



# Occurrence of arsenic in two large shallow freshwater lakes in China and a comparison to other lakes around the world



Nan Zhang<sup>a,b</sup>, Chaoyang Wei<sup>a,\*</sup>, Linsheng Yang<sup>a</sup>

<sup>a</sup> Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, China

<sup>b</sup> University of the Chinese Academy of Sciences, Beijing 100049, China

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## ABSTRACT

Elevated levels of arsenic (As) have been found in lakes and rivers due to intensive anthropogenic activities. However, information on the occurrence of As in large freshwater lakes has scarcely been available. This study aimed at integrated exploration of the occurrence of As in multiple media of the limnetic ecosystems, mainly waters, sediments, macrophytes, invertebrates and fish in two large freshwater lakes (Taihu and Dianchi) during 2009–2010. The dissolved As concentrations in lake water for Lake Dianchi (3.08–10.48  $\mu\text{g L}^{-1}$ ) were significantly higher than the dissolved As concentrations in Lake Taihu (1.39–5.65  $\mu\text{g L}^{-1}$ ). The As concentrations in sediments by dry weight in Lake Dianchi (12.49–169.25  $\text{mg kg}^{-1}$ ) were also significantly higher than the As concentrations in Lake Taihu sediments (4.66–10.85  $\text{mg kg}^{-1}$ ). The highest As concentration observed for invertebrates was registered in a mussel species (*Novaculina chinensis*) at 25.0  $\text{mg kg}^{-1}$ , and the lowest As concentration was observed in shrimp (*Exopalaemon modestus*) at 1.2  $\text{mg kg}^{-1}$  in Lake Taihu. Although different macrophyte species were sampled in the two lakes, a common pattern was observed for the As concentrations in the plants. Submerged plants had higher levels of As than floating plants. Arsenic concentrations in Lake Taihu *Ceratophyllum demersum* L. could be 12.0  $\text{mg kg}^{-1}$ ; in contrast, in *Nymphoides peltatum*, the As concentration was 1.4  $\text{mg kg}^{-1}$ . Six common fish species were collected in Lakes Taihu and Dianchi. No significant differences in As concentrations in fish muscles from the two lakes were found, although As bioaccumulation factors (BAFs) for fish muscles in Lake Taihu were much higher than the BAFs observed in Dianchi. This difference suggests that fish in Taihu and Dianchi have a balancing mechanism for As through metabolism, which might regulate As concentration in muscle tissues at relatively low levels. Overall, the occurrence of As in the two large shallow freshwater lakes in China is relatively lower in concentration than As levels in other lakes around the world, but human health risks from As contamination exist.

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## 1. Introduction

Arsenic (As) is a virtually ubiquitous metalloid element that ranks 20th in abundance in the earth's crust [1]. Arsenic exists in inorganic and organic forms and in different oxidation states (−3, 0, +3, +5) [2], and it is toxic to both plants and animals. Inorganic arsenicals have been proven to be carcinogens in humans [3]. Arsenic is released into the aquatic environment from both natural and anthropogenic sources [4]. Anthropogenic inputs such as mining and smelting activities, combustion of fossil fuels, and the use of As pesticides, herbicides and additives to livestock feed have been reported to contribute significantly to the elevation of the As levels in freshwater ecosystems

such as rivers and lakes around the world during the last century [5]. In natural waters, As can be accumulated by aquatic biota, especially by marine algae, fish and bivalves, and at the endpoint of the food-chain, contaminants can reach terrestrial animals and humans, resulting in adverse effects of varying extents [6]. The levels of As for marine organisms have been found to vary from several  $\mu\text{g/g}$  to more than 100  $\mu\text{g/g}$  [7]. However, the relative abundance of As in natural water ecosystems is largely unknown, especially in freshwater ecosystems [8]. In recent years, concerns have increased over As due to its high toxicity and widespread occurrence in the environment [9] because the occurrence of As in freshwater ecosystems (e.g., waters, sediments and biota) can pose a potential risk to human health [10].

Arsenic is one of the most toxic elements derived from the natural environment [11], and chronic As toxicity (arsenicosis) due to drinking As contaminated water is a major environmental health hazard throughout the world [12]. High levels of As in water lead to health problems such as melanosis, hyperkeratosis, black foot disease, lung disease, liver disease and even cancers of skin, lung and urinary

\* Corresponding author at: Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, A11 Datun Road, Chaoyang District, Beijing 100101, China. Tel.: +86 10 64889465.

E-mail address: [weicy@igsnr.ac.cn](mailto:weicy@igsnr.ac.cn) (C. Wei).

bladder [10,12]. The As in drinking water has been legally regulated up to  $10 \mu\text{g L}^{-1}$  by China, the USA and WHO for the protection of human health [13]. It is widely reported that aquatic organisms are directly affected through ingestion and inhalation or indirectly via the food chain in water ecosystems contaminated by As [8]. Arsenic toxicity to organisms depends on its concentration and speciation. Inorganic arsenite ( $\text{As}^{\text{III}}$ ) is more toxic than arsenate ( $\text{As}^{\text{V}}$ ) [3], while dimethylarsenous acid ( $\text{DMAA}^{\text{III}}$ ) and monomethylarsenous acid ( $\text{MMAA}^{\text{III}}$ ) are more toxic than inorganic As species [14].

Compared to numerous reports on As in the marine environment, the presence of As in freshwater ecosystems (e.g., lakes and rivers) has scarcely been reported around the world, especially in large lakes or rivers [8]. The limited literature on As occurrence in freshwater ecosystems has focused mainly on As in water, sediments and biota as well as As bioaccumulation and bio-transformations, with most of the studies conducted in small contaminated lakes or rivers. To our knowledge, an understanding of the relationship of As elevation in water and sediments to biota levels and trophic transfers in an independent freshwater ecosystem has rarely been documented. Foley et al. reported As concentrations in the waters of Lake Chautauqua, New York, as ranging between  $22.4$  and  $114.8 \mu\text{g L}^{-1}$ , exceeding the standard issued by the U.S. Public Health Service ( $10 \mu\text{g/L}$ ). They also found the ratio of As in fish to water ranging between 0.4 and 41.6 [15]. In another report in Lake Baikal, the average As concentration in sediment was recorded as  $8.25 \text{ mg kg}^{-1}$ , within the natural background value [16]. Whitmore et al. found that in little Lake Jackson in Florida, USA, which had been polluted by organic arsenical herbicides applied to a golf course and lawns nearby, the average total As concentration in surface sediments was  $47.3 \text{ mg kg}^{-1}$ , exceeding the consensus-based sedimentary concentration for probable toxic effects in freshwater benthic fauna [17]. Bioaccumulation and trophic transfer of As in a freshwater ecosystem was also studied in the Bravona River and its tributary, the Presa River, located on the east central side of Corsica, France [18]. Because the level of As in the rivers had been affected by past mining activities, the highest As concentrations in the waters and sediments of the Bravona River and its branch were measured as  $3000 \mu\text{g L}^{-1}$  and  $9000 \text{ mg kg}^{-1}$ , respectively [19]. The As levels in biotic and abiotic samples collected in the river followed the order of macroinvertebrates > bryophytes > water > fish [18].

Lake Taihu and Lake Dianchi are two large shallow freshwater lakes in China. Lake Taihu is the third largest freshwater lake in China, and the lake water has been used for agricultural and industrial applications as well as serving as the major drinking water source for several cities, including Wuxi, Shanghai and Suzhou [20]. Lake Dianchi is the largest plateau freshwater lake in China [21], which receives almost all of the wastewater from Kunming City, the capital of Yunnan Province, with a population of 3.58 million in the urban area. Both Lake Taihu and Lake Dianchi are among the three most seriously polluted large lakes in China and are the three priority lakes (another is Lake Chaohu) selected by the Chinese government for water pollution control [21,22].

The aim of this study was to investigate the integrated related occurrences of As in lake water, sediments, macrophytes, invertebrates and fish in the two large freshwater lakes of Taihu and Dianchi. By comparison of the As levels among various media from the two lakes and other lakes around the world, we expected to obtain a macroscopic panorama of the As distribution and occurrence status in typical large shallow freshwater lakes.

## 2. Materials and methods

### 2.1. The study area

Lake Taihu (Taihu) is located in the southern Jiangsu Province and northern Zhejiang Province, with an area of  $2338 \text{ km}^2$  and a mean depth of  $1.9 \text{ m}$  [23] (Fig. 1). The drainage basin of Taihu, covering

$36,500 \text{ km}^2$  in area, is one of the most developed areas in China, including 8 large cities, 31 cities in the county and a population of 34.5 million [24]. Taihu contributes greatly to the local economy in various functions such as water supply, aquatic breeding and products, flood control, water transport and recreation [25]. In recent years, Taihu has been impacted by serious pollution as a result of rapid economic growth in the surrounding region [20].

Lake Dianchi (Dianchi) covers an area of approximately  $300 \text{ km}^2$  and is located southwest of Kunming City, Yunnan Province, China [26]. The lake is also shallow with a mean depth of  $5.12 \text{ m}$  and a maximum depth of  $11.3 \text{ m}$  [27]. Dianchi is divided into two sections (Caohai and Waihai) by a dam in the upper north, with Caohai occupying only 3% of the total area of Dianchi. During the last three decades, with local economic development, increasing amounts of domestic sewage and industrial effluents have drained into Dianchi, especially into the Caohai section [28], resulting in extinction of many native living species and annual cyanobacterial bloom [29].

### 2.2. Sample collection and pretreatment

#### 2.2.1. Sample collection in Taihu

Water samples from the upper (20 cm to surface) and bottom layers (near the sediment-water interface) were collected using a water sampler (2 L in capacity) at 17 sites covering the whole area of Taihu in May, 2010 (Fig. 1). A portion of the collected waters was filtered in situ through a  $0.45 \mu\text{m}$  film. The unfiltered and filtered water samples were stored in polyethylene plastic bottles and acidified using 2–3 drops of super pure  $\text{HNO}_3$ . Samples were kept under low temperature conditions and were transported back to the laboratory for analysis within one week. The water quality in the lake water was measured using a YSI portable instrument in situ. Parameters measured included water temperature, pH, redox potential ( $E_h$ ), dissolved oxygen (DO), electric conductivity (EC) and chlorophyll content. Surface sediment samples were collected using a Peterson device at each site and stored in polyethylene plastic bags. The sediment samples were air dried and ground into  $150 \mu\text{m}$  powders for easy and accurate digestion.

Macrophytes were collected at several sites in southeastern Taihu (Fig. 1). Samples of the submerged and floating-leaved species were collected with stems and leaves but without roots. The samples were rinsed thoroughly with clean water, then kept in plastic bags. Phytoplankton samples in Meiliang Bay at the north end of Taihu were collected using a  $25^\#$  nylon (approximately  $70 \mu\text{m}$  in diameter) phytoplankton sampler. The samples were rinsed and kept in plastic bottles. Fish and zoobenthos were collected randomly at several ports along the shore of Taihu during the fishing season (Fig. 1), then dissected and rinsed in situ. All biological samples were stored in an incubator, covered with dry ice and transported back to the laboratory. The samples were further freeze-dried in a freeze drier and ground manually before analysis using an agate mortar.

#### 2.2.2. Samples in Dianchi

Samples in Dianchi were collected in October, 2010. Water and sediment samples were collected at 21 and 27 sites using the same procedure as described above for Taihu. The sites covered the whole area of Dianchi, including Caohai and Waihai (Fig. 1). Macrophytes were collected mainly in Caohai because they were found growing mostly there. The free-floating species were collected as the whole plant, and submerged species were collected with stems and leaves but without roots. Plankton samples were collected near the dam between Caohai and Waihai. Fish from Dianchi were purchased from a morning market near the shore of Lake Dianchi. In situ water quality monitoring and field sample treatment were performed the same as in Taihu.

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