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JES JOURNAL OF ENVIRONMENTAL SCIENCES www.jesc.ac.cn

Mercury in alpine fish from four rivers in the Tibetan Plateau

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ARTICLE INFO

Article history:
Received 14 July 2015
Revised 30 September 2015
Accepted 3 October 2015
Available online 8 December 2015

Keywords:
Mercury
Methylmercury
Fish
The Tibetan Plateau
Trophic level
Risk assessment

ABSTRACT

As a global pollutant, high levels of mercury (Hg) have been found in remote ecosystem due to the long range atmospheric transport. In this study, a total of 60 fish samples were collected from four rivers across the Tibetan Plateau to study the accumulation of Hg in remote and high-altitude aquatic environment. The total Hg (THg) and methylmercury (MeHg) in fish muscles ranged from 11 to 2097 ng/g dry weight (dw) (average: 819 ng/g dw) and from 14 to 1960 ng/g dw (average: 756 ng/g dw), respectively. Significantly positive linear relationships were observed between the THg ($r = 0.591$, $p < 0.01$, $n = 36$) and MeHg concentrations ($r = 0.473$, $p < 0.01$, $n = 36$) with the trophic level of fish from Lhasa River, suggesting trophic transfer and biomagnification of Hg in this aquatic ecosystem. Moreover, the THg levels in fish had significantly positive correlations with the length ($r = 0.316$, $p < 0.05$, $n = 60$) and weight ($r = 0.271$, $p < 0.05$, $n = 60$) of fish. The high levels of Hg were attributed to the slow growth and long lifespan of the fish under this sterile and cold environment. Risk assessment revealed that the consumption of *Oxygymnocypris stewartii*, *Schizothorax macropogon*, *Schizothorax waltoni*, *Schizopygopsis younghusbandi* and *Schizothorax o'connori* would lead to a high exposure to MeHg.

Introduction

Mercury (Hg) is a toxic and global pollutant. Hg in the environment majorly comes from two sources, natural sources and anthropogenic sources (Jiang et al., 2006). Natural emissions mainly originate from volcanoes (Zambardi et al., 2009), geothermal activity (Gustin et al., 2008) and emissions from soil and water. Mining, smelting, coal combustion and industrial usage of Hg in chlor-alkali production, batteries, fluorescent lamps, medical devices and so on comprise anthropogenic sources (Pirrone et al., 2010; Streets et al., 2011). Due to the long lifetime of Hg in atmosphere and the ability of long range transport, Hg emitted from both natural sources and anthropogenic sources can be deposited into remote lakes and rivers, resulting in serious pollution in aquatic system (Fitzgerald et al., 1998; Miller et al., 2005; Poissant et al., 2008).

The Tibetan Plateau has an average elevation of 4000 m above sea level. The population in the Tibet Autonomous Region was 2.21 million in 1990 and increased to 3.00 million in 2010 (Yu et al., 2012). The total area of the Tibetan Plateau is more than 1.22 million square kilometers including 64.57% farmland, 0.05% construction land and 35.38% unused land

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http://dx.doi.org/10.1016/j.jes.2015.09.009
1001-0742/

(Liao et al., 2008). The geological substrate in the Tibetan Plateau is mainly igneous rock (Ma, 1998). The vegetation on the plateau is the high-cold meadow, steppe and desert vegetation (Zhang, 1978). The Tibetan Plateau locates in Asia where the world's largest anthropogenic Hg emission source exits. In 2008, the global Hg emissions were 2000 tons and Asia was responsible for 64% of the total emissions (1280 tons) (Streets et al., 2011). The total anthropogenic Hg emissions in China were estimated at 609 tons in 2007 (Pirrone et al., 2010). Though local industrial activities in the Tibetan Plateau were rare, the long range atmospheric transport of Hg had resulted in Hg pollution in this plateau (Huang et al., 2012; Loewen et al., 2007; Yang et al., 2010). The backward air mass trajectory analysis suggested that dust storm containing particulate Hg originated in Iran and Afghanistan and finally reached Mount Nyainqentanglha in the Tibetan Plateau after traveling over thousands of kilometers (Loewen et al., 2007). Some trace metals (Cr, Ni, Cu, Zn, As) in aerosols from the Nam Co region, which is located in central Tibet and covering an area of 1980 km^2 at an elevation of 4718 m, might be from South Asia over the long range atmospheric transport (Cong et al., 2007).

Some studies have investigated the Hg contamination in the Tibetan Plateau. The results showed that the Hg concentrations ranged from 1.46 to 4.99 ng/L in water from Yarlung Zangbo River (Zheng et al., 2010). The average of total gaseous mercury concentrations in ambient air in the eastern slope of Mt. Gongga, south-eastern fringe of the Tibetan Plateau, was 3.98 ng/m^3 (from 0.52 to 21.03 ng/m^3) (Fu et al., 2008). Surprisingly, the maximum concentrations of total mercury (THg) and methylmercury (MeHg) in fish from lakes of the Tibetan Plateau reached 2384 and 1610 ng/g, respectively (Yang et al., 2011). However, there are still some questions remaining unclear about the bioaccumulation and biomagnification of Hg in the aquatic ecosystem in the Tibetan Plateau.

The toxicity, environmental behavior and bioavailability of Hg depend not only on its total concentration but also its species. As one of the organic mercurial species, MeHg is the most toxic. Due to its lipophilicity, MeHg can penetrate the blood–brain barrier and finally harm central nervous system (Cheng et al., 2005, 2015; Clarkson and Magos, 2006; Winship, 1986). MeHg constitutes more than 80% of THg in fish muscle (Akagi et al., 1995; Bloom, 1992; Campbell et al., 2005; Guentzel et al., 2007; Mohan et al., 2012). Consumption of fish with elevated MeHg is the principal pathway of human exposure to Hg (Liang et al., 2013; Tang et al., 2015). Hence, health risk assessment on MeHg through the consumption of fish is imperative and important.

The aim of this study was to investigate the pollution status of THg and MeHg in fish samples from rivers in the Tibetan Plateau, study the trophic transfer behavior of Hg in remote alpine aquatic ecosystem and further evaluate potential health risk posed *via* fish consumption.

1. Materials and methods

1.1. Sample collection

The map of the sampling area and the locations of sampling sites are shown in Fig. 1. A total of 60 fish samples were collected from four rivers (Yarlung Zangbo River (YZR), Lhasa River, Salween and Niyang River) in the Tibetan Plateau in August of 2010, 2011 and 2012. YZR, the longest alpine river in China, is regarded as the mother river by Tibetan. As the largest branch of the YZR, the Lhasa River originates from Nyainqentanglha Mountain on the Tibetan Plateau and flows through Lhasa city, the provincial capital of the Tibet Autonomous Region. Salween originates from Tanggula Mountains. The Niyang River is also one branch of the YZR, which originates from the Mila Mountain and finally goes into the YZR in Nyingchi.

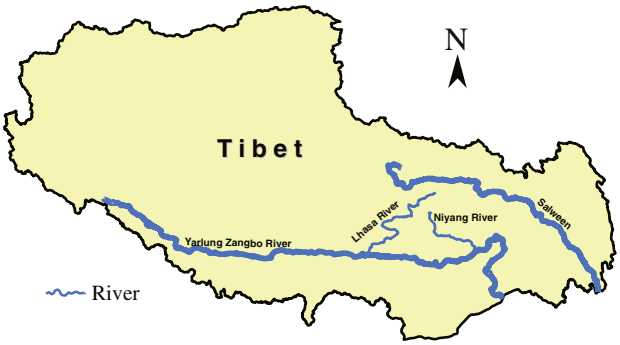

Fig. 1 – The map of the study area.

All fish species are unique in the Tibetan Plateau and these species belong to the family of *Cyprinidae* and subfamily of *Schizothoracinae*. Precautions were taken in order to avoid any contamination or loss during sampling. After collected, the fish samples were carried back to the laboratory as soon as possible in polyethylene zippered bags in boxes with ice. They were stored at −20°C before dissection. The dorsal muscles (50 g) were dissected from each fish using clean stainless steel scalpel after the length and weight were measured. The fish samples were freeze-dried, grounded and then stored at −20°C until analyzing.

1.2. Determination of THg, MeHg and trophic level

The concentrations of THg in fish samples were determined directly with a Hydra II-C atomic absorption spectrometry (Leeman Labs, Hudson, USA). Briefly, about 0.03 g of grounded fish samples was weighed in a nickel boat. A certain amount of diatomite was covered on the sample to prevent the lipid from entering the instrument. The sample was burned at high temperature and all forms of mercury were reduced to elemental mercury. Then, the elemental mercury was brought and trapped onto a gold amalgam. After decomposition from the gold amalgam, the concentrations of THg were determined.

For MeHg analysis, alkaline extraction was applied (Meng et al., 2014, 2015). About 0.1 g of sample was weighed into an 8-mL brown glass bottle and 4 mL 25% (M/V) KOH/CH_3OH solution was added. The bottle was then mechanically shaken for 4 hr at 37°C to achieve complete extraction. The solution was centrifuged at 3000 r/min for 10 min after cooling down. Before being analyzed, each sample was diluted tenfold. Then, 30 μL of diluents was injected into the auto sampler vial and ethylated by sodium tetraethylborate ($NaBEt_4$). The contents of MeHg were determined by purge and trap gas chromatography–atomic fluorescence spectrometry (GC–AFS) method