Resistive heating enhanced soil vapor extraction of chlorinated solvents from trichloroethylene contaminated silty, low permeable soil

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ABSTRACT
A 2D-laboratory box experiment (12 x 56 x 116 cm) was conducted to simulate the enhancement of soil vapor extraction by the application of low frequency electrical heating (joule heating) for the remediation of a low permeable, silty soil contaminated with trichloroethylene. Joule heating enlarged the flux of extracted TCE with a factor 19 (when boiling) in comparison to the removal rates achieved by soil vapor extraction alone (at 23°C). Within 45 days a removal of 99.8% of the TCE was accomplished.

DEUTSCHE ZUSAMMENFASSUNG
Durchgeführt wurde ein 2D-Laborversuch in einer Kammer (12 x 56 x 116 cm) zur Simulation einer erhöhten Boden-Wasserdampfextraktion. Zweck war die Sanierung schluffigen, mit Trichlorethylen kontaminierten Bodens geringer Durchlässigkeit durch die Anwendung einer Wechselstrom-Niederfrequenz-Erhitzung (Stromerhitzung). Die Stromerhitzung erhöhte den Fluß extrahierten TCEs im Vergleich zu den über reine Wasserdampfextraktion (bei 23 °C) erreichten Extraktionsraten um das 19fache (am Siedepunkt). In 45 Tagen wurden 99,8 % des TCE extrahiert.

RESUME EN FRANÇAIS:
Une expérience de laboratoire en 2D sur un modèle réduit de taille 12x56x116 cm a été mené pour simuler l'amélioration de l'extraction de vapeur du sol par l'application d'un chauffage électrique (résistive) à courant alternative à basse fréquence (chauffage par effet Joule) pour remédier à la faible perméabilité d'un sol silteux contaminé par TCE. L'effet Joule augmente le flux de TCE extrait d'un facteur 19 (en cas d'ébollution) par comparaison les taux d'extraction obtenu par l'extraction de vapeur du sol tout court (à 23°C). En 45 jours 99.8% du TCE a été extrait.

Introduction

Trichloroethylene (TCE) is a halogenated aliphatic compound which has been widely used, as a degreasing agent in the metal industry, on army bases and within the textile industry, for more than 50 years all over the world (Pankow et al., 1996). At present trichloroethylene is one of the most frequently detected halogenated organic compounds in aquifers (Pankow et al.1996). Trichloroethylene is a DNAPL with a density of 1.46 kg/L and a viscosity lower than water (Russell et al. 1992, CRC 1994). Downward migration of spilled TCE is impeded by layers and lenses with higher capillary resistance (in general lower permeability). On top of these layers and lenses TCE pools and spreads out laterally (Pankow et al. 1996, Schwille 1988), leading to deep and complex contaminant spreading throughout the aquifer. At the bottom of the pool TCE slowly enters the low permeable sediment by migration through cracks and by diffusion. The long term presence of these pools on top of low permeable layers and lenses leads to migration of significant amounts of TCE into the low permeable layers.

Based on the potential risks to human health, the drinking water standards for TCE are low (e.g. 5 µg/L US EPA, 1995, 1 µg/L Waterleidingbesluit, 1984). Due to its chemical and physical properties the presence of TCE in the subsurface forms a severe groundwater contamination and a serious threat to the environment, human health and drinking water reserves. Therefore, (rapid) remediation of TCE contaminated sites is required.

A large number of sites contaminated with TCE are located in urban areas or at industrial locations that are still in use. This restricts the accessibility of the sites for remediation of trichloroethylene severely. Consequently, in-situ treatment is most frequently applied. Common applied in-situ remedial technologies, like pump-and-treat and soil vapor extraction (SVE), remove the contaminant from the subsurface with an extraction fluid (liquid or gas) in which TCE is dissolved/partitioned. Subsurface permeability differences lead to preferential flow paths: more permeable zones are flushed significantly more than low permeable zones. Although the permeable zones are swept often and hence remediated relatively fast if no NAPL phase is present, long term remedial actions are common practice. This is caused by the slow release of TCE from the low permeable sediment into the mobile phases. Under natural conditions the contaminant flux generated by diffusion is low, therefore enhancement technologies are needed.

Enhancement of TCE removal by heating of soils

According to Udell (1996), Heron (1997), Davis (1997) and Heron et al. (1998b) the physical mechanisms governing the mobilization of TCE from low permeable soils (solubilization, diffusion, volatilization and sorption) are favorably influenced by increase of temperature. Increase of the soil temperature will lead to a higher flux of TCE to more permeable sediments.

Several methods for the heating of soils are described by Smith & Hinchee (1993), Heron (1997) and Davis (1997). Low frequency electrical (resistive) AC heating (joule heating) is one of the promising technologies for enhanced removal of volatile compounds from clay layers and lenses (Bergsman et al. 1993, Gauglitz et al. 1994, Buettner & Daily 1995, and Phelan et al., 1997). One of the main

advantages of joule heating is its preference for heating high electrically conductive sediments (low permeable clays) over sediments with low electrical conductivity (sands). Joule heating has relatively low heat losses to the more permeable layers, in contrast to steam injection, hot water injection and hot air injection, which primarily heat the clay layers by thermal conduction from the heated permeable zones.

According to Imhoff et al. (1997), Sleep & Ma (1997) and Davis (1997), thermal enhancement of VOC removal is much more effective when used in combination with gaseous phase extraction (SVE) than water extraction (e.g. pump and treat). Therefore the application of joule heating for TCE removal is preferentially conducted in combination with soil vapor extraction. Joule heating in combination with soil vapor extraction was applied in the field (Gauglitz et al., 1994), who showed a significant increase in removal of PCE and TCE from a 3 m thick clay layer and removed more than 99% based on soil cores.

To evaluate the relation between temperature and removal efficiency a laboratory experiment was conducted. This paper describes a two dimensional box-experiment simulating joule heating enhanced remediation of low permeable TCE contaminated soil.

Material and Methods

Box construction

Remediation of a soil section was simulated in a two dimensional test box (56 x 12 x 116 cm inner dimensions). Figure 1 shows the outline of the box. The soil section consisted of a 50 cm thick low permeable layer of silty fine sediment with a thin (10 cm thick) coarse sandy top layer in which the soil vapor extraction system was installed. Next to each electrode a 5 cm thick column of sand was situated to avoid drying of the soil next to the electrodes which would increase the box's resistance extremely. The soil was dry packed in thin layers which were intensively homogenized. After packing the soil, installing all equipment and sealing the box gas tight, CO_2 gas was flushed through the soil for several days to avoid entrapment of air when saturating the soil with spiked (almost TCE-saturated) water through the center inlet in the bottom plate. The average TCE concentration in the inlet water was 1120 mg/L. To avoid entrapment of gas during water addition, the driving force for saturation were the capillary forces; only a slight additional hydraulic head was applied. The total amount of TCE added was 35.5 grams.

The materials used to construct the box were selected on their physical and chemical properties. All materials needed to be resistant to aggressive conditions occurring: high TCE concentrations and temperature variations between 10 and 100°C. Furthermore many components needed to have a low electrical conductivity and low sorption of TCE. The dimensions of the box were determined by the needs to prevent interference of the sides with the electrical conduction and vapor transport, the minimal distance of the electrodes and the height required for simulating soil vapor extraction. The dimensions of the fume hood, in which the box needed to be placed, formed a constraint.

3/4 inch, 18mm, thick Teflon plates were used for the bottom and top. The end plates, 1 cm thick stainless steel, were used as electrodes. For the sides two types of material were used: glass (1 cm) and Teflon (also 1 cm tick). The glass plates were used initially to visualize the wetting of the soil during spiking. After the formation of cracks in the plates due to shear stress when tightening, the box was rebuild with the Teflon sheets.

A support structure was installed to fix the plates and the Viton sealing strips in place. To avoid heat loss, six inches thick of insulation was installed, surrounding the box and its support construction.
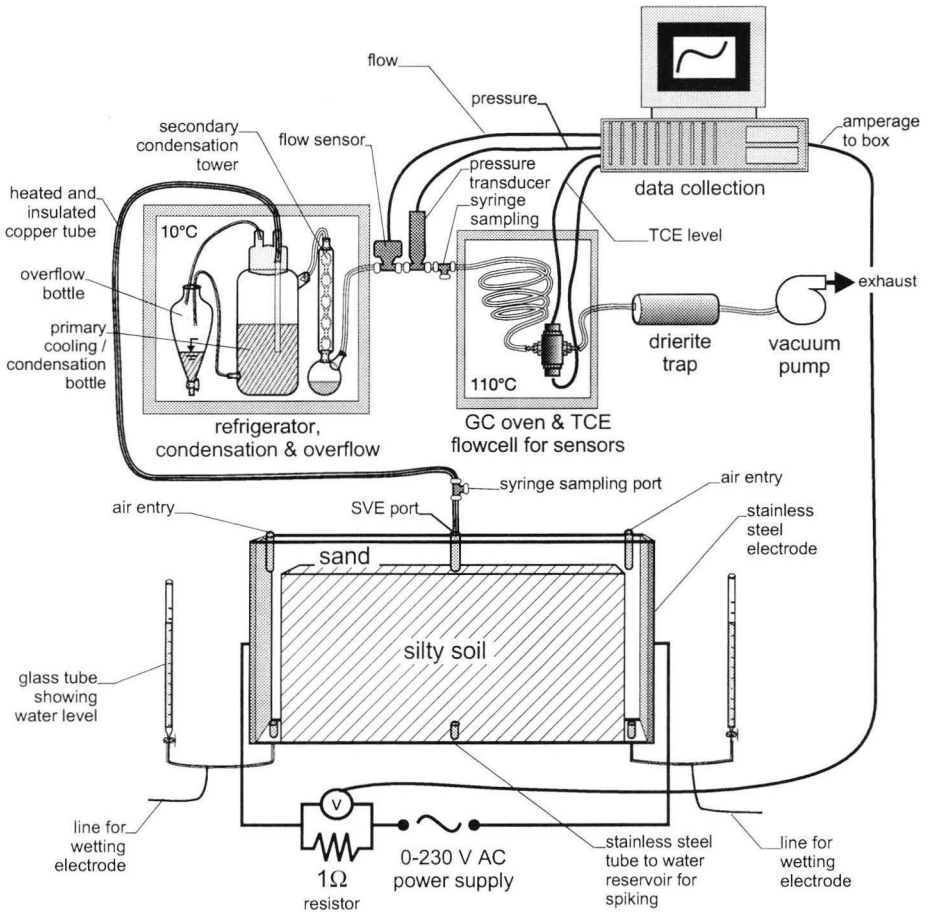

Figure 1 The full setup of the experimental box and vapor treatment

Measurements

Forty-eight thermocouples, with their sensitive tip in the middle of the box, were fixed in the back plate for temperature measurements and electrical conductivity measurements. In the front plate 40 sampling ports were installed for the collection of soil water and/or soil vapor samples for TCE measurements and soil moisture.

Other equipment was installed to control the applied power and the temperature of the electrodes. For a full, more detailed description we refer to Heron et al., 1998a.

The soil vapor extraction system, installed in the top layer, had inlets on each side and a central extraction port. The vapor was extracted with a vacuum pump. The vapor inlets were open to the ambient air. The direction of the vapor flow in the unsaturated zone (the sandy top layer) was from the inlets at the sides towards the central extraction port. The extracted vapor was led through a heated line to a treatment and detection system, consisting of a condensation step, a heating coil and a gas sensing unit with two tin dioxide gas sensors. Before leaving into the exhaust of the fumehood the vapor passed a Drierite trap for final determination of moisture content. The vapor treatment system is described in more detail in Heron et al., (1998c).

Using a LabView controlled data collection system highly frequent (intervals between 1 and 600 seconds) measurements were taken of the extracted vapor (TCE, temperature, pressure and flow rate), the applied power and temperatures in various points.

Frequent (with minimal 4 measuring rounds per day) gas samples of the extracted vapor were collected and directly analyzed on a GC with FID detector, and temperatures were read of a few thermocouples placed in the top and bottom parts of the box.

Less frequently all thermocouples were monitored, electrical resistivities determined and soil vapor and soil liquid samples were collected through all sampling ports and analyzed for TCE.

The vapor flow rate through the sand layer on both sides were determined with a methane tracer test. After completion of the experiment and slow cooling to room temperature, soil samples were taken and analyzed for moisture content and TCE concentration.

Remedial aspects of the experiment

The saturated box was, fully sealed, and left for equilibration for a period of 8 days. Before the start of the soil vapor extraction, a complete sampling of soil fluids for TCE, measurement of the electrical resistivity and temperature was performed. During the experiment five different remedial periods were applied. (1) The first 8 days of the experiment only soil vapor extraction was applied. The temperature in the box was around 23°C. A slight increase of temperature was detected around the outlet, caused by the thermal conduction from the heated vapor extraction line. (2) At day 8, the electrical heating was switched on heating the soil to an average temperature of 85°C reached at day 21. (3) This temperature was kept close to constant, by reducing the applied power, until day 35. (4) Within 4 days the temperature was raised to 99 to 100°C and (5) was kept constant for 5 days after which (at day 45) the power was switched off permanently and the box was cooled down slowly with the insulation still in place.

The application of joule heating is dependent on the soil moisture content and soil conductivity. A low moisture content causes high soil resistance and hence inefficient soil heating, and a high risk of overheating the electrodes. Because

vaporization of water lowered the moisture content, TCE-free water (with the same conductive properties as the water used for spiking) was added during the experiment. A constant hydraulic head was applied. The water was let into the box through two inlet ports at both sides of the bottom in the coarse sand columns.

Results and discussion

The fluxes of extracted TCE in vapor showed significant differences between the five applied soil vapor extraction periods. Figure 2 shows the mass of TCE extracted by the soil vapor extraction system and the soil temperature in the bottom center. The mass of extracted TCE is based on the interpolation of the GC analyses in combination with the data from the gas sensors, the vapor pressure and vapor flow measurements.

The results show that after the extraction of the initial TCE from the vadose zone, a constant TCE-flux of about 0.13 mg/day was extracted by the vapor extraction system at an average soil temperature of 23°C. The predicted duration of a full remediation of the soil based on this flux (not including a decrease of the flux over time) is minimal 270 days. In comparison, when the heating was switched on, and the box heated up with 4.8°C per day, the TCE-flux increased dramatically. Likewise, when the applied power was decreased to maintain a stable soil temperature the TCE-fluxes decreased.

During the two weeks when the temperatures were around 85°C the TCE-fluxes reached an average of 0.35 grams TCE per day (i.e. 3 times the extraction rates at 23°C). At day 35, just before the applied power was increased again, the total extracted mass of TCE was about 20 grams: a little over 40% of the initial added TCE still remained in the soil.

The rising temperature, during the 34th to 39th day, coincided with higher TCE-fluxes detected in the extracted vapors. After reaching boiling temperatures (99 to 100°C) in the box, the TCE-fluxes in the effluent fluctuated significantly over time, at an average rate of 2.5 g/d (i.e.19 times the TCE flux at 23°C).

Subsequently by reducing the temperature below boiling point by switching off the power, the detected TCE-fluxes diminished rapidly below detection limits within days.
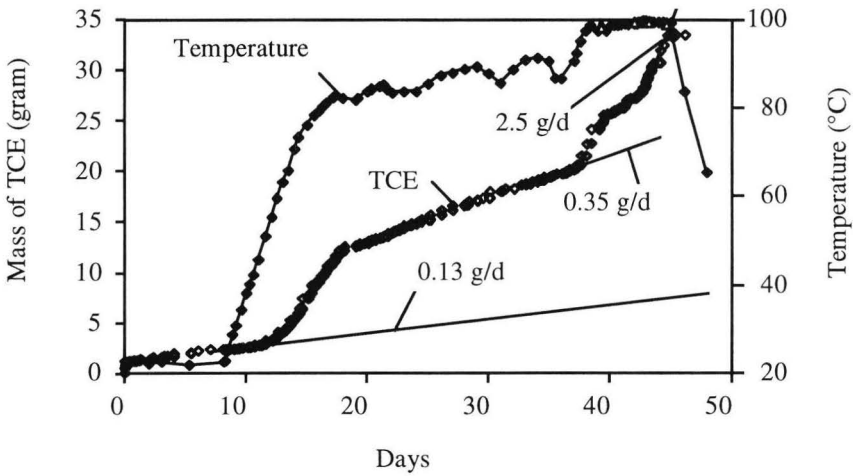
Figure 2 The mass of extracte TCE and the temperature in the bottom center

Analyses of the soil moisture samples and soil samples, collected after a two weeks period of cooling, indicated that even in the bottom center, TCE concentrations in soil and pore water decreased by 2 orders of magnitude, leading to concentrations below the soil criterion (5 mg/kg in Denmark). A total TCE mass of 70 mg remained in the soil after treatment. A removal of 99.8% of the total added TCE within 45 days was accomplished.

Based on the results over 94% of the total removed TCE mass was extracted by the SVE system. These results show the high effectiveness of joule heating, as a supplement to SVE in the permeable zones, to enhance significantlystrongly the cleansing of low permeable, clay/silt, layers which most remedial method fail to address.

Furthermore, the results demonstrate significantly higher removal rates for TCE when temperatures reached boiling point compared to temperature slightly below boiling. This suggests that steam formation and/or the volume expansion in every pore leads to very rapid removal of high concentrations of TCE in tight, almost saturated silt. Therefore, it appears that steam formation is essential for fast removal of volatile contaminants from low permeable soils.

The extraction of steam does however lead to drying of the soil. Since, Joule heating depends on the presence of groundwater for electrical conductance, additional water is required, especially around the electrodes, to maintain conductance.

With regard to the cost of thermal techniques, field applications of thermal techniques have been shown to be economically attractive due to the short treatment times needed for complete contaminant removal (Newmark & Aines, 1997, Bremser & Booth, 1996). In the 2D experiment the relevant aspects for field applications were revealed and related well to initial field tests (Bergsman et al.

1993; Gauglitz et al. 1994, Buettner & Daily, 1995). So was shown the importance of electrode wetting and moisture control to maintain high conductivity and vapor production. Furthermore, good control of the power input in relation to desired in-situ temperatures is essential. The high removal rates achieved in almost saturated soil indicates that large desaturation is not needed for effective remediation.

Although no DNAPLs were present in the remediation experiment and could be in field cases, DNAPL-pools on top of a low permeable layers does not seem to hold back effective soil remediation with the applied technology. On-going research strongly indicates that thermal techniques are even more efficient when NAPL phases are present, since NAPL areas boil preferentially at lower temperatures than 100 °C (Udell, 1996, 1997, Heron et al. 1998d).

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