

**2-D PHYSICAL MODELS OF THERMAL CONDUCTION HEATING
FOR REMEDIATION OF DNAPL SOURCE ZONES IN AQUITARDS**

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ABSTRACT

In-situ thermal remediation (ISTR) technologies are receiving increasing attention for remediation of dense non-aqueous phase liquid (DNAPL) source zones in soil and groundwater. A clear understanding of the primary mechanisms of ISTR is crucial to enable selection of appropriate sites and effective ISTR technologies for DNAPL source zone remediation. Thermal conductive heating (TCH) coupled with vapor extraction is an ISTR method that takes advantage of the invariance of thermal conductivity across a wide range of soil types to effect treatment of DNAPL in lower-permeability and heterogeneous formations. Under the sponsorship of the U.S. Strategic Environmental Research and Development Program (SERDP), bench- and larger-scale remediation experiments are being conducted at the facilities of VEGAS - the Research Facility for Subsurface Remediation at Universität Stuttgart, Germany to better understand the principal mechanisms that control TCH performance in the saturated zone. In parallel, a numerical model was optimized based on the experiments. This paper reports the results of various 2-D experiments that we completed, while the companion paper (Hiester et al. 2008) focuses on the related 3-D experiments.

A series of small-scale two-dimensional (2-D) experiments were carried out at VEGAS. The aim of these experiments was to investigate the principal processes of TCH in layered, saturated materials. The experiments were conducted in a stainless steel two-dimensional flume with the dimensions of 1100 mm [width] x 740 mm [height] x 85 mm [thickness]. The front panel consisted of a steel plate for homogeneous heating experiments, and Pyrex[®] glass for the remediation experiment. The flume was equipped with 100 Pt-100 discrete temperature sensors (10 rows x 10 columns) to measure the temperature distributions while heating to steam distillation temperatures, 100 °C. Additionally, an infrared camera was used periodically to obtain continuous temperature distributions during the remediation experiments, in which a known amount of tetrachloroethene (PCE) was released prior to heating. During heating, the mass of contaminant collected in the extracted gas was monitored. Following heating, soil samples were collected to estimate the mass of contaminant remaining in the soil.

The main TCH contaminant removal mechanism within the saturated porous media was vaporization. Nearly complete DNAPL removal from the source zone was accomplished by this method. No DNAPL

mobilization was observed through pool mobilization outside of the target treatment zone during or following TCH. These 2-D experiments set the stage for the more complex 3-D experiments presented in the companion paper (Hiester et al. 2008).

INTRODUCTION

Dense Non-Aqueous Phase Liquid (DNAPL) source zones have been among the most complex and difficult types of contaminated sites to remediate. Conventional clean-up methods like 'pump and treat' often fail due to low mass removal rates. Soil vapor extraction (SVE), multiphase extraction, in-situ air sparging, chemical oxidation, surfactant flushing and steam-air injection are all challenged by heterogeneity, primarily characterized by large variations in hydraulic conductivity and/or layers of low permeability within the source volume to be remediated. Because contaminants tend to reside in both high and low permeability layers, preferential flow and bypassing of fluid-based technologies around lower permeability zones can leave such residual contamination largely untreated.

In-Situ Thermal Remediation (ISTR) technologies are beginning to be widely applied to overcome these limitations, with results that are gaining attention. ISTR technologies include Steam-Enhanced Remediation (SER), Electrical Resistance Heating (ERH), and Thermal Conduction Heating (TCH). While SER has its greatest applicability to higher permeability zones beneath the water table through which injected steam can flow, ERH and TCH are most often applied within low and moderate permeability zones and in heterogeneous settings. ERH depends on the electrical conductivity of soil, which ranges over a factor of several hundred depending on soil type, from clay to sand, with higher electrical conductivity associated with zones that retain water. Furthermore ERH, like SER, is limited to achieving the boiling point of water. TCH, by contrast, is governed by the thermal conductivity of soil, which is one of the most invariant of all soil physical properties, varying only by a factor of about three over a wide range of soil types from clay to gravel. Capillary forces in a heat pipe system may act as an additional heat transfer mechanism (Udell and Fitch 1985). In addition, TCH can heat the soil to temperatures below, at, or if needed well above the boiling point of water. Thus TCH has applications in heterogeneous subsurface settings with layers of low permeability, and for contaminants with moderate to high boiling points.

Thermal wells used in TCH contain electrical heating elements operating at 400-700°C and can heat both high and low permeability media through a combination of thermal conduction and convection. The wells are typically operated in multi-well patterns that include both heater-only wells to inject heat and SVE wells to collect vapor for aboveground condensation and/or treatment. This combination of thermal wells and SVE is also known as In-Situ Thermal Desorption or ISTD (Baker 2006; LaChance et al. 2006).

Research Motivation and Scope. Recognizing the need to gain a better understanding of the underlying mechanisms of TCH/ISTD for treatment of DNAPL in the saturated zone, TerraTherm forged a research collaboration with VEGAS (a research facility for subsurface remediation located at the Universität Stuttgart, Germany) and MK Tech Solutions (numerical simulations). Our research project, funded by the U.S. Strategic Environmental Research and Development Program (SERDP), was intended to rigorously investigate and help optimize these processes and accompanying contaminant mobilization and removal.

Initially, bench-scale two-dimensional (2-D) remediation experiments were used to study the principal mechanisms that control the performance of TCH/ISTD below the water table. These experiments served as a benchmark for the numerical model, allowing testing of its reliability for representing such processes. The experimental data thus enabled calibration of the numerical model to improve the accuracy of subsequent simulations. The calibrated numerical model was then used to help design the more complicated experiments, including 3-D experiments conducted in two large-scale containers (75 and 150 m³), which are the subject of a companion paper (Hiester et al. 2008).

Objectives. The objectives of the overall project are to:

- 1) determine the relative significance of the various contaminant removal mechanisms below the water table (stream stripping, volatilization, in situ destruction, enhanced solubilization)
- 2) assess the percentage of the DNAPL source removal and accompanying change in water saturation at various treatment temperatures/durations through boiling; and
- 3) evaluate the potential for DNAPL mobilization, either through volatilization and recondensation, and/or pool mobilization outside of the target treatment zone during heating.

2-D EXPERIMENTS: HOMOGENEOUS HEATING

A series of two-dimensional (2-D) experiments were carried out at VEGAS. The experiments were conducted in a stainless steel 2-D flume with the dimensions of 1100 x 740 x 85 mm. The front panel

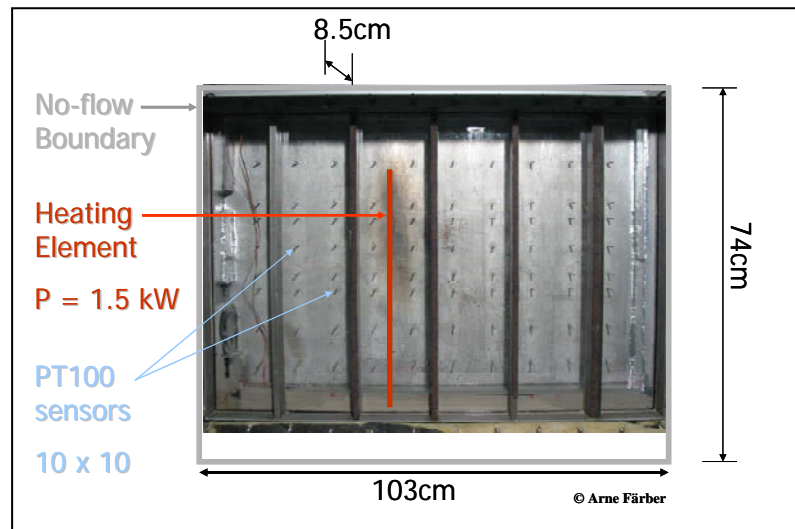
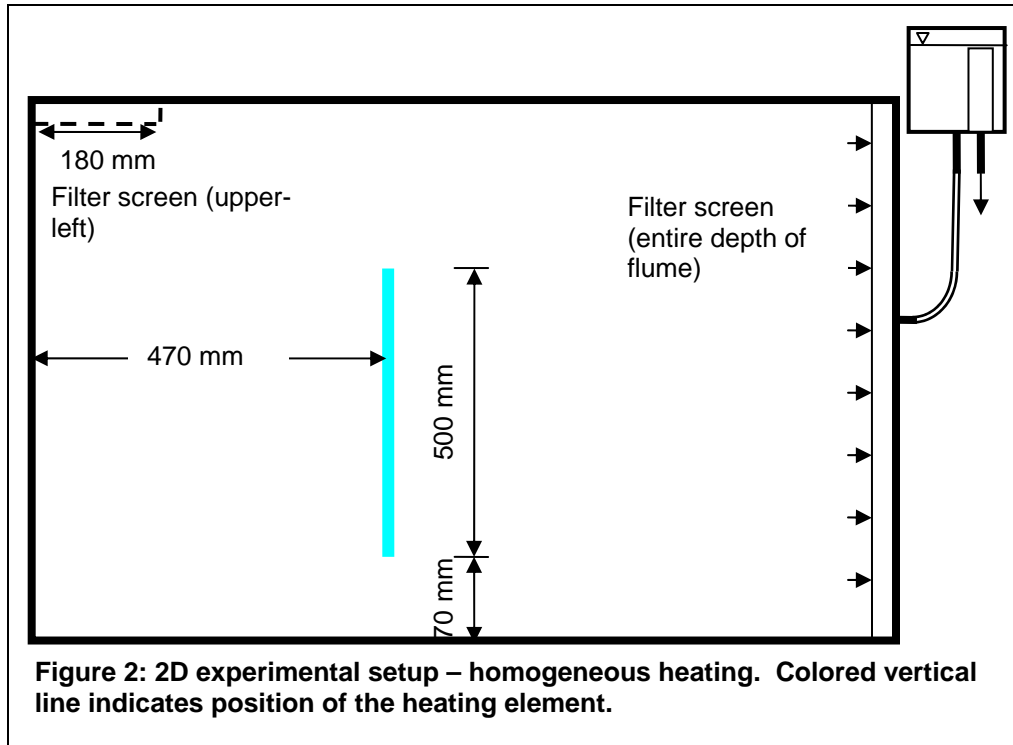


FIGURE 1. 2-D flume heat transport experiments.

consisted of a steel plate for homogeneous heating experiments (Figure 1), while for the subsequent remediation experiments the steel plate was replaced with Pyrex® glass. The flume was equipped with 100 Pt100 temperature sensors (10 rows x 10 columns, Fig. 1) to allow measurement of the temperature distributions with an accuracy of $\pm 1^\circ\text{K}$. Insulation surrounded the container, except during brief periods when it was peeled back to enable visual spectrum photographs to be taken. Additionally, an infrared camera was used to obtain temperature distributions during the remediation experiments. One heating element of 0.5 m length was located approx. in the center of the flume. Two homogenous heating experiments with different levels of power input were carried out to investigate the heat front propagation in the low permeable material (Li, 2004). The energy input to the heater was controlled and kept constant.

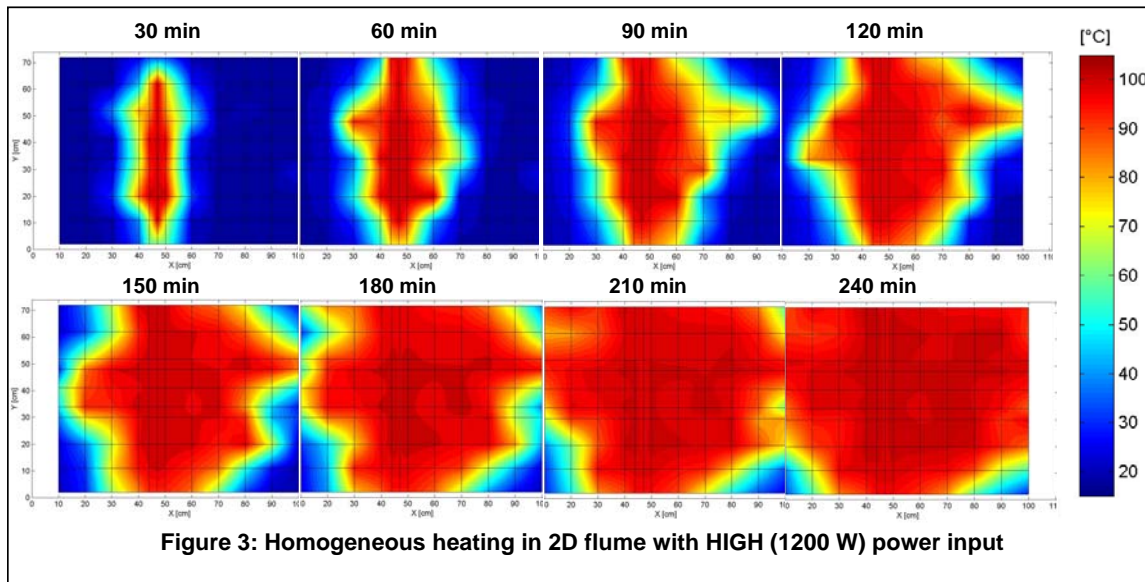
We designated the soil material used in these experiments “FSG” (German for “fine grained mixture”). It consisted of mixtures of GEBA – a well-sorted fine sand, and DORSILIT 2500 – a very fine sandy very coarse silt (Mark 2005). These components were mixed dry and then slightly moistened with 10% (weight) water. The FSG mixture was filled into the flume layer by layer (around 3 cm each) and compressed with the help of a wooden stick, which was the reason why a steel plate was used for the front panel instead of the Pyrex® glass. Its initial porosity after filling was 0.39. The saturated hydraulic conductivity, K_s at the in-place density of the two homogenous heating experiments was 4.6×10^{-3} m/s and 3.0×10^{-4} m/s, respectively. Since it was quite difficult to work in such a thin flume with many sensors distributed all around, it can be expected that the intended “homogenous” filling may not really have been that homogeneous.

The experimental setup is shown again in Figure 2. The vertical heating element was installed a bit to the left of the centerline of the flume. The right side of the flume was fully screened to provide a constant head boundary condition. The upper-left corner was also screened to allow water passage, preventing it from being trapped there during the experiments. The entire flume was



fully saturated before each of the experiments. During the experiments, the entire flume was insulated with glass wool and foam blocks, which were only removed temporarily to take infrared photographs.

Homogeneous heating with HIGH power input. For this experiment, the flume was heated at the full power of the heating element (1200 W). The temperatures were measured by Pt100 sensors with the help of an automated PC data collection system.



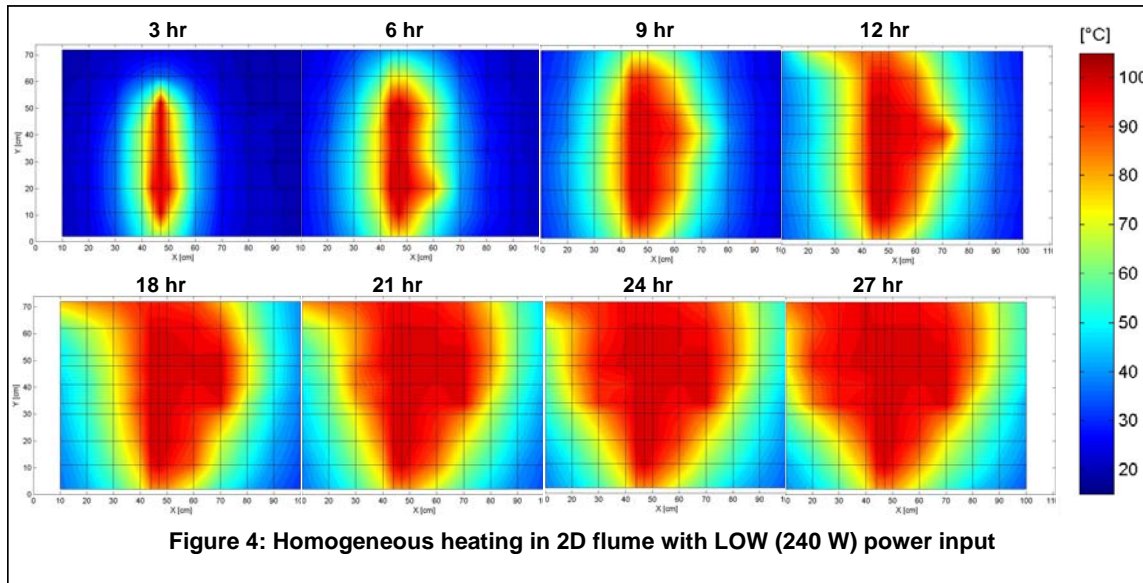
The results are presented in Figure 3. Early in the heating period, a steam zone formed around the heating element. As the experiment proceeded, the steam zone grew laterally until the breakthrough at the right side that is evident at 120 min. The difficulty of achieving uniform packing of soil in the experimental unit is evident. Nevertheless, viscous forces were dominant throughout the experiment.

The influence of buoyant forces appeared only at a very late stage. This experiment was repeated once more, and the results were quite similar.

An attempt to measure saturation values with a gamma source failed due to settlement of the fine material.

Homogeneous heating with LOW power input. For this experiment, the flume was heated at 20% of the full power of the heater (240 W). As with the previous experiment, temperatures were measured by Pt100 sensors with the help of a PC.

The results are presented in Figure 4. Again a steam zone formed around the heating element early during the heating. However, buoyant forces became dominant half-way through the experiment. Movement of water out the overflow at the upper left corner of the flume caused the temperature to be



higher there after about 12 hr. A V-shaped steam zone formed by the end of the experiment, a typical sign of buoyancy dominance. An attempt to use even lower power input (120 W) failed due to heat losses, which could not be ameliorated by adding more insulation.

Simulated and observed data for the two 2-D heating experiments matched well (Baker et al. 2006).

2-D REMEDIATION EXPERIMENTS

Next, two remediation experiments were carried out within the 2-D flume apparatus for the purpose of quantifying and visualizing the removal of PCE during TCH. The experiments were also designed to enable visualization of the potential for PCE migration during heating. The experimental setup for the first of the two experiments is shown in Figure 5.

The upper 50 cm consisted of a fine-textured layer termed "Mixture B" ($K_s = 1.0 \times 10^{-6}$ m/s) (Mark 2005), which is composed of GEBA, DORSILIT 2500 – a very fine sandy very coarse silt, and DORSILIT 4900 – a very coarse silt, at a ratio of 30:50:20, respectively. The lower 24 cm consisted of a coarse sand ($K_s = 1.0 \times 10^{-3}$ m/s). A 10 cm high x 20 cm wide coarse-textured sand lens was installed within the middle of the upper, finer-textured layer to serve as zone into which DNAPL could be released in a controlled fashion. A thin transition zone consisting of GEBA, a well-sorted fine sand was placed around the sand lens to prevent eluviation of the fine-textured material into the sand lens while the water table was raised. Both the upper and lower layers were saturated with water, after which dyed PCE was infiltrated into the

coarse sand lens in the middle. Two heating elements were installed to either side of the coarse lens and parallel to each other, each with a power input of 500 Watts. A blower was used to extract the vaporized PCE collected at the top of the flume, for capture by an activated carbon filter.

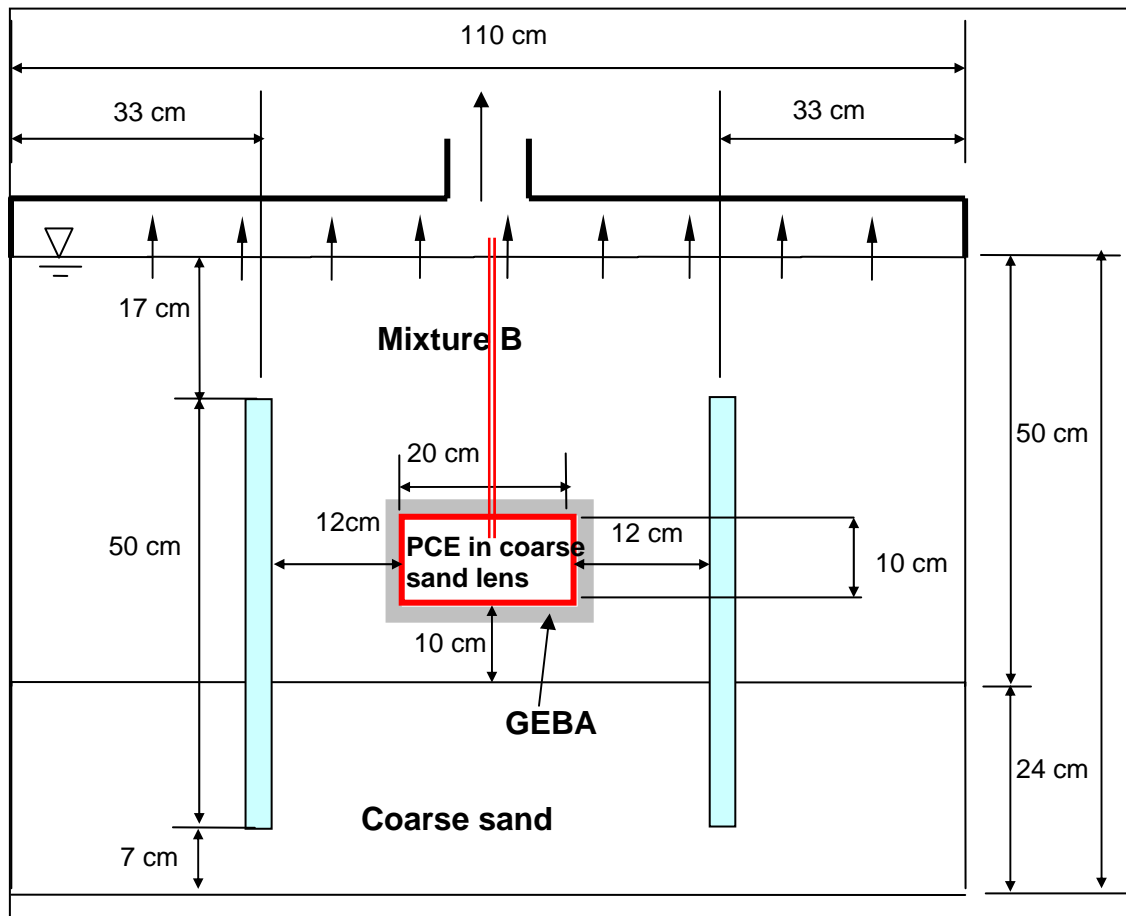


Figure 5: Experiment setup for first 2D remediation experiment. The light blue vertical bars are twin 500 W heaters. PCE was infiltrated into the coarse sand lens at the center.

The PCE was dyed with methyl red to improve visualization. A total of 320g of PCE was allowed to infiltrate into the coarse sand DNAPL storage lens through a pre-embedded tube. After infiltration, the tube was sealed to prevent short-circuiting of vaporized PCE. The surrounding soil served as a capillary barrier, temporarily trapping the PCE within the coarse lens. As is evident in Figure 6, a very small amount of the emplaced PCE found its way into the GEBA layer due to the presence of air at the inner surface of the glass.

The results of the first remediation experiment are summarized in Figure 7. The infrared (IR) photos were taken by briefly removing the insulation and taking an IR photo through the Pyrex® glass front panel, then replacing the insulation.

At the initial stage of the experiment, the steam zone propagated largely within the underlying high permeability coarse sand layer. When steam had occupied most of the high permeability layer, it started to expand into the overlying lower permeability zone. At about 60 min, the steam zone entered the coarse sand lens and started to vaporize the PCE. At 120 min, all the PCE had vaporized, but still no PCE was captured from the activated carbon filter. At this stage, all vaporized PCE was encapsulated within the steam zone, with the gas phase having no outlet to the outside. At about 180 min the steam

zone first broke through the top of the upper layer, and PCE started to be recovered. This can be seen from the PCE recovery curve in Figure 8. The heating was stopped after 6 hours, but vapor extraction remained on until 14 hours. Overall, 92% of the PCE was recovered by vapor extraction.

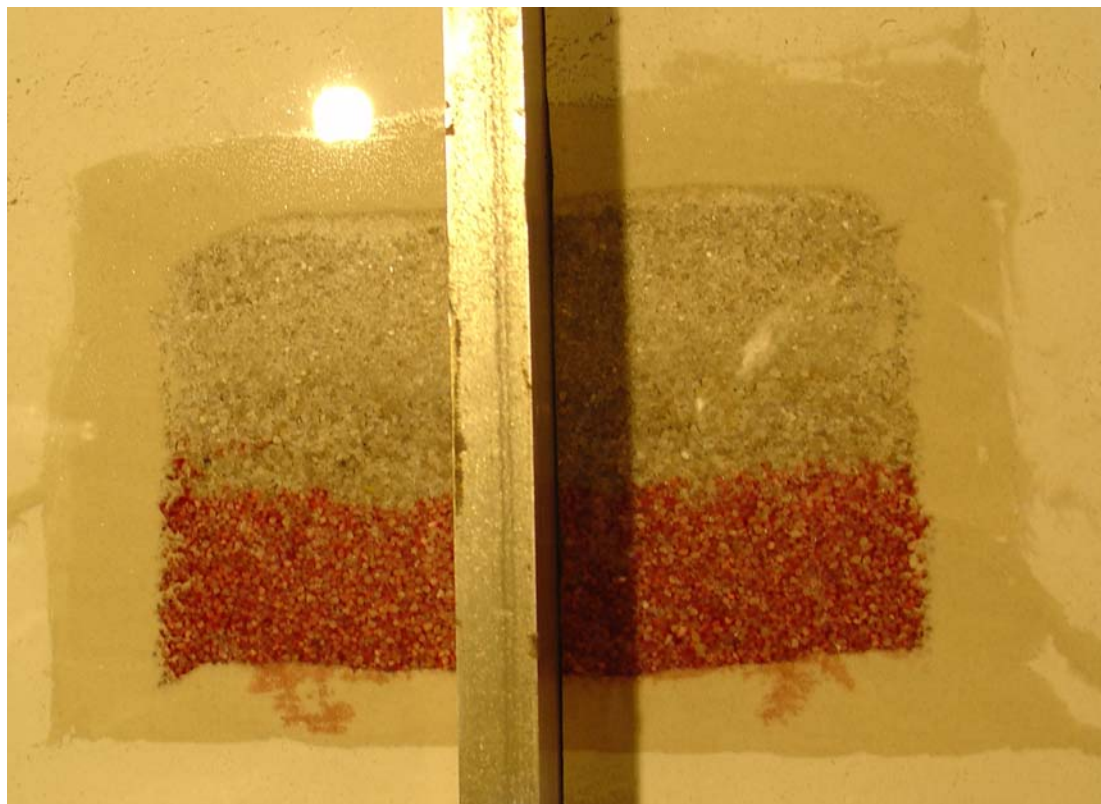


Figure 6: Closeup of PCE placed in coarse sand lens

Due to the low capillary pressure in the high permeability layer, the heat pipe effect (Udell 1985; Hiester et al. 2003) was destroyed after 60 min of heating. “Dry” zones were formed near the bottom of the heating elements, where all residual water was vaporized and the heat transfer was solely due to thermal conduction. The temperature gradient became very high, with the temperature at the heating elements rising to approximately 500°C.

After the experiment, the acetonitrile eluents from soil samples were analyzed with HPLC. The PCE distribution is depicted in Figure 9. The total PCE remaining in the soil amounted to less than 1 g. The 8% PCE unaccounted (around 25g) might have been desorbed from (i.e., broken through) the activated carbon filter.

The second 2-D remediation experiment was similar to the preceding, except that this time, the coarse sand layer ($K_s = 1.0 \times 10^{-3}$ m/s) was above the finer-textured layer ($K_s = 1.0 \times 10^{-6}$ m/s), rather than below it. The coarse-textured DNAPL storage lens was again situated within the finer-textured layer. A single 500 W electric heating element (simulating an angled thermal well) was oriented diagonally within the finer-textured layer, passing below but just beneath the DNAPL storage lens. After saturating the two layers with water, and flushing the storage lens with CO₂, 200 g of dyed PCE was infiltrated into the DNAPL storage lens.

Again heating progressed through boiling. Temperature sensors inside the DNAPL storage lens indicated that boiling of the DNAPL and water-filled sand occurred at ~90°C, close to the 88°C eutectic point of an azeotropic PCE-water mixture. By about 240 minutes, DNAPL had been boiled off, evident by the fact that the temperature then increased there to 100°C for the remainder of the experiment. Of the 200 g of PCE that was infiltrated, a total of less than 1 g of PCE remained within the soil.

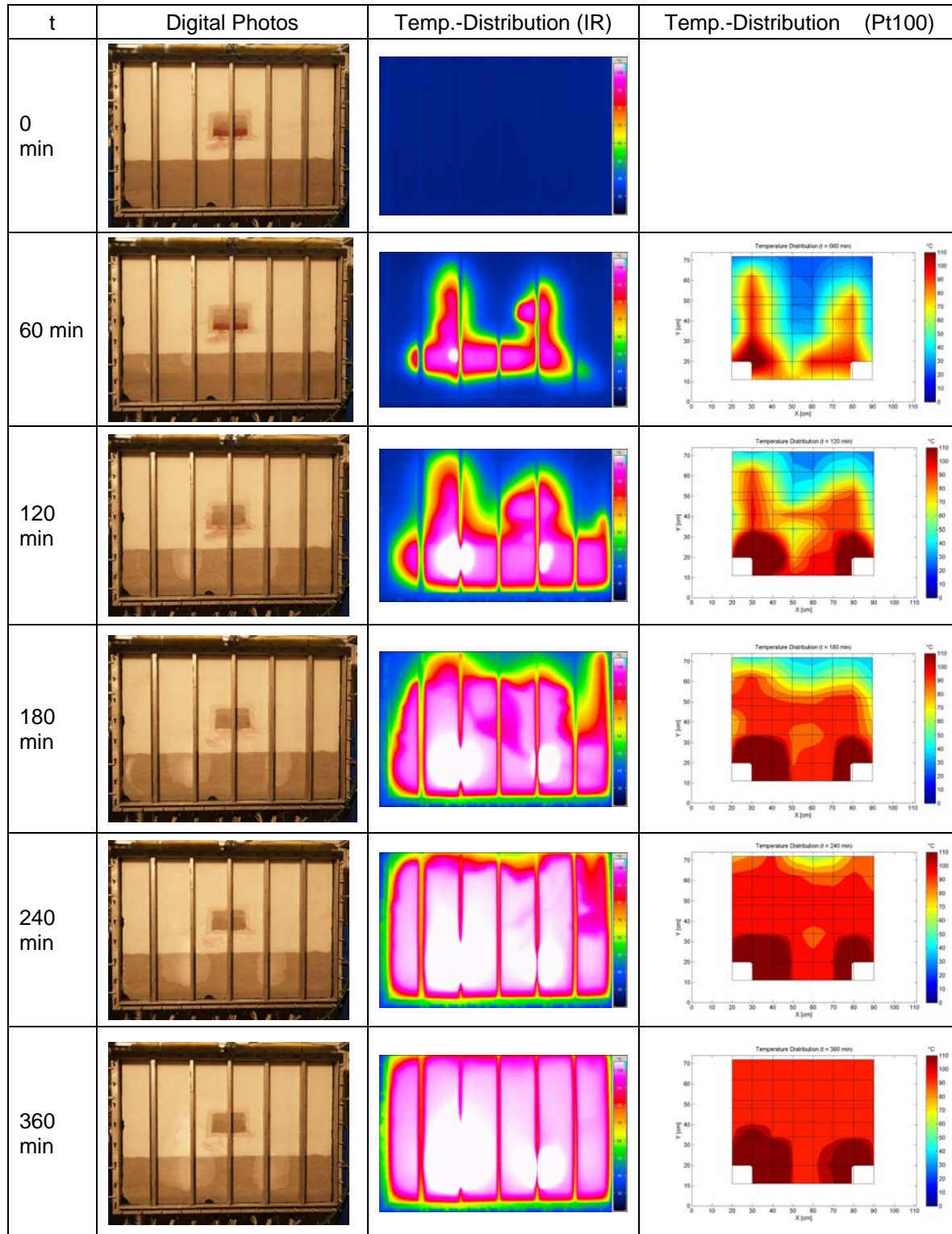


Figure 7: Results of the first 2D remediation experiment, based on visual, infrared and discrete temperature monitoring.

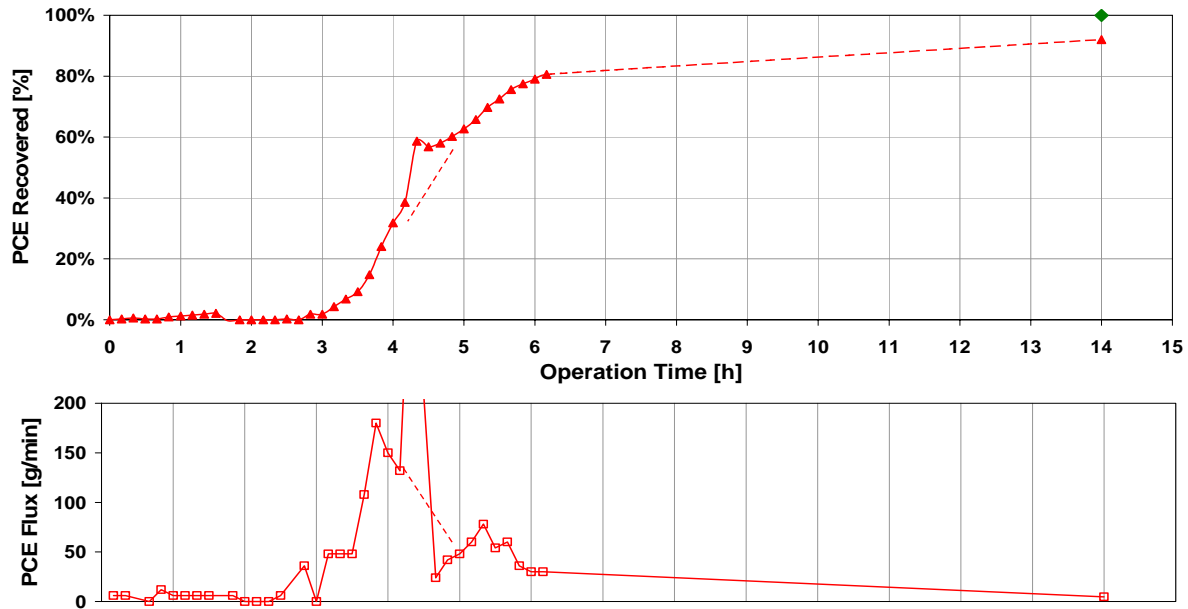


Figure 8: PCE recovery curve and PCE flux curve from the first 2D remediation experiment. Contaminant recovery could not begin until hour 3, at which time a gas phase pathway to the extraction point first occurred.

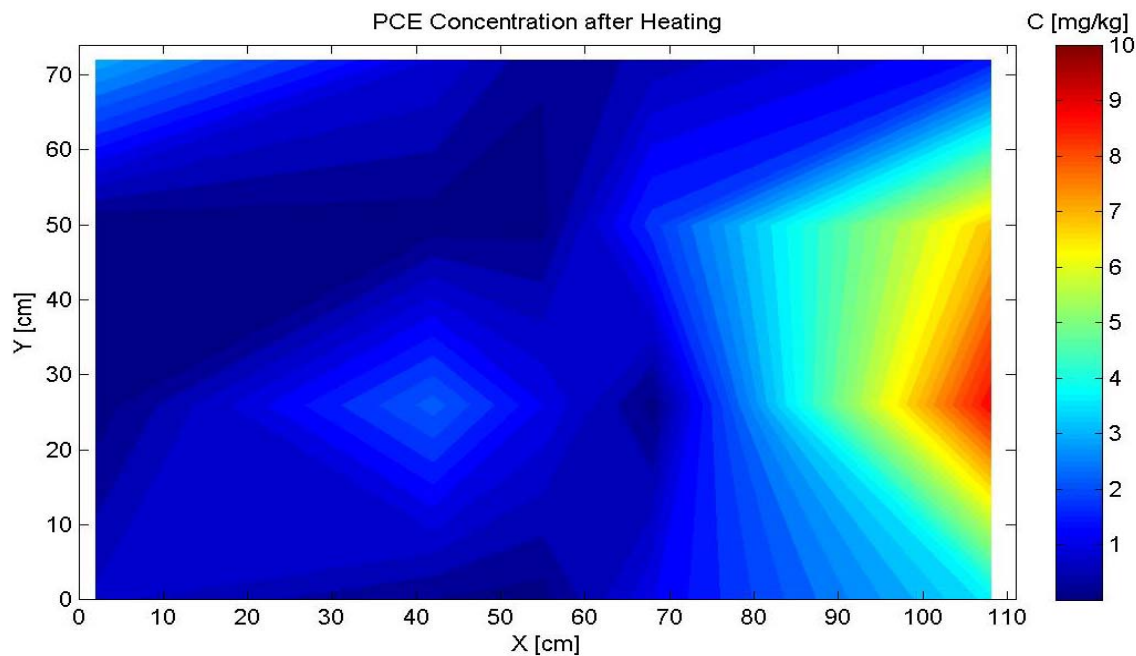


Figure 9: PCE distribution after the first 2D remediation experiment. The PCE remaining in the soil totaled less than 1 g, relative to 320 g infiltrated initially, representing a removal of >99.7%.

The progression of heating and contaminant recovery for the second 2-D remediation experiment was reported elsewhere (Li 2007). Efforts to model the results of these experiments numerically are ongoing.

CONCLUSIONS

The main TCH contaminant removal mechanism within the saturated porous media during both 2-D remediation experiments was vaporization. Nearly complete (>99.5%) DNAPL removal from the source

zones was accomplished by the TCH remediation method. No DNAPL mobilization was observed through pool mobilization outside of the target treatment zone during or following TCH. These 2-D experiments have set the stage for the more complex 3-D experiments presented in the companion paper (Hiester et al. 2008).

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