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Use of green washing fluids in a washing process for dioxin contaminated soils

Siwalee Yotapukdee[1)], Chitsan Lin*[1)], Jr-Ping Wang[2)], Wen-Yen Huang[1)], Acharee Kaewlaoyoong[3)], Wen-Ming Mao[1)] and Shun-Hsyung Chang[4)]

[1)]Department of Marine Environmental Engineering, National Kaohsiung Marine University, Nanzih District, Kaohsiung City 81157, Taiwan
[2)]Department of Naval Architecture, National Kaohsiung Marine University, Nanzih District, Kaohsiung City 81157, Taiwan
[3)]Department of Safety, Health and Environmental Engineering, College of Engineering, National Kaohsiung First University of Science and Technology, Kaohsiung City 82445, Taiwan
[4)]Department of Microelectronics Engineering, National Kaohsiung Marine University, Nanzih District, Kaohsiung City 81157, Taiwan

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Abstract

High levels of dioxin contamination in soil have significant environmental challenges. Soil washing is a successful remediation process that is primarily used to treat coarse soils. Several literature studies have used various kinds of chemical washing liquids to remove dioxins from soils, though there are secondary environmental effects. This study intends to develop environmentally friendly soil washing methods that are effective in dioxin removal at an acceptable cost. Sugarcane wine, compost leachate, and ground fish broth were chosen as potential washing liquids. Each washing liquid was analyzed to determine its content of semivolatile organic compounds (SVOCs) and volatile organic compounds (VOCs). These compounds are related to their bio-surfactant content. Several of the identified compounds had properties to help remove dioxins from contaminated soil. In the experiments, high removal efficiencies were observed, up to 70%~95% after five to six washes. Although effective removal was observed, a significant amount of wastewater was produced and the problems were not completely resolved. Thus, the optimal washing conditions are necessary to minimize the overall costs, while improving the process effectiveness. Moreover, an appropriate treatment method is required for wastewater containing dioxins.

Keywords: Hazardous waste treatment, Green remediation, Sustainable remediation, Green and sustainable remediation, Soil washing, Dioxins contaminated soil

1. Introduction

Dioxins are among the most toxic man-made chemicals on earth. Their structures are chlorinated dibenzo-p-dioxin (PCDD) and dibenzofuran (PCDF). Dioxins are inadvertently produced by industrial, municipal and domestic incineration, combustion processes, including the manufacture of chlorophenols and phenoxy herbicides, chlorine bleaching of paper pulp and smelting [1]. The major sources of PCDD/DFs are incineration and agrochemicals [2-4]. However, current exposure levels and dioxin accumulation in the food chain, particularly in animal fat, are a serious concern.

Recently, there has been much concern about dioxins in contaminated soil in southern Taiwan. A contaminated site was declared as a soil contaminated remediation site in 2004 and the remediation plan started in May 2009. The remediation approach uses a thermal desorption treatment to remedy high concentrations of dioxins in contaminated soil. However, this method is a process that uses high temperatures (usually below 400° C) [5] that can impact applicability or costs at specific sites.

As an alternative, soil washing is a process to separate contaminants from soil that use physical and/or chemical techniques. This method is relatively inexpensive and has been reported to function well on contaminated soils [6]. It uses a detergent that desorbs the pollutants from contaminated soils. Different kinds of detergents are applied for soil washing depending on the contaminants being removed. For example, PCDDs and PCDFs are removed by ethanol [6-8]. Natural oils (e.g., olive oil) can be used for oily contaminants [9]. Consequently, the chosen detergents in soil washing process are sugarcane wine, anaerobic compost tea, and ground fish broth. Anaerobically composted tea and sugarcane wine contained bio-surfactants. This is a significant key for toxic chemical removal from soils. Dioxins have extremely low solubility in the water. So, this study used ground fish broth to rinse dioxins from the soil. Moreover, this material is also the environmentally friendly. Therefore, the aim of this study was to apply soil washing for

*Corresponding author. Tel.: +886-9-84268732 [a)], +886-7-3651472 [b)]
Email address: mo_siwalee06@hotmail.com[a)], ctlin@webmail.nkmu.edu.tw[b)]
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dioxin contaminated soils for remediation purposes and develop an effective washing procedure using various natural reagents, i.e., sugarcane wine, anaerobically composted tea, and ground fish broth.

2. Materials and methods

2.1 Soil sample

Approximately 200 kg of dioxin contaminated soil was collected from southern Taiwan. It was sieved in to 7 fractions (No. 10 (2.00 mm), No.18 (1.00 mm), No. 35 (500 µm), No. 60 (250-µm), No. 140 (106 µm) and No. 270 (53 µm) sieve)) using a wet screening method, ASTM E276-13. The soil fraction with the largest proportion had a particle size range 0f 0.25-0.105 mm) and it had a lower concentration of t-dioxins (20,080 ng-TEQ/kg) (TEQ, toxicity equivalency quantity). In the current soil washing experiment, soil of this more abundant particle size was chosen.

2.2 Soil washing experiment

Portions (200 g) of contaminated soils were added to 500 mL of each of the washing liquids (sugarcane wine, anaerobic compost tea, and ground fish broth) in 600-mL glass bottles. The washing liquids were purchased from a local market. The ratio of soil and solution was 1:2.5. Soil washing was done in a 40 kHz ultrasonic bath. The temperature of water in ultrasonic bath was held constant at 30 ^{o}C throughout the washing process. Stainless steel double impellers served as an effective stirrer to admix the materials and increase particle contact while operating at 700 rpm. Each washing time was one hour. After the first washing, washing liquids were poured into a 250 mL glass bottle and about 10 g of washed soil was collected in a 100 mL beaker for analysis. Washes with sugarcane wine and anaerobically composted tea were done six times. Washes using ground fish broth were done five times. A schematic diagram of the soil washing process is depicted in Figure 1.

2.3 Instrument analysis

The Taiwan NIEA M801.13B method [10] was adopted for polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) analyses using gas chromatography/mass spectrometry. Each of the extracted 1 mL samples in small vials was analyzed using an Agilent 6890N Gas Chromatography coupled with an Agilent 5973N Mass Spectrometer (Agilent Technologies, Avondale, PA, USA), operating in the electron impact and scan system modes. Injection port temperature was set to 250°C using helium as a carrier gas at a rate of1 mL min^{-1}. The splitless mode was used for GC. A HP-5 MS with a 30 m × 0.25 mm × 0.25 µm capillary column in the GC was used for this analysis.

3. Results and discussion

The effect of the various washing liquids on the removal of dioxins contaminated soil was examined. Initially, the total concentrations of dioxins in soils washed by with sugarcane wine, anaerobically composted tea and ground fish broth were 8902, 22080 and 22080 ng-TEQ kg^{-1} on a dry weight basis, respectively. In the first wash, anaerobically composted tea rapidly reduced the concentration of dioxins better than other washing liquids. However, the dioxin concentration showed a more steady rate of reduction in the last wash. The dioxin concentration was not rapidly reduced by the first wash using either sugarcane wine or ground fish broth. It decreased linearly thought the entire series of washes. When using the ground fish broth washing, the purification efficiency of the each consecutive washing decreased. Removal efficiencies of 78.7% and 86.6% were obtained for washing with sugarcane wine and anaerobically composted tea, respectively, after the sixth wash (Table 1) and 94% with that of fish broth washing.

The dioxins were most effectively removed by washing with ground fish broth. About 94% of the dioxins were removed from soil samples after five washing cycles. This result is considered very good. Previous studies reported that vegetable oil was very effective in removing PAHs from soils. The vegetable oils used had typical fatty acid compositions that were similar to fish oil. Palmitic, stearic, myristic and oleic acids were present [11-15]. For example, Pannu et al. [12] used peanut oil for extraction of PAHs from soil and achieved an extraction efficiency of 90%. In this study, the residual t-dioxins in the spent sugarcane wine, anaerobically composted tea and ground fish broth were 1,901, 2,961 and 1,298 ng-TEQ kg^{-1}, respectively (Table1), which are still higher than the required value (1,000 ng TEQ kg^{-1}). It is possible that a nondegradable portion of dioxins remained irreversibly bound in soil under these conditions.

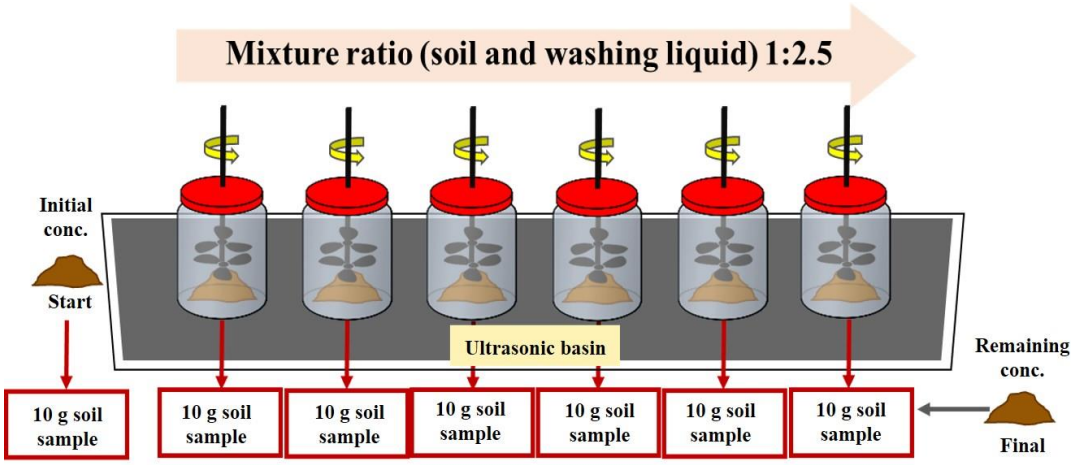

Figure 1 Schematic diagram of the soil washing process

Table 1 The concentration of dioxins in each washing liquid and percentage removal after the fifth wash using ground fish broth and sixth wash using sugarcane wine and anaerobically composted tea.

Washing liquid	Dioxin Conc. (ng-TEQ kg^{-1})	
	Initial	**Remaining**
Sugarcane wine (6 washes)	8,902	1,901
Removal efficiency (%)		78.65
Anaerobic compost tea (6 washes)	22,080	2,961
Removal efficiency (%)		86.59
Ground fish broth (5 washes)	22,080	1,298
Removal efficiency (%)		94.12

4. Conclusions

A set of parameters (washing time of 1 hr, mixing speed of 700 rpm min^{-1}, soil-to-solution ratio of 1:2.5, and a temperature of 30 ◦C) were selected for washing dioxins from contaminated soils containing 8,902 and 22,080 ng-TEQ kg^{-1} dioxins on a dry weight basis. High removal was achieved. Starting with a sample of 0.2 kg of dioxin contaminated soils, more than 90% removal was achieved by washing with ground fish broth. Further studies should consider the feasibility of this on a larger, as well as the secondary effects of dioxins on soil structure. Furthermore, before such procedure can be used to remediate soils, practical methods to remove the contaminants from the liquids used in the process must be developed.

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