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Species-specific bioaccumulation and correlated health risk of arsenic compounds in freshwater fish from a typical mine-impacted river

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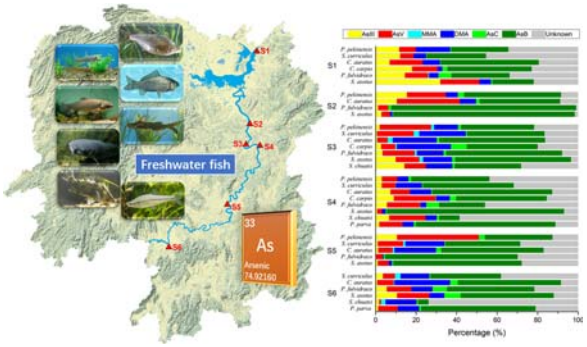
HIGHLIGHTS

- AsB was the predominant As species in the studied freshwater fish muscles.
- The percentage of iAs decreased with tAs concentration in a hyperbolic pattern.
- Spatial variability of As contamination results in large variation of As species in fish.
- Chronic exposure to iAs from fish consumption poses a considerable carcinogenic risk.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic (As) speciation and bioaccumulation in fish muscle tissues have been intensively investigated in marine ecosystem. However, little is known about these in freshwater fish. In this study, freshwater fish including 120 specimens and 8 species were collected from the Xiang River, a typical mine-impacted river in China. Six As species including arsenite (AsIII), arsenate (AsV), monomethylarsonic acid (MMA), dimethylarsinic acid (DMA), arsenocholine (AsC) and arsenobetaine (AsB) were simultaneously separated and determined using HPLC-ICP-MS. The mean (±SD) concentration of total As (tAs) in the dried fish muscle was 0.748 ± 0.651 mg·kg^{-1}. AsB was found as the predominant As species in most of the studied fish samples, in accordance with the reports in marine fish. However, the diversity of inorganic/organic As proportion observed in the studied freshwater fish species was larger than that in marine fish species due to greater spatial variability of As contamination, mobilization and origination in the studied catchments. The percentage of AsB (AsB%) in fish muscle was irrelevant to tAs concentration, while the percentage of iAs (iAs%) decreased with tAs concentration in a hyperbolic pattern. This can be attributed to restricted assimilation and accumulation of toxic iAs with increasing tAs concentration in fish. Chronic non-carcinogenic and carcinogenic health risks were evaluated through Monte-Carlo simulation. The result indicated that consuming freshwater fish in the Xiang River could cause considerable carcinogenic risk to local inhabitants.

1. Introduction

The trace element - arsenic (As) is ubiquitously present in earth crust. Except natural existence, As in contaminated areas is mainly

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originated from anthropogenic sources such as mining and smelting activities, fuel combustion, waste incineration and herbicide/pesticide usage (Ma et al., 2016; Zhao et al., 2015). The biotoxicity and mobility of As are dependent on its chemical forms (Ma et al., 2017a). Among all the As species, the inorganic As (iAs) including arsenite (AsIII) and arsenate (AsV) are the most toxic forms which are regulated as Group 1/A-known human carcinogen (IARC, 1987). Chronic exposure to low level iAs could lead to carcinogenesis and other health risks to human beings (Chen et al., 1992). Joint FAO/World Health Organization Expert Committee on Food Additives (JECFA) determined the $BMDL_{0.5}$ values for lung and urinary cancers in human associated with dietary exposure to iAs ranging from 3.0 to 5.0 μg/kg bw per day and 5.2 to 11.4 μg/kg bw per day, respectively (WHO, 2015). Monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA), considered as the metabolites of iAs, are far less genotoxic. However, DMA has been identified as a promoter in the carcinogenesis of iAs (Kenyon and Hughes, 2001). Other organic arsenicals such as arsenobetaine (AsB) and arsenocholine (AsC) are considered to be non-toxic (Borak and Hosgood, 2007).

Distributions of As species in marine organisms including fish, shellfish and seaweed have been widely investigated in last decades (Dahl et al., 2010; Krishnakumar et al., 2016; Rahman et al., 2012). Although these organisms were found to contain high concentrations of total As (tAs), over 90% of the arsenicals were in organic forms. AsB known as the end product of As metabolism is the predominant species in seafood. The biotoxicity of AsB is very low or negligible to human beings. Compared to marine fish, there are much fewer studies focusing on As speciation in freshwater fish (Rahman et al., 2012). In fact, high levels of tAs and large proportions of iAs were reported in the freshwater fish muscle from Thailand and Pakistan (Jankong et al., 2007; Shah et al., 2010). Fish and related food contribute an important part of diet for human due to their supplies of high value proteins and essential fatty acids which are benefit to human development and health (Domingo et al., 2007). In China, the output of freshwater production accounts for half of the total aquatic output. Freshwater fish is one of the most popular food especially for inhabitants in inland areas. Consumption of As contaminated fish will significantly increase exposure to toxic As species. On account of this, identification and determination of As species in freshwater fish muscle are important for further studies on biotoxicity and bioaccumulation of As.

The Xiang River as the main tributary of Yangtze River is mainly located in Hunan Province, China. In recent decades, the aquatic environment of the Xiang River has been severely polluted by various toxic substances including heavy metals and As due to long-term mining and smelting activities in the watershed (Li et al., 2013; Zhang et al., 2010). Elevated concentration of As in sediments from the Xiang River poses considerable ecological risk to the living organisms (Li et al., 2013). Fish located in the top of the aquatic food chain is susceptible to hazardous materials present in the aquatic environment (Alibabić et al., 2007). The As contamination can be transferred into fish body through direct contact by skin, ion exchange by gill and oral ingestion (Voigt et al., 2015). The distributions of tAs in three tissues (muscle, gill and liver) of freshwater fish collected from the Xiang River have been investigated in previous studies (Jia et al., 2016, 2017). The results showed no significant difference among tAs concentrations in different tissues. It means the muscle tissue can serve as an indicator for monitoring As contamination in fish. In the current study, fish samples were collected from six sites along the Xiang River. The aims of this work were to (1) extract and determine six As species including AsIII, AsV, MMA,

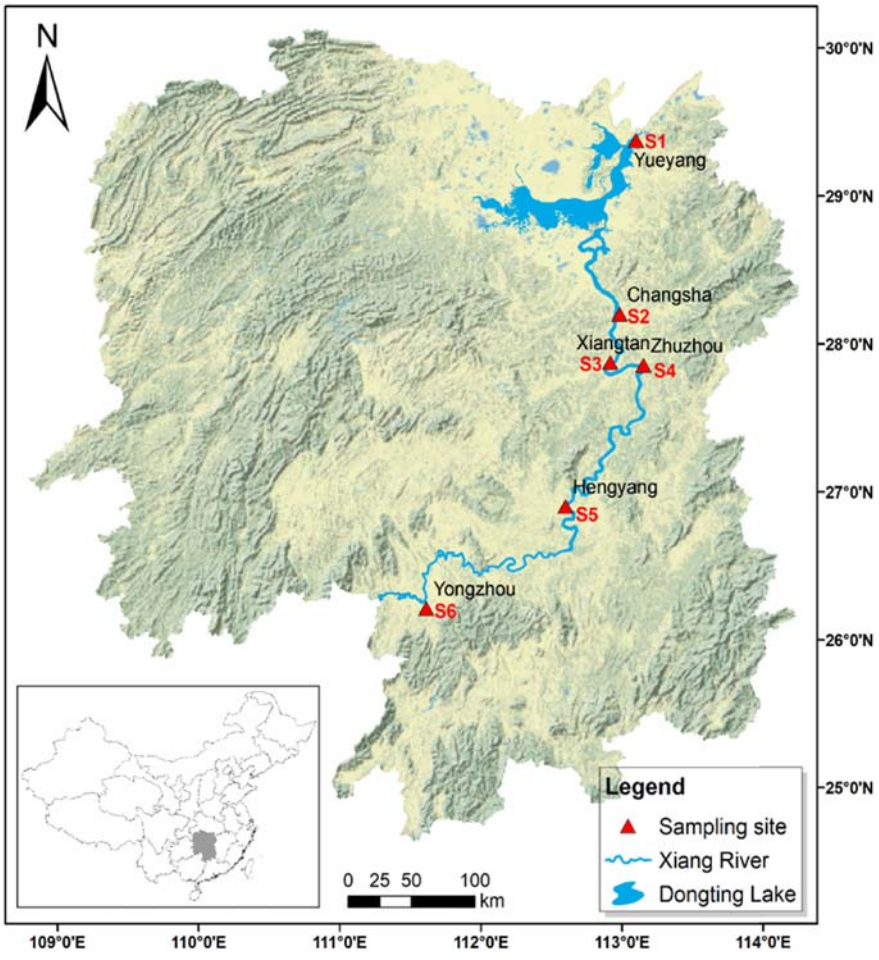

Fig. 1. Sampling sites in the Xiang River, China.