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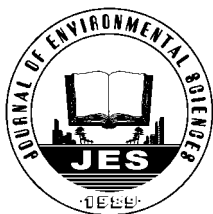
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Tannic acid and saponin for removing arsenic from brownfield soils: Mobilization, distribution and speciation

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ABSTRACT

Plant biosurfactants were used for the first time to remove As and co-existing metals from brownfield soils. Tannic acid (TA), a polyphenol, and saponin (SAP), a glycoside were tested. The soil washing experiments were performed in batch conditions at constant biosurfactant concentration (3%). Both biosurfactants differed in natural pH, surface tension, critical micelle concentration and content of functional groups. After a single washing, TA (pH 3.44) more efficiently mobilized As than SAP (pH 5.44). When both biosurfactants were used at the same pH (SAP adjusted to 3.44), arsenic mobilization was improved by triple washing. The process efficiency for TA and SAP was similar, and depending on the soil sample, ranged between 50%–64%. Arsenic mobilization by TA and SAP resulted mainly from decomposition of Fe arsenates, followed by Fe^{3+} complexation with biosurfactants. Arsenic was efficiently released from reducible and partially from residual fractions. In all soils, As(V) was almost completely removed, whereas content of As(III) was decreased by 37%–73%. SAP and TA might be used potentially to remove As from contaminated soils.

Introduction

Arsenic is a fairly common pollutant. According to the U.S. Environmental Protection Agency (US EPA, 2002), it is the second most common contaminant, after Pb, at National Priorities List sites. Moreover, arsenic was ranked as the most hazardous pollutant of 275 substances by the Agency for Toxic Substances and Disease Registry (ATSDR, 2011). The greatest environmental threat is posed by sites connected with mining, refining, grinding and concentrating As and metal ores and disposal of their tailings (Kwon et al., 2012). These activities are the main reason for extremely high As concentrations in the surrounding soils. For example, in mine dump soils around the Dalsung Cu-W mine in southeast Korea they ranged between 539 and 9380 mg/kg (Jung et al., 2002). Smith et al. (2008) found that in soils from former gold

mining areas in Victoria (Australia), As concentration was over 11,000 mg/kg. Maximum As concentration from an abandoned mining area in Zlata Idka (Slovakia) exceeded 13,000 mg/kg (Rapant et al., 2006). An even higher As content (up to 26,500 mg/kg) was detected in mining soils in Spain. In Poland, soil contamination with As affects the lower Silesia district as a result of former gold and arsenic mining and processing (Krysiak and Karczewska, 2007). Arsenic-contaminated soils must be treated using effective and safe methods due to the high toxicity of As.

Arsenic concentration in soil can be permanently lowered below permissible levels by soil washing. Many chemical agents have been used to increase the efficiency of arsenic removal. They can be classified into three groups: (1) inorganic salts, (2) inorganic and organic acids, and (3) alkaline agents (Giacomino et al., 2010). Recently, non-toxic and biodegradable washing agents have been tested, including cyclodextrins (Chatain et al., 2004), humic acids (Wang and Mulligan, 2009a, 2009c) and biosurfactants (Wang and Mulligan, 2009b, 2009d).

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Biosurfactants are amphiphilic compounds synthesized extracellularly or as part of the cell membrane by a wide variety of microorganisms or produced by plants (Mulligan, 2009). Almost all biosurfactants are either non-ionic or anionic, they reduce surface and interfacial tension and form micelles. Consequently, they can effectively solubilize, disperse and desorb both organics (e.g. petroleum hydrocarbons) and metals (e.g. Cd, Cu, Pb) from soils or sediments (Mulligan, 2009).

However, only a few investigations have been performed on arsenic removal by biosurfactants. Until now, only microbial rhamnolipids were examined (Wang and Mulligan, 2009b). As anionic agents they are able to enhance the removal of arsenic and heavy metals from mine tailings under alkaline conditions. On the other hand, plant biosurfactants represented by glycosides and polyphenols, commonly found in many plant species, are potentially attractive for metal removal because they naturally show acidic character and contain numerous functional groups. In the literature, there are some evidences indicating that saponin efficiently removes some heavy metals (e.g. Cu, Cd, Zn, Pb) from different soils (Hong et al., 2002, Gusiatin and Klimiuk, 2012). However, these plant biosurfactants have not been used to remediate arsenic contaminated soils.

Therefore, the objective of this study was to examine and compare how effectively two plant biosurfactants (tannic acid (TA) and saponin (SAP)) mobilize arsenic from soils affected by mining and smelting, using two washing modes (single and triple). Both biosurfactants were characterized in terms of their surface properties. Moreover, changes in arsenic distribution and speciation after soil washing were analyzed.

1 Materials and methods

1.1 Study site, soil sampling and characterization

Soil surface samples (0–30 cm) were collected in an area formerly used for arsenic mining and smelting in Zloty Stok (50°26'47"N, 16°52'59"E) lower Silesia (Poland). Three sites were chosen for soil sampling (**Fig. 1**), marked as soil 1 (S1): the foreground of the tailings reservoir in the former arsenic ore processing plant near the valley of the Trujaca stream in Zloty Stok, soil 2 (S2): the area affected by tailings reservoirs in the valley of the Trujaca stream between Zloty Stok and the village of Blotnica, soil 3 (S3): a forest area of mine spoils and slag dumps in the valley of Zloty Potok.

From each site, about 10 kg of representative soil was collected. In the laboratory, the samples were air-dried (2 weeks) and ground to pass through a 1 mm sieve, then homogenized and characterized in terms of their physicochemical properties. Particle size was determined using

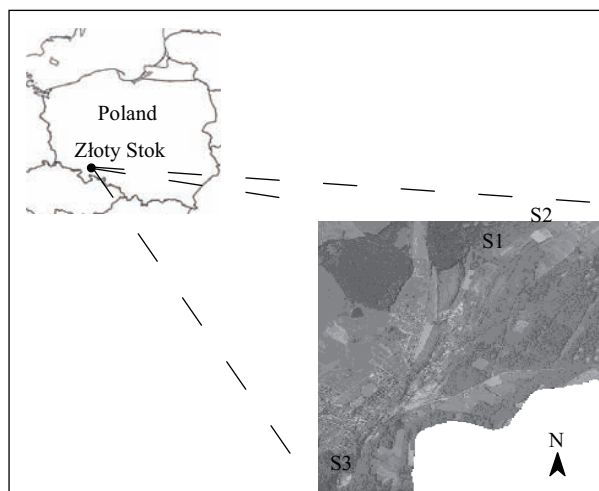


Fig. 1 Soil sampling area in Zloty Stok, South Poland.

a Mastersizer 2000 (Malvern Instruments, UK) analyzer and soil organic matter was determined by the Tiurin method (Ostrowska et al., 1991). The cation exchange capacity (CEC) of the soils was calculated as a sum of the hydrolytic acidity (in 1 mol/L $\text{Ca}(\text{CH}_3\text{COO})_2$) and the exchangeable bases (in 0.1 mol/L HCl) (Ostrowska et al., 1991). The equilibrium pH of the soil in distilled water (1:2.5 ratio, W/V) was measured with a pH meter (Hanna Instruments, Germany). Total As and metal (Cu, Fe, Ni, Pb, Zn) content in the soils was measured using a flame atomic absorption spectrometer (AA 280FS, Varian, Australia) after microwave digestion of samples (MARSSXpress, CEM, USA) using a HCl:HNO₃ mixture at a 3:1 ratio (V/V). The operational conditions for As and metal analysis are given in **Table 1**.

1.2 Plant biosurfactants

Two biosurfactants, TA (Product No. 16201) and SAP (Product No. 84510) were purchased commercially (Sigma-Aldrich, Poland) as powders and used without further purification as washing agents.

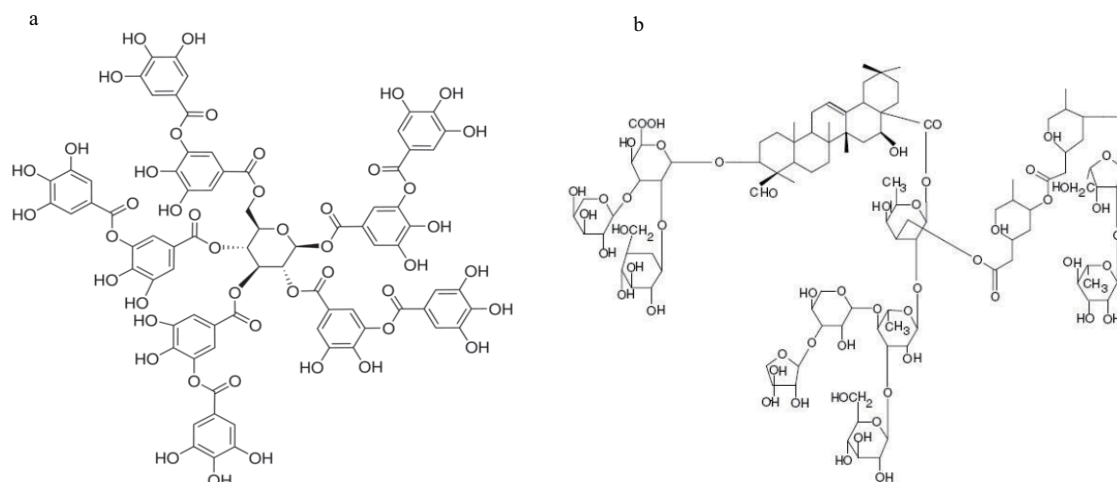
TA is a type of polyphenol consisting of a hydroxyl group (–OH) bonded directly to an aromatic hydrocarbon group. TA is usually extracted from *Caesalpinia spinosa* or *Rhus semialata*. It consists mainly of gallic acid residues linked to glucose via glycosidic bonds (**Fig. 2a**). The chemical formula is $\text{C}_{76}\text{H}_{52}\text{O}_{46}$ and its molecular weight is 1701.2 g/mol. For soil washing, tannic acid with puriss grade was used.

SAP is a mixture of triterpeneglycosides extracted from the bark of the tree *Quillaja saponaria*, in which the hydrophilic part is composed of sugar chains with functional groups (**Fig. 2b**). In the present study SAP of premium quality was used, in which the maximum sapogenin content (non-sugar hydrophobic part) was 25%.

The biosurfactants were characterized in terms of their ability to reduce surface tension and their critical micelle concentration (CMC). Surface tension was measured with

Table 1 Operational conditions for As and metal analysis by flame atomic absorption spectrometry (FAAS)

Parameter	As	Cu	Fe	Ni	Pb	Zn
Wavelength (nm)	193.7	324.8	248.3	232.0	217.0	213.9
Lamp current (mA)	10.0	4.0	5.0	4.0	10.0	5.0
Silt width (nm)	0.5	0.5	0.2	0.2	1.0	1.0
Calibration solutions range (mg/L)	25–100	1.0–6.0	2.0–8.0	1.0–6.0	2.5–15	0.5–2.0
Oxidant (air) flow (L/min)	0			13.5		
Oxidant (N ₂ O) flow (L/min)	11.0			0		
Fuel (acetylene) flow (L/min)	5.93			1.5		
Burner type	Mark 7 (0210164100)			Mark 7 (0210164000)		

**Fig. 2** Plant biosurfactants. (a) tannic acid (TA), (b) saponin (SAP).

a K100 tensiometer (Krüss, Germany) employing the Wilhelmy plate method in aqueous biosurfactant solutions, at concentration range of 0.005%–10% (W/V). For both biosurfactants, total acidity (barium chloride method) and carboxyl acid groups (calcium acetate method) were determined (Kononova, 1968). Hydroxyl (or phenolic OH) groups content was calculated by the difference between total acidity and carboxyl groups content.

1.3 Batch soil washing experiments

The experiments were performed in two ways as single and triple washings. Biosurfactant concentration was 3% (W/V) (Hong et al., 2002; Gusiati and Klimiuk, 2012). In single washing, TA and SAP solutions were used at their natural pH. In triple washing, fresh biosurfactant solution was added each time. In the latter case, the pH of SAP was adjusted with 0.5 mol/L H₃PO₄ to the same value as for TA to compare biosurfactant efficiency under the same conditions. As a control, distilled water was used.

All the soil washing experiments were conducted in 50 mL capacity polyethylene tubes with a soil to biosurfactant solution ratio of 1/40 (W/V). The samples were shaken in a mechanical shaker at 150 r/min for 24 hr at room temperature (22–24°C). At the end of the extraction, the suspensions were centrifuged at 8000 r/min for 20 min

and the supernatants filtered and analyzed for As and other metals. In the soil residues, total As, its distribution and speciation forms were measured.

1.4 Arsenic distribution and speciation in soils before and after washing

Arsenic distribution was established using a modified Community Bureau of Reference (BCR) sequential extraction procedure (Rauert et al., 2000). Four operationally defined fractions were determined: exchangeable and acid soluble (F1), reducible (F2), oxidizable (F3), and residual (F4).

The sequential extraction was performed by shaking 1 g soil (or correspondingly less for samples after washing) with reagents of increasing reactivity in a mechanical shaker. All operational conditions for the BCR procedure are given by Gusiati and Klimiuk (2012). Between the extractions of the F2 and F3 fractions, the soil residue was washed with distilled water (20 mL), shaken for 15 min. and centrifuged for 10 min at 8000 r/min. The supernatants were carefully decanted and filtered into polyethylene tubes. Washing with water was necessary to remove any remaining reagents and metals present in the soil residues from the previous extraction step. After extraction of the oxidizable fraction, the soil residue was dried at 60°C for

4 hr and finally digested in a microwave oven (MARSS-press, CEM, USA) using a HCl:HNO₃ mixture at a 3:1 ratio (V/V) and analyzed by FAAS (Table 1).

For As speciation, three basic species were determined using Chappell's method (Chappell et al., 1995): As(III), As(V) and As_{org}. In this procedure, arsenic is initially extracted from soil by concentrated HCl. As(III) is subsequently extracted into an organic phase (CHCl₃), then into water. Determination of total inorganic As (As_{inorg}) requires reduction of As(V) to As(III) via 50% (W/V) KI. After that As(III) is extracted into an organic phase, and then an aqueous phase. Finally, As(V) is calculated as As_{inorg} – As(III), whereas As_{org} = As_{total} – As_{inorg}. The concentrations of As(III) and As_{inorg} were analyzed by the FAAS method (Table 1).

1.5 Statistical analysis

All the data reported are the averaged values of three repetitions ($n = 3$). Data were statistically evaluated using STATISTICA 9.0 (StatSoft, Inc.). Duncan's test was used, and $p < 0.05$ was considered to be significantly different.

2 Results and discussion

2.1 Soil properties

Selected physicochemical properties are presented in Table 2 for the soils collected in the former arsenic mining and smelting area in Zloty Stok. All soil samples were classified as silty loam (20%–39.7% sand, 58.2%–76% silt, 1.3%–3.6% clay). This texture, especially the low content of clays indicates the soils are suitable to be remediated via soil washing (Mulligan et al., 2001). Depending on the site sampled, the soils varied in pH and organic matter content. The pH for soils S1 and S2 ranged from slightly alkaline to neutral. This was due to the direct influence of the tailings, which were alkaline (pH 7.4–7.6) byproducts of ore processing (Krysiak and Karczewska, 2007). In

contrast, soil S3 developed on mine spoils and slug dumps was acidic and contained the highest content of organic matter (Table 2), which is typical for forest soils due to the contribution of leaf drop. In all soils the CEC was relatively high (30.8–33.6 cmol/kg), which might affect As mobilization by plant biosurfactants.

The investigated soils were highly contaminated with As, but its total concentration depended on the site sampled (Table 2). In all soils, the limit value of As concentration for industrial areas (60 mg As/kg) (OME, 2002) was significantly exceeded (from 34- to 71-fold). Apart from As, the soils contained other metals (Cu, Ni, Pb, Zn, Fe) (Table 2). Among them only Pb in soil S3 was above the limit (600 mg/kg) for industrial areas (OME, 2002).

2.2 Plant biosurfactants characterization

TA and SAP, representing polyphenols and glycosides respectively, revealed different chemical properties (Table 3). SAP more effectively reduced surface tension of water at a notably lower CMC than TA. The CMC is defined as the concentration of a surfactant solution at which the molecules self-aggregate to form spherically shaped micelles (Savaroglu and Yurt, 2011). In contrast to SAP, TA aggregates into micelles at two different concentrations. This is because it forms two types of micelles. Usually, at low concentration spherical micelles are formed, but when the concentration is high enough rod-shaped micelles began to form. Therefore, the CMC given in Table 3 might refer to the concentration at which only spherical micelles occur for SAP, but spherical (CMC 1) and rod-shaped micelles (CMC 2) for TA. According to Urum and Pekdemir (2004), TA and SAP decrease efficiently also interfacial tension that reduces bonding forces between contaminant and soil.

In addition, both biosurfactants differed in total acidity, which is affected by the acid functional groups. The presence of these groups can be important when removing metal cations from soil. For example, carboxyl groups gradually dissociate between pH 2.5 and pH 7 to form carboxylate groups (COO[−]) (Coles and Yong, 2006). For

Table 2 Physicochemical characteristics of soils (standard deviation shown in parentheses, $n = 3$)

Characteristic	Soil S1	Soil S2	Soil S3
Texture	Silty loam	Silty loam	Silty loam
pH _{H2O}	7.9	6.9	4.4
CEC (cmol(+)/kg)	31.5	30.8	33.6
Organic matter (%)	5.95 (± 0.01)	5.88 (± 0.02)	18.1 (± 0.44)
As (mg/kg)	3574.8 (± 183)	4294.2 (± 264)	2041.1 (± 21.1)
Cu (mg/kg)	65.6 (± 0.05)	69.4 (± 2.4)	35.8 (± 3.0)
Zn (mg/kg)	143.5 (± 3.9)	146.9 (± 4.8)	87.4 (± 0.8)
Ni (mg/kg)	26.9 (± 0.05)	31.7 (± 0.81)	25.5 (± 0.5)
Pb (mg/kg)	378.8 (± 1.2)	409.2 (± 18.9)	661.2 (± 11.3)
Fe (g/kg)	35.9 (± 4.9)	48.6 (± 3.3)	34.1 (± 1.2)

CEC: cation exchange capacity.

Table 3 Properties of plant-biosurfactants used in soil washing

Property	TA	SAP
Surface tension (mN/m)	41.8	36.9
CMC (g/L)	0.1 (CMC 1) 12.7 (CMC 2)	0.93 (CMC 1)
COOH groups (meq/100 g)	34.6	366.6
OH groups (meq/100 g)	3351.4	1356.5
Total acidity (meq/100 g)	3386.0	1723.1
pH (for 3% solution)	3.44	5.44

both biosurfactants in the present study, the quantity of hydroxyl groups was higher than that of carboxyl. TA contained 2.5 times more OH groups than SAP. As a result it is characterized by a higher total acidity (lower pH) than SAP.

2.3 Arsenic mobilization in soils by plant biosurfactants

2.3.1 Single washing without pH adjustment

The efficiency of As removal from soils is presented in Fig. 3. The results showed that using distilled water alone removed only minimal amounts of As (less than 4%). Lee et al. (2007) also observed that water extracted little arsenic from stream sediments in the vicinity of an abandoned mine when used at a wide range of pH 2 and pH 12.

Both biosurfactants were used at the same concentration (3%), which exceeds their CMCs. This means that in SAP solution, spherical aggregates occurred, and in TA they were rod-shaped. Although SAP more efficiently reduces the surface tension of water (Table 3), which potentially may favor As mobilization, it removed less arsenic in a single washing than TA. The greatest differences were observed with soils S1 and S2, for which the efficiency of TA was 1.9- and 2.3-times higher than SAP (Fig. 3). These results indicate that the efficiency of As removal depends

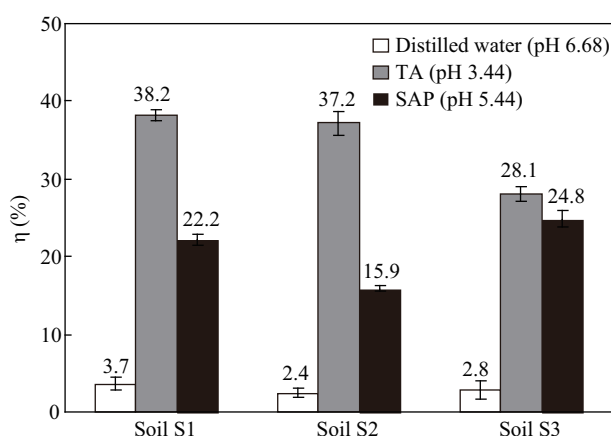


Fig. 3 Efficiency (η) of As removal from soils (S1, S2, S3) with distilled water (control), TA and SAP during a single washing without pH adjustment (error bars represent standard deviation, $n = 3$).

more on the total acidity of the biosurfactant than on its surface properties.

2.3.2 Triple washing with pH adjustment

Since the acidity of washing agents influenced arsenic mobilization in soils, during triple washing the pH of distilled water (control) and the SAP solutions was decreased and adjusted to 3.44 (± 0.2), the same value as the natural pH of TA. The changes in As mobility (as distribution coefficient, K_d) and process efficiency (η) after each washing are presented in Fig. 4.

In the present study, the K_d was defined as the ratio between the residual concentration of As in the soil after washing (mg/kg) and its concentration in the supernatant (mg/L). The K_d is often used to quantify metal mobility in soils (Antoniadis et al., 2007). Higher K_d values indicate more metal retention in soil through sorption (lower mobility), whereas lower values indicate higher metal concentration in solution (higher mobility) (Anderson and Christensen, 1988). It is desirable for soil washing to maximally decrease the K_d .

It was found that distribution coefficients decreased in subsequent washings, but their values varied considerably depending on biosurfactant type and washing number. For distilled water at pH 3.44, K_d values were an order of magnitude higher than for biosurfactants, despite increasing the number of washings (Fig. 4A–C). This means that more As remained in the soil. For distilled water, pH adjustment with H_3PO_4 increased the process efficiency, but only by a few percent; after triple washing it was below 15% in all soils (Fig. 4A–C). It is known that phosphate improves arsenic desorption by a ligand exchange mechanism and prevention of its readsorption (Yang et al., 2009). Overall, the use of water was ineffective, despite decreasing the pH and increasing the number of washings. This contrasts with Tokunaga and Hakuta's (2002) results. They removed almost 100% As with 1.6 mol/L H_3PO_4 . The differences can result from concentration of phosphate groups. In Tokunaga and Hakuta's research concentration of PO_4^{3-} in washing solution was much higher (around 0.38 mol/L) than in present study for water acidified to pH 3.44 (3.6×10^{-5} mol/L).

For SAP, decreasing pH from 5.44 to 3.44 increased arsenic mobilization after just the first washing (Fig. 4G–I); arsenic removal increased by 21.4% (soil S1), 17% (soil S2) and 6.8% (soil S3). For distilled water, despite adjustment to the same pH, the increases were lower; 3.8% (soil S1), 2.5% (soil S2) and 2.5% (soil S3). These results may suggest that arsenic mobilization by SAP (pH 3.44) was improved directly by the presence of biosurfactant rather than by exchange of phosphate oxyanions, because their concentration in SAP solution was low (7.64×10^{-3} mol/L). As the number of washings with SAP increased, the K_d values visibly decreased, resulting in a further increase of process efficiency (Fig. 4G–I).

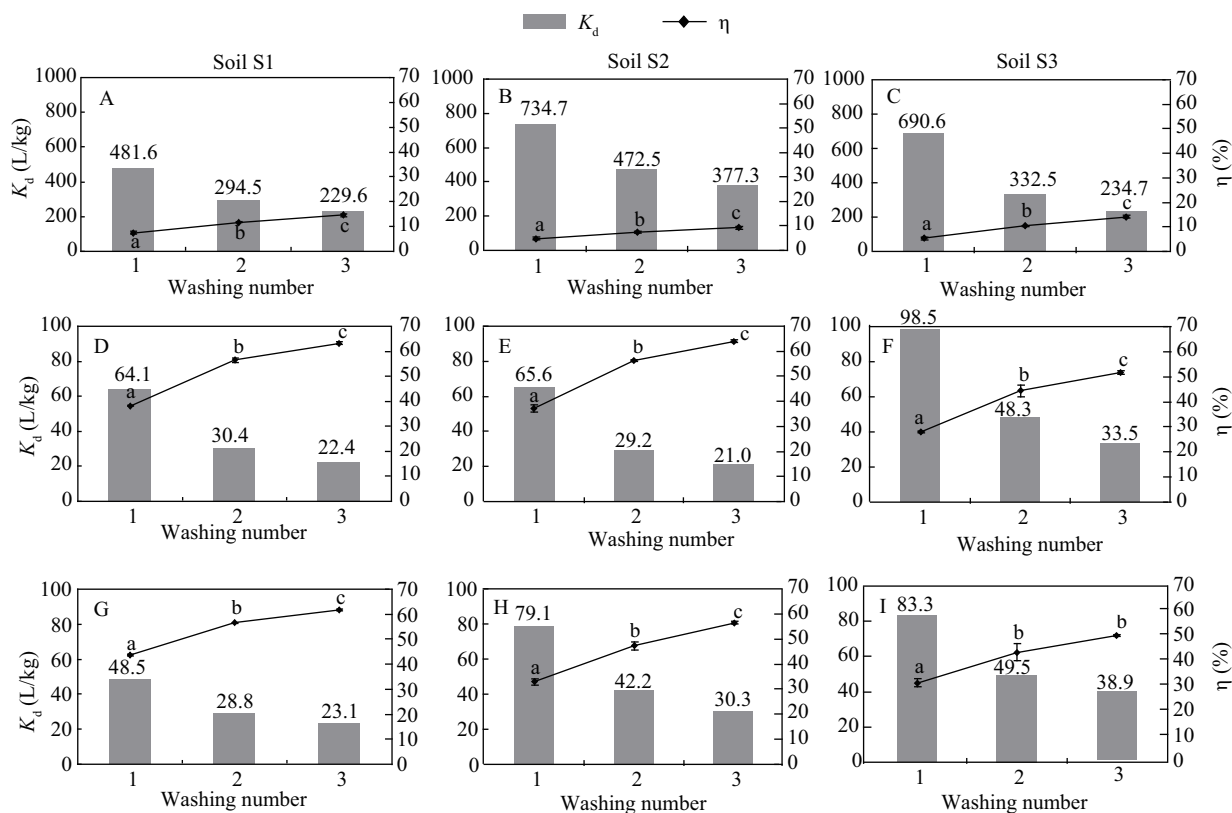


Fig. 4 Distribution coefficients (K_d) and efficiency (η) of As removal from soils during triple washing with distilled water (A, B, C), TA (D, E, F) and SAP (G, H, I). Mean values ($n = 3$) followed by the same letters do not differ significantly at $p < 0.05$.

For TA, As distribution coefficients and process efficiency changed with subsequent washings in a similar manner to SAP (**Fig. 4D–F**). After the third washing the efficiency of As removal by SAP and TA was comparable, despite the fact that they aggregate into different types of micelles. In soil S1 it was 61.8% and 63.2% ($p > 0.05$) and in soil S3 – 49.6% and 51.7% ($p > 0.05$), respectively. In soil S2, TA mobilized more As (63.9%) than SAP (56.4%) ($p < 0.05$).

In soils from old mining sites, arsenic forms mostly secondary minerals like simple As oxides or complexes of As(V) or As(III), oxygen and various metals, usually iron (Drahota and Filippi, 2009). Most frequently reported at these sites (about 60%) are amorphous Fe arsenates (e.g. pitticite, $\text{Fe}_x(\text{AsO}_4)_y(\text{SO}_4)_z \cdot n\text{H}_2\text{O}$) and crystalline Fe arsenates (e.g. scorodite, $\text{FeAsO}_4 \cdot 2\text{H}_2\text{O}$). The sorption of As(III) and As(V) to Fe (hydro)oxides contributes to As immobilization in soils. The solubility of these phases can vary widely, depending on redox and pH conditions, that affects As mobility in soil. The minimum solubility of well crystallized Fe arsenates occurs at a pH around 2.5. At $\text{pH} \geq 3.0$, crystallized Fe arsenates are likely to decompose to arsenate oxyanions (AsO_4^{3-} or AsO_3^{3-}) and to Fe^{3+} (iron (oxy)(hydro)oxides) (Henke, 2009). Under neutral or mildly acidic conditions arsenic form precipitates with iron.

In the present study, the pH of both biosurfactant solutions used for soil washing was acidic (pH 3.44).

Therefore, it could have affected decomposition of some mineral phases in soil (i.e. Fe arsenates). Due to decomposition, Fe, As oxyanions and other metals are released. The concentrations of As, Fe and co-existing metals in the supernatants from the subsequent washings are given in **Fig. 5**. The presence of Fe in supernatants apart from As, confirms decomposition of Fe arsenates. In all effluents Fe and As were prevailing ions and their concentrations were significantly different ($p < 0.05$). In contrast, the maximum concentration of Cu, Ni, Pb and Zn (as a sum) in TA effluents did not exceed 1.5 mg/L, whereas in SAP effluents was higher (6.7 mg/L). SAP contains 10 times more COOH groups than TA (**Table 3**), which react readily with metals (Buschmann et al., 2006).

After decomposition of Fe arsenates, arsenic oxyanions potentially could be sorbed again and/or coprecipitated with iron (oxy)(hydro)oxides. However, a competition of dissociated functional groups of TA and SAP with arsenic oxyanions towards iron prevents As coprecipitation with Fe oxides. The carboxylic and phenolic groups can react with Fe^{3+} by complexation or H^+ exchange. As a result, As oxyanions can be directly released into solution. The evidence of iron complexation by plant biosurfactants, especially by TA, was a color reaction. After each washing, the color of TA supernatant changed from brown (before washing) to intense blue, but no precipitates were observed. According to Tonello et al. (1997), intense blue

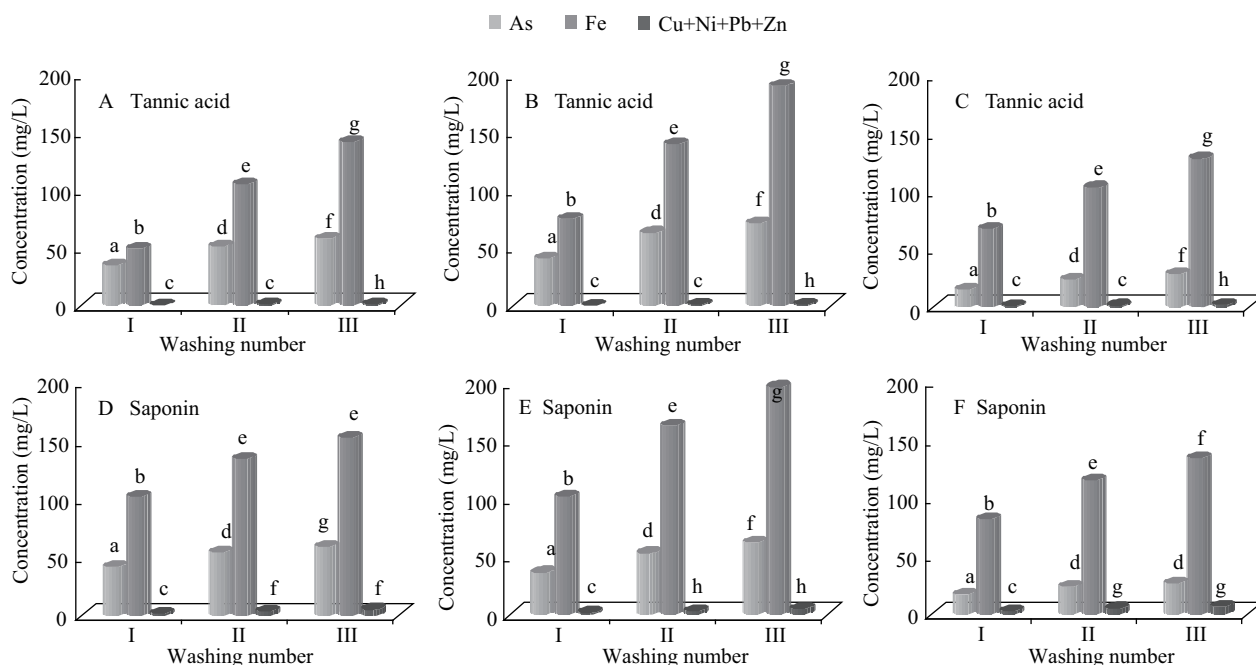


Fig. 5 Concentrations of As and metals in the effluents during triple soil washing with plant biosurfactants: (A), (B) soil S1, (C), (D) soil S2, (E), (F) soil S3. Mean values ($n = 3$) followed by the same letters do not differ significantly at $p < 0.05$.

chelates in the presence of TA occur at Fe concentration below 0.4%. At higher concentrations, blue-black precipitates are generated. In this study, Fe concentration in effluents after washing with TA was well below this value in all effluents. Saponin does not give color reaction with iron, but the presence of Fe in SAP effluents could confirm its complexation. The ability of plant biosurfactants to complex iron after decomposition of Fe arsenates might explain higher efficiency of As removal in successive washings compared to water alone, despite the same pH.

The results obtained in the present study are consistent with literature data. Mobilization of As is closely related with Fe mobilization because in contaminated soils most of As is usually bound to oxides. According to Wang and Mulligan (2009b), the mobilization of As from mine tailings by rhamnolipids was found to be not only closely correlated with the mobilization of Fe, but also well correlated with mobilization of Cu, Pb and Zn. However, these metals occurred at considerably higher concentrations than in the present study. Co-mobilization of the metals helped transfer arsenic into solution by metal-bridging mechanism. In such mechanism, dissolved metal cations (i.e. Fe^{3+} , Cu^{2+}) are attached to dissociated functional groups in biosurfactant micelles. In consequence, the outer surfaces of biosurfactant micelles take a positive charge, with one or two positive charges in excess. Next, the net positive charge is compensated by counter-ions, i.e. arsenic. In the present study, in contrast to Fe the effect of co-existing metals (i.e. Cu, Ni, Pb and Zn) on As mobilization through metal-bridging mechanism was negligible due to their low concentration in soils. However, it is highly probable

that As oxyanions released from the soils could have been incorporated further into Fe-biosurfactant complexes in supernatants.

The same mechanism as for plant biosurfactants was given earlier by Chatain et al. (2004). The authors demonstrated that arsenic mobilization from aged-contaminated soil from a French gold mining site by carboxymethyl- β -cyclodextrin (CMCD) under acidic conditions resulted from iron and copper complexation by CMCD firstly, due to presence of carboxymethyl ($\text{CH}_2\text{-COO}$) groups. This caused next the indirect release of arsenic that was initially associated with iron oxides.

In the present study, using of plant biosurfactants under acidic conditions resulted in a relatively high As mobilization. However, the results from other studies indicate that alkaline conditions favor much more As mobilization. Wang and Mulligan (2009a) found that at pH 3.0, As mobilization by humic acid (HA) was decreased probably as a result of its sorption to mine tailings and As retention to the sorbed HA. This would mean that TA and SAP are less sorbed to soil than HA, and facilitate As mobilization after decomposition of Fe arsenates. Due to dissociation of HA at higher pH ($\text{pK}_a = 3.92$ for strong acid groups, $\text{pK}_b = 8.22$ for weak acid groups), As mobilization was promoted mainly by co-mobilization of heavy metals (e.g. Fe) and formation of soluble As-HA or As-metal-HA complexes. This process was the greatest at pH 11. Moreover, under alkaline pH As can be additionally released from organic matter (Wang and Mulligan, 2009a, 2009c). Other investigations by Wang and Mulligan (2009b) performed for rhamnolipid JBR425 revealed visible increase of As

mobility in mine tailings with pH increase from 7 to 11. This process is complex and includes both physical reactions like reducing the interfacial tension or increasing the wettability of the solid phase, and also chemical reactions, i.e. As incorporation in biosurfactant micelles through the metal-bridging mechanism. Considering that the pK_a for rhamnolipids is 5.6, the negative charge due to ionization of carboxyl groups would increase above pH 6.0. The negatively charged biosurfactant would replace As oxyanions adsorbed onto mine tailings. In addition, with pH increase interfacial tension for rhamnolipids can be further decreased that enhances As mobilization.

To sum up, the present study and others investigations have shown that arsenic can be mobilized under both acidic and alkaline conditions, depending on biosurfactant type. Therefore, it was hypothesized that in both conditions, metal (i.e. Fe) mobilization in soil correlates with As mobilization.

2.4 Arsenic distribution and speciation in soils before and after treatment with plant biosurfactants

Arsenic fractionation and speciation in soils before and after triple washing with plant biosurfactants is given in **Fig. 6**. Arsenic distribution in the contaminated soils (before washing) was similar despite differences in its total concentration. In all three soils, most of the As existed in the residual (F4) and reducible (F2) fractions (**Fig. 6A, C, E**). In soils S1 and S2, the concentrations of As in the exchangeable (F1) and oxidizable (F3) fractions were lower, but roughly equivalent. In soil S3, the concentration of F1 was much lower than F3. This distribution pattern indicates relatively low mobility of As in the soils. However, the results of sequential extraction should be carefully interpreted. In the BCR procedure, hydroxylamine hydrochloride in nitric acid is used to mobilize the reducible fraction (F2). This solution leaches

As mainly from amorphous Fe and Mn (hydro)oxides, not from crystalline Fe oxides. The As that remains in the crystalline Fe oxides can be removed in the last step along with the residual fraction (F4). Thus, the concentration of As in the F2 fraction can be underestimated, and in the F4 fraction it can be overestimated. Despite this, the BCR extraction is commonly used for determination of As in soils (Pueyo et al., 2008). The distribution pattern for As found in this study resembles that found by other authors. Krysiak and Karczewska (2007) have used Wenzel's sequential extraction procedure to show that the concentration of the easily mobilized As fraction in soils from Zloty Stok (Poland) does not exceed 2% of its total content, whereas the amounts of Fe-Al oxides and residual fractions, depending on the site sampled account for 23%–96% and 54%–73% of total As, respectively. Similar As fractionation has been reported by Yang et al. (2009d) in mine tailings in South Korea, where Fe-Mn oxides and residual forms predominated, despite a wide range of total As concentrations (1941–37145 mg/kg). These results confirm the crucial role of Fe oxides and primary and secondary silicates in immobilizing As.

In contrast to sequential extraction, arsenic speciation enables assessment of its toxicity. It is known that inorganic species of As are more toxic and mobile than organic ones, As(III) being more toxic than As(V). In the present study, As(V) predominated in soils S1 and S2 due to oxidizing conditions in the soils, whereas there was nearly 1.5 times more As_{org} than As(III) and As(V) combined in soil S3 (**Fig. 6B, D, F**). The fraction of As(III) amounted to 22%, 17% and 26% of the total As in S1, S2 and S3, respectively. On contrary, Wang and Mulligan (2009b) found only As(V) in mine tailings when they used electrophoresis to separate As species.

Triple washing with plant biosurfactants resulted in removal of As from all individual fractions, especially from

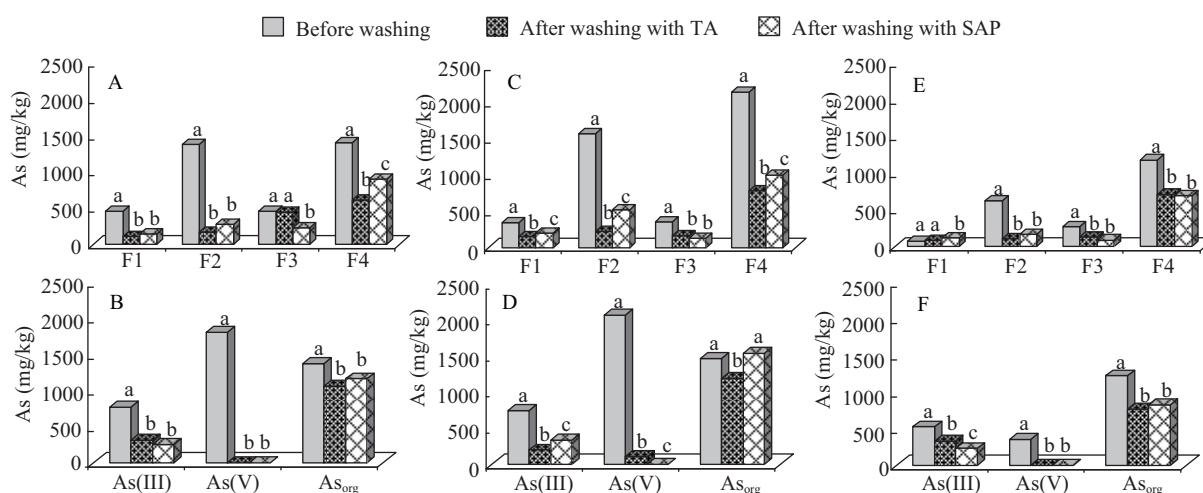


Fig. 6 As distribution and speciation in soils before and after triple washing with plant biosurfactants: (A, B) soil S1; (C, D) soil S2; (E, F) soil S3. As fractions: F1 – exchangeable, F2 – reducible, F3 – oxidizable, F4 – residual; As speciation forms: As(III), As(V), As_{org} . Mean values ($n = 3$) followed by the same letters do not differ significantly at $p < 0.05$ within a given fraction/speciation form.

the reducible, which that confirms decomposition of Fe arsenates. Surprisingly, As also appeared to be effectively removed from the residual fraction, especially in soil S2 (**Fig. 6C**). This is surprising, because arsenic in the residual form is considered to be more stable than in other fractions. In fact, the As may not have been removed from silicates, but from crystalline Fe oxides, which are part of the F2 fraction, but difficult to mobilize with the solution of hydroxylamine hydrochloride in nitric acid used to determine the content of the F2 fraction. Indeed, plant biosurfactants are more effective in As mobilization from stable fractions compared to other washing agents. Wang and Mulligan (2009) noticed that at total As concentration in mine tailings equal 2180 mg/kg, ramnolipids mobilized As mainly from water soluble and exchangeable fractions. The percentage of As removal from reducible, organic and residual fractions was 12.5%, 2.5% and 1.6%, respectively. However, humic acid used for the same mine tailings was less effective in As removal from reducible fraction and it did not affect residual form (Wang and Mulligan, 2009a). For comparison, plant biosurfactants decreased As concentration in reducible fraction by 82%–88% for TA and 67%–80% for SAP, whereas in residual fraction by 40%–63% for TA and 36%–53% for SAP.

In the present study, plant biosurfactants also diminished As toxicity. TA and SAP removed As(V) completely or nearly completely. In soils As(V) tends to be adsorbed mainly onto clay minerals and metal oxides. Therefore, its removal under acidic conditions is possible. The content of As(III) was decreased by 60%–68% (soil S1), 54%–73% (soil S2) and 37%–56% (soil S3). However, As_{org} was removed less efficiently (**Fig. 6B, D, F**).

3 Conclusions

Arsenic mobilization in brownfield soils by TA and SAP depended more on their acidity than surface properties. Triple washing with plant biosurfactants at the same pH and concentration improved As mobilization. Through decomposition of Fe arsenates in soils and iron complexation by biosurfactants, As oxyanions could be directly released into solution. The role of co-existing metals was negligible due to their low concentration in the soils. Plant biosurfactants reduced arsenic toxicity in the soils by efficiently removing As(V) and As(III).

The ability of these biosurfactants to complexate iron and increase arsenic mobility from soil is the main reason why they might be recommended for remediation technologies other than soil washing in brownfields, e.g. enhanced phytoextraction. Their biodegradability, low toxicity to plants and possibility of reuse makes plant biosurfactants attractive candidates for field-scale use in these processes, however, further investigations are needed before they can be recommended with certainty.

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