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Prey-specific determination of arsenic bioaccumulation and transformation in a marine benthic fish

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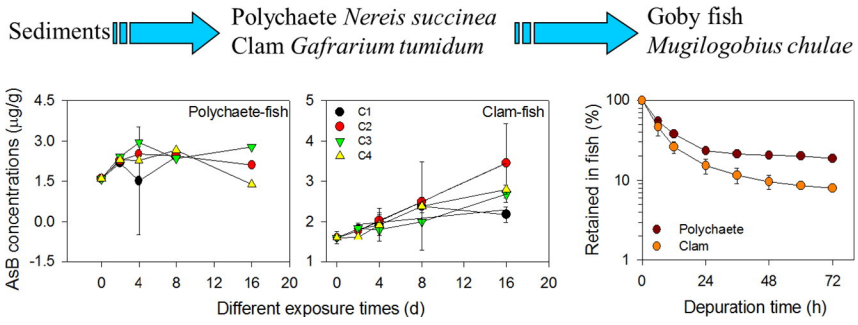
HIGHLIGHTS

- Biotransformation rate was higher in the goby fish fed on the clams than on the polychaetes.
- Low As bioaccumulation in the goby fish was caused by low dissolved uptake, dietary assimilation, and high efflux.
- Bioaccumulation of As in the goby fish fed on different prey types was comparable.
- As bioaccumulation in the goby fish was controlled both by biotransformation rate and As biokinetics.

GRAPHICAL ABSTRACT



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ABSTRACT

The sediments from Chinese coastal waters contain relatively high concentrations of arsenic (As), mainly arsenate As(V), which may be transferred along the marine benthic food chain. The prey-specific determination of As bioaccumulation and transformation in marine benthic fish remains little known. In this study, we focused on a typical marine benthic food chain comprising of sediments, deposit-feeding invertebrates (polychaete *Nereis succinea* and clam *Gafrarium tumidum*) and goby fish *Mugilogobius chulae*. Graded exposed experiments using different As exposure durations and concentrations were conducted to examine their transformation rate and efficiency. Radiotracer techniques were used to determine the rates of As uptake (as arsenate) from seawater, assimilation from two prey and its subsequent efflux in the goby fish. We demonstrated that the two prey (polychates and clams) displayed different As biotransformation in the goby fish. Biotransformation rate was higher in the goby fish fed on the clams than on the polychaetes, and biotransformation efficiency was lower with increasing inorganic As concentration in the prey. The As overall bioaccumulation in the goby fish was very low, mainly because of the low dissolved uptake and dietary assimilation and high efflux. Combining the biotransformation and biokinetics measurements, our findings highlighted that different prey containing different As concentrations and As species resulted in the comparable As bioaccumulation in the goby fish.

1. Introduction

Arsenic (As) is the most common toxic metalloid in the environment, ranking first on the superfund list of hazardous substances

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(http://www.atsdr.cdc.gov/cercla/07list.html), and has created worldwide human health problems. Arsenic pollution is one of the most serious problems in the coastal waters in China. Arsenic from various sources enters into the sea and is either stored in sediments or transferred along the food chains. One identified important factor modifying the As-related health effects is its biotransformation (Vahter, 2002). Biotransformation could be the primary source of arsenobetaine (AsB) in marine fish. Arsenic is represented by many different chemical species, with AsB being the single or main As compound in many marine organisms, while inorganic As and other organoarsenic compounds usually occur as trace constituents (Francesconi and Edmonds, 1997; Amlund et al., 2006; Fatrorini et al., 2006; Berges-Tiznado et al., 2013; Krishnakumar et al., 2016; Zhang et al., 2016a). In our recent study, we found that the bioaccumulation may be explained by the biotransformation ability (Zhang et al., 2016b).

Over the past decade, biokinetic measurements had provided important information for predicting the bioaccumulation of trace metals (Wang and Fisher, 1996, 1999; Luoma and Rainbow, 2005; Casado-Martinez et al., 2010; Kalman et al., 2014). Kalman et al. (2014) used the biodynamic modeling to investigate the bioaccumulation of trace metals (Ag, As, and Zn) by an infaunal estuarine invertebrate, the clam *Scrobicularia plana*. However, most previous studies only focused on the As biotransformation in the organisms (Erickson et al., 2011; Zhang et al., 2012; Zhang et al., 2016a), or on the As biokinetics (Williams et al., 2010; Casado-Martinez et al., 2010; Zhang et al., 2011; Kalman et al., 2014).

We hypothesized that the As bioaccumulation ability in marine fish may not only be correlated with the biotransformation, or the biokinetics, but also with the interaction between the biotransformation and biokinetics.

For As, the critical factors affecting the fate and transport are the transformation processes in food chains, which are not well understood under realistic environmental conditions. For example, As(V) was the main form in marine sediments (Ellwood and Maher, 2003), while inorganic As (61–74%) was the predominant As species in the deposit-feeding polychaete *Arenicola marina* (Geiszinger et al., 2002; Casado-Martinez et al., 2012). In contrast, AsB constituted 85–98% in the clams collected from the intertidal zone in Zhanjiang estuary, South China (Zhang et al., 2013). However, whether prey variances determine As bioaccumulation and biotransformation by marine benthic fish are still not well understood.

This study focused on a typical marine benthic food chain (sediment — polychaete/clam — goby fish). We specifically examined whether prey selection affected the As bioaccumulation and biotransformation in marine benthic fish by using As speciation analysis and radiotracer techniques. To investigate their transformation rate and efficiency, we studied the effects of different exposure times and concentrations on the goby fish, which could better characterize the time course and dose response of As bioaccumulation and biotransformation. We then used radiotracer methodology to measure the As biokinetics (uptake rate constant, assimilation efficiency and efflux rate constant) in the goby fish. Although of great importance, little information is available on the influence of transformation rate and efficiency on the behavior of As in marine fish.

2. Materials and methods

2.1. Fish, prey and radioisotope

Goby fish *Mugilogobius chulae* (2–3 cm in length) were obtained from Guangdong Laboratory Animals Monitoring Institute, China, maintained in the laboratory in circulating seawater (20 °C, 30‰) collected from Clearwater Bay, Hong Kong, and fed brine shrimp twice per day at about 3% of their body weights. The goby fish is a benthic and carnivorous species, primarily feeding on invertebrates. This fish has a potential of being an experimental model animal and pollution bioindicator. The polychaete *Nereis succinea* and the clam *Gafrarium tumidum* were purchased from Zhanjiang and Shenzhen, China, respectively. Both species were deposit-feeders, and As was primarily available through sediment ingestion.

Gamma radioisotope ^{73}As (as As(V), $t_{1/2} = 80.3$ d, specific activity: 1184 kBq/μg, in 0.1 M HCl) used in this study was purchased from Los Alamos National Laboratory. We chose to work on ^{73}As(V) mainly because the oxidized form of As(V) was the primary chemical form in oxygenated seawaters.

2.2. Arsenic bioaccumulation and biotransformation in the goby fish

The sediments were used in the dietary exposure experiments. They were collected from Daya Bay, China, and were sieved through the 180 μm mesh with seawater and kept at room temperature. To study the As biotransformation rate and efficiency, As(V) was added to the sediments as an aqueous solution of arsenate ($Na_2HAsO_4 \cdot 7H_2O$, 1000 mg/L, Sigma, USA), at four different As concentrations (C1, C2, C3, and C4) for 10 d. Nominal concentrations in the sediments of C1, C2, C3, and C4 were 10, 30, 90, and 300 μg/g, as compared to the detected As levels of 27, 55, 107, 195 μg/g, respectively. After 10 d of As spiking, the sediments were centrifuged at 5000 rpm for 15 min, and the overlying water was decanted. Seawater was added to the pellets, and sediments were re-centrifuged. After another centrifugation, the sediments were used as the food for the polychaetes and clams.

Before the exposure, the polychaetes and clams were acclimated in unspiked sediments for 7 d. Then they were exposed to different sediments containing different As concentrations. There were a total of 16 treatment beakers [4 As concentrations, each concentration had 4 time points] with 6 polychaetes or clams per beaker. The beakers were placed under 12:12 h light:dark cycle. The polychaetes and clams were exposed for 2, 4, 8, 16 d to the sediments containing different As concentrations. At each exposure time, the polychaetes and clams ($n = 6$) from each beaker were then sampled into a plastic bag, and later subjected to total As and As species analysis. In addition, another treatment (12 polychaetes or clams) was directly exposed for 16 d at each exposure concentration and was used as the food of goby fish. After exposure, the clams were also collected and finally dissected to acquire the soft tissues. The polychaetes and clams were minced using a soybean milk machine, using methods described in Zhang et al. (2016b).

The goby fish (10 individuals per beaker) were fed polychaetes or clams twice per day for 1 h at about 3% of their body weight. Afterwards, any remaining food was cleared to avoid the waterborne As exposure. The wet weight (g) and length (cm) of the goby fish were 0.23–0.39 g and 2.1–3.2 cm; 0.26–0.42 g and 2.5–3.4 cm; respectively, before and after intake of the two prey. There was no significant difference in the wet weight and length before and after intake of the two prey. At different exposure times (2, 4, 8, 16 d), the fish were removed from the exposure medium and placed in a sealed polyethylene bag, and then frozen at −80 °C for later analyzing the total As and As species.

2.3. Arsenic biokinetics

Arsenic uptake from solution was quantified by exposing individual goby fish ($n = 3$) to each of four concentrations of dissolved As(V) (0.5, 5, 15, 50 μg/L) added with 8 μCi/L of ^{73}As. Each concentration had 4 replicated beakers. The fish were removed from the beakers, carefully washed with nonradioactive filtered seawater, measured for radioactivity, and then immediately placed back to the uptake medium. The radioactivity in the seawater was determined at the beginning and end of As uptake period. Radioactivity of seawater decreased by <5%, therefore As radioactivity was relatively constant in the seawater. By the end of exposure, the whole fish were completely dried at 80 °C to determine their dry weights.

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