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Arsenic biokinetics and bioavailability in deposit-feeding clams and polychaetes

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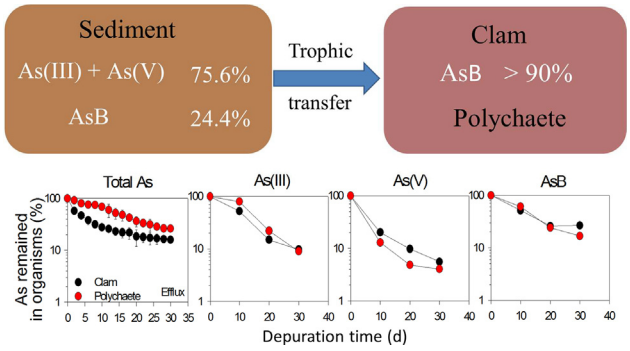
HIGHLIGHTS

- As biokinetics and biotransformation in clams and polychaetes were quantified.
- Sediments with different inorganic/organic As speciation resulted in different assimilation.
- Arsenobetaine (AsB) was more efficiently assimilated and retained than inorganic As.
- As(III) was less efficiently assimilated and more rapidly eliminated.
- As speciation caused different bioavailability to clams and polychaetes.

GRAPHICAL ABSTRACT



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ABSTRACT

In the present study, the arsenic (As) biokinetics and bioavailability in two deposit-feeding invertebrates (clams *Gafrarium tumidum* and polychaetes *Nereis succinea*) were quantified. Radiotracer techniques were applied to measure the dissolved uptake rate, dietary assimilation efficiency and efflux of As by the clams and polychaetes. Simultaneously, arsenic species analysis was conducted to examine the As biotransformation following dietary uptake. The radiotracer results showed that the uptake rate constant and efflux rate constant were 0.068 L/g/d and 0.07 d^{-1}, and 0.173 L/g/d and 0.09 d^{-1}, in the clams and polychaetes, respectively. Sediments labeled for different times (1.5–60 d) with different inorganic/organic As percentages led to diverse assimilation efficiencies of As (35.1–56.1% in the clams, and 51.6–72.6% in the polychaetes). Modeling calculations showed that sediment was a significant source for As bioaccumulation in the two deposit-feeders. After feeding on the spiked sediments, inorganic As (75.6%) was initially the predominant form, but arsenobetaine (AsB) became the predominant compound (>90%) in the clams and polychaetes during depuration, suggesting biotransformation of inorganic As. Combined with the biokinetics and biotransformation measurements, we showed that AsB was more efficiently assimilated and tended to be accumulated, whereas As(III) was less efficiently assimilated and more rapidly eliminated by the two invertebrates. This study demonstrated that As speciation in the sediments as a significant source for As bioaccumulation caused different bioavailability in deposit-feeding clams and polychaetes.

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1. Introduction

Arsenic (As) is a well-known metalloid and has been recognized as a global toxicant with considerable human health concern (Rossman and Klein, 2011). Arsenic concentrations in contaminated marine sediments can be as high as 200 μg/g (Toevs et al., 2006), while typical As concentrations in sediments from the coastal areas of South China were in the range of 0.7–42.3 μg/g dry weight (Wang et al., 2013). One of the unique aspects about As pollution is its transformation in sediments. It is difficult to predict the As bioavailability based on total concentration in sediments given its very different geochemistry, and the mobility and availability of As in sediment strongly depend on its speciation (Smedley and Kinniburgh, 2002). There is thus a clear need to understand the controls of different As species in sediments on its bioaccumulation and bioavailability to benthic organisms.

The clams and polychaetes are the dominant benthic macrofauna in coastal intertidal zones. Sediments are often an important exposure route of As to benthic invertebrates (Casado-Martinez et al., 2010; Neff, 1997; Rainbow et al., 2011). For example, Bryan and Langston (1992) reported that sediments are the major sources of As to many deposit-feeding invertebrates in the field, such as the polychaete *Nereis diversicolor*, and the clams *Macoma balthica* and *Scrobicularia plana*. Laboratory studies with radiolabelled As also demonstrated that sediments were the main sources of As to these species. Different biokinetic processes have provided important information for predicting the bioaccumulation of As (Luoma and Rainbow, 2005; Zhang et al., 2011). Several earlier studies examined the As biokinetics using radiotracer technique in green alga *Chlorella salina*, shrimp *Lysmata seticaudata*, oyster *Crassostrea virginica*, polychaete *Arenicola marina*, juvenile fish *Terapon jarbua* and killifish *Fundulus heteroclitus* (Casado-Martinez et al., 2010; Dutton and Fisher, 2011; Gómez-Batista et al., 2007; Karadjova et al., 2008; Zhang et al., 2011). However, these early studies did not directly measure the As species in the organisms/prey, which were all radiolabeled with ^{73}As(V), thus the control of As speciation on As bioavailability from contaminated sediments remains elusive.

Conversely, a few studies investigated the As species in the deposit-feeding clams and polychaetes without directly considering the biokinetics. For instances, the most common form of As presented in the clam *Mya arenaria* collected from relatively uncontaminated environments was arsenobetaine (AsB), and only an average of 8.2% (range of 1.8–19%) of total As was in inorganic form (Gagnon et al., 2004). In contrast, an unusually high proportion of inorganic As (>40%) was found in the clams living in a contaminated site in Seal Harbour, Nova Scotia, Canada (Koch et al., 2007). Likewise, some polychaete species contained the majority of total As as AsB (57–98%), with trace concentrations of other simple methylated species (<7.5%) and inorganic As (<1%) (Waring and Maher, 2005). However, the polychaete *A. marina* contained primarily inorganic As as high as 61–74% of total As (Casado-Martinez et al., 2012; Geiszinger et al., 2002). These earlier measurements suggested that the speciation of As in deposit-feeding invertebrates can be rather variable. In this study, we hypothesized that the As species in the sediments may cause different bioavailability to deposit-feeding clams and polychaetes. Very few earlier studies have simultaneously contrasted the biokinetics of As (uptake, assimilation efficiency and efflux) and biotransformation (As species) in deposit-feeding invertebrates.

Therefore, the purposes of the present study were to explore the As biokinetics and biotransformation in two deposit-feeding invertebrates (clams *Gafrarium tumidum* and polychaetes *N. succinea*), and to examine the bioavailability of different As species to these invertebrates. We conducted a series of experiments by simultaneously employing gamma-emitting radioisotopes and As speciation analysis to evaluate the bioaccumulation and bioavailability to deposit-feeding clams and polychaetes. We radiolabeled the sediments for different periods of time, and quantified the assimilation efficiencies of As to the two invertebrates. Simultaneously, As species in the sediments as well as in the animals feeding on sediment (labeled for 3 d) were determined to facilitate the interpretation of experimental results.

2. Materials and methods

2.1. Clams, polychaetes and radioisotopes

The clams *G. tumidum* (2–3 cm) were collected from Shenzhen, China, in August 2015. They distribute widely in the coastal soft sediments and are important fishery species along the Shenzhen coast. During the acclimation periods, the clams were kept at 20–25 °C in the laboratory under a photoperiod of L12:D12, with sediments collected from a mudflat in Sai Kung, Hong Kong. They were maintained under the tested conditions for six weeks. The polychaetes *N. succinea* (7–9 cm) were collected from Zhanjiang, Guangdong Province. They are the ecologically key species in the estuaries, serving as a vital bioturbator and an integral part of benthic estuarine mudflats, and representing an important food source for fish and crustaceans. The polychaetes were transported in cool boxes and arrived in the laboratory within 12 h. Once in the laboratory, the polychaetes were maintained in sediments with seawaters at 33 psu and 20–25 °C. The animals and sediments were collected from different locations based on their availability and convenience. They were also maintained under the laboratory conditions for six weeks.

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

2.2. Arsenic biokinetics

2.2.1. Dissolved uptake

We studied the As uptake by the clams and polychaetes utilizing the radiotracer in the presence of stable As(V). The uptake media were prepared 1 d ahead of schedule and kept overnight for equilibration. The corresponding As solution was added to the exposure beakers for 72 h in order to saturate the adsorption sites. Arsenic uptake from solution was quantified by exposing the clams and polychaetes individually to four concentrations of dissolved As(V) in filtered seawater (1, 5, 15, 50 μg/L) labeled with 10 μCi/L ^{73}As. The use of the gamma radioactive isotope and live counting permitted tracking the uptake of As in the same individual over time. Four groups of individuals were used in each treatment. At 2, 4, 6, and 8 h, four exposed clams and polychaetes were removed from the exposure beakers, carefully washed with non-radioactive filtered seawater, quantified with the radioactivity, and then directly returned to the uptake medium. Radioactivity of seawater showed no substantial decrease. By the end of exposure, the clams and polychaetes were completely dried at 80 °C, and then their dry weights were determined.

2.2.2. Assimilation experiment

The transfer of As from sediments to the two invertebrates was quantified. The sediments were sieved through a 45 μm mesh with seawater and kept at room temperature. To examine the assimilation efficiencies (AEs) of As species in sediments, sediments (20 g wet wt) were radiolabeled with ^{73}As (17.9 μCi/L) and spiked with stable As(V) for different times (1.5, 3, and 60 d) to reach a nominal concentration of 30 μg/g. Another batch of sediments was simultaneously spiked only with As(V) for the same period of time (1.5, 3, and 60 d) for subsequent total As and As species analysis. Following the radiolabeling, the sediments were centrifuged for 15 min at 5000 rpm, and the overlying water was decanted. This procedure was repeated three more times (to ensure the removal of most unabsorbed As) through the addition of filtered seawater. The radiolabeled sediments were finally placed in a 50 mL centrifuge tube before the radioactive feeding.

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