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Health risk assessment of heavy metals in freshwater fish in the central and eastern North China

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ABSTRACT

The distribution and potential health risks of eight heavy metals (Copper (Cu), Chromium (Cr), Zinc (Zn), Lead (Pb), Arsenic (As), Cadmium (Cd), Manganese (Mn), Nickel (Ni)) in 16 freshwater systems from central and eastern North China, were investigated. The fish were divided as wild fish, which grew naturally without artificially feeding, and farmed fish. The total concentrations of the eight heavy metals ranged from 82.9 to 226 μg/L in the surface water samples and 3.32–27.6 mg/kg dw in the fish samples. There was no significant difference in the heavy metal concentrations between natural and farmed water systems. The concentrations of toxic metals, including Pb, As, Cd, Cr, are similar in all kinds of fish. However, the essential metals (Zn, Cu, Mn, Ni) in crucian carp (15.9 mg/kg) was much higher than other kinds of fish. Comparing the wild and farmed fish, the average concentrations of each heavy metal in wild crucian carp, bighead carp, grass carp were higher than those in farmed fish. The average log BCFs (bioconcentration factor) of Zn, Cr and Cu were the highest (2.14, 2.04, 2.00 L/kg) while that of Cd (0.65 L/kg) was the lowest. The non-carcinogenic and carcinogenic health risks to adults and children resulting from consuming the fish were assessed based on the target hazard quotients (THQ). The results indicated that the non-carcinogenic health risk to humans by consuming fish products, no matter wild or farmed fish, was relatively low. The carcinogenic risk of inorganic As was 5.11×10^{-6}–1.95×10^{-4} for children and 2.71×10^{-6}–1.04×10^{-4} for adult, which are within the acceptable range. The results indicated that the concentrations of heavy metals in the freshwater fish in central and eastern North China were relatively low, and did not cause considerable human health risks.

1. Introduction

Aquatic products serve as an excellent source of protein, omega-3 fatty acids, vitamins, selenium and calcium, and are increasingly consumed by humans (Kalantzi et al., 2016; Bosch et al., 2016). The American Heart Association recommends two servings of fish per week as part of a healthy diet (Neff et al., 2014). Despite the positive health effects of aquatic products, they may accumulate contaminants due to the high contents of fat and proteins, which display harmful effects on human health (Usydus et al., 2009).

Heavy metals are one of the most significant pollutants in aquatic ecosystems due to their toxicities, persistence and bioaccumulation potentials. It is well known that heavy metals pose significant health risks when human exposure dose exceeds safe consumption levels (Djedjibegovic et al., 2012; Tao et al., 2012; Ahmed et al., 2015; Bosch et al., 2016; Saha et al., 2016). For example, previous studies demonstrated that Arsenic (As), Cadmium (Cd), Lead (Pb) and Chromium (VI) (Cr^{6+}) have harmful effects on human health even at low concentrations (Makedonski et al., 2017; Ahmed et al., 2015). As could cause central and peripheral nervous system damage, cardiovascular disease, birth defects, placental development disorders and other reproductive problems (Navas Acien et al., 2005; Jomova et al., 2011; Mandeep et al., 2015). Cd is associated with hepatic, skeletal, renal and reproductive effects (Shaheen et al., 2016; Bosch et al., 2016). Pb may affect nervous system, disrupt skeletal hematopoietic function, digestive and male reproductive systems (Levin and Goldberg, 2000; Needleman, 2004; Diamond, 2005). Cr^{6+} is carcinogenic for humans (Varol et al., 2017). For other heavy metals, such as copper (Cu), Zinc (Zn), Manganese (Mn), Nickel (Ni), they are classified as essential or probably essential metals, but they may also cause toxicity effects when the intake exceeds the safe consumption levels. High intake of Zn, Mn, Ni could cause neurotoxicity to human and Cu is associated with immunotoxicity and developmental toxicity (ATSDR, 2005). Thus, many concerns have been raised on the potential risks of human health associated with the consumption of aquatic products containing heavy metals.

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Freshwater fish is an important source of proteins for inland residents. However, the production amount of wild fish which growth in natural water system without feeding artificially can not meet the consumption demand in recent decades. Thus, the aquaculture industry has grown rapidly in China (Wang et al., 2005). According to the Food and Agriculture Organization (FAO), China is the largest producer of aquatic products in the world, accounting for approximately 60% of global aquatic production (FAO, 2005). Central and eastern North China is important region for aquaculture in China, providing a variety of aquatic products to the surrounding population (NBSC, 2010). Many consumers believe that wild fish can rich more nutrients because they obtain food naturally and grow much slower than the farmed ones, which are fed with abundant artificial feed accompanied with some additives and drugs (Nettleton and Exler, 2010). In recent years, great concerns have been raised from the government and general population on the health risks of the farmed fish, since many previous studies indicated that the surface water in central and eastern North China was heavily polluted by heavy metals (Chen et al., 2008; Liu et al., 2009, 2016; Li et al., 2010; Tang et al., 2013; Zhang et al., 2017). Therefore, the heavy metals in waters could be accumulated by aquatic products, and affect human health due to consumption of aquatic products (W. Li et al., 2015; Liu et al., 2016). Sparse information is available on the associated human health risks due to consumption of aquatic products contaminated with heavy metals.

This study aimed to assess human health risks of heavy metals in freshwater fish collected from central and eastern North China. Surface water, and fish, including wild and farmed fish, were collected at 16 sampling sites. The concentrations of eight heavy metals (Cu, Cr, Zn, Pb, As, Cd, Mn, Ni) were measured in the samples, and the non-carcinogenic and carcinogenic health risks to adults and children associated with consuming fish were calculated respectively.

2. Materials and methods

2.1. Sampling

Surface water and fish samples were collected from 16 freshwater sites in central and eastern North China during April to May 2016, including six natural water sites (W1-W6) and ten farmed water sites (F1-F10). Fish sampled in natural water sites were classified as wild fish, while which sampled in farmed water were classified as farmed fish. The detailed information for sampling was listed in Table 1. Two surface water samples were collected at each site using polyethylene bottles which were pre-washed with nitric acid (HNO_3). One sample from each site was filtered through a 0.45 μm polyether sulphone membrane, while the other was acidified to pH < 2 using concentrated HNO_3 (65–68%, AR grade, Tianjin chemical reagents Co., Ltd.). Fish samples were collected by fish net and transported alive to the laboratory using plastic packing box. Upon arriving at the laboratory, surface water samples were stored at 4 °C. The organisms were dissected immediately to obtain muscle (> 200 g) which were immediately frozen and stored at − 80 °C.

2.2. Sample pretreatment and analytical procedure

All unfiltered water samples and fish samples were treated using microwave digestion and analyzed by inductively coupled plasma mass spectrometry (ICP-MS).

To measure the total concentration of each metal in water, 45 mL of unfiltered water was shaken and placed in a microwave digestion tank, to which 4 mL of concentrated nitric acid (65–68%, AR grade, Tianjin chemical reagents Co., Ltd.) and 1 mL of hydrochloric acid (36%, AR grade, Tianjin chemical reagents Co., Ltd.) were added. Water samples were digested following the digestion procedure (the temperature was raised to 170 °C in 10 min and maintained for 10 min), and then analyzed by ICP-MS. To measure dissolved heavy metals in water, 15 mL of the water sample which was filtered through a 0.45 μm polyether sulphone filter was analyzed directly by ICP-MS.

After freeze-drying, 0.2–0.5 g of the organism muscles were accurately weighed and placed in a digestion tank, to which 10 mL of concentrated nitric acid and 2 mL of 30% hydrogen peroxide were added. The samples were digested following the digestion process, for which the temperature was raised to 175 °C over 10 min and maintained for 30 min.

Physico-chemical measurements of pH, dissolved oxygen (DO) were carried out following standard protocols of China (GB 6920, GB 7489). The total carbon (TC) and total organic carbon (TOC) were measured by TOC analyzer (multi N/C3100, Analytikjena Ltd.)

2.3. Quality assurance/quality control

To assess the reliability of the preparation methods, blank experiments, blank spike experiments, matrix spike experiments and parallel experiments (n = 3) were conducted. No heavy metal was detected in the blank samples. The recoveries of the eight heavy metals in the matrix spike samples ranged from 85.4% to 112% (RSD% from 4.42% to 7.63%) in the water samples, from 83.6% to 122% (RSD% from 11.2% to 13.9%) in biological samples. The limits of quantification of the eight heavy metals were 0.48 μg/L for As, 0.2 μg/L for Cd, 0.32 μg/L for Cu, 0.44 μg/L for Cr, 0.48 μg/L for Mn, 0.24 μg/L for Ni, 0.36 μg/L for Pb and 2.68 μg/L for Zn.

2.4. Bioconcentration factors (BCFs)

The bioaccumulation factor (BCF) is defined as the ratio of its concentration in organism (dry weight basis) to its concentration in water. (Eq. (1)) (Wang et al., 2017)

$$BCF = C_{bitoa}/C_{water} \quad (1)$$

where BCF is the bioaccumulation factor, L/kg; C_{bitoa} is the heavy metal concentration in organism, mg/kg dw; C_{water} is the heavy metal concentration in water, mg/L.

2.5. Human health risk assessment

2.5.1. The estimated daily intake (EDI)

The estimated daily intake (EDI) of heavy metals was defined as the following equation (Griboff et al., 2017):

$$EDI = (C \times IRd)/BW \quad (2)$$

where C is the concentration of heavy metals in biological samples (mg/kg dw), IRd is daily average ingestion rate (52.5 g/day for children, 55.8 g/day for adults) (Tong et al., 2016), BW is the average body weight (60 kg for an adult and 30 kg for a child). EDI was expressed as μg/kg bw/day.

2.5.2. Non-carcinogenic risks

The non-carcinogenic health risks depended on the consumption of aquatic products were evaluated by the target hazard quotient (THQ), which was defined as the ratio between the estimated dose of a contaminant and the reference dose (RfD) as following equation (USEPA, 1989):

$$THQ_{non-carcinogenic} = (EF \times ED \times IRd \times C)/(RfD \times BW \times AT) \quad (3)$$

where $THQ_{non\text{-}carcinogenic}$ is the target hazard quotient for non-carcinogenic risk, EF is exposure frequency (365 days/year), ED is the exposure duration (70 years for adults and six years for children), IR is the ingestion rate (52.5 g/day for children, 55.8 g/day for adults), C is the heavy metal concentration in aquatic products (mg/kg dw), RfD is the oral reference dose (μg/kg/day, 1 for Cd, 4 for Pb, 1500 for Cr, 40 for Cu, 20 for Ni, 300 for Zn, 140 for Mn, 0.3 for As) (Varol et al., 2017), Bw is the average body weight (60 kg for an adult and 30 kg for a child)