



Health risk assessment of heavy metals in fish and accumulation patterns in food web in the upper Yangtze River, China



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ABSTRACT

This study aims to concern the distribution of As, Cr, Cd, Hg, Cu, Zn, Pb and Fe in surface sediment, zoobenthos and fishes, and quantify the accumulative ecological risk and human health risk of metals in river ecological system based on the field investigation in the upper Yangtze River. The results revealed high ecological risk of As, Cd, Cu, Hg, Zn and Pb in sediment. As and Cd in fish presented potential human health risk of metals by assessing integrated target hazard quotient results based on average and maximum concentrations, respectively. No detrimental health effects of heavy metals on humans were found by daily fish consumption. While, the total target hazard quotient (1.659) exceeding 1, it meant that the exposed population might experience non-carcinogenic health risks from the accumulative effect of metals. Ecological network analysis model was established to identify the transfer routes and quantify accumulative effects of metals on river ecosystem. Control analysis between compartments showed large predator fish firstly depended on the omnivorous fish. Accumulative ecological risk of metals indicated that zoobenthos had the largest metal propagation risk and compartments located at higher trophic levels were not easier affected by the external environment pollution. **Capsule:** A potential accumulative ecological risk of heavy metal in the food web was quantified, and the noncarcinogenic health risk of fish consumption was revealed for the upper reach of the Yangtze River.

1. Introduction

Heavy metals from discharged industrial and agricultural wastewater are discharged into rivers and are absorbed by suspended sediment and then precipitated to be a part of surface sediment in riverbed (Fang et al., 2016; Zhu and Zang, 2002). Part of them absorbed by sediment in turn release into the water column as the potential secondary source of contaminants in case of certain disturbances, and this can be a potential threat to ecosystems (Fang et al., 2016; Varol, 2011). Consequently, analyzing the distribution of heavy metals in sediment was a potential strategy for investigating anthropogenic impacts on ecosystems and assessing risks posed by human activities (Ke et al., 2017).

Heavy metals are inert in sediment and considered as conservative pollutants (Oliveras-Rieumont et al., 2005). Sediment provides food and habitat for fish and benthos, which promotes the bioaccumulation of heavy metals from water and sediment to aquatic organisms through the prey relationships in the aquatic food webs (Demirak et al., 2006; Vicente-Martorell et al., 2009). Fish is a rich source of high-quality

proteins and long-chain polyunsaturated omega-3 fatty acids (Sadiq et al., 2003), therefore freshwater fish is particularly frequently consumed in China (Liu and Chen, 2014). Consequently, quality and safety of freshwater products are strictly limited. However rapid industrial and agricultural development have led to heavy metals pollution, which are a significant environmental hazard to invertebrates, fish, and humans (Fang et al., 2016; Varol, 2011; Vicente-Martorell et al., 2009). The heavy metals risk of human health by fish consumption can be carcinogenic or noncarcinogenic (Peng et al., 2016). Comparison between exposure concentrations and thresholds for adverse effects, as determined by dose–effect relationships, is conducted to represent the carcinogenic effects (Solomon et al., 2013). The probability risk assessment technique (Solomon et al., 2013), target hazard quotient (THQ) and toxic equivalent factor (Jiang et al., 2014) have been adopted by several researchers to characterize the exposure and toxicity. However, these methods have been used only to quantify the health risks of carcinogenic pollutants. Current noncarcinogenic risk assessment methods are based on the THQ. Although, the THQ-based risk assessment method does not quantitatively estimate the probability

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of experiencing adverse health effects for an exposed population, it provides a pragmatic indication of the risk level associated with pollutant exposure. This risk estimation method has been used by many researchers and has been proved to its validity and usefulness (Chien et al., 2002; Zhang et al., 2017). It was also applied in this study to assess the human health risk of heavy metals from fish consumption.

The accumulative ecological effects of metals on aquatic organism via the food web can be implemented by employing the ecological network analysis (ENA). The ENA is an ecosystem risk assessment method that is based on the input–output theory and the transfer process of material and energy (Fath et al., 2007). It can assess the ecological risks of human activities on the natural environment from a system perspective (Chen et al., 2015; Guesnet et al., 2015). Thus, the ENA is also used to quantify the ecological risk of heavy metals from the external source of the river ecosystem based on the prey–predation relationships in this study.

Although analyses of heavy metals in river sediment have been extensively monitored and used to identify pollution (Fu et al., 2013; Wang et al., 2014; Yi et al., 2011), few studies have focused on the routes of transmission of metals in the food web and from a system perspective for the upper Yangtze River. This study aims to identify the distribution of As, Cr, Cd, Cu, Pb, Zn, Hg and Fe in sediment and fishes in the upper Yangtze River. The noncarcinogenic risk of metals from fish consumption was assessed by judging if THQ and total THQ exceeded 1. And also, relationships among compartments were explained in a food web and the accumulative ecological risk of heavy metals was assessed based on the ENA. Furthermore, pollution sources of heavy metals in sediment were identified.

2. Materials and methods

2.1. Study sites

The Yangtze River, with a length of 6300 km, is the longest and the largest river in China and covers 1.80-million-km² watershed (Yi et al., 2011). It consists of the upper, middle and lower reaches. The upper reach extends from the river source to Yichang (the Three-Gorges Dam) (Fig. S1). According to the statistics of the Yangtze River Yearbook, the amount of Pb and Cr in sewage discharged from provinces in the Yangtze River basin was 84.31% and 68.84% of China in 2012, respectively (YRYCC, 2013). This indicates that the Yangtze River has been suffering severe heavy metals pollution.

2.2. Sample collection and analysis

Surface sediment and fish samples were collected from the four sampling sites, including Yibin (YB), Banan (BN), Chongqing (CQ) and Wanzhou (WZ) along the upper reach of the Yangtze River in July 2014 (Fig. S1). Surface sediments (0–10 cm) were collected by a Peterson grab sampler and brought back to the laboratory to be air-dried in the shadow. Then they were milled and filtered through a 100-mesh sieve for further analysis. After that, the surface sediment samples (0.2000 g) were allowed to microwave digestion with a HF–HNO₃–HClO₄ acid mixture. Concentrations of As, Cr, Cd, Hg, Cu, Zn, Pb and Fe were measured by inductively coupled plasma-atomic emission spectrometry (ICP-AES). Quality control included the reagent blank, parallel sample and standard substance (GBW07302; National Institute of Metrology, Beijing, China). The recoveries of metals were listed in Table S1 based on the measured results of standard reference soil samples (GSS-8, GSS-9, GSS-14, and GSS-15), which showed the reliable equipment.

Fish samples were collected using a seine by local fishermen at four sites. All samples were frozen and stored at –18 °C immediately after they were collected from the field. Fish muscles generally contain a lower accumulation capacity for heavy metals than that of other tissues except Cu (Demirak et al., 2006). As people normally intake fish muscles, this research mainly focuses on the heavy metals concentration in

muscles of fish. Therefore, about 100–300 g of dorsal muscle was dissected from each fish and was freeze-dried in a vacuum freeze-dryer to detect the heavy metals concentrations.

Tissue samples (0.5 ± 0.01 g) were digested using acid-washed Teflon digestion vessels. Then ten milliliters of ultra pure nitric acid were added and heated to 100 °C, until most of nitrogen dioxide was emitted. Finally, 4 mL mixed solution of HNO₃:HF (1:1 v/v) was added to microwave digestion. The digestion included three heating steps: 0.5 MPa for 1 min, 1.0 MPa for 2 min, and 1.5 MPa for 3 min. After completely cooling, the digested sample was transferred to a volumetric flask and Milli-Q water was added to 100 mL solution (Yi et al., 2011).

2.3. Tolerable daily intake and estimated daily intake

The estimated daily intake (EDI) depends on the metal concentration, food consumption, and body weight. To evaluate the risk of heavy metals from fish consumption at the extreme, we made the following assumptions in this research: the ingested dose was equal to the absorbed pollutant dose (USEPA, 1989); cooking had no effect on the pollutants (Chien et al., 2002); the average adult body weight of Chinese people was 55.9 kg (Ge, 1992); people who lived in coastal area of China would eat 105 g fish and crayfish per day (Jiang et al., 2005), and people living in the Yangtze River basin would consume the same dose just like the coastal area of China. Therefore, the EDI of heavy metals for adults was calculated as follows:

$$EDI = \frac{C \times C_{cons}}{B_w} \quad (1)$$

where C is the concentration of heavy metals in fish (mg/kg wet weight), C_{cons} is the average daily consumption of fish in the local area (105 g/day bw), and B_w represents the body weight (55.9 kg).

2.4. Determination of THQ

The THQ, the ratio of the exposure dose to the reference dose (RfD), represents the risk of noncarcinogenic effects. If it is less than 1, exposure level is less than the RfD. This indicates the daily exposure at this level is unlikely to cause adverse effects during a person's lifetime, and vice versa. The dose calculations were performed using standard assumptions from the integrated US EPA risk analysis (USEPA, 2000). The model for estimating THQ was determined by the following equation (Chien et al., 2002):

$$THQ = \frac{EFr \times ED_{tot} \times FIR \times C}{RfDo \times B_w \times ATn} \times 10^{-3} \quad (2)$$

where EFr is the exposure frequency (350 days/year); ED_{tot} is the exposure duration (30 years); FIR is the food ingestion rate (g/day), and 10^{-3} is the unit conversion factor; C is the heavy metal concentration in fish (mg/kg wet weight); $RfDo$ is the oral RfD (mg/kg-day); B_w is the average adult body weight (55.9 kg); and ATn is the average exposure time for noncarcinogens (365 days/year × number of exposure years, assuming 30 years).

In this study, the total THQ was expressed as the arithmetic sum of the individual metal THQ values according to the method of Chien et al. (2002):

$$\text{Total THQ (TTHQ)} = THQ(\text{toxicant } 1) + THQ(\text{toxicant } 2) + \dots + THQ(\text{toxicant } n) \quad (3)$$

2.5. Ecological network analysis

Metals adsorbed by sediment were transferred to zoobenthos, plankton, aquatic plants and demersal fish through nutrients intake from water and sediment. Heavy metals were transferred in a food web based on the control relationship of compartments (Supplementary

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