

STEAM-AIR-INJECTION FOR IN-SITU GROUNDWATER AND SOIL REMEDIATION: PILOT APPLICATION AT THE FORMER INDUSTRIAL SITE IN ZEITZ, GERMANY

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Introduction

In-situ thermally enhanced remediation methods (steam-air injection and thermal wells), developed and verified in several field applications by the Research Facility for Subsurface Remediation, VEGAS, can be used for source zone remediation of NAPL in the saturated and the unsaturated zone. Contaminants can be effectively removed within a few months if hydro-geological conditions are suitable. Since steam-air propagation in the (initially) saturated zone depends on the local geology and stratification it is advisable to run a pilot trial for a safe design of the full-scale thermal in-situ remediation.

At a former hydrogenation plant in Zeitz, Germany on a site which is part of the “Ökologisches Großprojekt ÖGP Zeitz” (ecological large-scale project Zeitz) a pilot-scale trial of thermal in-situ remediation using steam-air injection was carried out within the research project SAFIRA II. The main objective of the pilot was the removal of a benzene source (partial source removal) and in consequence the reduction of the downstream contaminant flux in the groundwater (plume). This was monitored by an accompanying project of the UFZ. The source area is located underneath a former benzene distillation unit of the hydrogenation plant.

The pilot trial started in May 2007. After a short period of a “cold soil-vapor extraction” and following air-sparging the steam-air injection started. During several phases steam was injected into both the unsaturated and the saturated zone. Steam-air injection at different wells and depths ensured a safe extraction of benzene from the different layers particularly from a silt layer located above the capillary fringe. Steam/heat propagation was continuously monitored by direct temperature measurements and by innovative surface geophysical methods (carried out by UFZ). The steam injection was terminated in November 2007. The cooling phase lasted through December 2007.

Site Description

The pilot site is in the area of the former benzene plant. Damages and spills during World War II as well as leakages and overflow during benzene production are the reasons for the subsurface contamination. Benzene concentrations in the groundwater can be found in the range of the solubility limit. The source area is located underneath the former benzene distillation unit.

During the demolition of the old plant the top soil layers (2 - 3 m) were excavated and exchanged. Therefore the upper layer consists of a 2 - 2.5 m thick filling. Below this layer a fine to medium sand / silty layer ($K \sim 0.5 \times 10^{-4}$ m/s) of approximately 2.5 m thickness follows (see Figure 1). The thickness of the underlying silt layer is approximately one meter. Below this the aquifer consists of sandy fine and coarse gravel ($K \sim 5 \times 10^{-4}$ m/s) from 6.5 - 8.5 m b.g.s., followed by medium sand layers ($K \sim 1 \times 10^{-4}$ m/s). The aquifer bottom is about 11.5 - 12.5 m b.g.s. The groundwater water table is at about 8.5 meters b.g.s.

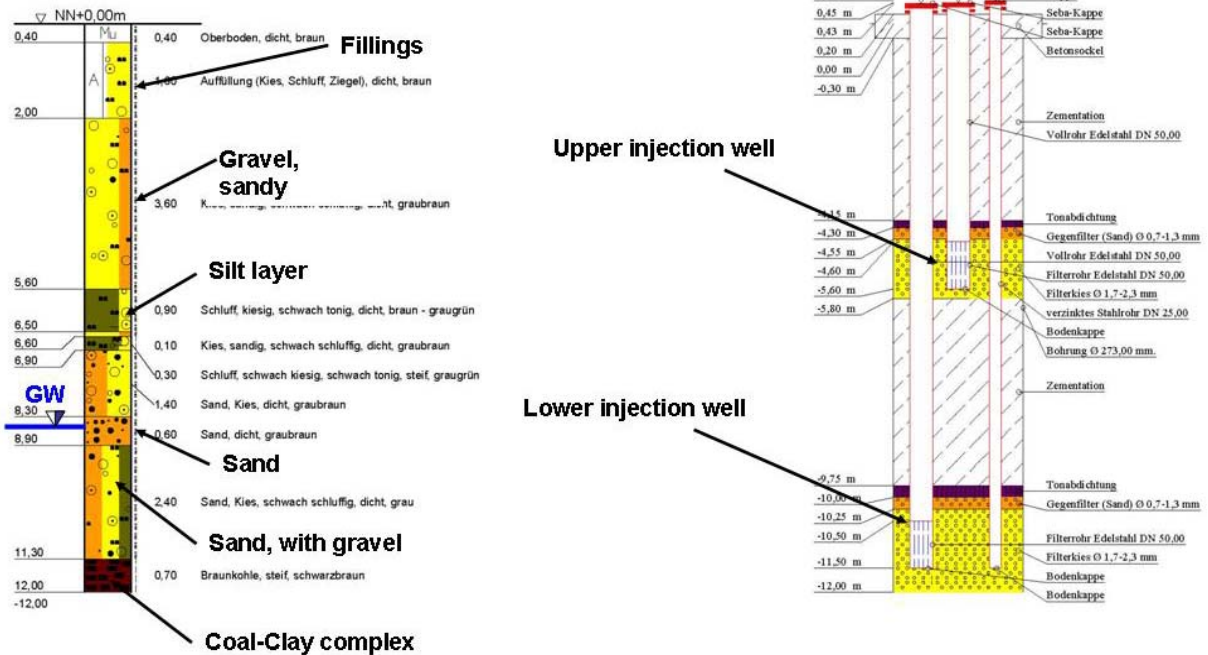


Figure 1: soil profile and set up of injection wells (I3)

Benzene concentrations in both the unsaturated and the groundwater fluctuation zone in the range of several g per kg soil were determined. During drilling for wells installation 28 soil samples from different depths were taken. In the upper part of the aquifer (9 - 10 m b.g.s.) high benzene concentrations were found. Most of the samples from the silt layers (depth 5 - 7 m) also showed high values. The mean benzene concentration was about 55 mg/kg of soil (BTEX: 61 mg/kg). Taking into account the losses during sampling due to the high volatility of benzene, the pollutant mass can be estimated to be about 1.700 kg of benzene in about 1.500 m³ of soil volume of the pilot site.

Groundwater samples from the surrounding monitoring wells show benzene concentrations between 370 mg/L (upstream) and 1.3 g/L (maximum solubility of benzene) downstream the pilot site. Hence, it could be estimated that the contamination was mainly located in the unsaturated zone and the capillary fringe in the silt layer.

Remediation Concept

Two thermally enhanced remediation techniques were applied - steam-air-injection and thermal wells (electrical heating elements, THERIS) - to heat soil to the "eutectic temperature" of benzene (75°C). The organic pollutants in the soil and groundwater evaporate during heating and are extracted as gas via a soil vapor extraction (SVE) system (Figure 2).

During steam-air injection into the soil layers with medium permeability the propagating steam condensation heats the saturated and unsaturated zone, while the air component acts as an inert carrier gas and transports the pollutants to the extraction wells. THERIS is used in the unsaturated zone in the soil layers with low permeability to evaporate the pollutants by conductive heating.

To differentiate the contaminant mass extracted from the unsaturated and saturated zone during the first step the steam-air mixture (90 - 110°C) was injected in the unsaturated zone (upper filter, Fig 2). After steam breakthrough at the extraction wells and a significant decrease of the pollutant concentrations to values <1 g/m³ of extracted soil vapor, steam-air was injected in the saturated zone. A remediation time of four months of steam-air-injection was estimated.

The pilot site area is divided into three roughly equal parcels, which were subsequently treated, starting in the North East (I1), continuing southwest (I2), and finishing in the center (I3). The site is equipped with:

- three injection wells with two injection filter sections at 4.5 - 6 m and 10 - 11.5 m b.s.g., respectively
- six soil vapor extraction wells, some with moderate groundwater extraction screened from 1 to 10 (11) m b.g.s.
- eight electric heating elements, each with 1.5 kW power, heated depth: 4.5 - 6 m b.g.s.
- 20 temperature lances (Pt100) at different distances to the injection wells and close to the extraction wells (121 Measuring points)
- Geo-electrical temperature monitoring system (M. Hirsch, UFZ) with 128 surface and 48 borehole electrodes (up to 16 m depth)
- additionally, 20 small-scale monitoring wells surrounding the pilot site were installed to control emissions.



Figure 4: mobile remediation unit (MOSAM) and pilot site

The pilot site included a data acquisition system and GSM modem for the online monitoring of essential process parameters (pressure, temperature, flow rates of steam, air, soil vapour and water, etc.). Contaminant concentrations in the extracted soil vapor were detected online using a PID (ex-protected) and BTEX concentrations were measured hourly using a process gas-chromatograph. Additional safety measures and equipment were required due to explosion hazard (benzene).

Implementation and Testing Phases

The pilot trial started in May 2007 and was divided into several phases:

- Phase 1: soil vapor extraction (SVE) (50-200 kg/h) and groundwater extraction at GWL1 and EK5, duration 3 weeks, cleaning of extracted air by thermal afterburning
- Phase 2: SVE, groundwater extraction and air sparging, 3 weeks
- Phase 3: steam-air injection in I1, I2 and I3 (s. fig. 3) and THERIS, about 19 weeks
- Phase 3.1: unsaturated zone (6 weeks) injection between 5 to 6 m b.g.s with injection rates of 80-150 kg/h steam (up to 100 kW) and 40 - 70 m³/h air.
Objective: medium temperature in the target zone > 75 °C
- Phase 3.2: saturated zone and silt layers (7 weeks): injection between 10 to 11 m b.g.s. with maximum injection rate (120 kW and 70 m³/h air).
Objective: medium temperature in the target zone > 60 °C
- Phase 3.3: saturated and unsaturated zone (3 weeks) in wells I1u, I2u, I1o and I3o with about 170 kg/h steam (120 kW) and 30 - 60 kg/h air.
Objective: average temperature in the pilot field > 85 °C (boiling point of benzene), extraction of benzene from unsaturated zone (silt layer),

Phase 3.4: unsaturated zone (3 - 4 weeks): in wells I1o, I2u, I3o and EK3 with about 170 kg/h steam (120 kW) and 40 - 80 kg/h air
 Objective: extraction of benzene from the unsaturated zone, treatment of the area around EK3

Phase 4: cooling phase, approximately 4 weeks.

To accelerate the cooling process compressed air was injected with 50-70 m³/h at various locations: unsaturated zone (air venting), saturated zone (air sparging). The cooling phase was finished after temperatures below 55 °C were reached.

Heat Propagation

The main focus of the pilot trial was the steam injection (phase 3) which started after 6 weeks. The propagation of the steam front in the unsaturated zone (Phase 3.1) was homogeneous around the injection wells in the area of I2 and I3 (radial symmetric behavior). In the region around I1 steam flux was mainly directed to EK5, EK2 and GWL1, (see Figure 5). EK3 was not affected by a steam breakthrough. The injection of steam (50 kW) into EK3 (32 days after the start of the injection) resulted in reaching the desired temperature of 85 °C (day 44 in Figure 5, day 84 in Figure 8) in the unsaturated zone.

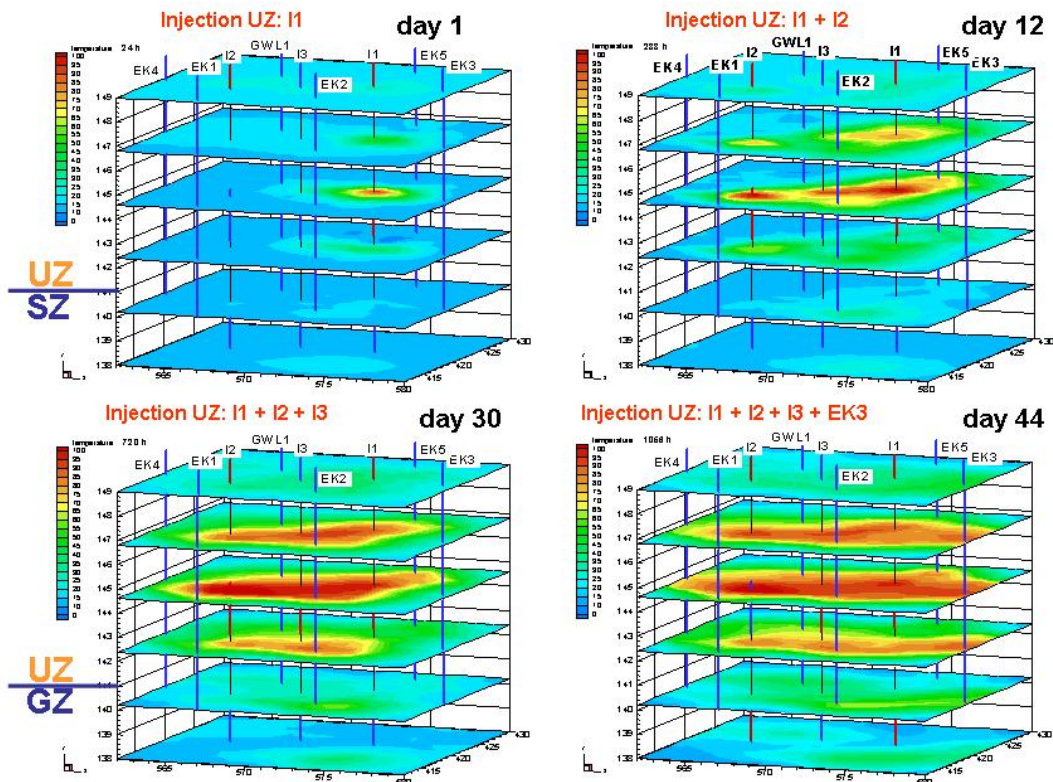


Figure 5: heat propagation during phase 3.1 (unsaturated zone)

At the beginning of phase 3.2 the steam-air mixture (150 kg/h steam + 80 kg/h air) was injected via I1u into the saturated zone on top of the clay-carbon complex (9.5 - 10.5 m b.g.s.). After a few days the steam breakthrough was observed in the saturated zone (8 to 10 m b.g.s) in the extraction wells GWL1 (6 days), EK2 (6 days), EK5 (12 days) and at lower temperatures at EK3 (12 days). The "thermal range of influence" in the saturated zone was more than 5.5 m. The temperatures in the saturated zone steeply increased (gray line Figure 8).

After 20 days of phase 3.2 the steam-air injection (power 50 kW) in the well I2u was added to the injection in I1u. The steam front propagation was fast around I2 and steam breakthrough first occurred on the level of the capillary fringe (8 ft b.g.s.) at EK1 and EK4. Due of the injection of "only" 50 kW at I2u the heating of the saturated zone up to 85 °C on a level of 10 m b.g.s. was slower and it took 29 days to reach EK1 and 34 days for EK4, see Figure 6.

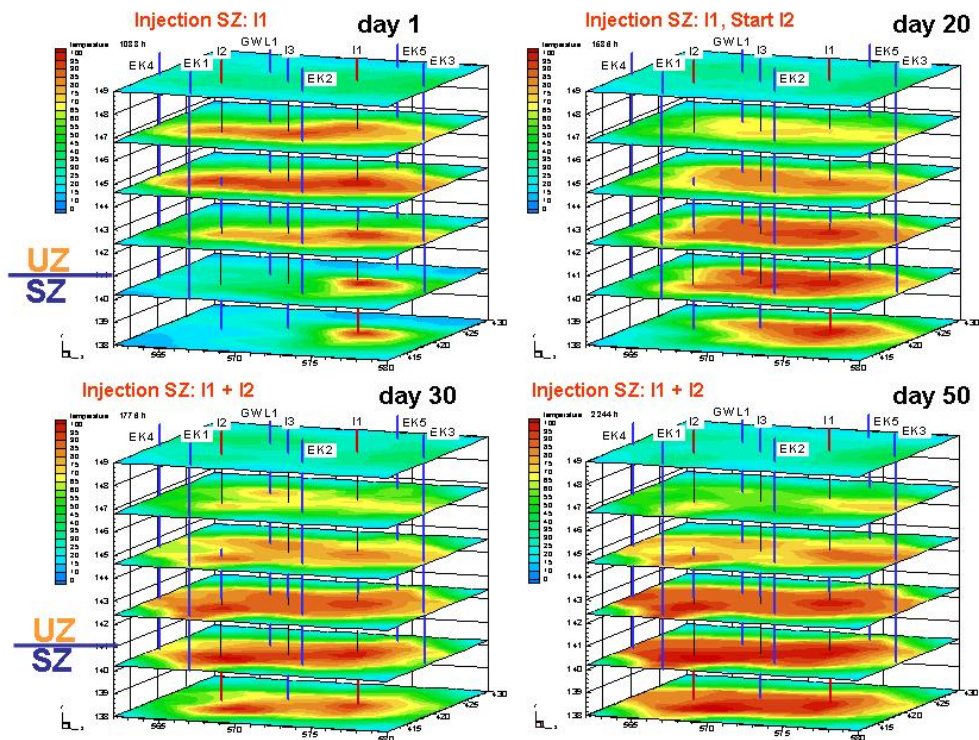


Figure 6: heat propagation during phase 3.2 (saturated zone)

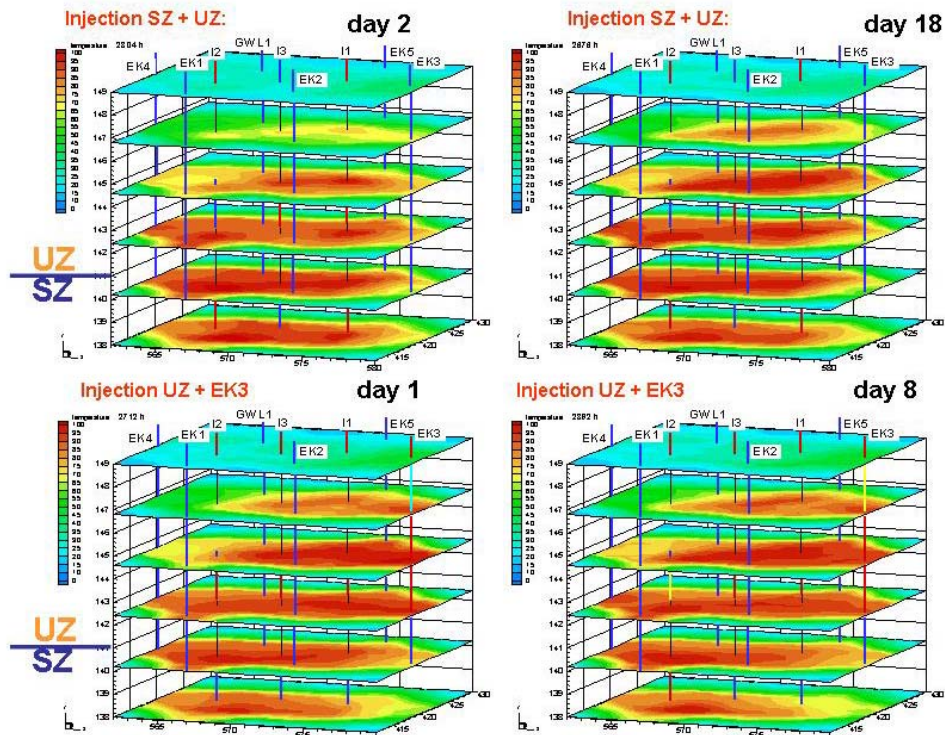


Figure 7: heat propagation phase, 3.3 and 3.4 (SZ + UZ, UZ only)

During the following injection (phase 3.3) of steam-air in the saturated (80 KW) and in the unsaturated zone (40 KW) the silt layer was heated to above 90 °C (yellow line in Figure 8). The average site temperature exceeded the boiling temperature of benzene (85 °C). After 18 days the unsaturated zone and especially the upper area of the unsaturated zone (about 7 to 3 m b.g.s.) reached the boiling temperature of benzene, see Figure 7, top right.

During Phase 3.4 the unsaturated zone, particularly the area around EK3, was heated to the boiling point of benzene. As a result of local low permeability zones around I1o the unsaturated zone in this area had to be heated by advective heat transport from the saturated zone (injection in I2u) and increased power (80 kW) at I3o, Figure 7, bottom right.

Energy Consumption

At the end of the steam-air injection into the unsaturated zone (Phase 3.1) approximately 90 MWh of energy (costs: 70 EUR/MWh) were consumed. Due to SVE about 10 MWh were “extracted” from the pilot trial (see Figure 8). When the average temperature in the unsaturated zone (yellow line, Figure 8) reached 85 °C (boiling point of benzene), exceeding the target temperature of 75 °C, phase 3.2 started.

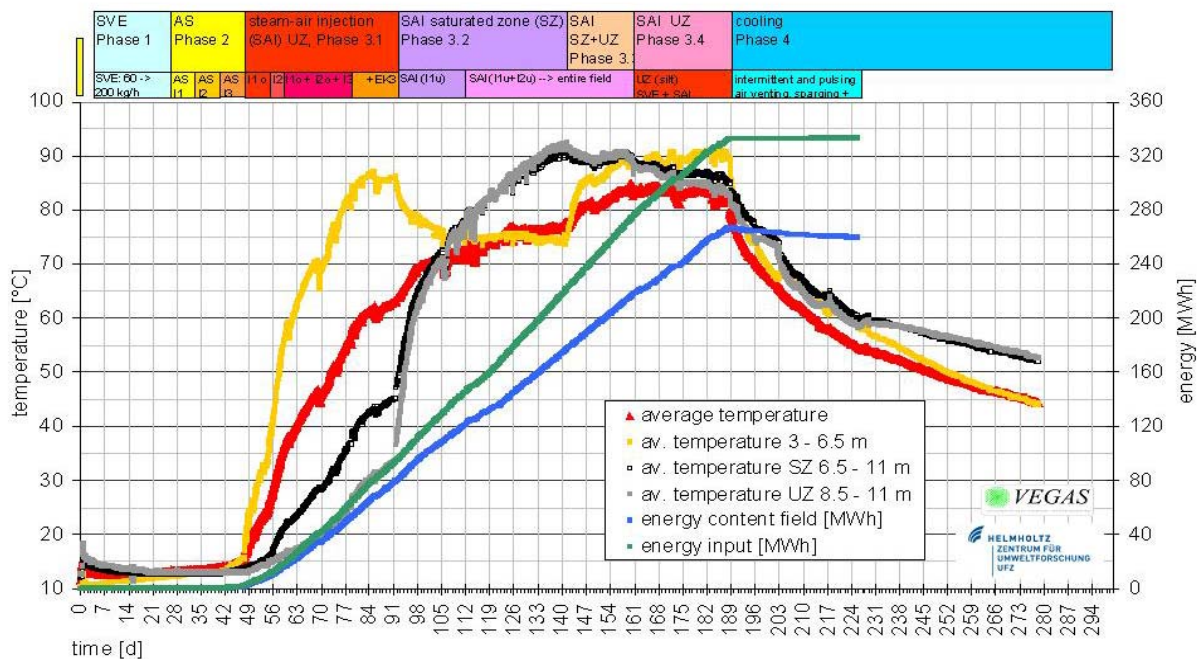


Figure 8: heating of the pilot site and energy consumption

Due to energy losses through groundwater, 130 MWh of energy were needed additionally for the heating of the saturated zone (phase 3.2) and the area underneath the silt layer. With an average temperature in the field of 77 °C and a temperature of more than 90 °C in the saturated zone (8.5 - 11 m b.g.s.) the target values (mean temperature in the aquifer 60 °C) were significantly exceeded.

For the heating of the silt layer (phase 3.3) another 60 MWh of energy was consumed. Due to simultaneous heating of both zones, the average temperature of the entire field reached values of around 85 °C (boiling point of benzene). At the end of the steam-air injection (11/08/07, day 189) in total 330 MWh of thermal energy had been injected, while 66 MWh of energy was extracted via the soil air and groundwater extraction. With an average temperature of 85 °C and an estimated heated volume of soil of about 1.500 m³ the stored energy in the subsurface was approximately 54 MWh. The difference of about 210 MWh meaning 65% of the energy was lost due to groundwater flow and due to convective heat transport across the test field boundaries.

Contaminant Extraction

During phase 1 a large amount of benzene (2.130 kg) was removed (see Figure 9) This was considerably more than estimated from the soil samples. The additional mass may have been extracted from the boundary areas of the field as a result of the range of influence of SVE wells during the cold SVE. The typical logarithmic decay behavior of the soil-air concentrations can be seen. Starting with an initial benzene concentration of 70 g/m³ in the soil vapor the concentration declined during the first three weeks to about 25 g/m³ (see Figure 9, blue line).

The decrease of the benzene concentrations in the wells continued during the air-sparging (phase 2). During intermittent air injection in field 1 and 2 the benzene concentration decreased almost linearly. Values of about 16 g of benzene per m³ of soil air were measured before switching to field 3. Starting air-sparging in field 3 the concentrations rapidly increased to values of 25 g/m³ for a period of several days but dropped again to 15 g/m³. In total 1.920 kg benzene were extracted during three weeks. Since air-sparging was less efficient than expected it is assumed that most of the contaminant mass is located in the unsaturated zone.

During Phase 3.1 (steam-air injection in the unsaturated zone) the concentrations of benzene significantly increased over several days at the beginning of each injection in the single field areas (I1 → I2 → I3). In general the concentrations decreased continuously, but slower than predicted based upon the extrapolation of logarithmic decay curve of phase 1 (see Figure 9). On 07/02/07 (day 61), the air injection rate was doubled resulting in a significant increase of extracted mass. By the end of the injection in the unsaturated zone (91 days, 6 weeks steam injection) approximately 2.280 kg of benzene were extracted. Compared to the extrapolated mass extraction based on the regression curve of cold soil vapor about 630 kg of benzene were extracted additionally due to the thermal enhancement. When switching the injection into the saturated zone (phase 3.2) the benzene concentrations in the extracted soil air were less than 1 g/m³.

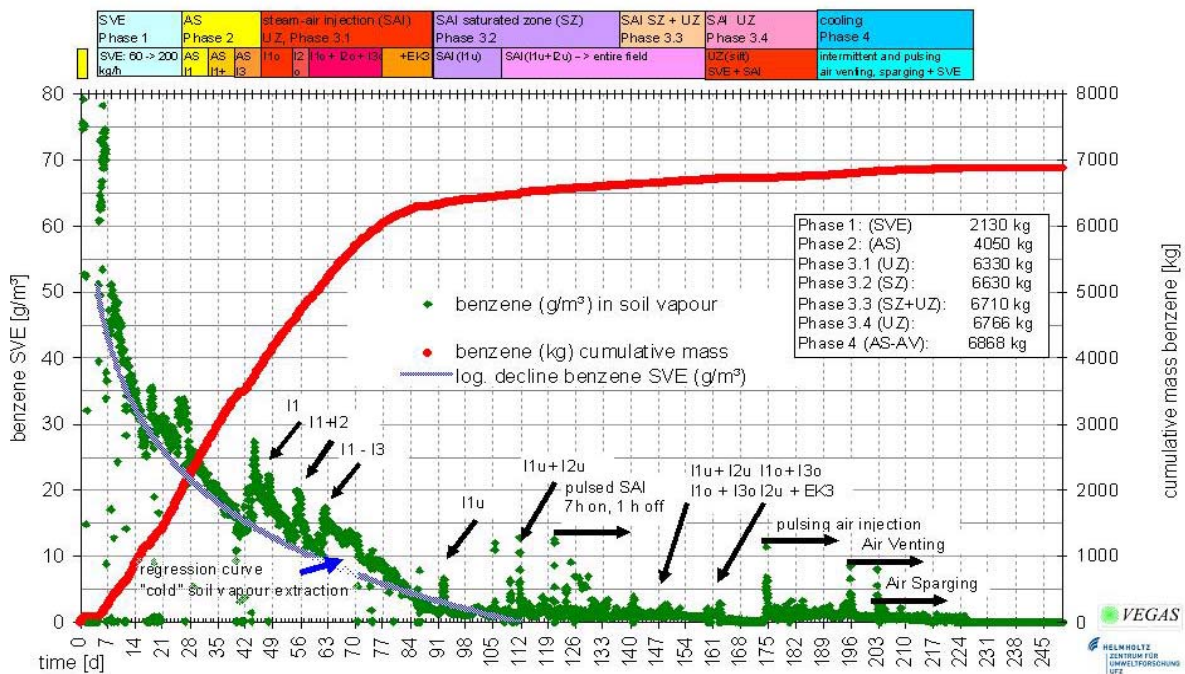


Figure 9: benzene concentrations and mass balance Phase 1 – 4

During phase 3.2 the injection of steam-air into the saturated zone resulted in the removal of 300 kg of benzene within seven weeks. At the end of this phase an increase in concentrations was observed indicating the expected vaporisation of benzene from the silt layer into the unsaturated zone. In order to avoid condensation of the vaporised benzene, a spreading of pollutants and to enhance contaminant removal from the silt layer steam-air was injected during phase 3.3 into both zones. The steam-air mixture was injected in the unsaturated zone with a power of approximately 40 kW. The heating of the silt layer was resumed by conductive heat transfer due to injection into the saturated zone underneath (80 kW). Increased benzene concentrations (peaks) were observed following the variation of steam injection rates and the operation of different wells. The contaminant mass extraction further decreased; benzene concentrations fluctuated between 1 and 2 g/m³ of soil vapour (see Figure 9). The extracted mass of benzene was relatively small (80 kg). Temperatures between 92 and 95 °C in the silt layer indicated the completion of the remediation of this zone.

During the final phase 3.4 (steam-air-injection, unsaturated zone) particularly the area around EK3 was treated. At the end of phase 3.3 benzene concentrations of 6 g/m³ in the soil air of EK3 were observed. At the beginning of the injection in EK3 benzene concentrations in the soil vapor significantly increased followed by a rapid decline after 4 days to values of about 400 mg/m³.

Continuation with discontinuous air injection resulted in an enhanced benzene extraction (0.8 - 3.3 g/m³) and after reaching the target value of 1 g/m³ benzene the steam-air injection was stopped.

To enhance the cooling of the unsaturated zone a high injection rate of air (90 m³/h) was applied (air venting), so residues of condensed contaminant could be extracted (peak behavior, day 196, Fig. 5). The concentration of benzene reached steady state values of about 600 mg/m³ in the soil vapor. To enhance cooling the saturated zone air was injected at different levels (air sparging, day 203). For a short-time benzene extraction increased but quickly returned to 600 mg/m³. Switching again to air venting and pulsating air injection resulted in a further reduction of contaminant concentration. One week before turning off the active cooling phase the benzene concentration was below 200 mg/m³ in the extracted soil vapor.

Summary

At a former hydrogenation plant in Zeitz, Germany, on a site which is part of the "Ökologisches Großprojekt ÖGP" of Zeitz (ecological large-scale project of Zeitz) a pilot-scale trial of thermal in-situ remediation using steam-air injection was carried out within the research project SAFIRA II. The source area is located underneath a former benzene distillation unit of the hydrogenation plant. The pilot trial had a volume of approx. 1.500 m³ and extended to a depth of 12 m b.g.s. Three injection wells were installed surrounded by six extractions wells.

The main objective was the removal of a part of the contaminant source zone (partial source removal) of benzene and in consequence the reduction of the downstream contaminant flux in the groundwater (plume). This was monitored by an accompanying project of the UFZ.

The pilot trial started in May 2007. Due to explosion protection reasons the amount of initially extracted soil vapour from the field site was kept very small (max. 10% of total flux) during the initial "cold" soil vapor extraction phase. The maximum daily discharge rate was about 110 kg benzene. Within the first three weeks, 2.130 kg of benzene were removed. This was considerably more mass than had been estimated based upon soil sampling. In addition, during the next phase (air-sparging), 1.920 kg of benzene were extracted.

During the following thermally enhanced phase by steam-air injection into the unsaturated zone, another 2.280 kg of benzene could be removed. The benzene concentration in the soil vapour significantly decreased from 25 g/m³ to less than 1.5 g/m³. During the following steam-air injection into the saturated zone approximately 300 kg of benzene were extracted. Heat propagation and small benzene concentrations indicated the completion of the thermal remediation in the saturated zone after seven weeks of operation. The benzene concentration was below 0.6 g/m³ in the extracted soil vapour.

After 6 days of injection steam breakthrough at the extraction wells was observed on a depth of 8-10 m b.g.s. The "thermal radius" (horizontal, radial steam propagation) of more than 5 m for an injection rate of approx. 220 kg/h of steam-air was larger than predicted (2 m) based upon gravity number related type curves (Ochs, 2006). This indicates an anisotropic permeability distribution.

In order to complete the removal of benzene from a silt layer located in the unsaturated zone above the capillary fringe the steam-air mixture was injected in both zones simultaneously. Locally high concentrations of benzene in one extraction well caused by inhomogeneous steam propagation in a small part of the unsaturated zone were removed using this well for steam-air injection during the final operational phase (unsaturated zone).

The end of the injection was in November 2007. The final cooling phase ended in December 2007. In total 6.780 kg of benzene were extracted, the final benzene concentration in the soil vapor was below 0.2 g/m³, indicating a removal of more than 99%. Benzene concentrations in the groundwater were reduced by a factor of 10.

Conclusions

The goal of the remediation was reached in the unsaturated and saturated zone, in the capillary fringe and especially in the silt layer. With regard to the total extracted mass it has to be emphasized that about 97% of the removed mass of benzene was extracted from the unsaturated zone and the capillary fringe.

The reason that only 3% of the total mass removed came from the saturated zone was not due to the method used but simply due to the fact that only 3% of the contaminant mass was located in the saturated zone in the first place. The energy consumption and operation time for steam-air injection in the saturated zone would not have changed appreciably if there had been a much larger contaminant mass, since the energy input is in the first place a function of the bulk soil volume that has to be heated and only in the second place a function of the contaminant in place.

For the site under consideration the sensible full scale remediation should therefore consist of three steps: First, cold soil vapour extraction should be used to remove as much as possible of the contaminants from the unsaturated zone. In a second step, the SVE should be thermally enhanced to ensure the cleanup of the silt layer, the capillary fringe and the to complete the remediation of the unsaturated zone. However, due to the relatively small amount of contaminant mass in the saturated zone, a cost-benefit-analysis might advise to concentrate on the unsaturated zone only.

VEGAS developed and built a modular mobile thermal remediation plant (MOSAM) for pilot trials in the framework of several R&D projects. The owner of MOSAM is the UFZ. VEGAS carried out the pilot trial in Zeitz on behalf of the UFZ. Simultaneously, in the framework of different projects (SAFIRA II, BEOQUE), various in-situ remediation technologies (RF heating, air sparging) for partial source removal were applied at the site to compare their effectiveness.

In close co-operation between the UFZ and the authorities (LAF, Landesanstalt für Altlastenfreistellung, Sachsen-Anhalt) responsible for the ÖGP, the results will be integrated in the future site development concept for the ÖGP.

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