



# Quicklime-induced changes of soil properties: Implications for enhanced remediation of volatile chlorinated hydrocarbon contaminated soils via mechanical soil aeration



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## HIGHLIGHTS

- TCE-contaminated soil was treated by mechanical soil aeration with quicklime.
- Changes in soil physico-chemical properties after quicklime addition were investigated.
- Increasing soil temperature facilitated volatilisation of TCE.
- Addition of quicklime increased macropores and reduced mesopores and micropores.
- Effects of soil pH and organic matter on TCE removal were analysed.

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## ABSTRACT

Mechanical soil aeration is used for soil remediation at sites contaminated by volatile organic compounds. However, the effectiveness of the method is limited by low soil temperature, high soil moisture, and high soil viscosity. Combined with mechanical soil aeration, quicklime has a practical application value related to reinforcement remediation and to its action in the remediation of soil contaminated with volatile organic compounds. In this study, the target pollutant was trichloroethylene, which is a volatile chlorinated hydrocarbon pollutant commonly found in contaminated soils. A restoration experiment was carried out, using a set of mechanical soil-aeration simulation tests, by adding quicklime (mass ratios of 3, 10, and 20%) to the contaminated soil. The results clearly indicate that quicklime changed the physical properties of the soil, which affected the environmental behaviour of trichloroethylene in the soil. The addition of CaO increased soil temperature and reduced soil moisture to improve the mass transfer of trichloroethylene. In addition, it improved the macroporous cumulative pore volume and average pore size, which increased soil permeability. As soil pH increased, the clay mineral content in the soils decreased, the cation exchange capacity and the redox potential decreased, and the removal of trichloroethylene from the soil was enhanced to a certain extent. After the addition of quicklime, the functional group –COO of soil organic matter could interact with calcium ions, which increased soil polarity and promoted the removal of trichloroethylene.

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## 1. Introduction

The addition of lime to the soil has long been practiced in agriculture, as such addition increases soil pH (Anderson et al.,

2013) and soil fertility (Jawad et al., 2014). Lime has also been used in water and wastewater treatment to reduce hardness (ASCE/AWWA, 2012) and to remove metals and phosphorus (Morse et al., 1998; Fu and Wang, 2011). The principle for these applications is that at high pH levels and in the presence of Ca<sup>2+</sup>, both of which lime provides, the precipitation of compounds, such as CaCO<sub>3</sub>, Pb(OH)<sub>2</sub>, and Ca<sub>5</sub>(PO<sub>4</sub>)<sub>3</sub>(OH) occurs. In wastewater treatment, lime

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is often added to dewatered sludge to make it safe for land application, as lime stabilises heavy metals and significantly reduces the faecal coliforms and viable helminth ova present in the sludge (Fytli and Zabaniotou, 2008). In addition, lime is the most cost-effective and widely used option for neutralising acids (National Lime Association, 2000), for flue-gas desulphurization (National Lime Association, 2007), and in asphalt construction applications (Little and Epps, 2006). The application of lime has been extended to the removal of contaminants, including persistent organic pollutants (Hall et al., 1996) and hexabromocyclododecane (Zhang et al., 2014b) in ball milling processes.

Furthermore, lime has been used to treat non-hydrocarbon oily wastes (British Lime Association, 2006), remove heavy metals from abandoned mines (Lee et al., 2007), remove the explosives in aqueous solution (Heilmann et al., 1996), and solidify/stabilise heavy metals (Saeed et al., 2012). Moreover, adding CaO has been extended to the remediation of contaminated soil, such as heavy metal-contaminated sites (Gray et al., 2006); 2,4,6-trinitrotoluene (TNT)-contaminated soils (Hansen et al., 2003; Davis et al., 2006; Waisner et al., 2008); polychlorinated biphenyls (PCBs)-contaminated soils (Waisner et al., 2008); explosive cyclotrimethylenetrinitramine and cyclotetramethylenetrinitramine-contaminated soils (Davis et al., 2006); and petroleum hydrocarbon-contaminated soil (Schifano et al., 2007). Furthermore, this technique is used for the hydrodechlorination of dioxin and contaminated soil, with the addition of nano-Ca/CaO (Mitoma et al., 2016), and for the immobilisation of radionuclide  $^{133}\text{Cs}$  in soil with nano-Ca/CaO (Mallampati et al., 2015). It appears that lime is the most cost effective and efficient option for the remediation of heavy metal- and/or organics contaminated soils/sites.

Limited studies have shown significant contaminant removal in volatile organic compounds (VOCs) and petroleum contaminated sites (soils) with the addition of quicklime. For example, quicklime is a useful modifier of polycyclic aromatic hydrocarbon (PAH), PCBs, and TNT in soil treatment. The treatment of highly TNT- and PCBs-contaminated soil in burning ground at the former Ordnance Works by using lime achieved a PCBs concentration of  $6.8 \text{ mg kg}^{-1}$  (70% removal), with the final treatment concentration for TNT being  $67 \text{ mg kg}^{-1}$  (96% removal) (Waisner et al., 2008). The results for using quicklime in petroleum hydrocarbon-contaminated soil in the laboratory indicated that its hydration released heat that promoted volatilisation of light gasoline fractions ( $\text{C}_5\text{--}\text{C}_{12}$ ) (Schifano et al., 2007). The effectiveness of quicklime in remediating contaminated soil appears to reside in its exothermic reaction. The released energy increases the soil temperature, resulting in changes in various soil and contaminant properties.

Hydrated lime treatment has been found effective for the destruction of explosive compounds, which are sensitive to alkaline hydrolysis. Waisner et al. (2008) suggested that alkaline reactions caused by contaminant desorption could lead to the substantial removal of PCB in soil slurry reactors. In addition, the PCB and PAH are merely encapsulated by entrapping the contaminants in a coating of precipitated  $\text{Ca}(\text{OH})_2$  (Waisner et al., 2008; Cassidy, 2010), which is produced from the reaction of quicklime with water. Moreover, soil, quicklime, and water pre-set tense react to form aggregates to which a large number of hydrocarbon molecules adheres, such that the total concentration of hydrocarbon in the soil decreases. These authors attributed the formation of aggregates to the improved conditions after the addition of CaO had increased the soil temperature and, to some extent, decreased the soil moisture.

Success has been achieved in the remediation of VOC-contaminated soil by combining the addition of lime with mechanical soil aeration (Ma et al., 2015). Mechanical soil aeration involves the agitation of contaminated soil through tilling or other

means (e.g. air) to volatilise contaminants at sites contaminated with VOC. The mechanical soil aeration system has long been recognised by United States Environmental Protection Agency as an alternative for removing VOCs from contaminated sites; however, apparently this technique was not being used often at full scale (Federal Remediation Technologies Roundtable, 2006). Nevertheless, the technique has received considerable attention in China because of its cost effectiveness in removing VOCs, particularly in view of the significant soil pollution caused by VOCs in China (Shi et al., 2012). The changed physicochemical properties of the soil measured include only the gross parameters of pH, moisture content, liquid limit, and the plastic limit (Waisner et al., 2008; Cassidy, 2010; Ko et al., 2011). However, the changes in the soil microstructure were not investigated to elucidate their effect on the removal of VOCs.

In view of the above, this study was undertaken to observe the enhanced VOC removal after adding quicklime to several artificially contaminated soils in a reactor specifically constructed for the present study. Physical/chemical changes after the addition of quicklime were quantified to enhance understanding and to determine the removal mechanisms. In particular, a model VOC compound, trichloroethylene (TCE) was selected to evaluate the removal efficiency after the addition of CaO. Essentially, a simple setup was designed to simulate mechanical soil aeration, to which different quantities of quicklime had been added. The strengthening effects of quicklime on the relevant physical and chemical mechanisms were discussed. In addition, the soil temperature, particle size distribution, pore size, pH, and organic matter content were measured to determine the changes in the soil structure after the addition of quicklime. These changes can interact in ways that can enhance VOC removal. This study can certainly fill the knowledge gap in regarding to the use of CaO in removal of contaminant in contaminated soils. However, the dominant means of removal of VOCs could not be determined and requires further study.

## 2. Materials and methods

### 2.1. Selection and pre-treatment of the tested soil

The soil samples were collected from Beijing (two typical soils, namely, silt and silty clay) and Heilongjiang Province (clay with high organic matter content). The collected samples were pre-treated by removing large particles and plant residues, after which the soils were air-dried, ground, and passed through a 40 mesh (0.47 mm) sieve.

### 2.2. Chemicals and reagents

Highly pure TCE (Sinopharm Chemical Reagent, > 99% pure) and quicklime (Sinopharm Chemical Reagent, 98% pure) were used in the study. TCE (Ultra Scientific,  $100 \mu\text{g mL}^{-1}$  in methanol) was used as the standard. Methanol (Fisher Scientific, GC grade) was used to collect and extract the products and the ultrapure water was produced by the Milli-Q system (Millipore, USA).

### 2.3. Experimental apparatus

A small experimental device (Fig. 1) was constructed to monitor the changes in the soil properties after the combination treatment with CaO and mechanical soil aeration. The setup included a CaO injection port, a cylinder of highly pure nitrogen, a gas flowmeter, a reactor consisting of impellers for adequate mixing of the soil with CaO, an exhaust-gas absorption device, and a temperature probe and recorder. The inner diameter of the glass reactor was 60 mm, with the height of 130 mm. In order to ensure system airtightness,

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