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RESEARCH ARTICLE

Removal of mixed contaminants from landfill leachate–contaminated soil by flushing with bio-surfactant: laboratory column tests

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Abstract

Landfill leachate–contaminated soil is widespread all over the world. In order to study the removal of mixed contaminants from landfill leachate–contaminated soil by flushing with bio-surfactant, soil column test was conducted to select an optimum concentration of bio-surfactant saponin (SAP) at first. Then, the removal efficiencies of organic contaminants, ammonia nitrogen, and heavy metals from landfill leachate–contaminated soil by flushing with SAP were studied. At last, the toxicity of contaminated soil before and after flushing was estimated by sequential extraction of heavy metals and plant growth test. The test results showed that the SAP solution with the concentration of 2.5 CMC could effectively remove the mixed contaminants from soil and would not introduce excessive pollutants of SAP in soil. Specifically, the removal efficiencies of organic contaminant and ammonia nitrogen were 47.01% and 90.42%, respectively. And the removal efficiencies of Cu, Zn, and Cd were 29.42%, 22.55%, and 17.68%, respectively. During flushing, hydrophobic organic compounds as well as physisorption and ion-exchange ammonia nitrogen in soil were removed by the solubilization effect of SAP, and heavy metals were removed by the chelation of SAP. After flushing with SAP, the reduced partition index (I_R) value of Cu and Cd increased, and the mobility index (M_F) value of Cu decreased. In addition, flushing with SAP reduced the plant toxicity of contaminated soil, and the residual SAP in soil promoted the plant growth. Therefore, flushing with SAP offered great potentials in remediating the landfill leachate–contaminated soil.

Keywords Landfill leachate · Soil flushing · Bio-surfactant · Heavy metal · Phytotoxicity

Introduction

Landfill is one of the most widely used methods for municipal solid waste disposal all over the world. However, the landfill has caused serious soil contamination due to the leakage of leachate (Han et al. 2016), which has drawn public attention. Contaminants from municipal solid waste landfills are diverse, primarily consisting of organic contaminations, inorganic matter, and heavy metals (Ye et al. 2019). For example, in a municipal solid waste site in Jiangxi province of China, the concentration of Cr(VI), Ni, Cd, As, Cu, Zn, and Pb was 265.08, 37.41, 0.53, 19.97, 51.5, 128.68, and 247.88 mg/kg (Li et al. 2019). Furthermore, in a municipal solid waste site in Zhejiang province of China, the concentration of organic contaminations ranges from 1000 to 10,000 mg/L. The removal of organic contaminations, inorganic salts, and heavy metals from landfill leachate–contaminated soil has become a major challenge. Thus, various technologies, such as contaminant isolation, soil flushing, soil vapor extraction, chemical oxidation reduction, bioremediation, and phytoremediation, are developed to remediate the contaminated soil (Khan et al. 2004; Steliga and Kluk 2020; Tran et al. 2022). Among these technologies, soil flushing, which is to in situ remove contaminants from soil by passing an surfactant or chelating agent solution through the soil, offers the great advantage of highly effective removal of contaminations in contaminated soils and is frequently used in soil remediation due to characteristics of rapid cleanup of a contaminated site, reduction of long-term liability, and cost-effectiveness (Gusiatin and Klimiuk 2012; Ramadan et al. 2018).

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In the past, most soil flushing focused on the soil solely containing organic contaminants or heavy metals. A few studies were conducted on the mix contaminants of organic contaminations, inorganic salts, and heavy metals (Liu et al. 2018; Saeedi et al. 2019). Furthermore, for the purpose of simultaneous removal of organic contaminants and heavy metals, chemical surfactants were usually used (Bezza and Chirwa 2015; Chaprao et al. 2015; Rodriguez-Cruz et al. 2007; Wei et al. 2015). However, due to its refractory characteristics, the retained chemical surfactants in soil always caused other environmental problems. Different from chemical surfactants, bio-surfactants were isolated from plants or produced by microorganisms. And it shows excellent performance in soil flushing owing to their low toxicity, good surface activity, biodegradability, and huge environmental compatibility (Mesbaiah et al. 2016; Wu et al. 2014). As a kind of bio-surfactants, saponin (SAP) could enhance solubilization of hydrophobic organic compounds (Kaczorek et al. 2008; Kobayashi et al. 2012; Song et al. 2008) and promote the removal of heavy metals (Chen et al. 2008; Song et al. 2008; Ye et al. 2015). Previous studies found that SAP had a significant solubilization effect on organic matter and heavy metals in soil by batch soil washing tests (Zhang 2021). Therefore, it is necessary to comprehensively study the simultaneous removal efficiency of mix contaminants from soil by flushing with SAP.

The objective of this study was to evaluate the simultaneous removal of organic and inorganic contaminants as well as heavy metals from landfill-contaminated soil by flushing with SAP. Firstly, an optimum concentration of SAP for flushing was selected through soil column flushing tests. Then, the removal efficiency of organics, ammonia nitrogen, and heavy metals by flushing with SAP was studied through soil column flushing experiments. At last, the speciation distribution of heavy metals and toxicity of soil before and after flushing were estimated.

Materials and methods

Materials

Saponin (SAP, CAS: 8047-15-2, 60%wt), one of bio-surfactants and non-ionic surfactants, was used as the surfactant in this research. Its critical micellar concentration (CMC) was 0.05 g/L. It was obtained from Shanghai Yuanye Bio-Technology Co., Ltd. Cadmium nitrate tetrahydrate ($Cd(NO_3)_2 \cdot 4H_2O$, CAS: 123318-82-1, AR), copper nitrate trihydrate ($Cu(NO_3)_2 \cdot 3H_2O$, CAS: 10031-43-3, AR), and zinc nitrate hexahydrate ($Zn(NO_3)_2 \cdot 6H_2O$, CAS: 10196-18-6, AR) were purchased from Macklin (Shanghai, China). In addition, distilled water was used as liquid phase to prepared aqueous surfactant solution. Landfill leachate was collected from the Woqishan landfill (Wenzhou, Zhejiang province, China), and its composition is shown in Table S1. The collected leachate was stored in refrigerator at 7 ℃. Two kinds of soils which were also collected from Woqishan landfill were used in soil column test. The specific gravity (Gs), particle size distribution, pH, and organic matter content as well as contaminant concentrations of soil I (SI) and soil II (SII) are shown in Table S2. The tall fescue and pea seeds were from Lvbao Seeds (Taiyuan, China).

Soil column test I

Soil column test I was conducted to select the optimum SAP concentration for flushing. In the experiment, a column with a dimension of 60 mm in inner diameter and 200 mm in height was used. The soil of SI was compacted in the column in ten layers, and the height of each soil layer was 10 mm. The total soil bed height in the soil column was 100 mm. The 50-mm inlet and outlet part of the column were packed with quartz sand to prevent loss of soil. When the soil columns were filled, they were saturated with distilled water for 24 h.

SAP solutions with different concentrations of 1, 2.5, 5, 7.5, and 10 CMC were prepared with SAP and distilled water. Each SAP solution was applied on separated soil column. During flushing, the SAP solution or distilled water was delivered at a flow rate of 0.5 mL/min to the top of the soil column by a peristaltic pump and the effluent was collected. The flushing lasted 312 h. After flushing, the chemical oxygen demand (COD) as well as the concentration of ammonia nitrogen and Zn of effluent was analyzed.

Soil column test II

Soil column test II was conducted to comprehensively estimate the contaminant removal efficiency from soil by flushing with SAP. In the experiment, synthetic-contaminated soil and SAP solution with concentration of 2.5 CMC which was determined based on soil column test I were used. The pH value of the SAP solution was 8.43. In order to obtain the synthetic-contaminated soil, certain mass of $Cd(NO_3)_2 \cdot 4H_2O$, $Cu(NO_3)_2 \cdot 3H_2O$, and $Zn(NO_3)_2 \cdot 6H_2O$ were dissolved into the leachate at first. Then, the soil of SII was saturated with the landfill leachate containing 1000 mg/L of Cu^{2+}, 1000 mg/L of Zn^{2+}, and 1000 mg/L of Cd^{2+} for 72 h. Afterwards, the saturated contaminated soil was air-dried for 50 days. At last, it was crushed and prepared for soil column test II. The contaminant concentration of synthetic-contaminated soil is shown in Table 1.

The column used in the experiment had an inner diameter of 100 mm and length of 320 mm. The 25-mm inlet and outlet part of the column were packed with glass beads (300 μm). Two thousand five hundred–gram dry

Table 1 The contaminant concentration of the synthetic-contaminated soil

Cu (mg/kg)	Zn (mg/kg)	Cd (mg/kg)	Organic contaminants (mg/kg)	Ammonia nitrogen (mg/kg)
934	1430	532	2100	1100

synthetic-contaminated soil was compacted into the column in ten identical steps. The soil bed height in column was 200 mm. The pore volume in soil column was 805 cm^3. After flushing, soil columns were saturated with distilled water for 48 h. The soil columns which were flushed by distilled water were used as control column. The pH value of the distilled water was 8.33. In experimental columns, SAP solution with concentration of 2.5 CMC flowed upwards through the soil column with a constant hydraulic head, and the flow rate was about 8.7 mL/min. During flushing, the effluent samples were collected, and the COD as well as the concentration of ammonia nitrogen, Zn, Cu, and Cd in the effluent was measured, respectively. The flushing took about 92 h. The experiment method of the control columns was the same as that of the experimental column. After flushing, the soil was collected from column and air-dried. The chemical speciation of Zn, Cu, and Cd in soil was tested and the soil was used to plant growth assays.

Analytical methods

The COD and ammonia nitrogen concentration of liquid were measured colorimetrically according to Standard (China 2010; 2017). The content of ammonia nitrogen in soil was measured colorimetrically according to the Standard (China 2012). The content of organic contaminants in soil was measured in terms of the previous studies (Zhang 2021). Specifically, 2 g of soil was mixed with 20 mL deionized water at first. Then, the mixture was shaken for 24 h at the rate of 220 r/min. Afterwards, the centrifugation was performed on the mixture at 4000 r/min for 30 min. At last, the supernatant was collected and measured colorimetrically. The content of organic contaminants in soil was calculated by the COD of supernatant. The concentration of Cu and Zn in soil was measured using a flame atomic absorption spectrometer according to Standard (China 2019). The concentration of Cd in soil was measured using a graphite furnace atomic absorption spectrometer atomic absorption spectrophotometer according to Standard (China 1997). The concentration of Cu, Zn, and Cd in liquid was measured using an inductively coupled plasma mass spectrometry (ICP-MS) according to Standard (China 2014). All analytical measurements were done in triplicate.

The removal efficiency of organic contaminants or ammonia nitrogen was calculated by Eq. (1):

$$\text{organic contaminants or ammonia nitrogen removal} = \frac{CV}{C_S m_S} \times 100\% \tag{1}$$

where C is the average concentrations of COD or ammonia nitrogen in effluent (mg/L); C_S is the concentrations of organic contaminants or ammonia nitrogen in soil (mg/kg); V is the total volume of effluent (L), and m_S is the dry mass of the soil (kg). The removal efficiency of metal was calculated by Eq. (2):

$$\text{metal removal} = \frac{C_0 - C_1}{C_0} \times 100\% \tag{2}$$

where C_0 is the initial metal concentration of contaminated soil (mg/kg), and C_1 is the final metal concentration of soil after flushing (mg/kg).

Synthetic-contaminated soil with or without flushing was subjected to sequential extraction using the three-step Community Bureau of Reference (BCR) procedure (Huang et al. 2018; Mossop and Davidson 2003).

Plant seed germination and growth test are rapid and practical techniques in assessing the change of soil properties and the toxicity of residual metals in soil (Jelusic et al. 2014). In this research, tall fescue and pea were planted to estimate the toxicity of soil before and after flushing. Specifically, the water content of synthetic-contaminated soil with or without flushing was adjusted to 80% by adding distilled water at first. Afterwards, one hundred undressed seeds of tall fescue or fifteen undressed seeds of pea were sown in soil. Then, the plant growth tests were executed in a laboratory for 28 days and under a natural light. The temperature of laboratory was 20–25℃. Each pot was irrigated by weight method every 2–4 days according to the amount of soil moisture lost during the experiment. The tall fescue and pea were harvested at the 28th day and the plant height was measured.

Results and discussion

Efficiency of SAP concentration on removal of contaminants from soil

Figure 1 shows the removal efficiency of organic contaminants, ammonia nitrogen, and Zn from soil by SAP solution and the COD of SAP solutions. It can be seen from Fig. 1 that by different concentrations of SAP, removal efficiencies of organic contaminants or ammonia nitrogen were almost

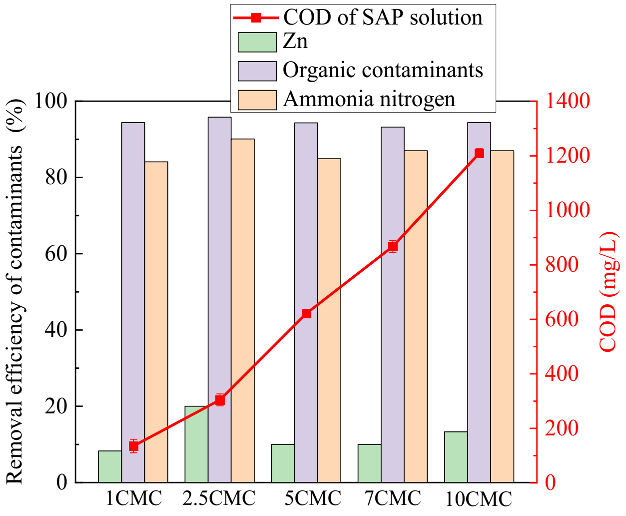


Fig. 1 Removal efficiency of contaminants by SAP solution and the COD of SAP solution

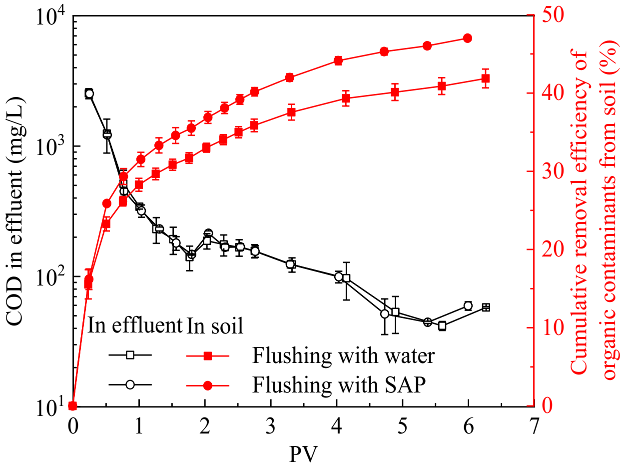


Fig. 2 COD in effluent and cumulative removal efficiency of organic contaminants from soil during flushing

the same. However, for Zn, the maximum removal efficiency was achieved in soil which was flushed with 2.5CMC SAP. In addition, it also can be seen from Fig. 1 that the COD of SAP solution increased with the increase in SAP concentration. To prevent the introduction of pollution by excess surfactant, the optimum SAP concentration seems to be approximately 2.5 CMC.

Removal of mix contaminants from landfill leachate–contaminated soil by flushing with SAP

The COD in effluent and the cumulative removal efficiency of organic contaminants from the soil during flushing are shown in Fig. 2. Before 2PV, the COD in effluent decreased with the PV increase, and then it kept about the same values after 2PV. It suggested that the elution of organic contaminants mainly occurred before 2PV. In addition, the COD value in effluent after 2PV might be attributed to the COD value of SAP solution. As shown in Fig. 1, the value of COD in SAP solution with 2.5 CMC SAP was 305 mg/L. Furthermore, after 2PV, the COD value of 100 mg/L in effluent was less than that in flushing solution. It was because that the soil with high organic matter content had strong adsorption capacity. A part of SAP in flushing solution was adsorbed on soil during flushing, which reduced the concentration of SAP in effluent and further led to low COD value in effluent. Figure 2 also shows the removal efficiency of organic contaminants from soil by water or SAP solution. As a whole, the cumulative removal efficiency of organic contaminants increased with the increase of PV. And organic contaminants in soil were removed to a certain extent by flushing with water, which revealed that organic matter desorption with water was achieved, even without surfactants. This could be due to the presence of easily accessible hydrocarbons in landfill leachate–contaminated soil that could be desorbed by water (Huguenot et al. 2015). At the end of flushing, the organic contaminant removal efficiency of 47.01% by SAP was higher than that by water. It was mainly due to hydrophobic organic compounds in soil being removed by desorption of SAP. Specifically, SAP improved the solubility of hydrophobic compounds by enhancing hydrophobic compounds and distributing them to the hydrophobic core of micelles. SAP also reduced the interfacial tension between water and hydrophobic organic compounds to accelerate the transfer of pollutants to aqueous solutions (Juhasz et al. 1997; Liu et al. 2017). Furthermore, compared with the removal efficiency of organic contaminants in soil column test I (shown in Fig. 1), it was lower in the experiment (shown in Fig. 2). This was because the synthetic-contaminated soil used in this experiment had more organic matter than the soil of SI which was used in soil column test I. It was difficult to elute organic contaminants from soil with high organic matter content.

Figure 3 shows the ammonia nitrogen in effluent and its cumulative removal efficiency from the soil during flushing. In Fig. 3, the variation tendency of ammonia nitrogen in effluent was similar to that of COD. The cumulative removal efficiency of ammonia nitrogen from soil by water was 64.5%. According to Deng et al. (2022), the ammonia nitrogen in soil can be divided into physisorption, chemisorption, and ion-exchange ammonia nitrogen. The transport ability of ammonia nitrogen was highest for physisorption followed by ion-exchange and lowest for chemisorption. The physisorption ammonia nitrogen in soil could be removed by flushing with water. Therefore, the cumulative ammonia nitrogen removal efficiency of 64.5% by water was the result of the removal of physisorption ammonia nitrogen.

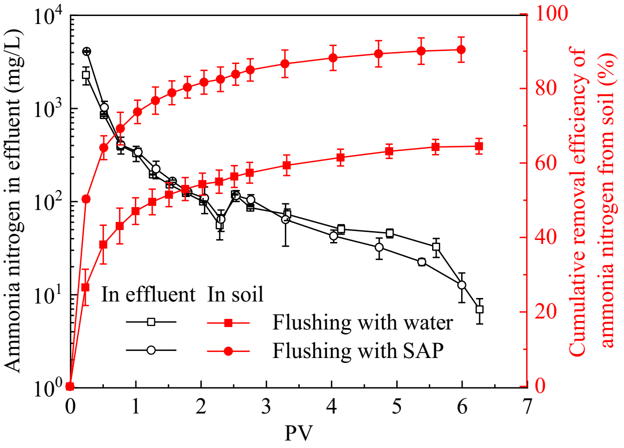


Fig. 3 Variation of ammonia nitrogen in effluent and cumulative removal efficiency of ammonia nitrogen from soil during flushing

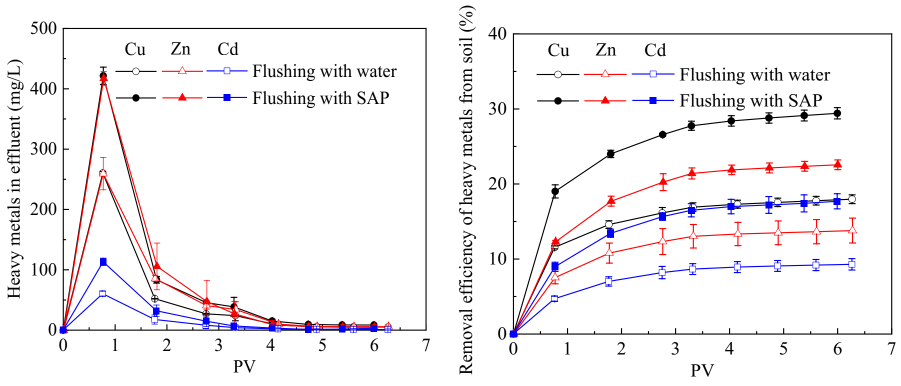
The cumulative removal efficiency of ammonia nitrogen by SAP was 90.42%, which was far higher than that by water. There were two reasons for this phenomenon: On the one hand, while the physisorption ammonia nitrogen was removed, SAP with solubilization effect removed the ion-exchange ammonia nitrogen from the soil particles. On the other hand, dissolved oxygen value of the SAP solution was 7.12 mg/L. The soil column was under aerobic condition during the whole duration of the flushing. Chemisorption ammonia nitrogen in soil was nitrified during flushing (Castaldelli et al. 2018; Huang et al. 2021). Therefore, the removal efficiency of ammonia nitrogen was outstanding by flushing with SAP.

The variation of heavy metals (Cu, Zn, and Cd) in effluent and removal efficiencies of heavy metals from soil during flushing are shown in Fig. 4. In effluent, the concentrations of Cu, Zn, and Cd all increased significantly before 1PV, and decreased thereafter until 5PV. While the COD or ammonia nitrogen in effluent all decreased before 2PV (shown in Fig. 2 and Fig. 3). That is, it took more time to remove heavy metals from soil than to remove organic contaminants or ammonia nitrogen. In addition, as shown in Fig. 4b, the removal efficiencies of heavy metals by flushing with SAP were higher than that by water. This was because that SAP molecules contain carbonyl groups and ionizable carboxyl groups; thus, they could chelate cationic metals and remove them (Ye et al. 2015). Specifically, there were two ways for SAP to enhance desorption of heavy metals from the soil. The first one was that SAP chelated with heavy metal ions in solution, which reduced the activity of liquid phase of heavy metal ions and induced the desorption of heavy metals in soil. And the second way was to reduce the interfacial tension, then SAP accumulated on the solid-liquid interface and directly contacted with adsorbed heavy metals, which led to the desorption of heavy metal from soil (Liu et al. 2017; Lu et al. 2010). Therefore, removal efficiency of heavy metals from soil by flushing with SAP was higher than that by water. In addition, it also can be seen from Fig. 4b that in soil by flushing with SAP, the removal efficiencies of Cu, Zn, and Cd were 29.42%, 22.55%, and 17.68%, respectively. This result was consistent with metal extraction yields typically following the sequence in research of Wuana et al. (2010): Cu> Ni> Zn> Cd> Pb.

Toxicity analysis of soil after flushing with SAP

Figure 5 shows the speciation distribution of heavy metals in soil before and after flushing. Chemical speciation distribution of heavy metals in soil includes exchangeable and acid soluble fraction (F1), reducible fraction (F2), oxidizable fraction (F3), and residual fraction (F4). The bioavailability and ecotoxicity of the four different chemical speciation of heavy metal varied in the following order: F1 > F2 > F3 > F4 (Wang et al. 2019). As shown in Fig. 5, Cu and Zn in soil without flushing occurred in different fractions of F1, F2, F3, and F4, while the Cd mainly occurred in F1 and F2. There were two reasons for this phenomenon. On the one hand, the soil aging was relatively short and it

Fig. 4 Variation of heavy metals during flushing



(a) Heavy metals in effluent (b) Removal efficiency of heavy metals from soil

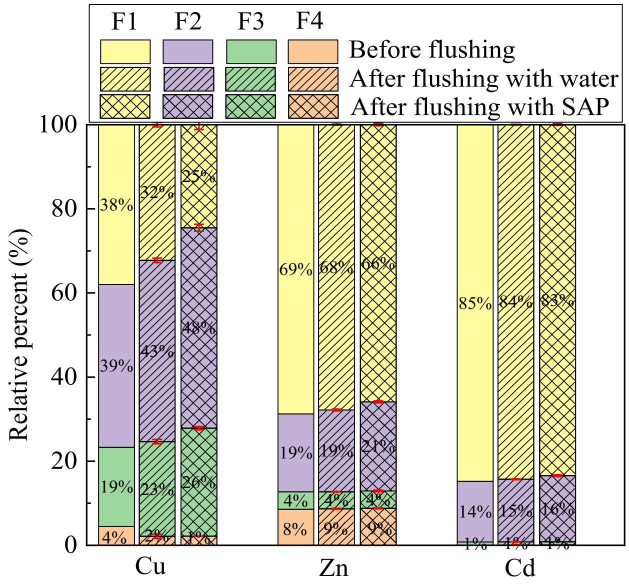
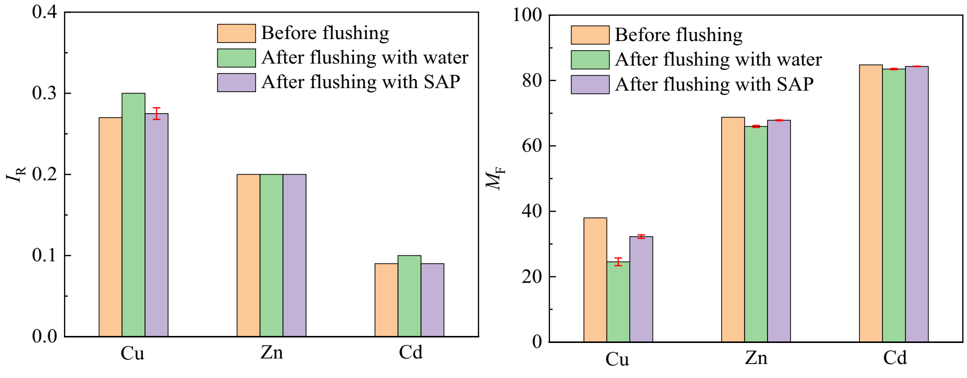


Fig. 5 Speciation distribution of heavy metals in soil before and after flushing

was only 50 days. On the other hand, the average bonding between Cd and soil particles was weakened (Gusiatin and Klimiuk 2012; Kashem et al. 2007). So Cd did not appear to form a strong complex with organic matter. According to Li et al. (1995), F1 and F2 can be removed by soil washing technique, whereas the F3 and F4 are very stable and not generally removed or changed by soil washing. It can be seen from Fig. 4 that after flushing with water or SAP, although the total content of heavy metals had a significant reduction, the variation of metals speciation distribution was not notable, particularly for Zn and Cd. According to Mulligan et al. (2001), the oxide phase containing Zn and Cu could be released under acidic conditions and alkaline conditions, respectively. As shown in Table S2 and Fig. S1, the soil and effluent remained alkalescence during flushing. Thus, the varieties of Zn speciation distribution were not notable, while that of Cu was remarkable. For Cu, the relative percent of F1 decreased, while the F2 and F3 relative fractions were increased. It was mainly attributed to the removal of exchangeable Cu during flushing with SAP. In addition, F4 of Cu decreased during flushing with SAP. That is, a part of residual Cu was removed. This result was consistent with the finding reported by Mulligan et al. (2001) that metals associated with the organic phase could be easily removed from sediments by bio-surfactants. For Zn and Cd, the variation of F1 was small. This might be attributed to the low removal efficiency of Zn and Cd during flushing with SAP. Besides, it also can be seen from Fig. 5 that there were relatively more exchangeable and acid soluble fraction Zn as well as Cd in soil after flushing with SAP. That is, the Zn and Cu in soil can be removed by multiple flushing.

The reduced partition index (I_R) could be used to describe the stability of metals based on sequential extraction (Han et al. 2003), and it could be defined by Eq. (S1). The I_R takes values between 0 and 1. High value of I_R represents metal stability in soil, and its low value indicates distributions with a high proportion of soluble forms (Gusiatin and Klimiuk 2012). Furthermore, mobility index (M_F), which was the ratio of metal concentration in the mobile fraction to the sum of all fractions, could be used to assess relative mobility and bioavailable form of metals in soil. It could be calculated by Eq. (S2). High M_F value means symptoms of relative high mobility and biological availability of heavy metals in soil (Gusiatin and Klimiuk 2012; Ma and Rao 1997). Figure 6 shows the value of M_F and I_R of heavy metals in soil before and after flushing. As a whole, the values of I_R for Cu, Zn, and Cd in soil were low before or after flushing, which indicated that metal binding intensity in soil was weak. After flushing with water or SAP, the I_R value of Zn kept about the same value whereas there were increases in Cu and Cd, which indicated the stability of Cu and Cd in soil was enhanced after flushing. In addition, I_R was strongly correlated with M_F. Generally, the lower the relative binding intensity, the higher the metal mobility. After flushing with

Fig. 6 The value of M_F and I_R of heavy metals in soil before and after flushing



(a) I_R

(b) M_F

water or SAP, the M_F value of Cu decreased. The decrease implied that the mobility and biological availability of Cu in soils became weak after the treatment of flushing with SAP. For Zn and Cd, its small variation of M_F value revealed that the mobility and biological availability of Zn and Cd have hardly changed by flushing. This phenomenon was mainly attributed to the small changes of speciation distribution of Zn and Cd during flushing (shown in Fig. 5).

Figure 7 shows growth situation of plant after a 21-day growth in synthetic-contaminated soil with or without flushing. The seed germination rate in soil after flushing was significantly higher than that without flushing. As shown in Fig. S1, the pH value of effluent in soil column during flushing with SAP or water was around 8.5 and 8.1, respectively. The influence of pH value variation on plant growth could be ignored. That is, high concentrations of contaminants in contaminated soil without flushing were found to be extremely toxic to plants, restraining plant growth and causing low germination rates. In addition, as shown in Fig. 7, compared with the plant in soil after flushing with water, it was luxuriant in soil after flushing with SAP. This was because that the removal efficiency of ammonia nitrogen in soil by water was less than that in soil by SAP. According to Vandijk and Roelofs (1988), an excessive amount of nitrogen could lead to yellowing and lower yields of plants.

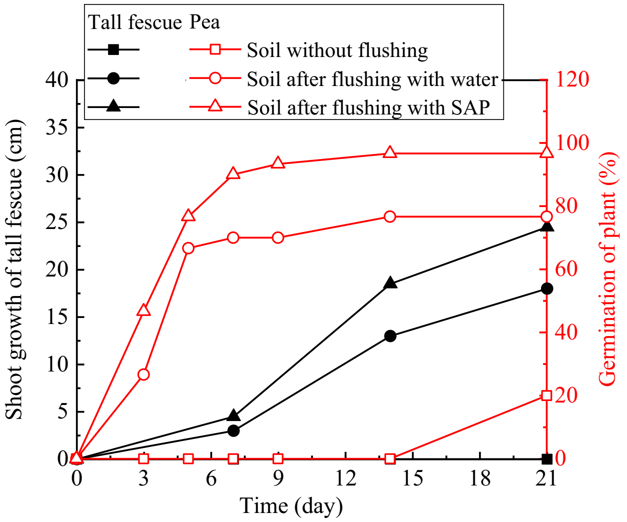
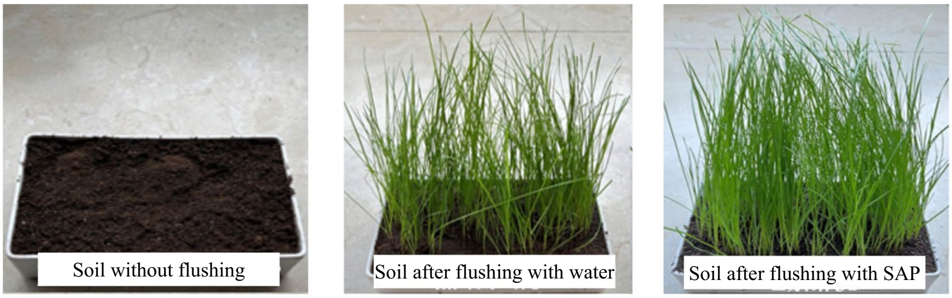


Fig. 8 Shoot growth and germination rate of plant during growth period in soil with or without flushing

Figure 8 shows the shoot growth and germination of plant during the growth period in soil with or without flushing. The germination rate of the plant can be calculated by Eq. (S3). The germination rate of tall fescue and pea in soil without flushing was 0% and 20%, respectively, which were significantly lower than that in soil after flushing with SAP of water. This was due to the fact that there was large amount of contaminants in the contaminated soil. Furthermore, compared with in soil by water, the germination rate of pea or shoot growth of tall fescue in soil after flushing with SAP was higher. Especially after the 7th day of sowing, the difference of germination rate or shoot growth of plant in soil between by SAP and by water became more marked. The average germination rate of pea and shoot growth of tall fescue was 96.7% and 24.5 cm, respectively, in soil after flushing with SAP at the 21st day. However, the plant growth situation was not consistent with the phytotoxicity of soil which was responded by M_F and I_R. This suggested that the residual SAP in soil might have a beneficial effect on plant growth. On the one hand, SAP could stimulate the activity of microorganism to accelerate the degradation rate of

Fig. 7 Growth situation of plant after a 21-day growth period in soil with or without flushing



(a) Tall fescue

(b) Pea

contaminants (Tao et al. 2020; Wang et al. 2016). On the other hand, SAP was suitable substrate for soil bacteria and further enzyme activities in soil (Greaves and Webley 1965; Soeder et al. 1996), which could enhance the decomposition of organic residues and the nutrient cycling in soil-plant systems (Giacometti et al. 2014). Therefore, the plant was luxuriant in soil after flushing with SAP.

Compared with the removal efficiency of organic contaminants in soil column test II (Fig. 2), it was higher in soil column test I (Fig. 1). Furthermore, greater removal efficiency of organic contaminants was found in research of Saeedi et al. (2018). They reported the polycyclic aromatic hydrocarbon removal efficiency of 75% from the mixture of sand and kaolinite by flushing with SAP solution. This difference of removal efficiency of organic contaminants was related to the organic matter content in contaminated soil. According to Wang et al. (2008), organic matter in soil was adverse to desorption of contaminants. Specifically, due to the hydrophobic tail of non-ionic surfactants and high organic matter content in soil, a part of the surfactant molecules adsorbed onto organic matter in soil. This increased the hydrophobicity and the retention of organic contaminants in the soil (Saeedi et al. 2019). Organic matter content of the contaminated soil used in soil column test I and II was 3.6% and 20.6% (shown in Table S2), respectively, while it was 5.7% in research of Saeedi et al. (2018). So, the removal efficiency of organic contaminants was low in soil column test II. Besides, according to Saeedi et al. (2018), co-present heavy metals adversely affect the removal of organic contaminants from soil. In this research, organic contaminants were co-existed with heavy metals in soil, which was another reason that there was low removal efficiency of organic contaminants in soil column test II. For removal efficiency of heavy metals, it was lower in this research than that in research of Gusiatin and Klimiuk (Gusiatin and Klimiuk 2012). This phenomenon was also mainly attributed to the organic matter content in soil. According to Strawn and Sparks (Strawn and Sparks 2000), bonds between organic matter and metals are stronger than that between soil mineral particles and metals. The great content of organic matter in soil resulted in less desorption of metals from soil. Therefore, the removal efficiency of heavy metals from soil by flushing with SAP was low in this research.

Although the removal efficiencies of organic contaminants and heavy metals in high organics soil by flushing with SAP were low, removal of contaminants from high organics soil is a common problem for soil flushing technique. In addition, as discussed in the "Toxicity analysis of soil after flushing with SAP" section, the phytotoxicity of soil decreased significantly by flushing with SAP. This demonstrated that flushing with SAP is practicable to remediate landfill leachate–contaminated soil. Besides, there were some means to improve contaminant removal efficiency in flushing with SAP. For example, Huang et al. (2021) found that the removal of diesel increased over 20% by flushing with SAP micro-bubble solution. In addition, it is also a feasible method to improve the removal efficiency of heavy metals from soil by flushing with mixed solution of SAP and chelating agent. The chelating agent is an effective choice for the remediation of metal-contaminated soil by flushing (Evangelou et al. 2007). Furthermore, multiple flushing is beneficial for enhancing the contaminant removal efficiency (Huang et al. 2021; Mulligan et al. 2001). Overall, with more development, flushing with SAP will be an effective, non-toxic means of remediation in landfill leachate–contaminated soil.

Conclusions

In this study, removal of mixed contaminants from landfill leachate–contaminated soil by flushing with bio-surfactant SAP was studied. Based on contaminant removal efficiencies and residual of SAP in soil, the optimum SAP concentration for flushing was 2.5 CMC. By flushing with 2.5 CMC SAP solution, the removal efficiency of organic contaminants and ammonia nitrogen was 47.01% and 90.42%, respectively. And the removal efficiency of Cu, Zn, and Cd was 29.42%, 22.55%, and 17.68%, respectively. With the solubilization effect of SAP, hydrophobic organic compounds as well as physisorption and ion-exchange ammonia nitrogen were removed by flushing. And some chemisorption ammonia nitrogen was nitrified during flushing. In addition, the heavy metals were removed by the solubilization effect and chelation of SAP. With the treatment of flushing with SAP, the stability of Cu and Cd in soil enhanced, and biological availability of Cu in soil decreased. And the phytotoxicity of contaminated soil reduced after flushing with SAP. Although the removal efficiencies of organic contaminants and heavy metals from high organic matter soil by flushing with SAP were low, the phytotoxicity of soil decreased significantly. Besides, the residual SAP in soil could stimulate the activity of microorganism to accelerate the degradation rate of contaminants and promoted the plant growth. Therefore, flushing with SAP is practicable to remediate landfill leachate–contaminated soil.

Supplementary information

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Author contribution Mei Bai: conceptualization, methodology, validation, writing—original draft. Zhibin Liu: conceptualization, writing—review and editing, supervision, project administration. Zhu Liu:

investigation, visualization. Haitao Yu: project administration. Liangliang Lu: resources.

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Data availability All data generated or analyzed during this study are included in this article (and supplementary information).

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

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References

Bezza FA, Chirwa EMN (2015) Biosurfactant from Paenibacillus dendritiformis and its application in assisting polycyclic aromatic hydrocarbon (PAH) and motor oil sludge removal from contaminated soil and sand media. Process Saf Environ Prot 98:354–364. https://doi.org/10.1016/j.psep.2015.09.004

Castaldelli G, Colombani N, Tamburini E, Vincenzi F, Mastrocicco M (2018) Soil type and microclimatic conditions as drivers of urea transformation kinetics in maize plots. Catena 166:200–208. https://doi.org/10.1016/j.catena.2018.04.009

Chaprao MJ, Ferreira INS, Correa PF, Rufino RD, Luna JM, Silva EJ, Sarubbo LA (2015) Application of bacterial and yeast biosurfactants for enhanced removal and biodegradation of motor oil from contaminated sand. Electronic J Biotechnol 18(6):471–479. https://doi.org/10.1016/j.ejbt.2015.09.005

Chen WJ, Hsia LC, Chen KKY (2008) Metal desorption from copper(II)/nickel(II)-spiked kaolin as a soil component using plant-derived saponin biosurfactant. Process Biochem 43(5):488–498. https://doi.org/10.1016/j.procbio.2007.11.017

China MEEPR (2017) Water quality-determination of the chemical oxygen demand-dichromate method. China Environmental Science Press, Beijiing

China MEEPR (1997) Soil quality-determination of lead, cadmium-graphite furnace atomic absorption spectrophotometry. China Environmental Science Press, Beijing

China MEEPR (2010) Water quality-determination of ammonia nitrogen-Salicylic acid spectrophotometry. China Environmental Science Press, Beijing

China MEEPR (2012) Soil-determination of ammonium, nitrite and nitrate by extraction with potassium chloride solution -spectrophotometric methods. China Environmental Science Press, Beijing

China MEEPR (2014) Water quality-determination of 65 elements-inductively coupled plasma-mass spectrometry. China Environmental Science Press, Beijing

China MEEPR (2019) Soil and sediment-determination of copper, zinc, lead, nickel and chromium-flame atomic absorption spectrophotometry. China Environmental Science Press, Beijing

Deng Z, Qin L, Wang G, Luo S, Peng C, Li Q (2022) Study on behaviors of adsorption and desorption of ammonia-nitrogen in kaolin. Chinese Rare Earths 43(2):32–41. https://doi.org/10.16533/J.CNKI.15-1099/TF.202202004. (in Chinese)

Evangelou MWH, Ebel M, Schaeffer A (2007) Chelate assisted phytoextraction of heavy metals from soil. Effect, mechanism, toxicity, and fate of chelating agents. Chemosphere 68(6):989–1003. https://doi.org/10.1016/j.chemosphere.2007.01.062

Giacometti C, Cavani L, Baldoni G, Ciavatta C, Marzadori C, Kandeler E (2014) Microplate-scale fluorometric soil enzyme assays as tools to assess soil quality in a long-term agricultural field experiment. Appl Soil Ecol 75:80–85. https://doi.org/10.1016/j.apsoil.2013.10.009

Greaves MP, Webley DM (1965) A study of breakdown of organic phosphates by micro-organisms from root region of certain pasture grasses. J Appl Bacteriol 28(3):454–000. https://doi.org/10.1111/j.1365-2672.1965.tb02176.x

Gusiatin ZM, Klimiuk E (2012) Metal (Cu, Cd and Zn) removal and stabilization during multiple soil washing by saponin. Chemosphere 86(4):383–391. https://doi.org/10.1016/j.chemosphere.2011.10.027

Han FX, Banin A, Kingery WL, Triplett GB, Zhou LX, Zheng SJ, Ding WX (2003) New approach to studies of heavy metal redistribution in soil. Adv Environ Res 8(1):113–120. https://doi.org/10.1016/s1093-0191(02)00142-9

Han Z, Ma H, Shi G, He L, Wei L, Shi Q (2016) A review of groundwater contamination near municipal solid waste landfill sites in China. Sci Total Environ 569:1255–1264. https://doi.org/10.1016/j.scitotenv.2016.06.201

Huang R, Zhang B, Saad EM, Ingall ED, Tang Y (2018) Speciation evolution of zinc and copper during pyrolysis and hydrothermal carbonization treatments of sewage sludges. Water Res 132:260–269. https://doi.org/10.1016/j.watres.2018.01.009

Huang Z, Chen Q, Yao Y, Chen Z, Zhou J (2021) Micro-bubbles enhanced removal of diesel oil from the contaminated soil in washing/flushing with surfactant and additives. J Environ Manag 290:112570. https://doi.org/10.1016/j.jenvman.2021.112570

Huguenot D, Mousset E, van Hullebusch ED, Oturan MA (2015) Combination of surfactant enhanced soil washing and electro-Fenton process for the treatment of soils contaminated by petroleum hydrocarbons. J Environ Manag 153:40–47. https://doi.org/10.1016/j.jenvman.2015.01.037

Jelusic M, Vodnik D, Macek I, Lestan D (2014) Effect of EDTA washing of metal polluted garden soils. Part II: can remediated soil be used as a plant substrate? Sci Total Environ 475:142–152. https://doi.org/10.1016/j.scitotenv.2013.11.111

Juhasz AL, Britz ML, Stanley GA (1997) Degradation of fluoranthene, pyrene, benz a anthracene and dibenz a, h anthracene by Burkholderia cepacia. J Appl Microbiol 83(2):189–198. https://doi.org/10.1046/j.1365-2672.1997.00220.x

Kaczorek E, Chrzanowski L, Pijanowska A, Olszanowski A (2008) Yeast and bacteria cell hydrophobicity and hydrocarbon biodegradation in the presence of natural surfactants: Rharnnolipides and saponins. Bioresource Technol 99(10):4285–4291. https://doi.org/10.1016/j.biortech.2007.08.049

Kashem MA, Singh BR, Kawai S (2007) Mobility and distribution of cadmium, nickel and zinc in contaminated soil profiles from Bangladesh. Nutr Cycl Agroecosyst 77(2):187–198. https://doi.org/10.1007/s10705-006-9056-4

Khan FI, Husain T, Hejazi R (2004) An overview and analysis of site remediation technologies. J Environ Manag 71(2):95–122. https://doi.org/10.1016/j.jenvman.2004.02.003

Kobayashi T, Kaminaga H, Navarro RR, Iimura Y (2012) Application of aqueous saponin on the remediation of polycyclic aromatic hydrocarbons-contaminated soil. J Environ Sci Health Part a-Toxic/Hazardous Subst Environ Eng 47(8):1138–1145. https://doi.org/10.1080/10934529.2012.668106

Li Y, Huang T, Xie Z, Wang X, Chu X, Zhang X, Wu D (2019) Characteristics and assessment of heavy metal pollution in soil and groundwater of informal landfills. Earth Environ 47(3):361–369

Li W, Peters RW, Brewster MD, Miller GA (1995) Sequential extraction evaluation of heavy-metal-contaminated soil: How clean is clean? Chemistry

Liu Z, Li Z, Zhong H, Zeng G, Liang Y, Chen M, Wu Z, Zhou Y, Yu M, Shao B (2017) Recent advances in the environmental applications of biosurfactant saponins: a review. J Environ Chem Eng 5(6):6030–6038. https://doi.org/10.1016/j.jece.2017.11.021

Liu L, Li W, Song W, Guo M (2018) Remediation techniques for heavy metal-contaminated soils: principles and applicability. Sci Total Environ 633:206–219. https://doi.org/10.1016/j.scitotenv.2018.03.161

Lu N, Yu J, Fang Y (2010) Study on the process and mechanism of heavy metals desorption from contaminated soil by saponin. Anhui Agric Sci Bull 16(9):36–39. https://doi.org/10.16377/j.cnki.issn1007-7731.2010.09.004. (in Chinese)

Ma LQ, Rao GN (1997) Chemical fractionation of cadmium, copper, nickel, and zinc in contaminated soils. J Environ Q 26(1):259–264. https://doi.org/10.2134/jeq1997.00472425002600010036x

Mesbaiah FZ, Eddouaouda K, Badis A, Chebbi A, Hentati D, Sayadi S, Chamkha M (2016) Preliminary characterization of biosurfactant produced by a PAH-degrading Paenibacillus sp under thermophilic conditions. Environ Sci Pollut Res 23(14):14221–14230. https://doi.org/10.1007/s11356-016-6526-3

Mossop KF, Davidson CM (2003) Comparison of original and modified BCR sequential extraction procedures for the fractionation of copper, iron, lead, manganese and zinc in soils and sediments. Analytica Chimica Acta 478(1):111–118. https://doi.org/10.1016/s0003-2670(02)01485-x

Mulligan CN, Yong RN, Gibbs BF (2001) Heavy metal removal from sediments by biosurfactants. J Hazard Mater 85(1–2):111–125. https://doi.org/10.1016/s0304-3894(01)00224-2

Ramadan BS, Sari GL, Rosmalina RT, Effendi AJ, Hadrah (2018) An overview of electrokinetic soil flushing and its effect on bioremediation of hydrocarbon contaminated soil. J Environ Manag 218:309–321. https://doi.org/10.1016/j.jenvman.2018.04.065

Rodriguez-Cruz MS, Sanchez-Martin MJ, Andrades MS, Sanchez-Camazano M (2007) Retention of pesticides in soil columns modified in situ and ex situ with a cationic surfactant. Sci Total Environ 378(1–2):104–108. https://doi.org/10.1016/j.scitotenv.2007.01.021

Saeedi M, Li LY, Grace JR (2018) Desorption and mobility mechanisms of co-existing polycyclic aromatic hydrocarbons and heavy metals in clays and clay minerals. J Environ Manag 214:204–214. https://doi.org/10.1016/j.jenvman.2018.02.065

Saeedi M, Li LY, Grace JR (2019) Simultaneous removal of polycyclic aromatic hydrocarbons and heavy metals from natural soil by combined non-ionic surfactants and EDTA as extracting reagents: Laboratory column tests. J Environ Manag 248:109258. https://doi.org/10.1016/j.jenvman.2019.07.029

Soeder CJ, Papaderos A, Kleespies M, Kneifel H, Haegel FH, Webb L (1996) Influence of phytogenic surfactants (quillaya saponin and soya lecithin) on bio-elimination of phenanthrene and fluoranthene by three bacteria. Appl Microbiol Biotechnol 44(5):654–659

Song S, Zhu L, Zhou W (2008) Simultaneous removal of phenanthrene and cadmium from contaminated soils by saponin, a plant-derived biosurfactant. Environ Pollut 156(3):1368–1370. https://doi.org/10.1016/j.envpol.2008.06.018

Steliga T, Kluk D (2020) Application of Festuca arundinacea in phytoremediation of soils contaminated with Pb, Ni, Cd and petroleum hydrocarbons. Ecotoxicol and Environ Saf 194:110409. https://doi.org/10.1016/j.ecoenv.2020.110409

Strawn DG, Sparks DL (2000) Effects of soil organic matter on the kinetics and mechanisms of Pb(II) sorption and desorption in soil. Soil Sci Soc Am J 64(1):144–156. https://doi.org/10.2136/sssaj2000.641144x

Tao Q, Li J, Liu Y, Luo J, Xu Q, Li B, Li Q, Li T, Wang C (2020) Ochrobactrum intermedium and saponin assisted phytoremediation of Cd and B a P co-contaminated soil by Cd-hyperaccumulator Sedum alfredii. Chemosphere 245:125547. https://doi.org/10.1016/j.chemosphere.2019.125547

Tran HT, Lin C, Hoang HG, Bui XT, Le VG, Vu CT (2022) Soil washing for the remediation of dioxin-contaminated soil: a review. J Hazard Mater 421:126767. https://doi.org/10.1016/j.jhazmat.2021.126767

Vandijk HFG, Roelofs JGM (1988) Effects of excessive ammonium deposition on the nutritional status and condition of pine needles. Physiologia Plantarum 73(4):494–501

Wang L, Niu J, Yang Z, Shen Z, Wang J (2008) Effects of carbonate and organic matter on sorption and desorption behavior of polycyclic aromatic hydrocarbons in the sediments from Yangtze River. J Hazard Mater 154(1–3):811–817. https://doi.org/10.1016/j.jhazmat.2007.10.096

Wang Q, Liu X, Zhang X, Hou Y, Hu X, Liang X, Chen X (2016) Influence of tea saponin on enhancing accessibility of pyrene and cadmium phytoremediated with Lolium multiflorum in co-contaminated soils. Environ Sci Pollut Res 23(6):5705–5711. https://doi.org/10.1007/s11356-015-5784-9

Wang X, Chi Q, Liu X, Wang Y (2019) Influence of pyrolysis temperature on characteristics and environmental risk of heavy metals in pyrolyzed biochar made from hydrothermally treated sewage sludge. Chemosphere 216:698–706. https://doi.org/10.1016/j.chemosphere.2018.10.189

Wei Y, Liang X, Tong L, Guo C, Dang Z (2015) Enhanced solubilization and desorption of pyrene from soils by saline anionic-nonionic surfactant systems. Colloids Surfaces a-Physicochem Eng Aspects 468:211–218. https://doi.org/10.1016/j.colsurfa.2014.12.026

Wu Z, Zhong H, Yuan X, Wang H, Wang L, Chen X, Zeng G, Wu Y (2014) Adsorptive removal of methylene blue by rhamnolipid-functionalized graphene oxide from wastewater. Water Res 67:330–344. https://doi.org/10.1016/j.watres.2014.09.026

Wuana RA, Okieimen FE, Imborvungu JA (2010) Removal of heavy metals from a contaminated soil using organic chelating acids. Int J Environ Sci Technol 7(3):485–496. https://doi.org/10.1007/bf03326158

Ye M, Sun M, Wan J, Fang G, Li H, Hu F, Jiang X, Kengara FO (2015) Evaluation of enhanced soil washing process with tea saponin in a peanut oil-water solvent system for the extraction of PBDEs/PCBs/PAHs and heavy metals from an electronic waste site followed by vetiver grass phytoremediation. J Chem Technol Biotechnol 90(11):2027–2035. https://doi.org/10.1002/jctb.4512

Ye J, Chen X, Chen C, Bate B (2019) Emerging sustainable technologies for remediation of soils and groundwater in a municipal solid waste landfill site - a review. Chemosphere 227:681–702. https://doi.org/10.1016/j.chemosphere.2019.04.053

Zhang J (2021) Study on the leaching mechanism of mixed-contaminated soil in sanitary landfill and the screening of efficient leaching reagents. Southeast University, Nanjing