (Mendil et al., 2009), 0.0039–0.0352 mg kg^{-1} (Pehlivan et al., 2008), 0–800 ng g^{-1} (Llorent-Martínez et al., 2011a, b), respectively. Our iron levels were in agreement with those reported in the literature (Cindiric et al., 2007). It is known that adequate iron in a diet is very important for decreasing the incidence of anemia (Ashraf and Mian, 2008). Fe deficiency is frequently associated with anemia and, thus, reduces working capacity and impaired intellectual

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