



# Influence of soil properties on vapor-phase sorption of trichloroethylene



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## H I G H L I G H T S

- Vapor intrusion is a major exposure pathway for volatile hydrocarbons.
- Certainty in transport processes enhances vapor intrusion model precision.
- Detailed understanding of vadose zone vapor transport processes save resources.
- Vapor sorption near-steady-state conditions at sites may take months or years.
- Type of clay fractions equitably affects sorption of trichloroethylene vapor.

## A R T I C L E I N F O

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## A B S T R A C T

Current practices in health risk assessment from vapor intrusion (VI) using mathematical models are based on assumptions that the subsurface sorption equilibrium is attained. The time required for sorption to reach near-steady-state conditions at sites may take months or years to achieve. This study investigated the vapor phase attenuation of trichloroethylene (TCE) in five soils varying widely in clay and organic matter content using repacked columns. The primary indicators of TCE sorption were vapor retardation rate ( $R_t$ ), the time required for the TCE vapor to pass through the soil column, and specific volume of retention ( $V_R$ ), and total volume of TCE retained in soil. Results show TCE vapor retardation is mainly due to the rapid partitioning of the compound to SOM. However, the specific volume of retention of clayey soils with secondary mineral particles was higher. Linear regression analyses of the SOM and clay fraction with  $V_R$  show that a unit increase in clay fraction results in higher sorption of TCE ( $V_R$ ) than the SOM. However, partitioning of TCE vapor was not consistent with the samples' surface areas but was mainly a function of the type of secondary minerals present in soils.

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**Abbreviations:**  $C_a$ , soil air permeability, cm/s;  $C_w$ , soil saturated hydraulic conductivity, cm/s; CRCCARE, cooperative research centre for contamination assessment & remediation of the environment; ECD, electronic capture detector; GC, gas-chromatograph; IGC, inverse gas chromatography; LOR, limit of reporting;  $R_t$ , retardation rate; SOM, soil organic matter; VI, vapor intrusion; VIM, vapor intrusion model;  $V_R$ , specific volume of retention; VOC, volatile organic compound;  $\mu_w$ , dynamic viscosity of water, centipoise (cP);  $\mu_a$ , dynamic viscosity of air, centipoise (cP);  $\rho_w$ , density of water, kg/m<sup>3</sup>;  $\rho_a$ , density of air, kg/m<sup>3</sup>.

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## 1. Introduction

The distribution of volatile organic compounds (VOCs) between soils and sediments and the soil solution aqueous phase underpins modeling of the fate and transport of such substances in the subsurface environment [1]. Sorption of VOCs onto the soil matrix as they migrate upwards from subsurface contaminated sources results in strong binding to SOM, and therefore retarding the transport of vapor to the ground surface [2–4]. Ignoring attenuation of vapor whether via microbial degradation or sorption processes can lead to incorrect predictions about the vapor transport and potential risk it poses to receptor. This may well lead to the wrong clean-up technology being chosen [4].

Current vapor intrusion models (VIMs) assume that the vapor source has been in place long enough to fully develop near-steady-

state conditions and sorption is assumed to occur instantaneously for modeling purposes. Therefore models consider no attenuation of vapor due to partitioning [5]. However, vapor phase partitioning could be significant at the contaminated subsurface with a finite source. VIMs such as that of Johnson and Ettinger [6] and BioVapor [7] are used by environmental agencies and practitioners to evaluate the health risks arising from vapor intrusion (VI). These models, however, assume steady-state conditions and do not consider vapor phase attenuation [4] and as a consequence prediction of risks when in reality vapor emanating from the subsurface environment may be significantly less than that at the source. Despite these limitations and given the lack of existing models that consider vapor attenuation, Cooperative Research Centre for Contamination Assessment and Remediation of the Environment (CRC CARE) in consultation with State EPAs in Australia and industry sector representatives have recommended the Johnson and Ettinger (J&E) model for the assessment of risks emanating from vapor intrusion [8]. In the absence of realistic information on subsurface fate and behavior of VOC, limited attenuation of vapor was considered a safe option because it minimizes liability resulting from exposure to VOC. As a consequence sites posing limited risks could end up being remediated costing owners of contaminated sites more than otherwise needed to manage such sites.

Sorption isotherms for VOCs on saturated soils or sediments have been demonstrated in a number of studies. However, vapor phase sorption isotherms have not been dealt with in sufficient depth [9] especially in light of subsurface soil heterogeneity. Vapor phase sorption has been studied using a range of soil organic matter (SOM) sourced from activated carbon and humic and fulvic acids by grafting these amendments onto clay minerals [10,11]. Rhue et al. [12] and Pennell et al. [13] have also investigated sorption on pure clay minerals such as smectite and kaolinite. However, in the present study, vapor attenuation was investigated using natural surface and subsurface soils varying in SOM content and mineralogy.

This study addresses the research gap created by the use of grafted organic amendments on surfaces of mineral soil for experimental purposes. The coating of organic substance on mineral soil has its own disadvantage since it is controlled by the chemical nature of the organic materials (presence of functional groups), nature of clay minerals and physical properties such as surface area and mesopores [14]. Thus sorption studies using natural soil have direct application to the generation of generic sorption factors for inclusion in risk assessment tools, as well as decisions regarding remediation action plans for managing hydrocarbon contaminated sites.

## 2. Experimental

### 2.1. Material

Trichloroethylene (TCE) (>99% purity) was purchased from Sigma–Aldrich (Germany) and used as supplied. The laboratory study was conducted using clean sand as a control and six soil types representing topsoil formations in various parts of Australia. Soil samples were selected to cover a range of factors such as soil pH, SOM, surface area and clay mineralogy that contribute to vapor phase sorption processes. Pertinent properties of the soils are summarized in Table 1. Soil physico-chemical tests were conducted as per the standard methods documented by Rayment [15].

The glass columns were made of 5 cm internal diameter and 15 cm length with three pieces, a bottom compartment separated from the middle column of repacked soil with a built-in fine perforated glass disc and capped with a glass lid on the top (Fig. 1). The glass column top and bottom compartment has the side port

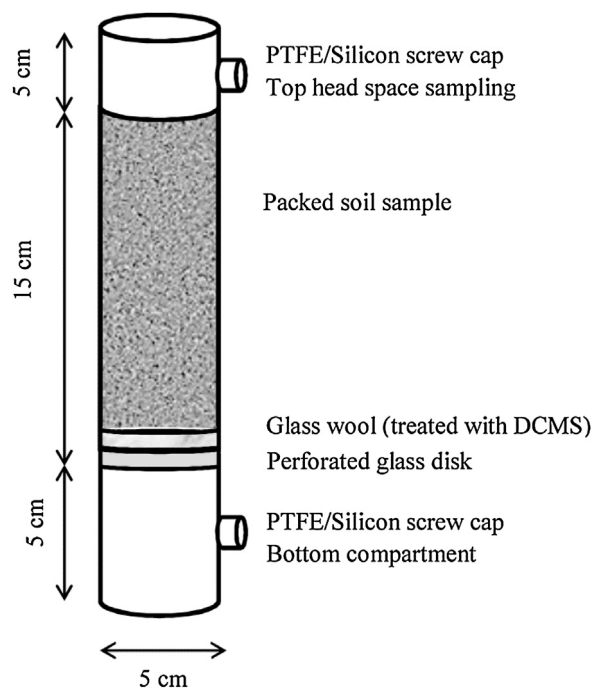


Fig. 1. Packed soil glass column set-up.

with a screw cap sealed with PTFE–silicon septum for injecting TCE solution. Septum with PTFE face is selected to prevent reaction with TCE vapor and silicon body minimizes vapor loss from syringe injections, resists coring and is recommended when multiple injections are required.

### 2.2. Apparatus and procedure

Surface area and total mesopore was determined by  $N_2$  adsorption at  $-196^\circ\text{C}$  using a Micromeritics Gemini-V Surface Area and Pore Size Analyzer, assuming a value of  $0.164\text{ nm}^2$  for the cross-section of the  $N_2$  molecule. The specific surface area was calculated by the BET method [16]. The clay size fractions ( $<2\ \mu\text{m}$ ) were extracted by treating the soils ( $<2\text{ mm}$ ) with sodium hexametaphosphate (1 M). Ultrasound (50 W and 24 kHz for 15 min) treatment was provided to ascertain complete dispersion of the soil particles [17]. The clay size fractions were then pipetted out using Stokes' law. Soil organic matter was removed by sodium hypochlorite treatment (1 M NaOCl and pH adjusted to 8 by adding 1 M HCl) [18]. After drying the samples at  $60^\circ\text{C}$ , soil mineralogy was characterized by X-ray diffraction (XRD) of the powdered samples using  $\text{CuK}\alpha$  radiation ( $\lambda = 1.5418\ \text{\AA}$ ) on a PANalytical, Empyrean X-ray Diffractometer (PANalytical, Australia) operating at 40 kV and 40 mA between  $2.0$  and  $80^\circ$  ( $2\theta$ ) at a step size of  $0.016^\circ$ .

Trichloroethylene concentrations were analyzed using a conventional gas chromatograph (Agilent 7980) equipped with electron capture detector (ECD) and J&W 122–1334, DB-624 GC-column ( $0.25\text{ mm i.d.}$ ,  $30\text{ m length}$ ). The gas chromatograph was operated with initial isothermal oven temperature ( $35^\circ\text{C}$ ) for 5 min and then raising the temperature to  $95^\circ\text{C}$  at  $10^\circ\text{C/min ramp-1}$  followed by an increase to  $200^\circ\text{C}$  at  $50^\circ\text{C/min ramp-2}$ . Injection port and detector temperatures were set at  $250^\circ\text{C}$  and  $300^\circ\text{C}$ , respectively. Helium carrier gas flow  $30\text{ ml/min}$  and Nitrogen makeup gas flow  $10\text{ ml/min}$  were used. Nitrogen and helium gas (extra pure >99.99%) was obtained from BOS Inc., Adelaide, South Australia. A chromatography data-handling system served to collect the data.

Trichloroethylene vapor that passed through the repacked soil was monitored in the top headspace using gas-tight syringes. A

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