



Potential benefit of surfactants in a hydrocarbon contaminated soil washing process: Fluorescence spectroscopy based assessment

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ABSTRACT

Soil washing is an ex situ soil remediation treatment process. The purpose of soil washing is to clean the major gravel and sand fractions, concentrating contamination into the fine silt and clay fractions. The addition of surfactants can improve the efficiency of this method. Here we report the use of UV fluorescence spectroscopy to assess the hydrocarbon cleaning process as a rapid and cost effective alternative to gas chromatography. Three wash solutions were tested on a total petroleum hydrocarbon contaminated soil: water, Sea Power 101 (SP101) at 1% (v/v) and Tween80 at 0.5% (w/v). The most effective to wash the gravel and sand was SP101 (54 and 65% improvement over the water only wash, respectively) which moved contamination to the silt fraction (94% of contaminants). Tween80 appeared not to enhance TPH removal efficiency from the gravel and sand fractions but did concentrate TPH in the effluent (95% more than water wash). In addition to TPH removal from gravel and sand, SP101 also showed potential benefit in the soil washing sedimentation process, enhancing sludge/water volume separation by 10% over the water only wash. In summary, fluorescence spectroscopy proved an effective technique to compare TPH removal efficiencies as part of soil washing laboratory based treatability testing.

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1. Introduction

Soils contaminated with aromatic hydrocarbons, including polycyclic aromatic hydrocarbons (PAH) are often slow to remediate by techniques such as bioremediation [1]. Soil washing, on the other hand is a comparatively rapid soil remediation technique [2–4], however, the hydrophobic nature of aromatic hydrocarbons can impede the effectiveness of the water only wash process to clean gravel and sand fractions and thereby concentrate contamination in the fine solid fractions [5]. Therefore, particularly where contaminants are coated or adsorbed onto gravel and sand fractions, surfactants are of potential interest to improve the cleaning process.

Many articles researching the use of surfactants in the soil washing process refer to removal efficiency as the ability to concentrate contaminants into the liquid phase (effluent) [6,7]; however, these are in the main based on soils consisting of a single (sand) fraction only. Contaminant removal efficiency in soils consisting of multiple fractions such as gravel, sand, silt and clay are more complicated. Specifically, contaminant removal efficiency can be considered in

terms of the removal from a particular solid fraction or, the transfer from the soil to the liquid effluent. Given that the objective of soil washing is to recover the gravel and sand fractions, contaminant removal efficiency should address these fractions specifically. In addition, as the silt and clay fractions are usually heavily contaminated and often require disposal, it may be better to concentrate contamination in silt and clay rather than the liquid effluent. As such, surfactants which primarily enhance the desorption of contaminants from gravel and sand fractions, without necessarily increasing contaminant water solubility are of interest.

Numerous studies have assessed the potential of chemical surfactants such as SDS, Brij 35 or Tween80 to improve removal efficiency of contaminants in the soil washing process; the results of which are that surfactants show varying contaminant removal efficiencies. With the relative performance of a particular surfactants being soil specific, laboratory based treatability tests are beneficial to decide the most appropriate surfactant for a specific soil and site. For example, Surfapcol has been shown to be more effective than Tween80 or SDS in sand [8], whereas, Tween80 and Brij35 had similar removal efficiencies in a sandy loam soil [9].

In addition to the choice of surfactant, many studies have focused on defining optimal washing parameters for the surfactant, for example, the influence of washing time, temperature, as well as surfactant concentration [7,10]. In these previous articles, washing times varied from 5 min to 48 h; however, in order to have

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a time-efficient treatability test and to mimic currently available soil washing plant, washing duration should last minutes rather than hours. Finally, artificial ageing of pollution has also been studied, although in reality, ageing of pollution is not a variable in soil washing treatability testing. There are few articles to date using soils which have not otherwise been artificially contaminated.

In recent years, biosurfactants have increasingly been considered for potential application in soil washing processes. Biosurfactants have been isolated from various sources including microorganisms (such as bacterial rhamnolipid), animals (frog saliva) and even plants [11,12]. In the relatively few studies to date, biosurfactants have shown similar or better results when compared with chemical surfactants in terms of contaminant removal efficiency. For example, rhamnolipid showed a better TPH removal than Tween80 [13] and a similar removal efficiency to SDS [14,15]. Furthermore, biosurfactants have a lower toxicity, excellent biodegradability, are less influenced by environmental conditions (such as pH and temperature) and are more cost-effective [16].

In addition to contaminant removal from the gravel and sand fractions, the use of surfactants in a soil washing process will have an influence on sedimentation of the fine solid component. Flocculating agents may be used as part of the sedimentation process and so the influence of surfactants on flocculating agents also needs to be considered. Amongst other properties surfactant can reduce suspension viscosity [17]. Interactions between surfactants and flocculants can be positive or negative depending on chemical type(s) [18,19]. Therefore, soil washing treatability testing involving surfactants should also consider effects on sedimentation processes.

Finally, in the majority of articles, assessment of surfactant performance has been made using gas chromatography [9,10] or gravimetric method [8,20]. As an alternative to gas chromatography in particular, Ultra-Violet fluorescence (UVF) spectroscopy is of interest as a comparatively rapid and cost-effective technique that retains a high degree of sensitivity.

The aim of this study was to determine the effects of two surfactants (the chemical surfactant Tween80 and a biosurfactant Sea Power 101) on soil washing of a historically contaminated soil. Specifically, their potential benefit on hydrocarbon removal from gravel/sand and, effects on the sedimentation process when compared to the water only wash process. The washing process efficiency was assessed using UV fluorescence spectroscopy as a rapid, cost effective technique of benefit for soil washing remediation treatability testing.

2. Materials and methods

2.1. Solutions for soil washing

The three wash solutions were:

- (1) Deionised water (hereafter referred to as 'water').
- (2) Sea Power 101 at 1% (SP101); a bio-stimulant and biosurfactant product (supplied by Sea-Chem Ltd, Shropshire, UK). The concentration was based on previous washing experiments by ourselves which demonstrated that 1% was effective when compared to higher concentrations. In addition, the critical micelle concentration (CMC), was determined with a UV fluorescence method using pyrene as described previously [21,22]. Briefly, emission (EM) scans were undertaken using a UV fluorescence spectrophotometer (see Section 2.6 for details) in order to excite pyrene at 334 nm and record its emission at 373 and 384 nm.

Table 1
Soil characteristics.

Moisture content	16.8%
pH	6.2
<i>Soil distribution</i>	
Silt/clay	23.6%
Sand	70.6%
Gravel	6.5%
<i>TPH aromatic – whole soil (mg/kg)</i>	
UVF	283.85
GC-FID	549.95
<i>TPH aromatic – gravel only fraction (mg/kg)</i>	
UVF	90.85
GC-FID	230.75

- (3) Tween80 at 0.5% (Tween80); a non-ionic surfactant (Fisher Scientific, Loughborough, UK). The concentration was chosen based on previous articles [8].

The solutions were prepared in a 2.5 L container with deionised water.

2.2. Solutions for flocculation test

The solutions were:

- (1) Deionised water (hereafter referred to as 'water'),
- (2) Ferric chloride at 0.5% (v/v) (FeCl_3); a flocculating agent,
- (3) Sea Power at 1% (v/v) (SP101),
- (4) Sea Power at 1% (v/v) and ferric chloride at 0.5% (SP101& FeCl_3),
- (5) Tween80 at 0.5% (w/v) (Tween80),
- (6) Tween80 at 0.5% (w/v) and ferric chloride at 0.5% (v/v) (Tween80& FeCl_3).

2.3. Soil

A historically hydrocarbon contaminated soil was taken from a former oil refinery located in Ayrshire, Scotland, UK. Soil characteristics can be found in Table 1.

2.4. Soil washing procedure

For each wash solution, three replicate experiments were conducted following the stages below:

- 1 kg of fresh soil, previously sieved through 8 mm, was mixed with 1 l of wash solution and stirred for 20 min. A wash time of 20 min was chosen based on previous reports [8]. This mixture was passed through two sieves (3.35 mm and 63 μm aperture) and washed with an additional 500 ml of wash solution.
- The <63 μm fraction was allowed to settle for 5 h (corresponding to time where no significant reduction in sediment height observed). After which, the resulting supernatant was removed. The three soil fractions were oven-dried.
- The three resulting solid fractions; gravel (>3.35 mm), sand (>63 μm) and silt/clay (<63 μm), as well as effluent were analyzed as indicated below (Section 2.6).

2.5. Procedure of flocculation test

For each wash solution, three replicate experiments were conducted following the stages below:

- Soil aliquots of 200 g, previously sieved through 3.35 mm, were mixed with each wash solution generating 500 ml of soil/wash solution mix; this mix was stirred for 20 min.
- Flocculating agent was added and stirred for 5 s.

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