A microemulsification approach for removing organolead and gasoline from contaminated soil

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Abstract

Remediation of soils contaminated with leaded gasoline due to leakage, spillage, and inappropriate disposal is an important environmental consideration. Columns of a loam soil initially saturated with saline solution (aqueous 0.01 M NaCl) were contaminated with 48 ml of leaded gasoline. The contaminated soil columns were then flushed sequentially with saline and surfactant/cosurfactant/water (S/CoS/W) solutions in order to investigate removal efficiencies for residual tetraethyl lead (TEL) and gasoline components. As expected, the saline solution immiscibly displaced only limited amounts of mobile gasoline and associated TEL (dissolving in gasoline) components from the soil columns. However, immobile or residual gasoline and associated TEL entrapped in the soil pores were removed primarily as the S/CoS/W solution produced leaded-gasoline-in-water (LG/W) microemulsions. The S/CoS/W solution removed 95% of the immobile gasoline and 90% of the immobile Pb from the soil columns when the initial saturation of leaded gasoline was approximately 30% (or 48 ml) in the soil columns. Mass balance analysis shows that one gram of surfactant (sodium lauryl sulfate) removed 0.6 g of immobile gasoline and 2 mg of immobile Pb from the soil. These immobile gasoline and immobile Pb were not removable by NaCl solution.

Keywords: Microemulsions; Surfactants; Gasoline; Organolead; Remediation

1. Introduction

Contamination of soil and groundwater resources with nonaqueous phase liquids (NAPLs) and organic forms of heavy metals is a current public concern. One of the most prevalent heavy metals found in contaminated subsurface environments is lead (Pb). The source of Pb contamination is due mostly to former emissions from automobiles using leaded gasoline during the period from 1925 to 1975. Tetraethyl lead

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(TEL) is a nonionic organic compound that has been selected as an antiknock component for leaded gasoline [1]. Over the past two decades in the United States, concern over the problem of Pb contamination of soils due to the use of leaded gasoline has greatly increased. This concern for Pb contamination exists although the use of leaded gasoline in autos has been greatly decreased within the past decade [2]. Leaded gasoline is, however, still used extensively as an aviation fuel. At present, no federal regulations address the acceptable level of Pb in soils for public health protection and remedial action. However, soil and drinking water with Pb concentrations >50 ppm and >15 ppb, respectively, are considered toxic waste by the United States Environmental Protection Agency (US EPA) [2].

TEL is very unstable in soil and undergoes a series of sequential dealkylation reactions [2]. TEL is first degraded to triethyl lead (TREL) cations which in turn are degraded to diethyl lead (DEL) cations, and finally to inorganic Pb^{+2}. Mobility of TEL and its ionic degradation products in soil contaminated with leaded gasoline is largely unknown because of the complexity of chemical reactions or sorption by clay minerals and organic soil components. As rainwater infiltrates into soils, the leachability of TEL and its degradation products should be determined largely by sorption and degradation reactions. An unknown but finite potential exists for leachability of TEL from soils previously used for disposal of fuel sludge. The degree of water solubility of TEL and its degradation products greatly influences the potential for transport during water flow in soil. The relatively low water-solubility for TEL provides low potential for transport through soil. However, the high water-solubility of ionic TEL degradation products such as TREL and DEL species makes these compounds particularly suspect for mobility in soil systems during periods of water infiltration into the soil.

In recent years, surfactants have been used as a remedial technique to remove NAPLs from contaminated soils [3–8]. A surfactant is a chemical substance that lowers the surface or interfacial tension of the medium in which it is dissolved. The important properties of a surfactant are its amphipathic structure, monolayer orientation at interfaces, and adsorption at interfaces. A surfactant molecule is made up of two functional groups, a hydrophilic head and a lipophilic carbon tail. The two groups line up between the gasoline and water phases with their opposing ends dissolved in the respective phases. This arrangement creates a monolayer at the interface, thereby, decreasing the interfacial tension between gasoline and water. The balance of the head group and carbon tail determines which phase the surfactant molecule dissolves into most easily. That is, if the head group is more heavily balanced, the surfactant as a whole will be more water soluble. In this case, it will tend to pull gasoline into solution as droplets encased in a shell of surfactant molecules.

Although the use of surfactants to clean up NAPLs from contaminated soils has shown significant potential for application, one of the major obstacles is the potential formation of unstable macroemulsions with unfavorable phase conditions such as a tendency to form gels. Macroemulsions are thermodynamically unstable and have droplet sizes ranging from 0.1 to 10 μm. Flow of macroemulsions has been reported to inhibit the transport of NAPLs through soil [4]. This inhibition occurs because macroemulsions are unstable and can be disemulsfied (or broken) into two

immiscible liquids such as NAPL and water. In addition, the relatively larger size of droplets in a macroemulsion has a tendency to clog micropores during transport in soils.

However, if a mixture of surfactant (S), co-surfactant (CoS), and water (W) is used to remediate NAPLs from contaminated soil, an oil-in-water microemulsion may result [7]. An oil-in-water microemulsion is a special kind of stabilized emulsion consisting of water, oil (or NAPL), surfactant, and co-surfactant components [9, 10]. Microemulsions tend to be transparent and thermodynamically stable. Droplet sizes for microemulsions range from 0.1 to 0.01 µm. Flow of microemulsion in soils has been reported to enhance the transport of gasoline and Pb species [5]. Enhanced flow occurs because of the stability and favorable phase behavior of microemulsions, as well as the small size of NAPL droplets dispersed in an aqueous phase relative to the soil pores. Therefore, miscible displacement of microemulsions through water-saturated or -unsaturated soil can greatly minimize clogging of pores with NAPL droplets.

Unfortunately, laboratory experiments have not been reported for the simultaneous removal of TEL, its ionic degradation products, and gasoline using the microemulsification approach. The objective of this study was to determine if components of leaded gasoline can be effectively removed from contaminated soil with a selected S/CoS/W solution that can generate gasoline-in-water microemulsions. The specific S/CoS/W solution was comprised of sodium lauryl sulfate as surfactant, n-pentanol as co-surfactant, and water.

2. Materials and methods

2.1. Reagents

Analytical grade TEL was purchased from All-Chemie Ltd. (Ft. Lee, NJ). Pb standard solution and sodium lauryl sulfate (surfactant) were purchased from Fisher Scientific (Orlando, FL). All other chemicals such as concentrated HCl, n-hexane, n-pentanol (co-surfactant), KI, KIO_3, NaCl, and concentrated HNO_3 were of analytical grade. Commercial-grade leaded aviation gasoline was purchased from the Regional Airport in Gainesville, FL, USA. The organolead species in this gasoline is TEL with a concentration of $700 \, \text{mg} \, l^{-1}$. Benchmark properties of TEL and sodium lauryl sulfate are given in Table 1.

Madison loam soil from Dobbins Air Force Base, Georgia was used for the experiments. Selected soil properties are given in Table 2. The specific S/CoS/W solution was made by mixing 4.3 g sodium lauryl sulfate, 8.7 g n-pentanol, and 87 g deionized (DI) water. The solution thus contained 4.3 and 8.7% surfactant and co-surfactant, respectively. The specific S/CoS/W solution was chosen with the aid of a pseudo ternary phase diagram for an oil-in-water microemulsion as reported by Rosano and Clausse [7]. This S/CoS/W solution has shown to generate stable LG/W microemulsions in pre-experimental testing. A detailed discussion on how to select optimal compositions of S/CoS/W can be found on Rosano and Clausse's report

Table 1
Benchmark properties of TEL and sodium lauryl sulfate

Parameter	Value	Reference
TEL		
Formula	$Pb(C_2H_5)_4$	[18]
Molecular wt.	323.4	
Density	$1.65 \, mg \, l^{-1}$ (20 °C)	[18]
Solubility		
in water	Insoluble ($< 0.1 \, mg \, Pb \, l^{-1}$)	[18]
in hexane	Soluble	[18]
Toxicity	LD_{50} (oral) $250 \, mg \, kg^{-1}$	[19]
Sodium lauryl sulfate		
Formula	$CH_3(CH_2)_{10}CH_2OSO_3Na$	
Molecular wt.	288	
Toxicity	None	
Ionic nature	Anionic	
Critical micelle conc.		
in Water	$0.0081 \, mole \, l^{-1}$	[9]
in 0.1 M NaCl	$0.00139 \, mole \, l^{-1}$	[9]

Table 2
Selected properties of Madison loam soil

Property	Madison soil
pH	4.7
Organic C (%)	0.61
Sand (%)	43
Silt (%)	40.3
Clay (%)	16.09
Soil depth (m)	0–0.15

[7]. Although the surfactant (4.3%) and co-surfactant (8.7%) compositions used in this study were high with respect to cost-effectiveness, the actual S/CoS/W mixture can be changed to reduce the amounts of surfactant and co-surfactant, depending on the estimated NAPL concentrations in a given contaminated site using the pseudo ternary phase diagram. Our overall focus was to evaluate microemulsification as a means for remediation of leaded gasoline-contaminated soil under controlled laboratory conditions.

2.2. Standards

Analytical grade TEL was dissolved in *n*-hexane to make working standards of TEL at concentrations of 0.01, 0.05, 0.1, 1, 5, and $10 \, mg \, l^{-1}$. Leaded aviation gasoline was dissolved in *n*-hexane to make standard solutions of gasoline with concentrations of 120, 480, 1500, and $3000 \, mg \, l^{-1}$. A 0.1 M ICl solution was prepared by

mixing 40 ml DI water, 44.5 ml concentrated HCl, 11.0 g KI, and 7.5 g KIO_3. This solution was used to convert the TEL into Pb^{+2} so that the total Pb content in the effluent could be measured with an atomic absorption spectrophotometer [11]. Working standards of Pb solution for measuring total Pb content in the effluent were made by adding standard Pb solution in 3:1 DI water:0.1 M ICl solution at concentrations of 1, 10, and 20 mg l^{-1}. Working standards of Pb for measuring total Pb content in soil samples before and after the experiments were prepared according to the US EPA method 3035 [12].

2.3. Column packing

Duplicate glass cylinders with 4.8 cm inner diameter were used to contain 16 cm length soil columns. Air-dried soil, which had been crushed and passed through a 0.5 mm screen, was poured into the cylinders in 2-cm depth increments, and stirred to prevent layering.

The columns were initially saturated with 0.01 M NaCl solution with an upward-flow mode using a peristaltic pump to establish a low liquid flow rate (6 ml h^{-1}) until the soil columns were approximately saturated. The choice of the diluted NaCl solution was based on a consideration that the presence of Na^+ ion favors the formation of gasoline-in-water microemulsion for the experimental conditions used in this study. A detailed discussion of the effects of Na^+ and Ca^{+2} on the formation of oil-in-water microemulsion is given by Schramm [10]. Our pre-experimental testing had shown that the dispersion of clay particles due to the use of 0.01 M NaCl solution is minimal.

After wetting, the soil columns were weighed to determine the pore volumes (i.e., the mass difference between the dry and water saturated soil columns). The column flow was then increased to provide a desired Darcy flow velocity of 1.5 cm h^{-1}, which is equivalent to a flow rate of 0.5 ml min^{-1} (1 ft day^{-1}). This relatively low flow rate was selected to allow enough time for interactions between S/CoS/W solution and residual gasoline to generate microemulsions. Experimental conditions for the columns are given in Table 3.

Table 3
Experimental parameters

Parameters	Madison soil
Bulk density, ρ_b (kg m^{-3})	1.02×10^{-3}
Darcy velocity, V (m s^{-1})	4.2×10^{-6}
Column length (m)	0.16
Cross section area (m^2)	0.0018
Applied leaded gasoline (ml)	48
Saturated water content (m^3 m^3)	0.54
Pore volume (ml)	154.5
Capillary number	1.4×10^{-7}

2.4. Column experiment

After steady state flow (1.5 cm h^{-1}) was reached, 48 ml of leaded gasoline (enough to provide about 30% saturation of the column pore spaces) was pumped through the soil columns with an upward-flow mode. The columns were then flushed with NaCl solution (1.6 pore volumes) at a velocity of 1.5 cm h^{-1} until gasoline was no longer detected in effluent samples. After overnight interruption of flow, the columns were washed with 6.4 pore volumes of the S/CoS/W solution using the same flow velocity.

Effluent was collected every 10 min in test tubes using an automatic fraction collector. During the collection of effluent, stoppers were immediately placed into the test tubes to reduce the evaporative loss of gasoline. All test tubes and soil columns were wrapped with aluminum foil before starting the experiment to prevent photodegradation of TEL by incoming light. For analyzing TEL and gasoline in the effluent flushed with NaCl solution, a 0.8 ml-sample of effluent was pipetted into 10 ml of n-hexane and shaken for 30 min. Similar procedures were used for analyzing TEL and gasoline in effluent washed with the S/CoS/W solution except that a small amount of NaCl salt was added to the samples to prevent the possible coalescence of hexane and effluent.

After termination of liquid flow in the columns, the soils were analyzed for residual gasoline, TEL, and total Pb contents. For analyzing TEL and gasoline contents in soil samples, 1 g of wet soil was added to 10 ml of n-hexane and shaken for 1 h. Then, the samples were centrifuged at 2000 rpm for 20 min and decanted. The clear supernatant was used to measure TEL and gasoline by GC and UV spectrophotometer, respectively. The total Pb contents in the column soil before and after the flow experiments were measured by the US EPA 3035 method [12].

Current methods for detecting gasoline contaminated sites include analysis for benzene, toluene, xylene, ethylbenzene, total petroleum NAPLs, or immunoassay [13]. In this study we used the UV spectrophotometer to measure components of aromatic and unsaturated bonds as an indicator for gasoline. We assumed that concentration of gasoline is proportional to concentration of aromatic and unsaturated components in effluent and soil samples. A preliminary study shows that the scanning diagrams measured by a UV spectrophotometer between the pure gasoline and the gasoline in the column effluent and soil samples extracted by hexane had similar distribution patterns. The recovery rate of gasoline spiked in the effluent samples was about 93% for our experimental conditions. This observation justified the use of the UV spectrophotometry to measure aromatic and unsaturated bonds as an indicator of gasoline.

A 5-ml aliquot sample of effluent was poured into a cuvette for analyzing gasoline contents using the UV spectrophotometer with the scanning wavelengths ranging from 240 to 300 nm. A 20-μl aliquot sample was injected into a GC (HP 5710A Gas Chromatograph) to analyze the TEL concentration with GC conditions as reported by Chakraborti et al. [14]. The method for measuring total Pb contents in the effluent samples was developed by Ouyang et al. [11].

2.5. TEL degradation and sorption experiments

Triplicate 5 ml samples of leaded gasoline with a TEL concentration of $700 \, mg \, l^{-1}$ and 5 g wet Madison soil were added to 50-ml test tubes and shaken for 2, 6, 24, and 48 h, respectively. The test tubes were rotated end-over-end manually for 2 min and put on the shaker to insure complete mixing. The soil samples were then centrifuged at 20 000 rpm for 10 min with a centrifuge. The clear supernatant was analyzed for TEL using GC. Differences between the initial and final concentrations for samples were taken as the combined amount of TEL degradation and adsorption by the soil at a given time. Results of TEL degradation from this study were used to compare the stability of TEL in soil between batch experiments and column experiments.

3. Results and discussion

3.1. Immiscible displacement of gasoline

Immiscible displacement of leaded gasoline provided a means to contaminate the water-saturated soils. Subsequent flushing of the soil columns with 0.01 M NaCl solution provided a means to remove mobile gasoline, thus leaving immobile or residual gasoline in the soil columns. The duplicate experiments were initiated by displacing 48 ml (0.3 pore volume) of aviation leaded gasoline through the soil columns, followed by 1.6 pore volume of NaCl solution flushing.
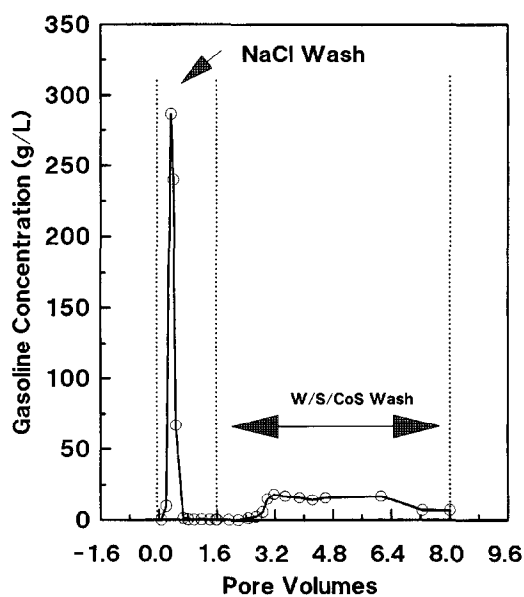

Fig. 1. BTC for gasoline in Madison loam soil experiments.

Analysis of the breakthrough curve (BTC) for gasoline in the column effluent (Fig. 1) indicated that 41% of the total applied gasoline was displaced with 1.6 pore volumes of NaCl solution leaving 59% as residual or immobile gasoline entrapped in the soil pores (i.e., intraparticle and interparticle voids). Concentrations of gasoline in the effluent ranged from a minimum of $0.17\,g\,l^{-1}$ to a maximum of $287\,g\,l^{-1}$. When the leaded gasoline and NaCl solution were sequentially applied to the soil columns, capillary attractive forces caused some gasoline to be displaced by NaCl solution from relatively large pores but not from small pores. Mobilization of gasoline droplets in this immiscible displacement process can be estimated by the capillary number. The capillary number is a dimensionless ratio of viscous to capillary forces, and provides a measure of how strongly residual gasoline is trapped within a given porous medium [15]. A capillary number for two immiscible liquids flow can be calculated by

$$N_c = \frac{v\mu}{\delta},\tag{1}$$

where v is Darcy flow velocity $(m\,s^{-1})$, μ is viscosity of the displacing phase $(1.01 \times 10^{-3}\,N\,s\,m^2)$, and δ is the interfacial tension between the displaced and displacing phases $(0.03\,N\,m^{-1})$. Gupta and Trushenski [16] reported that mobilization of gasoline entrapped in soil usually begins at a capillary number of about 10^{-5} (the critical capillary number), and complete oil recovery occurs at a high value of capillary number of about 10^{-2}. The capillary number for our experiments was 2.4×10^{-7} (Table 3), which was considerably smaller than the critical capillary number. Therefore, some of the gasoline applied to the soil was expected to be entrapped in the pores.

A steep breakthrough of gasoline occurred for both front and back sides of the BTC (Fig. 1) when the soil columns were flushed by 1.6 pore volumes of NaCl solution. This result indicates that convection flow rather than diffusion flow of gasoline was a dominant mechanism during the displacement process. This occurred because gasoline and NaCl solution are two immiscible liquids, thereby diffusion of gasoline into NaCl solution or vice versa was trivial.

3.2. Miscible displacement of gasoline

After saline flushing of the soil columns to remove mobile gasoline, the S/CoS/W solution was applied to the columns to remove residual or immobile gasoline. The process started by displacing 6.4 pore volumes of S/CoS/W solution through the soil columns. About 54% of the total applied gasoline was flushed from the soil columns leaving only 5% of the total applied gasoline as residue in the column soils (Fig. 1). The effectiveness of remediation with the S/CoS/W solution is primarily attributable to a high ability of the solution to dramatically lower the interfacial tension between gasoline and water [7], creating gasoline-surfactant-water micelles, and producing gasoline-in-water microemulsions.

Several important features of microemulsions are their thermodynamic stability, high solvent power [17], and relatively smaller size of droplets as compared to those

Table 4
Diameters of colloidal particles, molecules in solution, macroemulsion droplets, microemulsion droplets, and soil micropores

Parameter	Diameter	Reference
Colloid particles	0.1–2 µm	[20]
Molecules in solution	<0.001 µm	[20]
Macroemulsion droplets	1–10 µm	[20]
Microemulsion droplets	0.01–0.1 µm	[20]
Soil micropores	<30 µm	[21]

of macroemulsions. Table 4 gives the average diameters for the colloid particles, macroemulsion droplets, microemulsion droplets, and soil micropores. Unlike the colloid suspension and macroemulsion, the microemulsion has a much smaller droplet diameter than that of soil micropores. Therefore, capture of gasoline droplets by soil pores was minimized in the microemulsion flow process, resulting in the effective removal of gasoline from the soil columns.

In contrast to a steep initial breakthrough of gasoline in effluent from soil columns when the columns were flushed by NaCl solution, when the soil columns were flushed by the S/CoS/W solution the gasoline BTC was characterized by excessive tailing and smaller slope of the breakthrough (Fig. 1). This result indicates that dispersive transport of gasoline was an important process during the miscible flow of gasoline-in-water microemulsion due to the use of the S/CoS/W solution.
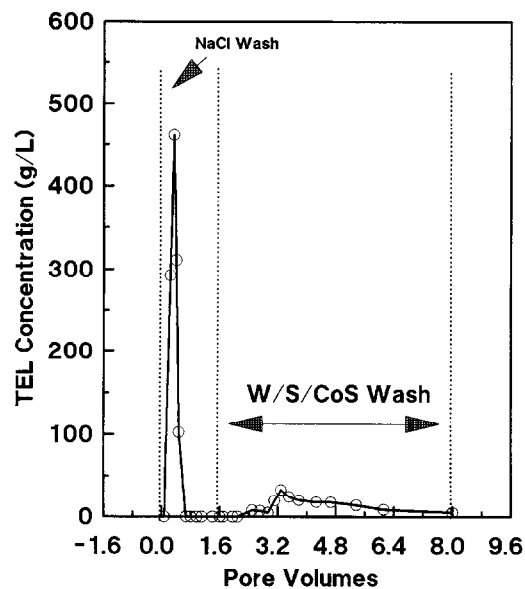

Fig. 2. BTC for TEL in Madison loam soil experiments.

3.3. Displacement of Pb species

The breakthrough pattern observed for TEL (Fig. 2) was similar to that for gasoline (Fig. 1). Approximately 51.5% and 33.6% of applied TEL were recovered from the soil columns by flushing with NaCl solution (immiscible displacement) and the S/CoS/W solution (miscible displacement), respectively. Only about 0.04% TEL was detected as residue in soil samples at the end of the experiments. An estimation of total TEL mass shows that only 83% of applied TEL was recovered. This low recovery rate of TEL was attributed to the biotic and abiotic degradation of TEL in the soils. This low rate was confirmed by our batch study results for the degradation of TEL in the same soil (Fig. 3). Approximately 74% of the applied TEL disappeared (or 26% of TEL remaining in the soil) by 48 h, which was the time period used for the column experiments (Fig. 3). The discrepancy between the mass recovery rates of TEL in batch (74%) and column (83%) investigations at 48 h was attributed to the time of TEL contact with the soil. The column flow involved a shorter contact time and possibly nonequilibrium reactions between TEL and soil due to the earlier breakthrough of some TEL from the soil columns, whereas the batch study involved a longer contact time and probably equilibrium reactions between TEL and soil due to the complete mixing of TEL and soil with shaking. Mass balance analysis shows that average total Pb recovery rate for the experiments was 93% indicating that experimental measurements were reasonable.
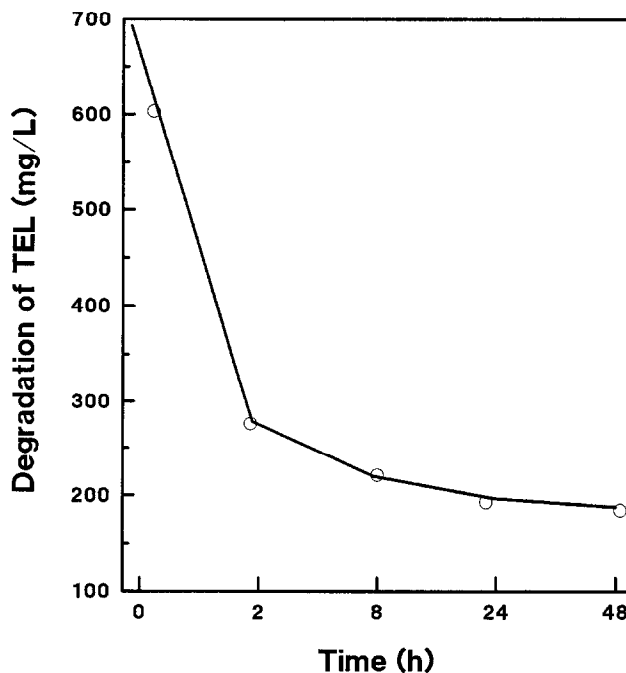

Fig. 3. Degradation of TEL in gasoline by Madison loam soil experiments at times of 2, 8, 24, and 48 h. Initial TEL concentration was 700 mg l^{-1}.
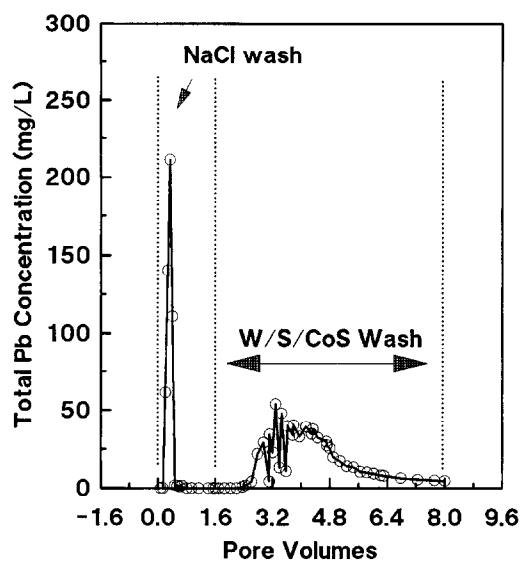
Fig. 4. BTC for total Pb in Madison loam soil experiments.

Comparison of BTCs for gasoline (Fig. 1) and for TEL (Fig. 2) shows that TEL was removed from the soil columns by flushing with NaCl solution due to the removal of the gasoline phase. This occurred because TEL is highly soluble in gasoline. The BTC for total Pb is shown in Fig. 4. It should be noted that inorganic ionic Pb^{+2} adsorbed on the soil cannot be easily washed out by diluted NaCl solution although a certain amount of Pb^{+2} adsorbed onto the surfaces of soil particles or organic matter may be washed out by the S/CoS/W solution. Therefore, the breakthrough of total Pb from the columns flushed by NaCl solution was mainly a result of the breakthrough of organic TEL dissolved in gasoline and its water-soluble degradation products such as TREL, DEL, and Pb^{+2} in solution phase. A mass balance analysis of TEL for the experiments may be inadequate due to the unstable nature of TEL in the soil. However, a mass balance analysis of total Pb for the experiments can provide information about the accuracy of the experiments. The equation used to estimate the mass balance of total Pb is

$$\text{Mass balance}\% = (Pb_{\text{effluent}} + Pb_{\text{residual}})/(Pb_{\text{input}} + Pb_{\text{back}}), \qquad (2)$$

Table 5
Mass balance of total Pb for the column flow experiments in Madison soil

Parameter	Madison soil
Mass in background soil (mg)	18.2
Mass in influent (mg)	21.5
Mass in effluent (mg)	20.4
Mass in soil (mg) at the end of the exp.	19.6
Mass balance	101%

where $Pb_{effluent}$ is the total Pb in the effluent (mg), $Pb_{residual}$ is the residual total Pb in the soil column at the end of the experiment (mg), Pb_{input} is the total Pb input to the soil column (mg), and Pb_{back} is total background Pb in the soil before the experiments (mg). Values for parameters in Eq. (2) are given in Table 5. The average total Pb recovery for the experiments was 101%. This high recovery rate indicates that the experimental error is minimal.

3.4. Remediation efficiency of gasoline and total Pb

A residual analysis was performed by estimating amounts of gasoline applied to the soil columns, flushed from the soil columns, and remained as residue in the soil columns at the end of experiments. Results show that 5.89% of gasoline was left in the columns after flushing with NaCl solution and the S/CoS/W solution when the column pore spaces were initially saturated with 30% of leaded aviation gasoline (about 48 ml). The remediation efficiencies of NaCl solution (1.6 pore volumes) and the S/CoS/W solution (6.4 pore volumes) for removing gasoline were 41% and 53%, respectively. Experimental results indicate that the NaCl solution can be used to displace only limited amounts of gasoline in the soil pores. In contrast, the use of the S/CoS/W solution alone removed about 95% of the immobile residual gasoline from the soil columns (i.e., $54\% \div [1 - 43\%] = 95\%$).

Remediation efficiencies of the S/CoS/W solution for removing the immobile residual gasoline (i.e., after the columns were flushed with NaCl solution) from the soil was 3.9 ml gasoline per pore volume (PV) of the S/CoS/W mixture. This indicates that one gram of surfactant (sodium lauryl sulfate) removed 0.6 g of immobile or residual gasoline (which was not removable by NaCl solution) in the soil. A similar analysis was performed to estimate the remediation efficiency of total Pb from the soil columns by the saline and S/CoS/W solutions. Results show that 6.5% of total Pb was left in the columns after flushing with NaCl and S/CoS/W solutions (Table 5). Remediation efficiency of the S/CoS/W solution for removing the immobile total Pb (i.e., after the columns were flushed with NaCl solution) from the soil column is 90%. This implies that one pore volume of the S/CoS/W solution removed 2 mg of total Pb from the soil columns. Further study is needed to test the remediation efficiency of gasoline and organolead with S/CoS/W solution under the hydraulic control conditions in unsaturated and heterogeneous soils with varying organic matter contents before a given solution is used in the field scale.

Acknowledgements

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