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Deriving water quality criteria for trivalent and pentavalent arsenic

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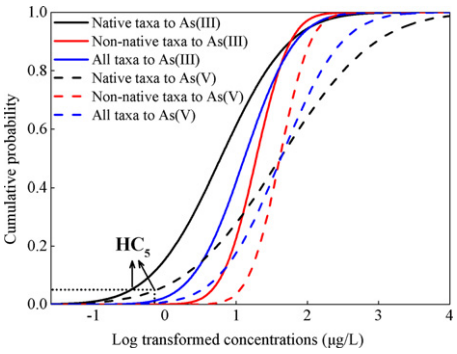
HIGHLIGHTS

- The water quality criteria for As(III) and As(V) were derived respectively.
- The influence of pH values on arsenic toxicity was complex and species-specific.
- There was difference in sensitivity between native and non-native species.
- The CMC and CCC of WQC for As(III) were 167 and 42 μg/L, respectively.
- The CMC and CCC of WQC for As(V) were 384 and 44 μg/L, respectively.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic (As) is a common trace element whose oxidation states mainly include four types (−3, 0, +3, and +5), and inorganic As(III) and As(V) are regarded as the most commonly existing forms in aqueous environments. Generally, As(III) has a higher toxicity than As(V) due to the different mechanisms in arsenic toxicity. However, there are few studies about the water quality criteria (WQC) of As(III) and As(V) respectively because of the deficiency of arsenic toxicity data coming from diverse taxonomic groups. In this research, eight native Chinese aquatic organisms were adopted to conduct toxicity tests for As(III) and As(V) to supplement the published toxicity data. The species sensitivity distribution (SSD) method on the basis of the Log-normal model which was the most optimal among eight models was applied to derive WQCs of As(III) and As(V). Results showed that crustaceans were the most sensitive to As(III) and As(V) among all tested species, thus they could be a biological indicator, and the influence of pH values on arsenic toxicity was complex and species-specific. Besides, the sensitivity differences between native and non-native species were observed. Finally, a criterion maximum concentration (CMC) of 167 and 384 μg/L for As(III) and As(V), and a criterion continuous concentration (CCC) of 42 and 44 μg/L for As(III) and As(V) were derived using native species, regardless of pH values. The WQCs were also verified by other two methods of ETX 2.0 and species sensitivity rank.

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1. Introduction

Arsenic (As) is a hazard environmental substance widely distributed all over the world (Sorg et al., 2014), and >50 different arsenic species exist in the environment (Nearing et al., 2014). Besides natural sources including weathering of minerals and rocks, volcanic eruptions and overflowed water of hot springs, anthropogenic sources such as mining, smelting, glass manufacture and pesticides also contributed to the global distribution of As (Moriarty et al., 2009). The arsenic toxicity is largely dependent on its chemical speciation. Generally, inorganic As (i.e., arsenite-As(III) and arsenate-As(V)) has a higher toxicity than its organic form (Adair et al., 2005; Sun et al., 2009; Zavala et al., 2008). In aqueous environments, inorganic As(III) and As(V) are the most commonly existing forms and could interconvert into each other under different pH and redox conditions (Cullen and Reimer, 1989; Kim et al., 2009; Oremland and Stolz, 2003; Yamani et al., 2012). Besides, previous studies have reported that As(III) was more toxic than As(V) due to their different toxic mechanisms (Hirano et al., 2003; Korte and Fernando, 1991; Styblo et al., 2000). As(III) has a high affinity to sulfhydryl groups of proteins and could react with it to inhibit the activity of enzymes (USEPA., 1984). Although As(V) doesn't react with sulfhydryl groups, it could uncouple oxidative phosphorylation resulting in the shortage of adenosine triphosphate (ATP) (Schiller et al., 1977; USEPA., 1984).

Water Quality Criteria (WQC) is an important tool to protect ecosystems from the immerging pollutants. Establishment of WQC and development of derivation method have been the research hotspots in the world (Paul et al., 2008; Wu et al., 2015; Yang et al., 2012). Currently, species sensitivity distribution (SSD) recommended by Netherland and European Union (Van Vlaardingen et al., 2005) and species sensitivity rank (SSR) recommended by United States Environmental Protection Agency (USEPA) (Stephen et al., 1985) are the most popular methods to derive WQC of the pollutants in the world. Unfortunately, it is difficult to perform water environment management of As(III) and As(V) because of the absence of WQC for As(III) and As(V).

WQC for As has been developed by USEPA (USEPA., 1984, 1995), Canadian Council of Ministers of the Environment (CCME) (CCME., 2007), Economic Commission for Europe (ECE) (ECE., 2003) and National Institute for Public Health and The Environment (RIVM) (RIVM., 2007). In USEPA, the recommended WQC that was applied to the management of total arsenic was actually derived only from the toxicity data for As(III) due to lack of As(V) data. The WQCs of As in CCMC, ECE and RIVM is 5 μg/L, 10-100 μg/L for Class II surface water and 10 μg/L, respectively, but these WQCs are all expressed as total arsenic without considering differences in arsenic valence. Up to now, no available data have been recognized to demonstrate whether the toxicities of different arsenic forms to aquatic organisms are additive (USEPA., 2009).

In China, similarly, the risk assessment of As only takes into consideration the total arsenic WQC. For example, in last ten years, at least 4 super-large arsenic contamination accidents happened in China. High concentrations of As (i.e., 0.62 mg/L in Xinqiang River, 0.57 mg/L in Duliu Creek, 0.18 mg/L in Yangzonghai Lake and 0.53 mg/L in Dashahe River) were detected in contaminated waterbody in these accidents (Chen et al., 2009; Wang, 2008; Wang et al., 2010; Zhang et al., 2008), and these detected concentrations of As were all higher than 0.05 mg/L which was the environmental quality standard of Chinese surface water (MEP., 2002). However, risk assessment of As in these accidents only relied on total arsenic WQC, which might not be appropriate to evaluate the potential risks of As in different forms. Therefore, it is very urgent and necessary to monitor arsenic concentration and develop WQC based on the arsenic valence due to the difference of chemical construction and mode of toxicity action. Besides the lack of WQC of As in different valence, there were no available WQCs for risk assessment of sudden arsenic pollution accident and day-to-day management in China. Therefore, the Criterion Maximum Concentration (CMC) and Criterion Continuous Concentration (CCC) should be established to resolve these two problems (Stephen et al., 1985).

In this research, 6 chronic and 15 acute toxicity experiments were performed using 8 Chinese native organisms, and the WQCs of As(III) and As(V) for freshwater aquatic organisms were derived according to the SSD method based on eight widely used distribution models (Chen et al., 2016; Dyer et al., 2008; Jin et al., 2013; Tatjana and Jana, 2002). The aquicolous organisms consisted of diverse taxonomic groups (4 Phyla and 8 Families), which mainly included three fishes (*Oryzias sinensis, Misgurnus anguillicaudatus* and *Pseudorasbora parva*), an annelid (*Limnodrilus hoffmeisteri*), two kinds of crustaceans (*Neocaridina denticulata sinensis* and *Daphnia magna*), a mollusk (*Cipangopaludina cahayensis*) and an insect (*Chironomus plumosus*). In addition, ETX 2.0 software suggested by Netherland and SSR method were used to verify the final WQCs. Furthermore, the sensitivity difference was compared between indigenous and non-indigenous organisms.

The purposes of this work are (1) to accomplish database for As(III) and As(V) toxicity, (2) to compare the difference in sensitivity between indigenous and non-indigenous organisms exposed to As(III) and As(V), and (3) to derive WQCs (CMC and CCC) of As(III) and As(V), respectively. Because arsenate and arsenite are weak acid strong alkali salts and pH could affect its existence form in waterbody, an attempt whether pH could affect the toxicity of As(III) and As(V) to aquatic species was done as well. This study would provide a scientific support for ecological risk assessment and contamination control for As(III) and As(V) in aquatic ecosystem in China.

2. Materials and methods

2.1. Chemicals and organisms

$NaAsO_2$ (99%, Analytical grade) and $Na_3AsO_4 \cdot 12H_2O$ (99%, Analytical grade) were purchased from Sinopharm Chemical Reagent Beijing Co., Ltd., and their concentrations were expressed as mg/L on the basis of the metallic ion (As(III) and As(V)) in the solution.

Eight Chinese local aquatic organisms were selected for the acute and chronic toxicity experiments. All tested organisms were acquired from Beijing Chao-lai Spring Flower Market. *D. magna* (age < 24 h) were obtained from State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences. Prior to the toxicity tests, all the species were acclimated in the laboratory for at least 7 days. All acute and chronic tests were performed following the standard guidelines of American Society of Testing Materials (ASTM) (ASTM., 1993; Wang et al., 2013).

2.2. Toxicity data collection and selection

Toxicity data of native species for As(III) and As(V) and the corresponding pH values were collected and selected from China Knowledge Resource Integrated Database (http://www.cnki.net/), the ECOTOX database (http://cfpub.epa.gov/ecotox/), government documents and published literatures. At least ten species from at least three taxonomic groups (e.g., fish, crustaceans, amphibians, worms and insects) were selected to describe the toxicity of As(III) and As(V) to aquatic species in surface waters in China.

As for acute toxicity data of As(III) and As(V), median effect concentration (EC_{50}) was applied as the measurement endpoint. If the EC_{50} couldn't be obtained, the median lethal concentration (LC_{50}) was used instead. As for chronic toxicity data, 10% effective concentration (EC_{10}) was the first choice, but no observed effect concentration (NOEC), maximum acceptable toxicant concentration (MATC) or lowest observed effect concentration (LOEC) were applied when EC_{10} was not available.

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