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Removal of selenium containing algae by the bivalve *Sinanodonta woodiana* and the potential risk to human health☆,☆☆

Chuanqi Zhou, Jung-Chen Huang*, Fang Liu, Shengbing He, Weili Zhou

School of Environmental Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, PR China

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ABSTRACT

Selenium (Se) is an essential micronutrient for animals and humans with a relatively narrow margin between nutritional essentiality and potential toxicity. Even though our previous studies have demonstrated algae could efficiently remove Se, mainly through volatilization, concern is raised about eco-risks posed by the remaining Se in algae. Here, *Sinanodonta woodiana* was investigated as a biofilter for the removal of Se-containing *Chlorella vulgaris* and for its potential risk to human health. Our results suggest filtration rates of *S. woodiana* were independent of Se levels in algal biomass, with a removal efficiency of between 60 and 78%. However, Se concentrations accumulated in mussels were significantly correlated with algal-borne Se levels, with a dietary assimilation efficiency ranging from 12% to 46%. Thus, a pilot biofiltration system was set up to assess uptake and depuration processes. The system was found to efficiently remove Se laden algae through the uptake by mussels, while 21% of Se in mussels could be depurated in 6 days. Among tissues, gills accumulated the highest Se concentration after assimilating algal-borne Se but shed Se compounds in the fastest pace during depuration. Health risks posed by consumption of mussels exposed to different sources of Se were further assessed. *S. woodiana* accumulated the highest Se concentration after exposure to waterborne SeMet, followed by dietary Se, selenite and control. The relatively higher Se levels were found in gills for all the treatments. After boiling, the most common method of cooking mussels, the greatest reduction in Se concentration occurred in mantle for the control and dietary Se groups and in muscle for the SeMet and selenite treatments. Therefore, within the safe limits, Se-containing mussels can be consumed as a dietary supplement. Overall, our research suggests incorporation of mussels into an algal treatment system can improve Se removal efficiency and also provide financial incentives for practitioners.

1. Introduction

Selenium (Se) is an essential micronutrient for animals and humans with a relatively narrow margin between nutritional essentiality and potential toxicity (Rayman, 2008; Young et al., 2010). High levels of dissolved Se in drainage water (mainly selenite and selenate) have been closely related to developmental defects, reproductive failure and even mortality in fish and waterfowl (Luoma and Presser, 2009; Stewart et al., 2004).

Compared to physico-chemical technologies developed for treating Se-containing wastewaters, bioremediation approaches may have lower cost for construction and operation (CH2M, 2013). Our previous study has shown freshwater algae assimilated Se very efficiently (Huang et al., 2013; Liu et al., 2016; Zhou et al., 2017), while *Chlorella vulgaris*, in particular, was able to remove 96% of selenite within 72 h, mainly through volatilization. This process has attracted interest because it leads to a net loss of Se from ecosystems mainly in volatile forms (e.g., dimethyl selenide) which are relatively non-toxic (Wilber, 1980). Nevertheless, there is still some concerns that the remaining Se in the algae might potentially expose wildlife to toxic effects through the aquatic food chain.

To alleviate the potential eco-risk posed by the algal treatment of Se, we explored the idea of biofiltration of Se laden algae using bivalves, which can filter great volumes of water, removing particulates from the water column as well as from deposited

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* Corresponding author. School of Environmental Science and Engineering, Shanghai Jiao Tong University, Environment Science Building, 800 Dongchuan Rd, Minhang District, Shanghai 200240, PR China.
E-mail address: ecojch@sjtu.edu.cn (J.-C. Huang).

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sediment. Through suspension- and deposit-feeding processes, bivalves can accumulate contaminants that are dissolved in water or adsorbed by particulate matters (Ward and Shumway, 2004).

Therefore, for the present study, we chose the native mussel, *Sinanodonta* (*Anodonta*) *woodiana*, as a natural biofilter to remove Se laden algae from water. *S. woodiana* is a widely distributed benthic, freshwater mussel species that daily filters large volumes of water, retains a wide size range of particles (Chen et al., 2015; Douda et al., 2012; Watters, 1997), and, thus, is able to improve water quality, including via removal of the phytoplankton biomass from the water column (Kim et al., 2011; Liu et al., 2010; Yang et al., 2008).

However, little is known about effects of Se species on filtration efficiency of Se laden algae by *S. woodiana* as well as the fate of Se in mussel tissues. Thus, we aimed to gain further insight into the concentration dependence of Se uptake by mussels and the correlation with organic and inorganic Se species present in the water column and algal diets. Moreover, *S. woodiana* is a traditionally edible species in its native range, including China (Chen et al., 2015) where many diseases have been linked to Se deficiency (Stone, 2009; Sun et al., 2016). Since mussel harvesting may offer an effective way to ensure the removed contaminants will not re-enter food chains via fecal matters or dead bivalves (Gifford et al., 2004; Rose et al., 2014), we further explored whether Se-containing mussels might be consumed as dietary Se supplements for humans.

Overall, our objectives in the present work were to (1) measure the filtration rate of *S. woodiana* supplied with Se-bearing algae; (2) determine the Se uptake and depuration processes by *S. woodiana*; (3) investigate effects of exposure to different Se sources (aqueous vs. dietary) and Se species supplied on Se accumulation by mussels; and (4) examine effects of boiling on Se-containing mussels. The results obtained from this research will be useful for subsequent development of an efficient and eco-friendly algal-mussel water treatment system for Se removal.

2. Materials and methods

2.1. Materials

The mussel *Sinanodonta woodiana* was purchased from a local market in Minhang District (Shanghai, China). Adult individuals of *S. woodiana* were used in our study because of their higher tolerance of varied water qualities than young mussels (Kim et al., 2011). *Chlorella vulgaris* (FACHB-8) was obtained from Freshwater Algae Culture Collection at the Institute of Hydrobiology (Wuhan, China).

Sodium selenite (Na_2SeO_3), seleno-$_{DL}$-methionine (SeMet; $C_5H_{11}NO_2Se$) and selenium standard (1000 mg/L±4 mg/L) were purchased from Sigma-Aldrich.

2.2. Mussel preparation and algal density determination

After cleaning of shells, mussels were maintained in groups in aerated tap water (temperature 18 ± 0.5 °C) under a natural light regime in a controlled environment room. The algal biomass rinsed twice with DI water was supplied regularly as food. Mussels were acclimatized in the laboratory for at least 2 weeks. Prior to experimental use, mussels were depurated in aerated tap water for 48 h, and their shells were cleaned and rinsed gently. Algal density reported in this study was expressed by the concentration of chlorophyll *a* (chl*a*) in algal suspensions, which was determined by PHYTO-PAM (WALZ, Germany).

2.3. Removal of Se-bearing algae by mussels

2.3.1. Preparation of Se-bearing algae

Cultures of *C. vulgaris* in the late exponential growth phase were centrifuged at 5000 rpm/min, rinsed twice with sterile DI water, followed by 72 h exposure to 1000, 2000 and 6000 μg Se/L, respectively, as selenite in a 1 L Pyrex glass bottle and bubbled with 0.22 μm filtered air to prevent algal settlement. According to our previous results (unpublished data), the concentration of 6000 μg Se/L was found to be the tolerance limit for *C. vulgaris*. Prior to dietary exposure experiments, Se laden algal cells were rinsed twice and concentrated with DI water to obtain algal suspensions at three different levels of biomass Se (311.03, 836.73, 1302.63 μg Se/g dry weight (DW)). Algal samples were collected and dried at 60 °C to determine total Se accumulated in the algal biomass.

2.3.2. Removal of Se-bearing algae

The length of *S. woodiana* used in experiments averaged 13.4 ± 0.2 cm. Prior to the experiments, mussels were supplied with clean algae yielding an initial concentration of about 400 μg chl*a*/L every day. After cleaning of shells, each individual mussel was placed in an aerated 2 L tank, which contained Se treated algal suspension (*C. vulgaris*) and was diluted with tap water to achieve a 56.83 mg DW/L concentration. Three replicates were prepared for each of three treatments whose Se levels corresponded to the supplied algal suspension low, medium, and high. Another three tanks containing Se free algae were set up as control. Mussels were exposed to dietary Se for 2 h, followed by an assimilation period of 10 h.

Since the ventilation of bivalves is strongly dependent on the algal density (Fournier et al., 2005b), the initial algal density of each group was controlled in the range of 300–500 μg chl*a*/L. After a 12 h uptake and assimilation period, each mussel was dissected into four parts: 1) mantle, 2) muscle (including foot), 3) visceral mass and 4) gill. Each tissue sample was minced and preserved under −20 °C for total Se analysis. The average filtration rate of individual mussel was estimated by measuring the clearance rate of algal cells that were 100% efficiently retained by gills (Møhlenberg and Riisgård, 1978; Riisgard, 2001). The clearance rate was calculated according to the equation by Riisgard (2001):

$$Cl = (V/t) \times ln(C_0/C_t)$$

where V is volume (L) of water in the tank, t is the time (h), and C_0 and C_t are algal concentration (μg chl*a*/L) at time 0 and time t.

2.4. Uptake and depuration processes

2.4.1. Se uptake by mussels

Mussels were maintained in groups receiving constant aeration in a natural light regime. The water treatment unit was conducted in a tank (62 cm × 37.5 cm × 15 cm) with a water depth of 10 cm (i.e., 23.25 L water). Each tank contained 21 mussels with an average length of 13.2 ± 0.2 cm at the beginning of the test.

Concentrated *C. vulgaris* was supplied with 2000 μg Se/L as selenite in DI water for 72 h. Details of Se laden algae preparation and collection are as described in section 2.3.1.

At the beginning of a 72 h treatment period, mussels were supplied with Se laden algae at a quantity of ~5000 μg chl*a*/mussel, which was less than the mussels could consume based on tests in section 2.3. Water samples (without filtration) were collected every 2 h for the first 16 h, and then collected at the 24th, 32nd, 42nd, 54th, 66th and 72nd h for total Se analysis. After 72 h, water was renewed, and 3 mussels were dissected, minced and then preserved under −20 °C for total Se analysis.