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Arsenic release: Insights into appropriate disposal of arsenic-loaded algae precipitated from arsenic contaminated water

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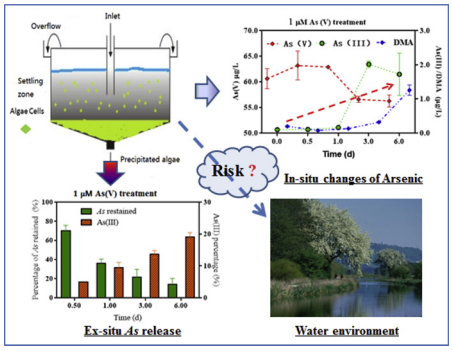
GRAPHICAL ABSTRACT



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ABSTRACT

Limited information is available on arsenic (*As*) release from *As*-loaded algae precipitated from *As* contaminated water and its subsequently appropriate disposal. In this study, selected *M. aeruginosa* as algal model, changes of *As* concentrations and its speciation were thus investigated in the in-situ treated algae water by optimal coagulation. Meanwhile, after ex-situ disposal, *As* release kinetics were also examined from its precipitated algae with living and heat-treated conditions. Results showed that in the in-situ treated water, total dissolved *As* slowly decreased for 6 days, but arsenite increased largely after 3 days partly caused by its reduction status. While being disposed ex-situ, *As* release from precipitated algae depended not only on intracellular *As* content but also on the living or heat-treated status of algae. Additionally, potential risks arised from *As* release in short-term duration (24 h) from both the precipitated algae at 1.0 μM *As*(V) pre-exposure with the living and heat-treated conditions due to their higher release. Furthermore, *As* release in long-term (6 d) duration from heat-treated algae at 10.0 μM *As*(V) pre-exposure also resulted in potential risks. Accordingly, this study offers insights into the appropriate methods at a proper time of disposing precipitated algae with *As*-contamination.

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1. Introduction

Arsenic (*As*) contamination in freshwater is a widespread problem with serious human health implications. Being a toxic and carcinogenic chemical element, *As* resides in the environment in its common inorganic arsenate (*As*(V)) and arsenite (*As*(III)) forms as well as their methylated metabolites including monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA) (Uppal et al., 2017). In general, inorganic forms are more toxic than their organic counterparts, and *As*(III) is more toxic than *As*(V). Freshwater *As* concentrations can range from less than 0.5 mg/L to greater than 5000 mg/L, depending on the contamination source (Rahman et al., 2014; Bissen and Frimmel, 2003). The standard guideline for drinking water from the World Health Organization (WHO) was set at 10 μg/L (Sharma and Sohn, 2009), validating that *As* is considered an important contaminant that necessitates precise global management.

Algae have long been considered a suitable species for the removal of numerous heavy metals from waters by means of bioremediation. For example, *Chlorella* sp., *Scenedesmus* sp., *Lessonia nigrescens*, and *Microcystis aeruginosa* (*M. aeruginosa*) have been identified for their high heavy metal and *As* accumulation rates and their possible usage for water treatment (Pal and Paknikar, 2012; Wang et al., 2015). Specifically, *M. aeruginosa* is an algae species abundant in many environments. Ongoing research shows the potential of this organism for removing and detoxifying *As* by means of accumulation and transformation to less toxic inorganic or methylated *As* species (i.e., *As*(V), MMA^V, DMA^V, and trimethylamine-*N*-oxide (TMAO)) as well as their impacted factors (Wang et al., 2015, 2017a). Accordingly, *M. aeruginosa* can play critical roles in *As* bioremediation in aquatic ecosystems (Wang et al., 2015). Meanwhile, *M. aeruginosa* is also one of the most common algal bloom species found in freshwater ecosystems (Pan et al., 2006), often containing elevated *As* concentrations under eutrophic conditions (Yan et al., 2016). Under these circumstances, *M. aeruginosa* potentially accumulates arsenic, which could pose to healthy risks (Wang et al., 2014).

Conventional coagulation are frequently applied to remove algae from water phases, which is one of the most common methods but rate-limiting step used in algal removal (Henderson et al., 2008). The coagulation process is affected significantly by pH and the type of coagulant used, such as commonly applied inorganic iron salts and aluminum chloride, as well as the contaminants present in the water phase (Ghernaout et al., 2010). Consequently, not matter what arsenic loaded by means of *M. aeruginosa* bioremediation approach or by the co-presence of *M. aeruginosa* bloom with *As* contamination in eutrophic condition, important parameters of coagulation for algal removal include pH, the coagulant type and dosage, and *As* content. Therefore, when using coagulation to remove *As*-loaded algae from the water phase, there parameters should be optimized to understand its optimal condition. Meanwhile, additional budget should be allocated to the management of the toxic algae produced (Nicomel et al., 2016) since the precipitated *M. aeruginosa* may to a certain extent release *As* again into water phases. Thus, environmental safety of the coagulation technology for removing toxic algae should be assessed prior to its use, which has not yet been done so far (Li et al., 2016; Sun et al., 2018). In particular, changes in *As* content and *As* speciation in *As*-loaded *M. aeruginosa* were not addressed clearly after in-situ or ex-situ disposal in water phases after algal coagulation and precipitation.

The objectives of this study were 1) to explore to what extent in-situ changes occur in total dissolved *As* (TAs) and *As* species in a simulated sedimentation tank under the obtained optimal coagulation conditions, which selected *M. aeruginosa* as the algal model; and 2) to examine *As* release from precipitated algae after ex-situ disposal to further assess the environmental safety of off-site disposal of these *As*-loaded algae. Herein, we used two living algal statuses of living and heat-treated cells which cultured in ex-situ disposal water to assess *As* release from the corresponding precipitated algae. In order to obtain precipitated algae, coagulation experiments were conducted based on its optimal coagulation condition of four selected factors with coagulant type (and precise dosages), pH level, and *As* content. Moreover, the optimal coagulation condition was determined by the Taguchi method under its relevant statistical assumptions to determine optimal coagulation condition for removing *M. aeruginosa* (Wang et al., 2017a; Zolfaghari et al., 2011). This obtained information can pave a road to predict the potential risks of *As* release from precipitated algae in-situ in sedimentation tanks and from precipitated algae ex-situ disposal in waters and to understand its potentially appropriate disposal.

2. Materials and methods

2.1. Algae cultivation

M. aeruginosa were obtained from the Freshwater Algae Culture Collection at the Institute of Hydrobiology (FACHB-collection), Chinese Academy of Sciences, and then cultivated them in an axenic BG-11 medium at 25 ± 1 °C under fluorescent light (40 W, 16 h light/8 h dark) in an incubation shaking chamber at 125 rpm. We harvested algae for experiments during the early stationary phase. Additionally, we exposed approximately 2.0 L *M. aeruginosa* (approximately from 3 to 5×10^7 cells/mL) *As*(V) contaminated algal cells to an additional exposure of 1.0, 10.0, and 50.0 μM *As*(V) ($Na_3AsO_4{\cdot}12\ H_2O$) in a sterile nutrient solution (BG-11) for 96 h. The obtained algae were utilized to conduct our following algal coagulation tests.

2.2. In-situ changes of arsenic concentrations and speciation in sedimentation tank under optimal coagulation conditions

To understand whether *As* contaminated alga cause potential eco-environmental risks through the release of *As* to the water phase as well as changes in speciation within the water phase after coagulation treatments, we investigated in-situ changes in TAs and *As* species in the water phase in a simulated sedimentation tank under the optimal coagulation condition (shown in Supplementary information, SI). First, we validated algal removal efficiency at the optimal coagulation conditions, which applied the most suitable coagulant at its optimal dosage, pH level, and *As*(V) content. As shown in Section 2.1 and 2.2 of SI, the optimal coagulation condition is 25 mg/L PACl, 1 μM *As*(V), and pH 6 (Fig. S3). In order to further validate algal removal efficiency by coagulation using the optimal coagulant type, its dosage, and pH level, we selected another common *As* concentration (10 μM) to conduct our coagulation experiments (Yan et al., 2016). Also shown in Section 2.3 of SI, the predictive values of OD removal efficiency for *As* contaminated algae were determined to be 99.7% ± 0.2% and 96.6% ± 0.5% for the 1.0 μM and 10.0 μM treatments, respectively in the orthogonal test design. Obviously, the optimal condition was favorable to removal of algal cell number.

Following this, we prepared 500 mL of algae (with an initial cell density of approximately 5×10^7 cells/mL) from the 2.0 L algae solution pretreated with the two *As* concentrations (1.0 and 10 μM) for 96 h. The algal cell density was calculated from significant correlation between cell density and OD, which was measured at 680 nm. The used volume of the chosen coagulant was calculated by applying a simple volumetric analogy between the prepared coagulant stock solution and the used volume of the test algae. The TAs and *As* species in the supernatant were investigated at different time intervals (0, 0.5, 1, 3, and 6 d) over a period of 6 d, which started immediately at the end of the coagulation process (Wang et al., 2017a). For each interval, 10 mL of the sample was taken from a medium depth of the beaker (the simulated sedimentation tank) using a syringe. Three replications were conducted for each *As* pretreatment level.

2.3. Ex-situ arsenic release from precipitated algae under optimal coagulation conditions

To assess the environmental safety of off-site disposal of the *As* loaded algae, we further determined concentrations of *As* released from the precipitated algae and the corresponding speciation following coagulation during ex-situ disposal in water. After conducting the aforementioned coagulation experiments under the selected optimal coagulation conditions, we first collected approximately 200 mL precipitated algae using centrifugation, which we rinsed twice with deionized water, and then resuspended the algae into a new 100 mL diluted BG11 (1/10) medium, which was utilized to simulate the ex-situ disposal water with a relatively cleaner condition. The collected precipitated algae were first separated into two equal parts: the first part was resuspended in the diluted BG11 medium as living algae, and the second part was heat-treated in a water bath for 5 min at 50 °C, which was defined as heat-treated algae but keep intact cell (Wang et al., 2014; Miao and Wang, 2006). We then observed *As* release and its corresponding speciation from both the living and heat-treated algal cell suspensions under two time statuses: a short-term duration (24 h) and a long-term duration (6 d). The observational time intervals were 30 min, 3 h, 6 h, 12 h, 24 h for short-time duration, and 0.5 d, 1 d, 3 d, and 6 d for long-term duration. For each time interval, 10 mL aliquots were taken from the suspensions to determine intracellular *As* content and *As* speciation in algal cells (Wang et al., 2017a, 2014). Three replications were conducted for each experiment.

A simple first-order kinetic equation was then used to simulate *As* release from the precipitated algal cells (Bradac et al., 2009):

$$k_e = -1/t \times LnC_t/C_0$$

where C_0 and C_t represent the intracellular *As* content (μg g^{-1}) at the start and time t (h) of release, respectively; k_e is the release rate constant (h^{-1}).

2.4. Analytical methods

Total *As* concentrations in the water phase and algal cell were determined using a 7500a inductively coupled plasma-mass spectrometer (ICP-MS, Agilent Technologies Inc., United States of America). We also used the 7500a ICP-MS coupled with a HPLC 1100 (i.e., HPLC-ICP-MS, Agilent Technologies Inc.) to determine *As* species (*As*(III) and *As*(V), dimethylarsinic acid (DMA), and monomethylarsonic acid (MMA)). These species were separated using a 10 mm RP-X100 (Hamilton Robotics, Inc., United States of America), an anion-exchange column, fitted with a matched guard column (Hamilton Robotics, Inc., 11.2 mm and 12–20 mm) (Wang et al., 2013a).

2.5. Statistical analysis

All experiments were independently repeated three times, and data were recorded as means with their corresponding standard deviations (SD). SPSS 12.0 was used to perform statistical analysis on the data. We analyzed the Pearson correlation coefficients between arsenic contents and precipitation time. Additionally, One-way analysis of variance (ANOVA) was used to determine the differences between the resultant data among the different time.

3. Results and discussion

3.1. In-situ changes of arsenic concentrations and speciation in simulated sedimentation tank under optimal coagulation conditions

Concentrations of dissolved TAs in both the 1.0 and 10.0 μM *As* treatments decreased with increasing precipitation time (Fig. 1). For the 1.0 μM *As* treatment, *As* removal efficiency was 13.8% on day 6, but for the 10.0 μM treatment it was only 4.6% on day 6. Since algae can

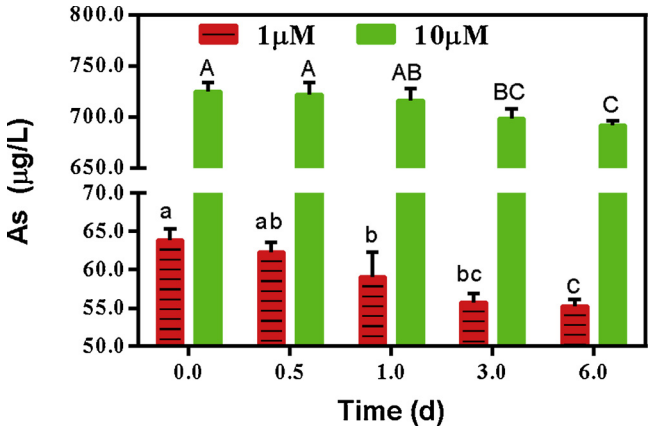

Fig. 1. Dissolved TAs concentrations of in situ supernatant in the simulated sedimentation tank for the 1 μM and 10 μM *As*(V) contaminated water treatments. Different letters on the bars represent significant differences ($P < 0.05$).

effectively bioremediate *As* contamination in water, *M. aeruginosa* could be utilized in more batches to accumulate more *As* from *As* contaminated water until reaching the appropriate removal efficiency of *As*. Furthermore, results from Pearson correlation coefficients yielded -0.70 ($P < 0.01$) and -0.82 ($P < 0.01$) between TAs and precipitation time for the 1.0 and 10.0 μM *As* treatments, respectively. This implied that the more favorable condition for algal removal is a slow rate of *As* decrease in an in-situ simulated sedimentation tank due to *As* adsorption onto algal cells and co-precipitation between PACl and algal cells during coagulation. In addition, as it pertains to *As* removal during algal coagulation, the uptake capacity of PACl reached 184 μM *As* /g, which was close to the maximum uptake capacity of polyaluminum granulates (i.e., 198 μM /g *As*(V) in synthetic water media) (Mertens et al., 2012). This indicates PACl used in this study exhibited comparable adsorption capacity of *As* with other aluminum-based sorbents (Mertens et al., 2012).

At the same time, Pearson correlation coefficients between *As*(V), *As*(III), and DMA concentrations and precipitation time were -0.77 ($P < 0.01$), 0.82 ($P < 0.01$), and 0.93 ($P < 0.01$) for the 1.0 μM *As*(V) treatment, and -0.62 ($P < 0.05$), 0.93 ($P < 0.01$), and 0.99 ($P < 0.01$) for the 10.0 μM *As*(V) treatment, respectively (Fig. 2). Along with decreasing *As*(V) concentrations in the simulated sedimentation tank, *As*(III) and DMA increased with precipitation time (Fig. 2), and we particularly observed a significant increase after day 3. This demonstrated that *As*(V) biotransformation continued in the sedimentation tank after algal precipitation removal, particularly after day 3. Specifically, the total *As* concentration comprised of approximately 3.7% and 2.2% *As*(III) and 2.3% and 0.7% DMA for the 1.0 and 10.0 μM *As*(V) treatments, respectively, on day 6. This differed from *As*(V) biotransformation by *M. aeruginosa* alone in culture media, where the conversion rate to DMA was 0.4% and only negligible *As*(III) was detected in media (Wang et al., 2013a). Additionally, moderately reductive conditions in the in-situ aquatic environment developed with precipitation time (33.5~-75.0 mV redox potential; Table S4). This implied that reductive environments after algal precipitation typically favor the presence of *As*(III) (Katsoyiannis and Zouboulis, 2004; Tang et al., 2019). Although *As*(V) remained the major dissolved *As* species under near-neutral conditions, the higher toxicity level of *As*(III) aggravated potential eco-environmental risks. Similarly, the presence of *As*(III) was observed in *Closterium aciculare* (Hasegawa et al., 2001). This phenomenon must therefore be taken into account when using the coagulation method with proper precipitated duration to remove algal cells for purposes of bioremediation in *As* contaminated water.

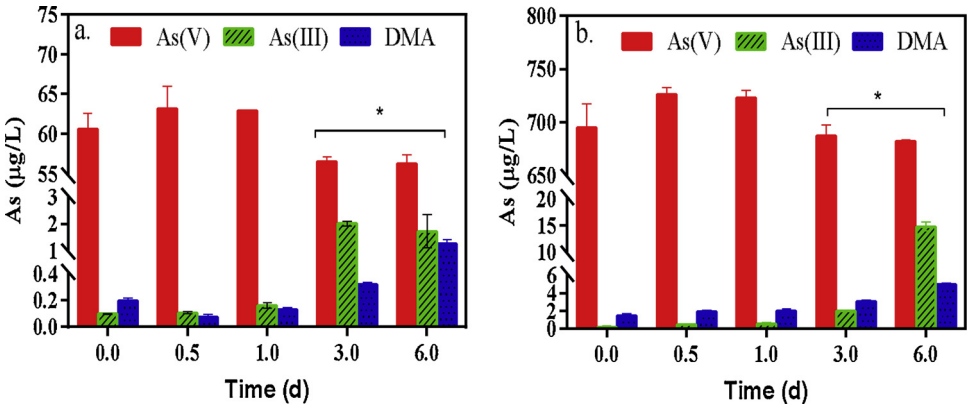

Fig. 2. Transformation of *As* species in water phase for the (a) 1.0 μM and (b) 10.0 μM *As*(V) contamination treatments in the simulated sedimentation tank with precipitation time. * on the bars represents significant differences while compared to other groups ($P < 0.05$).

3.2. Arsenic release from ex-situ disposed coagulated and precipitated algae

3.2.1. Short-term arsenic release

Precipitated *M. aeruginosa* accumulated *As* to average concentrations of 54.7 ± 6.2 and 384 ± 28 μg *As* g^{-1} (dw) in cells after pre-exposure to 1.0 μM and 10.0 μM *As*(V), respectively (Fig. 3). During the first 3 h release period, intracellular *As* concentrations exhibited a sharp decrease in both living and heat-treated algal cells (Fig. 3). Additionally, short-term *As* release rate constants (K_e) of precipitated algal cells after the 1.0 μM *As*(V) treatment was more than four times higher compared to the 10.0 μM *As*(V) treatment (Table 1). To some extent, short-term rapid *As* release from precipitated algae presented potentially higher risks at lower *As* levels. Furthermore, for the 1.0 μM *As*(V) treatment, the *As* release rate was a factor of 1.13 higher for living cells compared to heat-treated cells (Table 1). On the other hand, for the 10.0 μM *As*(V) treatment, *As* release occurred more rapidly for heat-treated cells than living cells (Table 1). Specifically, calculated K_e of intracellular *As* in heat-treated cells was higher by an approximate factor of 1.57 compared to living cells. Like our previous findings, *As* release from heat-treated cells was higher than living cells after 24 h exposure to 10 μM *As*(V) under ± phosphorus treatments (Wang et al., 2014). This indicated that changes in metal binding ligands on cell surfaces after heating could have a dramatic impact on *As* released from algal cells highly contaminated by *As*, but such changes were negligible for low *As* contaminated algal cells due to their relatively rapid release of *As* (Table 1). Additionally, *As* detoxification mechanisms, such as biotransformation, could play a more effective role in *As* release from living algal cells compared to heat-treated algal cells under conditions of low *As* contamination (Qin et al., 2009). Release rate constants determined in this study were higher than those of Cd^{2+} and Zn^{2+} (0.029–0.055 h^{-1}) in *M. aeruginosa* (Hajdu et al., 2010) and *As* in *Scenedesmus obliquus* (0.068–0.11 h^{-1}) and *Chlamydomonas reinhardtii* (0.10–0.12 h^{-1}) (Wang et al., 2013b). Collectively, we must take into account the high potential short-term risk of *As* release from heat-treated precipitated *M. aeruginosa* cells in water highly contaminated by *As*.

Table 1
Short-term release rate constants (K_e; h^{-1}) for living and heat-treated algal cells, ex-situ disposed after pre-exposure to 1.0 and 10.0 μM *As*(V), coagulation and precipitation. Data are mean ± SD ($n = 3$).

Parameters	Living algae		Heat-treated algae	
	1 μM	10 μM	1 μM	10 μM
K_e (h^{-1})	2.63 ± 0.63	0.35 ± 0.04	2.37 ± 0.58	0.55 ± 0.14
$T_{1/2}$ (h)	0.26	1.98	0.29	1.26
R^2	0.94	0.97	0.93	0.93

3.2.2. Long-term arsenic release

Intracellular *As* concentrations significantly decreased in both living and heat-treated cells during long-term release (6 d) (Fig. 4). Relative *As* percentages in algal cells retained at the end of the release period were 13.9% (living) and 7.8% (heat-treated) after the 1.0 μM *As*(V) treatment, and 4.4% (living) and 3.7% (heat-treated) after the 10.0 μM *As*(V) treatment (Fig. 4). Therefore, the higher risk of long-term *As* release at higher *As*(V) concentrations in the 10.0 μM *As*(V) treatment (compared to the 1.0 μM *As*(V) treatment) should certainly be of great concern.

Furthermore, during long-term *As* release processes in our study, the predominant species in algal cells was *As*(V), which accounted for a range of 74.8%–98.4% of TAs, and decreased with increasing release time (Fig. 4). Moreover, *As*(III) was found in all algal cells, increasing from 4.6% on day 1 to 25.2% on day 6 after the 1.0 μM *As*(V) treatment, and increasing from 1.3% to 15.9% after the 10.0 μM *As*(V) treatment (Fig. 4). In particular, we detected no methylated *As* species in algal

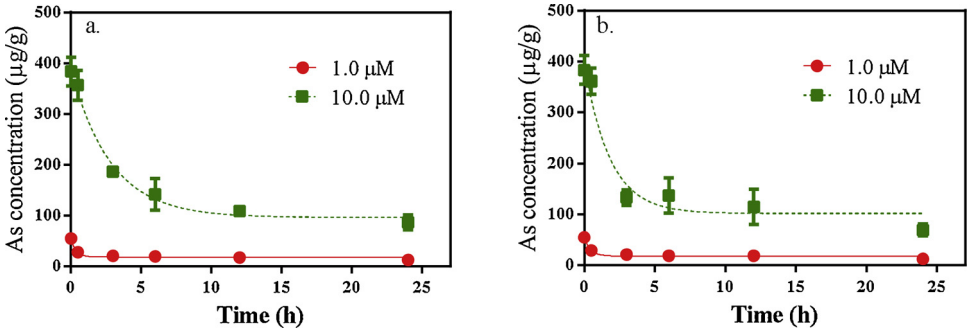

Fig. 3. Intracellular *As* changes in short-term (24 h) ex situ disposed living (a) and heat-treated (b) precipitated algal cells pre-exposed to 1.0 and 10.0 μM *As*(V).

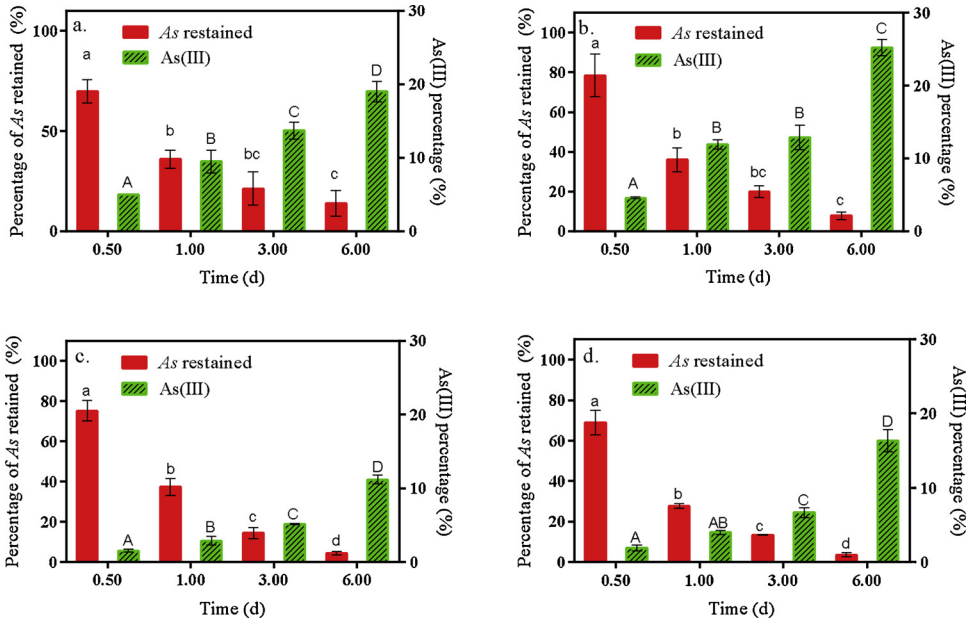

Fig. 4. Percentage of *As* retained and *As*(III) changed in long-term release of ex situ disposed living and heat-treated precipitated algal cells (6 d) pre-exposed to the 1.0 (a - living cell and b – heat-treated cell) and 10.0 (c - living cell and d – heat-treated cell) μM *As*(V). Different letters on the bars represent significant differences ($P < 0.05$).

cells possible owing to their restrictions under detected limits and rapid extracellular excretion (Hellweger and Lall, 2004). We know that *M. aeruginosa* can reduce *As*(V) to *As*(III) and methylate the *As* to MMA and DMA (Wang et al., 2013a; Zhu et al., 2017). Also, the rapid release of *As* (V) can cause an obvious increase in *As*(III) quotas inside algal cells. Although we observed rapid *As*(V) release and its active diffusion of its high concentrations into algal cells, *As*(III) generation and excretion was not easy, especially for heat-treated precipitated algal cells. Our previous study also confirmed that *As* release from heat-treated cells was higher than from living cells and was not influenced by different *As* speciation (Wang et al., 2014).

Table 2 shows the calculated long-term release rate constants (k_e and k'_e) and biological retention half-life ($t_{1/2}$) of both *As*(V) and *As*(III) species in both heat-treated and living algal cells after pre-exposure to the 1.0 μM and 10.0 μM *As*(V) treatments and subsequent coagulation. For the 1.0 μM *As*(V) treatment, calculated intracellular k_e values of *As* (V) and *As*(III) were both relatively higher in living cells than in heat-treated cells (by an approximate factor of 1.22 and 2.20, respectively). This showed that the intracellular reduction of *As*(V) to *As*(III) occurred more rapidly in living cells than heat-treated cells, and the high quotas led to significant *As*(III) excretion, especially under low *As* conditions. This was consistent with simulated results that showed that *As*(V) and *As*(III) release was higher in living cells than in heat-treated cells in media, and that *As* biotransformation and detoxification in living cells was superior to *As* release in heat-treated cells under conditions of relatively low *As* contamination (Hellweger and Lall, 2004). However, for the 10.0 μM *As*(V) treatment, calculated intracellular k'_e of *As*(V) and *As* (III) in heat-treated cells was higher by a factor of 1.28 and 1.78 compared to living cells, respectively. The higher *As* release rate from heat-treated cells after the high *As* treatment illustrated that the relatively high *As* concentrations in heat-treated algal cells underwent more rapid *As* dissociation and subsequently release than *As* biotransformation and detoxification in living cells during the 6 d long-term release treatment. This demonstrated that the precipitated *M. aeruginosa* with high *As* concentrations had a significantly higher risk of *As* release from heat-treated algal cells after longer terms compared to living cells.

Table 2
Long-term (6 d) release rate constants (k_e; d^{-1} and k'_e; d^{-1}) for living and heat-treated algal cells, ex-situ disposed after pre-exposure to 1.0 and 10.0 μM *As*(V), coagulation and precipitation. Data are mean ± SD ($n = 3$).

Parameters		Living algae		Heat-treated algae	
		As(V)	*As*(III)	*As*(V)	*As*(III)
1 μM	k_e (d^{-1})	1.15 ± 0.30	4.05 ± 2.72	0.94 ± 0.31	1.84 ± 0.82
	$t_{1/2}$ (d)	0.61	0.17	0.73	0.38
	R^2	0.92	0.85	0.90	0.80
10 μM	k'_e (d^{-1})	0.90 ± 0.24	1.78 ± 0.37	1.15 ± 0.33	2.05 ± 0.61
	$t'_{1/2}$ (d)	0.77	0.39	0.60	0.34
	R^2	0.93	0.95	0.94	0.90

3.3. Potential implications for appropriate disposal

To date, relevant studies have only reported on individual removal of algae or *As* in waters by means of coagulation (Wu et al., 2011; Kubinakova et al., 2017; Nidheesh and Singh, 2017), filtration (Swanepoel et al., 2017; Kwon et al., 2014), adsorption (Mertens et al., 2012; Wei et al., 2019), and other related approaches (Wang et al., 2017b). In contrast, this study presents the first combined evidence for *As* release from the precipitated algae after coagulation and its subsequent potential risks in the case of *M. aeruginosa* removal in *As*(V) contaminated water. Furthermore, the reduction status was found to be generated following algal cell precipitation, which was indicated by a redox potential of 33.5~-75.0 mV after 6 d precipitation. *As*(III) and DMA levels could be increased in solutions under this reduction status (Mertens et al., 2012; Tang et al., 2019). This result differed from our previous investigation that only trace amounts of *As*(III) were found after *As*(V) biotransformation in *M. aeruginosa* suspensions, and that *As* (V) reduction was the rate limiting step for *As* methylation to transpire in *M. aeruginosa* (Wang et al., 2013a). Additionally, *M. aeruginosa* could effectively be removed by PACl coagulation, while *As* concentrations in solutions decreased over time as species were transformed in the weakly reducing environment (Table S4). Furthermore, after ex-situ disposal, it exhibited that *As* released from precipitated *M. aeruginosa* depended not only on *As* content in the algal cells but also the living status of the algae during ex-situ disposal. Actually, high *As* release risks resulted from the facts of the rapid *As* biotransformation and detoxification of living precipitated algae under low *As* concentration and the passive, and rapid release of *As* from heat-treated precipitated algal cells under high *As* concentration. Collectively, continuous *As* removal within in-situ precipitated alga must be acknowledged; also, more attention should be paid to potential *As* release risks from the disposal of precipitated alga being stored ex-situ. Therefore, the appropriate methods of disposing these precipitated algae are necessary to take

account into their rational in-situ or ex-situ ways at given proper time when release of *As* and formation of arsenite are relatively lower.

Additionally, *As* metabolism in algae is quite complex, often involving *As* oxidation and reduction, *As* uptake, and excretion as well as their diverse influencing environmental factors including pH, oxygen, phosphorus, nitrogen and other potential co-existed contaminants in water phase (Wang et al., 2017a, 2019; Al Mamun et al., 2019). In practice, this study has still several limitations of insufficient data such as arsenic levels and potential influencing factors since water is diverse matrix. More studies on this subject are therefore necessary to understand how *As* release from precipitated algae as ex-situ disposal is affected by various influencing factors in natural aquatic environments.

4. Conclusions

Algal coagulation removal favored a continuous decrease of dissolved *As* in an in-situ coagulation environment. Meanwhile, *As*(III), which increased in such environments, could also aggravate potential risks due to its higher toxicity. Therefore, when using the coagulation method to remove *As*-contaminated algae, proper precipitated duration should be taken into account. Additionally, the *As* release from precipitated *M. aeruginosa* depended not only on the *As* concentration in algal cells but also on the living status of algae during ex-situ disposal. Living precipitated algae released *As* more rapidly than heat-treated precipitated algae under low *As* concentrations. However, the opposite was true for high *As* concentrations under long-term treatments. Our obtained knowledge can help understanding on the proper disposal of precipitated algae with *As*-contaminated water.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jhazmat.2019.121249.

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