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1 **Treatability of volatile chlorinated hydrocarbon-contaminated** 2 **soils of different textures along a vertical profile by mechanical** 3 **soil aeration: A laboratory test**

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A B S T R A C T

Mechanical soil aeration is a simple, effective, and low-cost soil remediation technology that is 21
 suitable for sites contaminated with volatile chlorinated hydrocarbons (VCHs). Convention- 22
 ally, this technique is used to treat the mixed soil of a site without considering the diversity 23
 and treatability of different soils within the site. A laboratory test was conducted to evaluate 24
 the effectiveness of mechanical soil aeration for remediating soils of different textures (silty, 25
 clayey, and sandy soils) along a vertical profile at an abandoned chloro-alkali chemical site 26
 in China. The collected soils were artificially contaminated with chloroform (TCM) and 27
 trichloroethylene (TCE). Mechanical soil aeration was effective for remediating VCHs (removal 28
 efficiency >98%). The volatilization process was described by an exponential kinetic function. 29
 In the early stage of treatment (0–7 hr), rapid contaminant volatilization followed a pseudo- 30
 first order kinetic model. VCH concentrations decreased to low levels and showed a tailing 31
 phenomenon with very slow contaminant release after 8 hr. Compared with silty and sandy 32
 soils, clayey soil has high organic-matter content, a large specific surface area, a high clay 33
 fraction, and a complex pore structure. These characteristics substantially influenced the 34
 removal process, making it less efficient, more time consuming, and consequently more 35
 expensive. Our findings provide a potential basis for optimizing soil remediation strategy in a 36
 cost-effective manner. 37

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53 Introduction

54 Volatile chlorinated hydrocarbons (VCHs), which are potential
 55 mutagens and carcinogens, are among the most commonly
 56 used industrial products. VCHs are introduced into the
 57

environment through their use as chemical intermediates. 58
 Such compounds are frequently found in contaminated soils 59
 in China and throughout the world (Wu et al., 2005). 60

Soil remediation technologies that are applied to sites con- 61
 taminated with VCHs include soil vapor extraction (Albergaria 62

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63 et al., 2012), soil flushing (Kujawski et al., 2007), thermal
64 desorption (He and Sulkes, 2011), bioremediation (Chang et al.,
Q4 Q3 2002; Williamson et al., 2009), microwaving (Acierno et al., 2001,
Q5 Acierno et al., 2004; Jones et al., 2002), and bioventing (Fukue
67 et al., 2006). However, economic constraints may restrict the use
68 of these technologies in developing countries.

69 Mechanical soil aeration was developed as a low-cost and
70 highly effective remediation technology for VCH-contaminated
71 sites and was mentioned in an annual report on remediation
72 technologies published by the United States Environmental
73 Protection Agency (U.S. EPA, 2007). This technology facilitates
74 the release of volatile organic compounds (VOCs) from the soil
75 by plowing and forced cross-ventilation. The released VOCs are
76 then collected and treated. The advantages of mechanical soil
77 aeration include ease of operation, significant remedial effi-
78 ciency, a short remediation cycle, and cost-effectiveness. How-
79 ever, there are some drawbacks associated with this technology
80 (Zhang et al., 2015). First, handling of contaminated soils is
81 associated with an increased risk of human toxic exposure. In
82 addition, the excavation process may pose structural risks to
83 neighboring buildings (Yang et al., 2013).

84 Mechanical soil aeration removes VOCs by volatilization
85 induced by mixing, cutting, and flipping the soils. Our previous
86 studies on soil remediation by mechanical soil aeration in the
87 field and laboratory revealed that agitation frequency, soil
88 temperature, and aeration affect the removal efficiency of VCHs
89 (Shi et al., 2012; Ma et al., 2015a, 2015b). Heterogeneity and
90 non-uniformity of soil media introduce many uncertainties
91 into the remediation process. Remediation efficiency is strongly
92 influenced by differences in soil characteristics.

93 In practice, mechanical soil aeration is commonly applied
94 after excavation of soil for *ex situ* remediation, without con-
95 sidering the effects of different soil layers along a vertical
96 profile. It may be more cost effective to treat each soil fraction
97 (i.e., clay, silt, and sand) separately to achieve optimal
98 remediation efficiency, assuming that separate excavation of
99 the soil texture types is feasible. The present study was
100 undertaken to conduct a laboratory treatability study of three
101 soils along a vertical profile of an abandoned chloro-alkali
102 chemical site. Our approach aimed to investigate the response
103 of different soil textures to mechanical soil aeration and
104 analyze the remediation efficiency of different treatments.
105 The results provide data to optimize decision-making for the
106 remediation of sites contaminated with VCHs.

108 1. Materials and methods

109 1.1. Chemicals and reagents

110 Chloroform (TCM) and trichloroethylene (TCE) were used
111 to simulate contaminated soils and were purchased from
112 Sinopharm Chemical Reagent (Beijing, China), with purities of
113 99%. Methanol (Fisher Scientific, GC Resolv, America) was used
114 to extract the above mentioned contaminants (TCM and TCE).

115 1.2. Preparation of contaminated soils

116 Original soil samples were collected from three soil horizons
117 of a vertical profile in a contaminated site: horizon 2

(2.5–5.5 m deep), horizon 3 (5.5–8.1 m deep), and horizon 4 118
(8.1–10.7 m deep) (Fig. 1). 119

120 Prior to the experiment, large particles and plant residues
121 were removed from the soil. The soil was air-dried for 20 days
122 ground, sifted using a #10 (2 mm) sieve, dried at 105°C for
123 12 hr, and stored for subsequent use (Ma et al., 2015a, 2015b). 124
125 This pretreatment was performed to obtain a fraction of soil
126 with uniform physical properties and without contaminants. 127

128 The artificially contaminated soils were prepared as follows. 129
130 The contaminant solution was prepared by adding 15 mL of
131 each of the two contaminants to 250 mL of methanol. Next,
132 2000 g of sieved soil was spread evenly on a polyethylene plastic
133 sheet; 500 mL of high-purity water was added to maintain soil
134 moisture content of 20% (W/W), and the mixture was stirred and
135 pressed. We immediately added 50 mL of contaminant solution,
136 covered the mixture with 1000 g of sieved soil and 100 mL of
137 high-purity water, and immediately wrapped the mixture with
138 plastic sheeting and sealed it with tape (Ma et al., 2015a, 2015b).
139 The contaminated soils were sealed in airtight boxes and stored
140 at 4°C for 4 days to ensure that the concentration of contami-
141 nants was even and stable (Shi et al., 2012). 142

143 1.3. Experimental procedure

144 We designed an apparatus to simulate mechanical soil
145 aeration. This apparatus allowed contaminants to be removed
146 via volatilization induced by mixing, cutting, and flipping
147 of the soil by rotating coulters. The released VOCs were
148 discharged after treatment with activated carbon. The appa-
149 ratus consisted of a main body, a temperature control system,
150 an automatic control system, and an exhaust gas treatment
151 system (Fig. 2). The following operation parameters were fixed
152 during the experiment: aeration rate (3 L/min), initial soil
153 temperature (20°C), agitation interval (2 hr), mixing speed
154 (200 r/min), and agitation time (10 sec). 155

156 Soil samples were collected every hour for the first 24 hr and
157 every 2 hr between hours 25 and 48. Each group of samples
158 included three replicates, and the final data were the average
159 values of the three replicates. Soil samples collected for VCH
160 measurement were placed in 40-mL closed glass vessels
161 pre-filled with 10 mL methanol. Additional soil samples were
162 collected from the area surrounding the VCH sampling sites and
163 used to analyze water content. All samples were stored in a
164 refrigerator at 4°C before analysis (Ma et al., 2015a, 2015b). 165

166 1.4. Analytical methods

167 The concentrations of VCHs in the soils were measured using
168 a gas chromatograph (7890A GC, Agilent Technologies, USA)
169 equipped with a mass spectrometer (5975C MS, Agilent,
170 America), in accordance with the USEPA-8260C method. The
171 GC was fitted with a DB-624 capillary column (60 m ×
172 250 μm × 1.4 μm; Agilent) and was operated with helium as
173 the carrier gas (flow rate, 1.2 mL/min). The oven temperature
174 was programmed as follows: 40°C for 2 min, increased to
175 200°C at a rate of 20°C/min, increased to 250°C at a rate of
176 10°C/min, and maintained at 250°C for 3 min. The Method
177 Detection Limits of target VOCs detected are presented in
178 Table S1. The relative percent difference between replicate
179 samples was ≤25%. 180

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