DNAPL Removal from the Saturated Zone using Thermal Wells

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ABSTRACT: In-situ thermal remediation (ISTR) technologies are receiving increasing attention for remediation of 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) 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. Bench- and larger-scale remediation experiments are being conducted 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, enabling numerical simulations to now be used to design the 3-D large-scale experiments. Several 2-D experiments have been conducted, examining heat propagation in various soil types. In addition, during 3-D heat transfer experiments in large (150m³) containers, the progression of heating to 100°C and accompanying desaturation have been monitored using 300 temperature sensors and 35 time domain reflectometry probes, respectively, allowing comparisons of the physical experiments and accompanying numerical simulations.

INTRODUCTION

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 characterised 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

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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 two 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, 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 heated vacuum 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 (ISTD).

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) and MK Tech Solutions (numerical simulations). Our research project, funded by the U.S. Strategic Environmental Research and Development Program (SERDP), is intended to rigorously investigate and help optimize these processes and accompanying contaminant mobilization and removal.

Initially, small-scale remediation experiments (2-D bench-scale) are being used to study the principal mechanisms that control the performance of TCH/ISTD below the water table. These experiments are serving as a benchmark for the numerical model, allowing testing of its reliability for representing such processes. The experimental data thus enable calibration of the numerical model to improve the accuracy of subsequent simulations. The calibrated numerical model is now being used to help design the more complicated experiments. Initial 3-D experiments are being conducted in a large-scale container (150 m^3) with controlled boundary conditions at *VEGAS*, where thermal well experiments in the unsaturated zone have been successfully conducted in the past (Hiester et al., 2002). Hundreds of temperature sensors and other measurement devices are being used to monitor the processes that occur during subsurface heating, including changes in temperature, phase, contaminant concentration, pressure and saturation. The large-scale experiments have begun with heat transport experiments to improve understanding of the propagation of heat in the subsurface. Further experiments will be conducted with carefully emplaced contaminant sources to quantify the removal efficiency as well as to investigate whether mobilization and spreading of the contaminants occurs or can be prevented using this remediation method.

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.

The objectives for the first year of the project, which is currently underway, were to undertake project startup; develop more detailed project execution plans; and begin the 2-D and 3-D experiments as well as the accompanying numerical modeling.

METHODS

The research methods reported on here are the precursor to experiments in which TCH accompanied by vacuum extraction will be employed under a range of conditions in large-scale containers (3m x 6m x 4.5m, and 6m x 6m x 4.5m) [width, length, height], in controlled-release, closed mass balance experiments with geologically-relevant layering. Modeling of experiments was conducted using STARS, a fully compositional, non isothermal, advanced process simulator developed and marketed by CMG, Ltd of Calgary, Alberta. STARS is regularly used to model ISTD field projects and other thermal processes such as laboratory and oil-field in-situ combustion projects.

2-D Experiments. Initial, smaller-scale experiments have been designed to enable examination of a larger range of experimental conditions than will be feasible in the large-scale experiments (Li, 2004). The experiments are conducted in a stainless steel 2-D flume with a Pyrex® (glass) front wall (Figure 1), which allows infrared photos of heat propagation at a fine resolution, and enables observation of the movement of tracer liquids in the porous medium. Inflow and outflow conditions are controlled, allowing no-flow and constant head boundary conditions to be maintained. Insulation surrounds the container, except during brief periods when it is peeled back to enable visual spectrum and infrared photographs to be taken. One heating element of 0.5 m length is located approx. in the center of the flume. One hundred PT-100 temperature sensors are installed in the flume to monitor the soil temperatures with an accurancy of ± 1°K. Additionally, in some experiments, infrared camera and saturation measurements using a gamma-source are used to focus on specific processes of interest. To obtain information of the interaction of soil permeability and heat transport, several

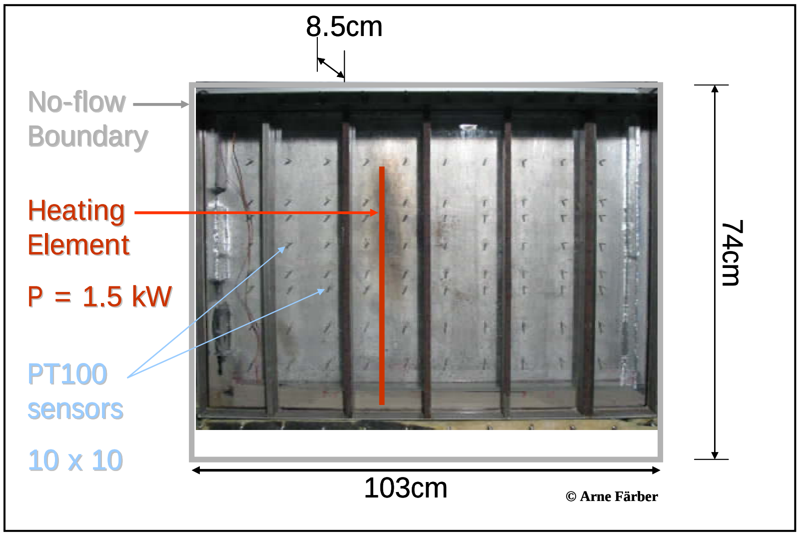

FIGURE 1. 2-D flume heat transport experiments.

experiments with different soil types are being conducted. In all experiments, the energy input of the heater is controlled and kept constant.

3-D Experiments. The existing large-scale container set-up from former experiments with Thermally-Enhanced SVE (TESVE) with thermal wells in the unsaturated zone (Hiester et al., 2002) has been modified so that it can be used for TCH heat-transfer experiments in the saturated zone (Figure 2). The base of the container is 6 m x 6 m and

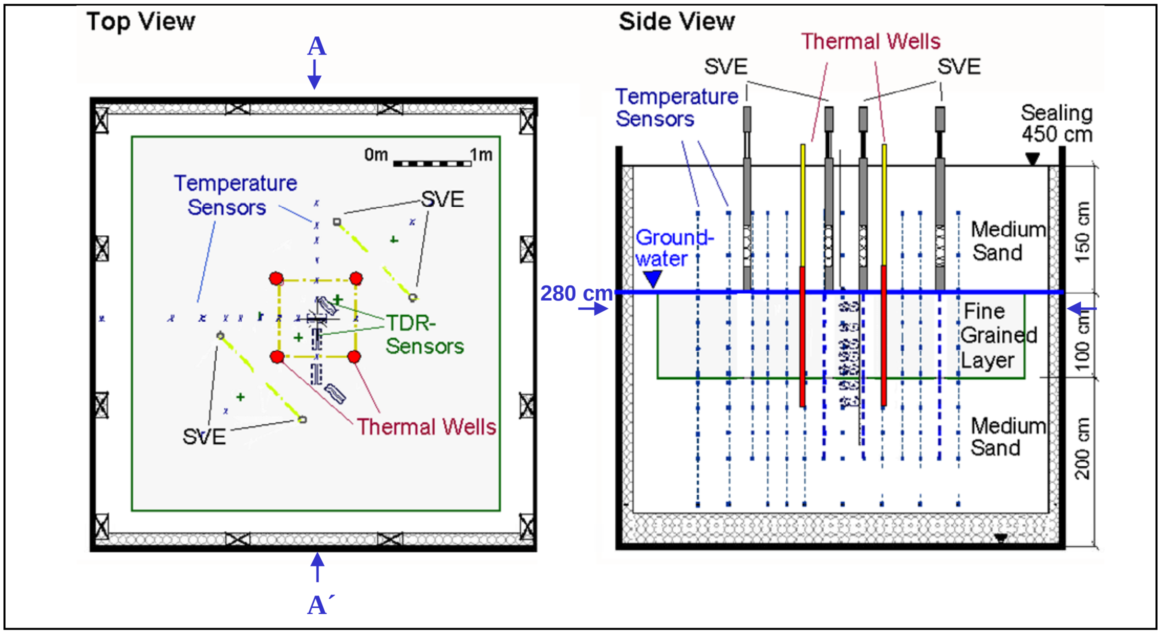

FIGURE 2. Plan view and side view of the 2nd 3-D heat transfer experiment (after Hiester et al. 2006).

its height is 4.5 m. A layer of a low permeability material is embedded in coarse sand. More than 300 thermocouples and Pt-100 temperature sensors enable monitoring of the temperature distribution in the subsurface. Additionally, 35 High Temperature- and Corrosion-resistant Time Domain Reflectometry (HTC-TDR) sensors are used to evaluate the water saturation in the subsurface. Furthermore, ground water level, discharges, pressures as well as temperatures in the soil vapor extraction (SVE) system are monitored. Non-isothermal numerical modeling performed in parallel with these experiments is being used to simulate the physically dominant mechanisms and processes of the experiments. Several options concerning the boundary conditions of the heating experiments were intensively discussed at an early stage of the project. During the first heating experiment the groundwater level was initially adjusted at 3 m above the bottom of the tank, which is at the top border of the fine-grained layer. The groundwater table was maintained at this position, creating a constant head boundary condition. In the top sand layer, each of the four SVE wells was operated with a constant flux of 5 m^3 SATP/h. Under this boundary condition, the lower tier of SVE wells positioned in the bottom sand layer are now in the saturated zone and are therefore not being operated. Heat propagation (temperatures), water saturation and SVE mass fluxes (water content and air) were measured throughout the experiments.

Design of New Container Set-Up. The upcoming 3-D large-container experiments that will include contaminants will be undertaken with a new stratigraphy, relative to what was used in the previous experiments that were originally set up to study TESVE. The new experiments will be conducted using lower-permeability materials more relevant to this project. Based on the existing container design (Hiester et al. 2002), detailed bench-scale experiments were conducted at Göteborg University, Sweden (Mark, 2005) in cooperation with our group to improve the understanding of the behavior of several fine sand and quartz flour mixtures being considered for this application. The layout and the realization of the 3-D experiment will be designed with the aid of the numerical model.

RESULTS AND DISCUSSION

2-D Experiments. Heating experiments were conducted in the 2-D flume, after establishing the water table at the top of the packed container. Figure 3 (left) shows the

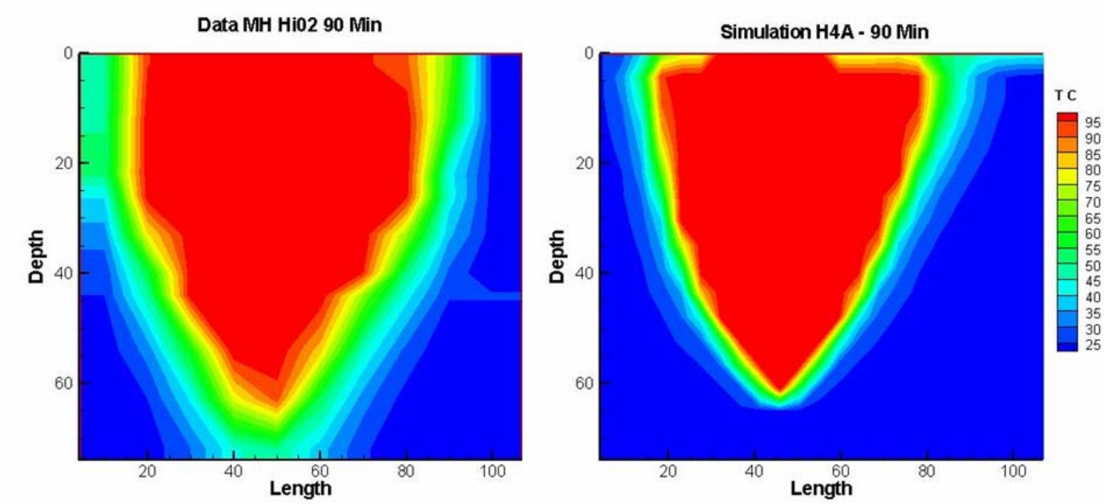

Figure 3. Temperature comparision: 2-D flume experiment (left) vs. numerical simulation (right). K_s=4.6 x 10^{-3} m/s; P = 1.5 kW; K_v/K_h=0.25; t=1.5 hr.

temperature achieved in the moderate permeability soil having a saturated hydraulic conductivity, K_s = 4.6 x 10^{-3} m/s after 1.5 hr of heating at a constant power, P = 1.5 kW. The corresponding numerical simulation is shown at the right. The STARS simulation assumed anisotropy (ratio of vertical to horizontal permeability, K_v/K_h) = 0.25. The simulations were conducted in a 35x5x20 model of the flume. The outer layers of the model represented the steel frame and glass front window, while the inner 33x3x18 cell region was water saturated sand. Inflow and outflow locations were included and heat loss was 2 W/cm^2/°K. The measured capillary pressure was used in the model but was extended to very low saturations and high capillary pressure (13 MPa) to facilitate movement of water toward the heater. As Figure 3 shows, the most evident difference between experiment and simulation is the much thinner transition between hot (red) and cool (blue) regions, the thermal boundary layer. Figure 4 presents similar results for the fine permeability experiment and simulation for Ks = 3.0 x 10^{-4} m/s after 27 hr of heating at a constant power, P = 0.24 kW. Experiments for each soil type (high, moderate, and low permeability) were repeated once, with good reproducibility. The difficulty of achieving uniform packing of soil in the experimental unit is evident in the lower permeability material (Fig. 4, left). In addition, movement of water out the overflow at the upper left corner of the flume causes the temperature to be higher there.

The coupled flow and heat transport processes in a multiphase system are complex with respect to the simultaneous range of influence. Nevertheless, using these experiments and models as a basis, simple approaches to determine the range of influence

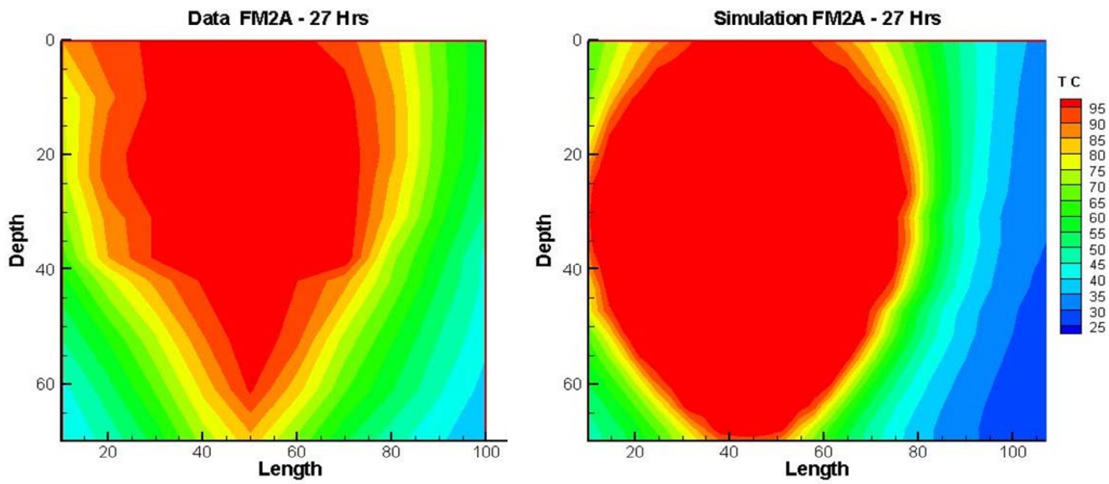

Figure 4. Temperature comparison: 2-D flume experiment (left) vs. numerical simulation (right). K_s=3.0 x 10^{-4} m/s; P=0.24 kW; K_v/K_h=0.25; t=27hr.

of a heater (propagation of the heat front as functions of power and time) in homogeneous soils of various hydraulic conductivities can be developed. Future experiments of this type will also be conducted with Trichloroethene (TCE) emplaced within the soil prior to heating.

3-D Experiments. The large-scale tank containing soil inherited from the earlier TESVE experiments was flooded to enable two heat transport experiments to be conducted, without contaminants being added yet. During the first experiment, a couple of 1-m long thermal wells were operated in the fine grained layer at 500°C. Within one week, temperatures of approximately 80°C were achieved in the central area between the thermal wells, at a spacing of 1 m. The heaters were switched off and the cooling phase of the heated subsurface was monitored (temperatures, pressures, fluxes). Prior to beginning a second large-scale heat transport experiment in the same container and with the same packing, the heaters were replaced by newly designed thermal wells of 1.5-m length (Figure 2). Rather than maintaining a constant head boundary condition, the water table was allowed to drop as water was evaporated off, to determine the effect of heating on the fluctuation of the water table and pressure conditions. Figure 5 displays the temperature distribution over time along A-A', and at elevation 280 cm (see Figure 2) along one of the HTC-TDR transects. It can be seen that for this packing, heating occurs predominantly within the top of the saturated zone (Fig. 5, Level 280). Examination of the various profiles indicates that the highest temperatures were achieved, as expected, in the midpoint between the heaters. Temperatures >100°C indicate that steam is produced in these regions, which effects a desaturation of the former saturated zone. With time, the extent of the heated zone increased (view Fig. 5 from left to right), with an increasingly larger fraction of the soil at Elev. 280 cm attaining the boiling point of water.

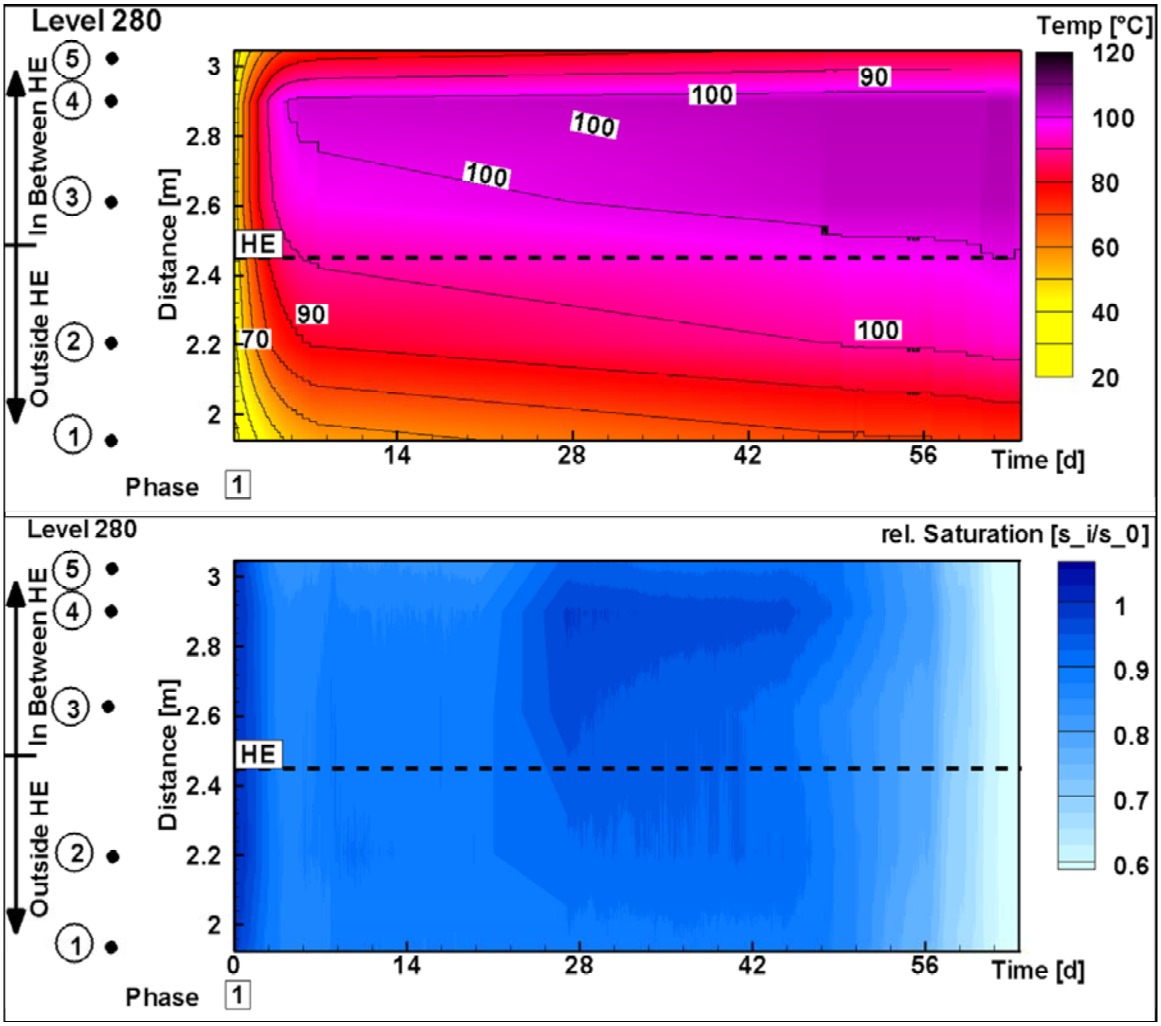

FIGURE 5. Progression of (a) Temperature and (b) Saturation Measured at Elevation 280 in Initial Large-Scale Container Experiment. This elevation corresponds to 20 cm below top of fine-grained layer. © Uwe Hiester

Concomitantly, as water evaporated and was extracted, the soil at that level was observed by means of the HTC-TDR sensors to have become desaturated, until at about 25 hr when it evidentally resaturated temporarily. This may be a result of condensation of water vapor being produced as heating extended into the underlying zone. Following about 42 hr, however, the entire volume of the fine-grained layer began to become desaturated, with the saturation at Elev. 280 cm being approximately 0.6 at 65 hr. This degree of saturation is comparable with completion of remediation, based on full-scale ISTD field results (LaChance et al., 2004, 2006). Data collected but not able to be presented due to the brevity of this paper provides a complete picture of the 3-D progression of temperature rise and saturation decrease throughout the container. Future reports will document this more completely.

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