Remediation of soils contaminated with trichloroethylene*

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ABSTRACT

Soil contamination is a common problem in our society. Several activities introduce contaminants in soils affecting their quality and future utilization. In order to invert this situation and to avoid further spreading of the contaminant several remediation technologies could be used. For soils contaminated with trichloroethylene (TCE), Soil Vapor Extraction (SVE) can be an efficient remediation technology. It involves the application of vacuum to the soil matrix creating an airflow that conduce the volatized contaminant to extraction wells that lead them to the exterior of the soil where they are properly treated. This work presents the results obtained in the soil vapor experiments performed in a laboratory using soils with different water contents contaminated with TCE. These experiments aimed the study of the influence of soil water content and the airflow used in the remediation time and on the process efficiency. The results showed that in sandy soils contaminated with TCE: a) for similar levels of contamination, the increase of the soil water content from 0 to 4% created an increase of the concentration of TCE in the gas phase of the soil, in some cases by almost 50%; b) the influence of water content is not significant being overlapped by influence of the airflow rate; c) SVE performed with higher airflow rates had shorter remediation times (in some cases four times shorter) without any significant impact on the process efficiencies (fluctuations of 1 to 3%); and d) for the experimented soils, the utilization of higher airflow rates led to shorter and consequently cheaper remediation.

INTRODUCTION

Trichloroethylene (TCE) is a man made chemical that does not exist naturally in the environment. It is commonly used in the electronic industry as a degreasing agent, extractant for some oils, solvent for cellulose esters or dry cleaning fluid. The incorrect handling and disposal of this chemical led to several cases of soil contamination. In the United States from 1982 to 2005, 977 soil remediation projects were documented and the compounds that were most often addressed were the halogenated volatile organic contaminants (that include the TCE) found in 410 remediation projects, followed by the group constituted by benzene, toluene, ethylbenzene and xylene (BTEX) and metals (USEPA 2007).

For the remediation of soils contaminated with halogenated volatile organic contaminants that have high volatility, namely TCE, soil vapor extraction (SVE) can be an appropriate technology. SVE is based on the application of vacuum to the soil matrix through the utilization of pumps. The vacuum creates a movement of air within the soil, dragging the vapors of the contaminants to extraction wells that lead them to the exterior of the soil where air treatment units guarantee legal emission levels (Khan et al. 2004; Suthersan 1996). In the last thirty years, SVE has been the most often used *in-situ* remediation technology in the United States (used in 60% of the all the *in-situ* remediation projects (USEPA 2007)).

The effectiveness of the SVE is influenced by several parameters such as the volatility of the contaminant, contents of organic matter or water, or the airflow rate used in pumping. A recent study performed in soils contaminated with benzene showed that soils with higher organic matter content adsorb higher quantities of contaminant what hinders the remediation turning it more time consuming and less efficient. When lower airflows were used, the process became even longer but with higher efficiencies (Soares et al. 2010). The volatility of the contaminant is closely related with the tendency of the contaminant to preferentially remain in the gas phase in the soil and consequently be more available and be easily extracted (Chai and Miura 2004).

Soil water content is another parameter that strongly influences the SVE process. The water in the soil matrix can be found covering the soil particles, filling the pores of the soil,

adsorbed in the different phases present in the soil, and moving vertically through gravitational forces (Alvim-Ferraz et al. 2006). The increase of the soil water content leads to the filling of the inter-particle pores, decreasing significantly the air permeability of the soil (Poulsen et al. 1999) and consequently hindering the airflow through the soil. This creates unreachable areas that remain untreated and act as a source of contamination to the rest of the soil (Reddy and Adams 2001). Experiments performed in sandy soils contaminated with cyclohexane showed that in soil with higher water contents the SVE showed higher remediation times and slightly lower efficiencies (Alvim-Ferraz et al. 2006). Yoon et al. (2002) studied the impact of the soil water content on the nonaqueous phase liquid (NAPL) volatilization and reported that at 61% water saturation a severe limitation on the mass transfer was observed and a decrease on the effluent gas concentrations causing long tailing and long remediation periods.

The airflow rate has also an important impact on the SVE process. For an efficient remediation process, the air should reach to all the contaminated area and should flow with a rate that is slow enough to be near equilibrium conditions and fast enough to reduce remediation costs. A previous study evaluated the airflow rate influence on soil vapor extractions, performed in sandy soils contaminated with BTEX, TCE and perchloroethylene and concluded that equilibrium between the pollutants and the different phases present in the soil matrix was reached and if slow diffusion effects did not occur, higher airflow rates exhibited the fastest remediations (Albergaria et al. 2008).

This work reports the results obtained in SVE experiments performed in a laboratory installation using different sandy soils with different water contents and contaminated with TCE. These experiments aimed to study the influence of the soil water content and the airflow used on the remediation time, volume of air used in the extraction and on the process efficiency.

MATERIAL AND METHODS

Material

Pro-analysis TCE was obtained from Fluka. Activated carbon 8-20 mesh (0.85-2.36 mm) was obtained from Riedel-de-Haen.

Apparatus and chromatography

The TCE analysis was performed by gas chromatography with a Shimadzu GC-2010 equipped with a flame ionisation detector (FID). The column used was a TRB-35 ($30m \cdot 0.53mm \cdot 0.52\mu m$). The injector and detector were set at 250°C and the column worked isothermically at 200°C. Flame gases were air, at $400ml \cdot min^{-1}$, and hydrogen at $40ml \cdot min^{-1}$. Helium was the carrier gas at $13ml \cdot min^{-1}$. TCE quantification was performed by direct calibration method being checked weekly. Under these conditions the retention time was 0.71min.

Soil preparation and characterization

The sandy soils used in this work were collected at 3m depth, from different places of a beach of Porto region in Portugal. The samples were stored in appropriate vessels in a clean area of the laboratory to prevent further contamination. These soils, mainly constituted by silica, shell debris and decomposed organic remains had a negligible amount of clay and were not contaminated with TCE, as checked by chromatographic analysis of the samples. The soil preparation to SVE experiments involved: a) water washing of the soil till a limpid water was obtained; b) drying, first at 35°C during five days and then at 110°C for 24 hours; c) sieving with a 2mm sieve, to obtain a granulometric fraction of the soils with uniform physical properties; and d) adding deionised water in order to induce the different soil water contents (2, 3 and 4%). These contents represented 40, 60 and 80% of the water saturation of the sandy soil, respectively. The prepared soil was identified as P_0, P_2, P_3 and P_4, the subscript indicating the soil water content of each soil.

International standard methodologies were used for the characterization of the prepared and real soils, including granulometric distribution, apparent density, particle density, porosity, pH, and the contents of sand, clay, natural organic matter and moisture (Albergaria 2003).

Equilibrium isotherms

The objective of the construction of equilibrium isotherms was to easily calculate the amount of TCE remaining in soil after the SVE and consequently to improve the process efficiency. The experiments to obtain these equilibrium isotherms were conducted in stainless steel columns of 37cm height and of 10cm internal diameter (Albergaria 2003) and involved three stages: i) introduction of the soil into the column; ii) induction of contamination; and iii) settling of equilibrium. The introduction of the soil into the column was performed adding fractions of 500g until a total of 4kg of soil. In order to guarantee the same soil porosity in the column, after each introduction, the soil was compacted using always the same procedure. The contamination induction ranged from 12.5 to $100.0mg \cdot kg_{soil}^{-1}$ that corresponded to the range of contaminations obtained in soil after the remediation with SVE. The concentration of TCE in the gas phase of the soil was monitored by gas chromatography in four levels of the column and when they were similar (less than 5% deviation) it was concluded that the equilibrium was reached. Equilibrium isotherms were constructed relating the concentration of TCE in the gas phase of the soil (C_{gas}) with the total amount of TCE induced to the soil matrix (m_{soil}) after the establishment of equilibrium conditions.

Soil vapour extraction

The soil preparation for the SVE experiments was similar to that described in the Equilibrium isotherms paragraph, however, the level of contamination was always $250mg \cdot kg_{soil}^{-1}$.

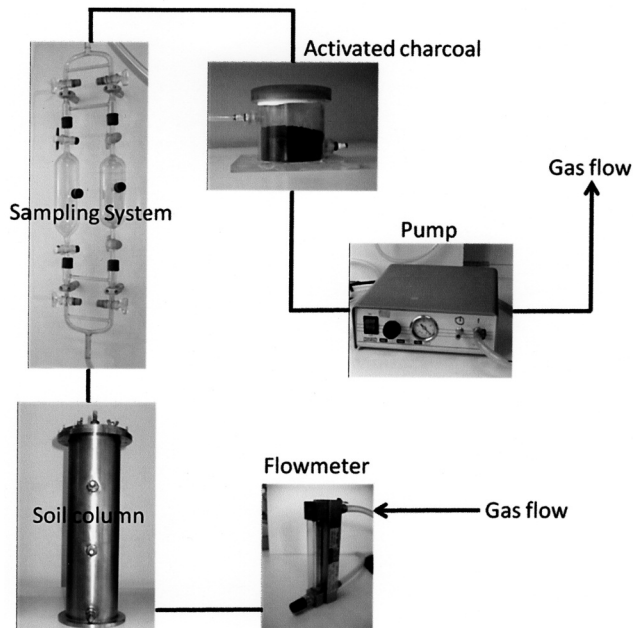The installation used to simulate SVE is shown in Figure 1.

Figure 1. Soil Vapor Extraction (SVE) installation.

After the equilibrium was reached, a vacuum pump was turned on creating in the column an airflow rate that was measured with a flow meter. The flux percolated through the soil column extracting the TCE and transporting it to the sampling system where its concentration was monitored. The monitoring consisted of the collection of a gas sample with a syringe that was analyzed by gas chromatography. To avoid atmospheric contamination and to protect the pump, a small container with activated carbon was placed between the sampling system and the vacuum pump. The remediation process was considered finished when the concentration of TCE in the gas phase was below $1.0 \text{g} \cdot \text{m}^{-3}$. The time needed to reach this clean-up goal was considered the remediation time.

RESULTS AND DISCUSSION

Soil characterization

The characteristics evaluated for the experimented soils are shown in Table 1. The clay content was under the detection limit, and the relatively high pH values were due to the presence of shell debris.

Equilibrium isotherms

Figure 2 shows the equilibrium isotherms that were constructed for each experimented soil (P_0, P_2, P_3 and P_4).
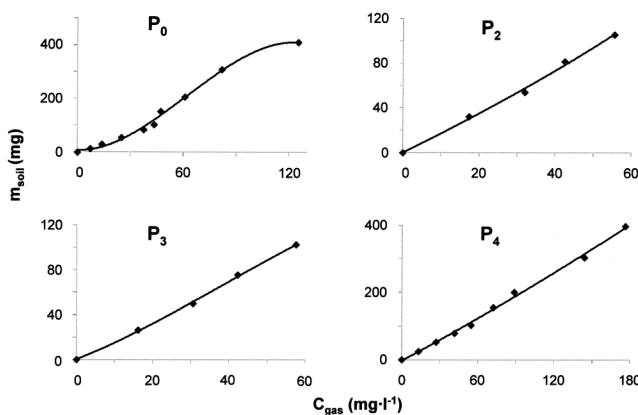

Figure 2. Equilibrium isotherms for each experimented soil (P_0, P_2, P_3 and P_4). C_{gas} – Concentration of trichloroethylene (TCE) in the gas phase of the soil ($\text{mg} \cdot \text{l}^{-1}$); m_{soil} – mass of TCE in the soil matrix (mg).

It was observed that the increase of the soil water content led to linear relations between the concentration of the contaminant in the gas phase of the soil and its amount on the soil matrix. The increase of the soil water content from 0 to 4% showed that for similar levels of contamination the concentration of TCE in the gas phase of the soil also increased, in some cases for almost 50%. This can be explained by the fact that higher amounts of water cover the soil particles decreasing its adsorption capacity and that the water fills the soil pores reducing the gas phase of the soil.

Table 1. Characteristics of the experimented soils (in each case particle size was <2mm, particle density 2.5g·ml⁻¹, pH 8.8, and natural organic matter content was <0.02%).

Soil	Apparent density (g·ml⁻¹)	Porosity (%)	Water content (%)
P_0	1.5	42	0.0
P_2	1.3	49	2.0
P_3	1.3	50	3.0
P_4	1.2	51	4.0

Soil vapor extraction experiments

For every SVE performed the remediation time was registered and the process efficiency and the volume of air used were calculated. The calculation of the volume of air needed (V) for each SVE used the following equation:

$$V = \int Q \, dt$$

where t represents the time (h), and Q the airflow rate ($l \cdot h^{-1}$) measured in standard temperature and pressure conditions (STP). The results obtained in the SVE experiments are presented in Figure 3.
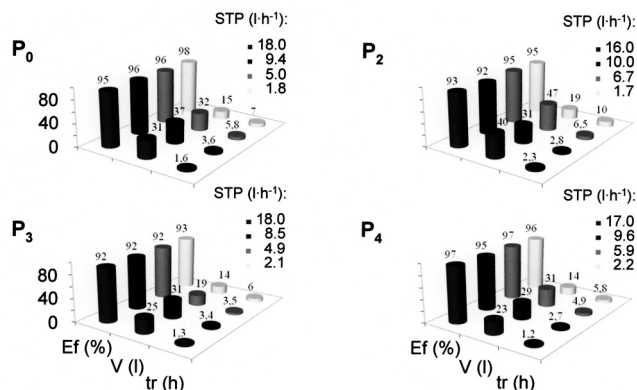
Figure 3. Results obtained in SVE experiments. Ef – Efficiency (%), V – Volume of air used (l), t_r – remediation time (h).

Figure 3 allows the evaluation of the influence of the soil water content and the airflow rate on the remediation time, volume of air used and SVE efficiency. The efficiency of the SVE ranged slightly from 92 to 98%. For sandy soils contaminated with TCE, the influence of water content was not significant. It was observed that, in some cases, an increase of the soil water content led to longer remediation time and higher volume of air used, but in other cases the opposite was observed. These observations could be explained by the fact that the airflow rate was influencing the SVE in the opposite direction. The increase of the soil water content hinders the remediation leading to longer treatments, however the airflow rate had higher impact on the SVE process inverting the water effect. As an example, we can use the SVEs performed in the four soils using the higher airflow rate. The experiments performed with P_0 and P_2 were conducted under similar airflow rates and the remediation time increased due the soil water content influence, however, the next experiments (P_3 and P_4) had higher airflow rates and consequently showed shorter remediation time inverting the influence of the soil water content.

As stated above, the airflow rates had an important impact on the SVE process. For the experimented soils contaminated with TCE, SVE performed with higher airflow rates had lower remediation times (in some cases four times less) without any significant impact on the process efficiencies (fluctuations of 1 to 3%).

Figure 4 shows how the process efficiency evolved with the volume of air that passed through the soil column during the remediation process.
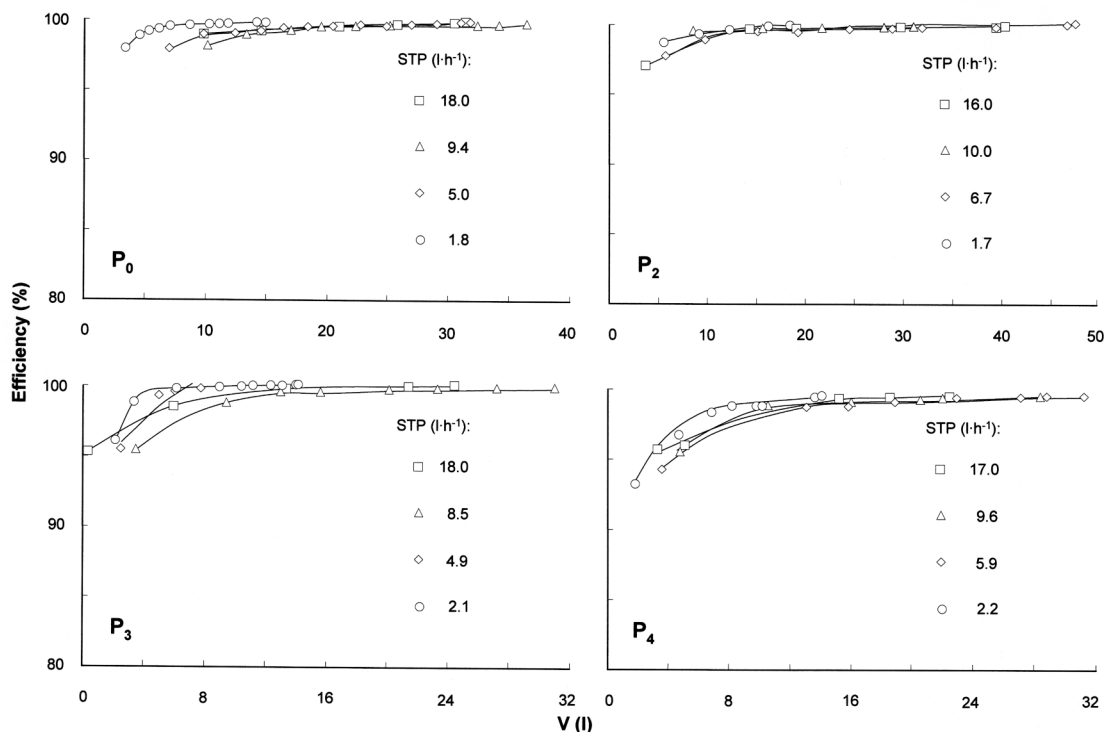
Figure 4. Influence of the airflow rate on SVE process for each experimented soil.

This study allows the identification of the most favorable operational conditions concerning the airflow rate. It can be concluded that, for the experimented soils, the air needed to reach the maximum SVE efficiency was not considerably different, what indicates that the utilization of higher airflow rates led to shorter and consequently cheaper remediation. This can be explained by the extremely low natural organic content of the soil that indicates that the TCE has not strong bonding to the soil what makes it more mobile and available to the extraction.

CONCLUSIONS

The results obtained in sandy soils contaminated with TCE allowed concluding that:
- For similar levels of contamination, the increase of the soil water content from 0 to 4% created an increase of the concentration of trichloroethylene in the gas phase of the soil, in some cases by almost 50%.
- The influence of water content was not significant being overlapped by influence of the airflow rate.
- SVE performed with higher airflow rates had lower remediation times (in some cases four times less) without any significant impact on the process efficiency (fluctuations of 1 to 3%).
- For the experimented soils, the utilization of higher airflow rates led to shorter and consequently cheaper remediation.

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