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1 Mobilization of arsenic from contaminated sediment by anionic 2 and nonionic surfactants

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ABSTRACT

The increasing manufacture of surfactants and their wide application in industry, 17
 agriculture and household detergents have resulted in large amounts of surfactant 18
 residuals being discharged into water and distributed into sediment. Surfactants have the 19
 potential to enhance arsenic mobility, leading to risks to the environment and even human 20
 beings. In this study, batch and column experiments were conducted to investigate arsenic 21
 mobilization from contaminated sediment by the commercial anionic surfactants sodium 22
 dodecylbenzenesulfonate (SDBS), sodium dodecyl sulfate (SDS), sodium laureth sulfate (AES) 23
 and nonionic surfactants phenyl-polyethylene glycol (Triton X-100) and polyethylene glycol 24
 sorbitan monooleate (Tween-80). The ability of surfactants to mobilize arsenic followed the 25
 order AES > SDBS > SDS ≈ Triton X-100 > Tween 80. Arsenic mobilization by AES and Triton 26
 X-100 increased greatly with the increase of surfactant concentration and pH, while arsenic 27
 release by SDBS, SDS and Tween-80 slightly increased. The divalent ion Ca²⁺ caused greater 28
 reduction of arsenic mobilization than Na⁺. Sequential extraction experiments showed that 29
 the main fraction of arsenic mobilized was the specifically adsorbed fraction. Solid phase 30
 extraction showed that arsenate (As(V)) was the main species mobilized by surfactants, 31
 accounting for 65.05%–77.68% of the total mobilized arsenic. The mobilization of arsenic was 32
 positively correlated with the mobilization of iron species. The main fraction of mobilized 33
 arsenic was the dissolved fraction, accounting for 70% of total mobilized arsenic. 34

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49 Introduction

50 Arsenic is ubiquitously distributed in aquatic environments,
 51 soil and sediment. Both natural phenomena including
 52 weathering and biological activity and anthropogenic pro-
 53 cesses such as mining activities, fossil fuel combustion and
 54 use of pesticides contribute to the input of arsenic in the
 55 environment (Cullen and Reimer, 1989). The release of arsenic
 56 from sediment has attracted much attention, as the process

could directly elevate arsenic concentrations in surface and 57
 ground water, causing severe risks to organisms and human 58
 health. The arsenic mobilized from contaminated sediment 59
 has become the major source of arsenic in groundwater in 60
 many countries and regions, where the concentrations have 61
 often exceeded the WHO drinking water limit of 10 µg/L (Islam 62
 et al., 2004; Nickson et al., 2000; Polizzotto et al., 2008). The 63
 predominant species of arsenic in contaminated sediment 64
 include arsenate (As(V)), arsenite (As(III)) and organic arsenic 65

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66 compounds, among which As(III) is more toxic and mobile
67 than As(V) and organic complex fractions. Mobilization of
68 arsenic by coexisting substances is an important factor
69 contributing to arsenic release to water. The most widely
70 studied substances relating to the mobilization of arsenic so
71 far have been natural organic matters. In addition, coexisting
72 anions such as PO_4^{3-} and SO_4^{2-} are well-known substances
73 contributing to the displacement of arsenic from sediment
74 (Bauer and Blodau, 2006; Guo et al., 2011; Mladenov et al., 2010;
75 Routh et al., 2007; Sharma et al., 2011; Wang and Mulligan,
76 2009a, 2009b).

77 Surfactants are groups of amphiphilic compounds that
78 consist of both polar hydrophilic groups and hydrophobic
79 hydrocarbon chains. Due to their ability to reduce interfacial
80 tension, surfactants are widely used in detergent products,
81 mining activities and enhanced oil recovery (Basar et al., 2004;
82 Bennie et al., 1997; Céspedes et al., 2008; Clara et al., 2007; Zhang
83 et al., 2013). This wide range of applications leads to large
84 amounts of surfactant residuals being discharged to water
85 treatment plants or directly into the water environment.
86 Different concentrations of surfactants have been detected in
87 wastewater, sewage treatment plant effluents, water bodies
88 and sediments. For example, the concentrations of some linear
89 alkylbenzenesulfonates (LAS) in wastewater and treated sludge
90 have been reported to reach 1090 $\mu\text{g/L}$ and 30,200 mg/kg ,
91 respectively (Lara-Martín et al., 2006, 2008; Ying et al., 2002). In
92 natural water, levels of LAS were found to be up to 416 $\mu\text{g/L}$
93 (Lara-Martín et al., 2006, 2008; Marcomini et al., 2000; Shalaby,
94 2007; Ying et al., 2002; Ying, 2006; Zgoła-Grzeskowiak et al.,
95 2009). In addition, surfactants are also efficient at removing
96 organic matters and heavy metals from contaminated soil by
97 solubilization and ligand exchange (Mulligan et al., 2001a;
98 Mulligan, 2005, 2009; West and Harwell, 1992). This indicates
99 the potential of surfactants to enhance the mobility of co-
100 existing organic matters and trace metals from sediments or
101 soil. Previous research has reported that arsenic in mine tailings
102 could be greatly mobilized by biosurfactants (Wang and
103 Mulligan, 2009c). In our previous research, the anionic surfac-
104 tant sodium dodecyl benzene sulfonate (SDBS) and nonionic
105 surfactant Triton X-100 were found to reduce arsenic adsorp-
106 tion onto ferrihydrite and enhance arsenic transport through
107 columns packed with ferrihydrite-coated sand (Liang et al.,
108 2016). However, there has been a lack of study on the potential
109 of commercial surfactants to mobilize arsenic. Thus, in the
110 current study, five commercial surfactants were chosen to
111 evaluate the potential of surfactants to mobilize arsenic from
112 contaminated sediment. SDBS, sodium dodecyl sulfate (SDS) and
113 sodium laureth sulfate (AES) are the most widely used anionic
114 surfactants. The nonionic surfactants phenyl-polyethylene gly-
115 col (Triton X-100) and polyethylene glycol sorbitan monooleate
116 (Tween-80) are also widely used in industrial applications.
117 Elucidating the effects of surfactants on arsenic mobilization
118 from sediment is of great importance in understanding the
119 processes causing arsenic release to water. In this investigation,
120 batch and column tests were carried out to study the potential of
121 the five surfactants to mobilize arsenic from contaminated
122 sediment. The effects of surfactant type, surfactant concentra-
123 tion, pH, coexisting cations and time on arsenic mobilization
124 were evaluated. Sequential extraction procedures (SEPs), solid
125 phase extraction (SPE), ultrafiltration and dialysis tests were

conducted to investigate the speciation of released arsenic. 126
Mechanisms relating to arsenic mobilization by surfactants were 127
also discussed. 128

1. Materials and methods 130

1.1. Sediment sample 131

The sediment sample was collected from the Zi River near a 132
tin mining site in Lengshuijiang City, Hunan Province, China. 133
The sediment sample was freeze-dried, ground and passed 134
through a 100-mesh sieve previous to use. For arsenic and 135
metal content measurement, the sediment was first digested 136
using the CEM Mars 6 digestion system. The total concentra- 137
tions of arsenic and metals in the solution from digestion 138
were measured using inductively coupled plasma mass 139
spectrometry (ICP-MS, 7500a, Agilent, USA). The specific 140
surface area of the sediment particles was obtained using 141
the BET- N_2 adsorption method. Content of organic matter was 142
determined according to the weight loss after calcination at 143
550°C for 4 hr. The pH of a mixture of 10.0 g of sample in 144
50 mL ultra-pure water was employed as the sediment pH. 145

1.2. Surfactants 146

Anionic surfactants SDBS ($\text{C}_{18}\text{H}_{29}\text{SO}_3\text{Na}$), SDS ($\text{C}_{12}\text{H}_{25}\text{OSO}_3\text{Na}$) 147
and AES ($\text{C}_{14}\text{H}_{33}\text{O}_3\text{SO}_3\text{Na}$) were purchased from Sino Pharm 148
Chemical Reagents Co., China. Nonionic surfactants Triton 149
X-100 ($\text{C}_{14}\text{H}_{22}\text{O}(\text{C}_2\text{H}_4\text{O})_{10}$) and Tween-80 ($\text{C}_{64}\text{H}_{125}\text{O}_{26}$) were 150
purchased from Sigma Aldrich, USA. All chemicals were of 151
analytical grade and used without further purification. The 152
contents of arsenic in the surfactants were negligible. 153

1.3. Batch experiments 154

Batch experiments were conducted to investigate arsenic 155
mobilization from sediment by surfactants at various concen- 156
trations, over a range of pH conditions and in the presence of 157
different metal ions. The mobilization of iron by surfactants 158
was also evaluated in batch experiments. 2.0 g sediment 159
samples and 40 mL solutions with prescribed surfactant 160
concentrations, pH values, and Na^+ or Ca^{2+} concentrations 161
were added to a batch of 50 mL centrifuge tubes. The tubes 162
were shaken at 150 r/min and 25°C for 24 hr. Then the 163
suspensions were centrifuged at 8000 r/min for 15 min and 164
the supernatants were collected for arsenic and/or iron con- 165
centration analysis. 166

The time dependence of arsenic mobilization was studied 167
in batch tests. 5.0 g sediment samples and 500 mL of 100 mg/L 168
surfactant solutions were added to 500 mL bottles. The 169
suspensions were continuously stirred at 25°C. The initial pH 170
of the suspensions was adjusted to 7.0 and readjusted to 7.0 at 171
prescribed time intervals using HCl or NaOH solutions. For 172
comparison, bottles containing 5.0 g sediment samples and 173
500 mL deionized water were stirred under the same condi- 174
tions. At prescribed time intervals, 2.0 mL aliquots of the 175
suspensions were taken and centrifuged at 8000 r/min for 176
15 min. The supernatant was collected for arsenic concen- 177
tration analysis. In the batch tests, all the solutions were 178

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