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Technical Note

Soil vapor extraction in sandy soils: Influence of airflow rate

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ABSTRACT

Airflow rate is one of the most important parameters for the soil vapor extraction of contaminated sites, due to its direct influence on the mass transfer occurring during the remediation process. This work reports the study of airflow rate influence on soil vapor extractions, performed in sandy soils contaminated with benzene, toluene, ethylbenzene, xylene, trichloroethylene and perchloroethylene. The objectives were: (i) to analyze the influence of airflow rate on the process; (ii) to develop a methodology to predict the remediation time and the remediation efficiency; and (iii) to select the most efficient airflow rate. For dry sandy soils with negligible contents of clay and natural organic matter, containing the contaminants previously cited, it was concluded that: (i) if 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, (ii) it was possible to predict the remediation time and the efficiency of remediation with errors below 14%; and (iii) the most efficient remediation were reached with airflow rates below 1.2 $cm^3\ s^{-1}$ standard temperature and pressure conditions.

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1. Introduction

The release of contaminants into the soil matrix produces a negative impact in soil quality, creating, in some cases, situations of risk to public health. For soil remediation different techniques are being used but soil vapor extraction (SVE) has been the most used to soils contaminated with volatile or semi-volatile organic compounds in the unsaturated zone (USEPA, 2007). The air circulation through the soil matrix produces mass transfer into the gas phase, due to free pollutant volatilization, desorption from the soil and dissolution from the aqueous phase. Before reaching the atmosphere, these vapors are usually treated to guarantee legal limits and air quality (Suthersan, 1996; Khan et al., 2004).

The success of a SVE project depends on several parameters including contaminant characteristics such as vapor pressure and solubility, soil properties such as natural porosity, permeability, organic matter or water contents and operational conditions like temperature or airflow rate. Contaminants with higher vapor pressures and high soil temperatures are favorable conditions to SVE utilization. These conditions enhance the volatilization of the contaminant becoming more available and easy to extract (Chai and Miura, 2004). High porosity and high permeability also benefit SVE, mainly, due to their positive impact on the airflow rate through the soil (Poulsen et al., 1999). On the opposite situation contaminants with high solubility and soils with high contents of water or natural organic matter create more difficult conditions for SVE (Yoon et al., 2002; Alvim-Ferraz et al., 2006), due to dissolution in the soil water or to the adsorption on the organic matter, which decreases the mass transfer between the gas phase and the soil matrix.

The induced gas flow towards the extraction wells causes the evaporation of the nonaqueous liquid phase, the volatilization of contaminants dissolved in the pore water and the desorption of chemicals from the porous material, increasing the mobility and availability of the contaminant, consequently increasing the remediation efficiency (Kaleris and Croisé, 1997). The air circulation into the soil matrix is not homogeneous, being influenced by soil heterogeneities. Due to this, the air flows through paths of lower resistance, avoiding regions of lower permeability. These regions will not be crossed by the airflow and consequently will be remediated only through diffusion. Furthermore, these untreated regions can act as sources of contamination to surrounded remediated soils and groundwater (Reddy and Adams, 2001) leading to higher remediation times, lower efficiencies and more expensive processes. The remediation time of soils using SVE is inversely proportional to the air flow rate, but to take full advantages of that relation, equilibrium between the pollutants and the different phases present in soil matrix is necessary, and slow diffusion effects should not occur. Even modest non-equilibrium and slightly slow diffusion effects can strongly affect the efficiency and cost of the remediation process (Alvim-Ferraz et al., 2006). Experiments

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previously carried out with two soils contaminated with cyclohexane demonstrated that for air flow rates smaller than 1.5 $cm^3 s^{-1}$ at standard temperature and pressure (STP), the remediation efficiency did not depend on the volume of gas passed through the soil, because the equilibrium between the pollutant and the different phases present in the soil was reached and the diffusion effects were avoided (Alvim-Ferraz et al., 2006). However these conclusions were only applied to cyclohexane, under specific soil conditions.

In this work the study was widened to six contaminants, typical of contaminations with petroleum products, allowing the establishment of operational SVE conditions for the remediation of contaminated soils. The airflow rate influence on SVE was evaluated for sandy soils contaminated with benzene, toluene, ethylbenzene, xylene, trichloroethylene and perchloroethylene. The objectives were: (i) to analyze the influence of airflow rate on the process; (ii) to develop a methodology to predict the remediation time and the remediation efficiency; and (iii) to select the most efficient airflow rate.

2. Experimental

2.1. Materials

Pro-analysis benzene and perchloroethylene were obtained from Riedl-de-Haën; toluene, xylene and ethylbenzene were obtained from Merck; trichloroethylene was obtained from Fluka.

2.2. Apparatus and chromatography

The quantification of the contaminants was performed by gas chromatography with an Ai Cambridge GC95 equipped with a flame ionisation detector. The packed column used was a Chrompack Hayesep Q 80–100 mesh (3m × 6.35 mm × 4 mm). The injector and the detector were set at 230 °C and the oven worked isothermically at 200 °C. Flame gases were air, at 270 $cm^3 min^{-1}$, and hydrogen at 30 $cm^3 min^{-1}$. The carrier gas was nitrogen at 30 $cm^3 min^{-1}$. Chromatographic data were recorded using Barspec Data System software (Barspec System, Inc., Israel). The direct calibration method was used. Under the above described experimental conditions, the contaminants showed retention times of 25, 51, 99, 101, 25 and 51 min, respectively, for benzene, toluene, ethylbenzene, xylene, trichloroethylene and perchloroethylene.

2.3. Soil preparation and characterization

The sandy soil was collected at 3 m deep in different places of a beach of Oporto region in Portugal, being stored in appropriate vessels. This sandy soil was mainly constituted by silica and shell debris, with negligible amounts of clay and natural organic matter. The preparation of soils involved four steps: (a) washing till clean water was obtained, (b) drying, first at room temperature for 5 d and then at 110 °C for 24 h, (c) sieving in a 2 mm sieve to obtain a granulometric fraction of the soil with uniform physical and chemical properties; and (d) adding of water in order to induce 2% of water content that corresponded to the average water content of the studied soils in unsaturated conditions. The real sandy soils studied in this work were collected in a different beach of the same region at different depths. These sand samples were just sieved in the 2 mm sieve, stored in stopped vessels and identified as $R_{0.5}$, $R_{0.6}$, $R_{0.8}$, $R_{1.0}$, $R_{1.2}$ and $R_{2.4}$, the subscript indicating the water content (%).

International standard methodologies were used for the characterization of the prepared and real soils, including the determination of apparent density (ASTM D4531-86), particle density (DIN 18124), pH (US-EPA 9045 D), and the contents of water (ASTM D 2216) and natural organic matter (Wakley–Black method) (Albergaria, 2003). Porosity was calculated through soil apparent and particle densities.

2.4. Equilibrium isotherms

The equilibrium isotherms, related the contaminant concentration in the gas phase with the concentration in the soil matrix (solid, aqueous and non aqueous liquid phases), were used to quantify the total amount of contaminant in the soil matrix, through the experimental determination of contaminant concentration in the gas phase.

To obtain these equilibrium isotherms for the prepared and the real soils, experiments were performed in a stainless steel column with 37 cm height and 10 cm of internal diameter. These experiments involved the following stages: (i) introduction of the soil in the column, (ii) soil contamination, (iii) equilibrium settling; and (iv) determination of the concentration of the contaminant in the gas phase through gas chromatographic analysis. In the first stage, 4.00 kg of soil were introduced in the column in fractions of 500 g. After each introduction, the soil was compacted using always the same procedure in order to guarantee similar soil porosity. The second stage consisted of the addiction of different amounts of chilled/frozen contaminant (from 0.025 to 0.400 g) on the top of the column allowing the percolation and distribution of the contaminant into the soil matrix. The soil was then left isothermically at 296 K. To evaluate if the equilibrium was reached, the concentration of the contaminant in the gas phase (C_{gas}) was monitored in different levels of the column along time. When the concentration was similar at different levels (less than 5% deviation), equilibrium was considered to have been reached; all contaminants reached within 24 h.

Through Eq. (1) the amount of contaminant in this phase (m_{gas}) was calculated, where V_{gas} is the volume of the gas phase.

$$m_{gas} = C_{gas} V_{gas} \tag{1}$$

With Eqs. (2) and (3), the concentration (C_{aq}) and the mass (m_{aq}) in the aqueous phase was determined, where V_{aq} is the volume of water present in soil and H the constant of Henry's Law.

$$C_{aq} = \frac{C_{gas}}{H} \tag{2}$$

$$m_{aq} = C_{aq} V_{aq} \tag{3}$$

To calculate the amount in the solid and non aqueous liquid phases (m_{s+napl}) Eq. (4) was used, where m_{tot} is the total amount of contaminant.

$$m_{s+napl} = m_{tot} - m_{gas} - m_{aq} \tag{4}$$

The concentration of contaminant in the soil matrix (C_{mat}) (namely in aqueous, solid and non aqueous liquid phases) was calculated using Eq. (5), where m_s is the total amount of soil.

$$C_{mat} = \frac{(m_{aq} - m_{s+napl})}{m_s} \tag{5}$$

The equilibrium isotherms were elaborated relating the concentration of the contaminant in the gas phase (C_{gas}) with its concentration in the soil matrix (C_{mat}) (Albergaria, 2003).

2.5. Soil vapor experiments

To perform the SVE experiments with the prepared and real soils, columns were prepared in a similar way to the equilibrium isotherms: introduction of the soil, induction of contamination (in this case, always 1.00 g) and equilibrium establishment. After

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