3c.2 Thermal technologies for site remediation

STEAM-AIR INJECTION IN FRACTURED BEDROCK: COMPLETION OF A CHC-REMEDIATION AT THE SITE BISWURM (VILLINGEN-SCHWENNINGEN, GERMANY)

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Introduction

In situ thermally enhanced remediation methods (steam-air injection and thermal wells) may lead to a cost effective source zone remediation of NAPL in the saturated and the unsaturated zone. Contaminants can be effectively removed within several months if hydrogeological conditions are suitable.

In 2009, *VEGAS* successfully completed a pilot trial of steam-air injection to remove chlorinated hydrocarbons from a fractured sandstone aquifer for the city of Villingen-Schwenningen in south-west Germany [5]. The test field extended over 2,000 m³ of fractured rock including the upper of two aquifers. The thermal radius of steam propagation was 5 m in the target zone between 3 - 15 m bgs. The heating period by steam-air injection lasted for 19 weeks. The contaminants were thermally desorbed from the sandstone matrix during the conductive heating of the bedrock while steam-air was flowing through the fractures. More than 91% of the total extracted mass (560 kg chlorinated hydrocarbons, CHC) was removed from the groundwater fluctuation and unsaturated zones via the soil vapour extraction system, less than 6% thereof (34 kg of CHC) via the groundwater containment. The CHC values in the soil vapour and the groundwater were decreased by 95% and 85%, respectively.

Consequently, the steam-air enhanced remediation of the groundwater fluctuation zone and of the unsaturated zone (2,900 m², 15 m thickness) for the entire site was designed. The site was divided into nine treatment sections in sizes of 400 – 600 m², meaning 4,500 – 6,000 m³ of fractured bedrock each. The duration of the steam-air injection phase (steam injection power of 400 kW) was calculated to last 33 months requiring 31 two-level injection wells and 34 soil vapour extraction wells (SVE). After awarding the contracts in 2012, the costs were estimated to be 3.5 million EUR to treat ca. 43,000 m³ of sandstone during four years of operation.

Site description and remediation method

The former incineration plant for liquid organic waste (CHC, BTEX) of Biswurm extends over an area of 2,900 m². Organic liquid wastes were stored and incinerated in six storage and incineration ponds covering a total area of about 800 m². Throughout the concrete ponds liquid contaminants migrated through several meters of clay into the underlying fractured sandstone formation. The contaminant source zone extended over ca. 2,000 m².

During the demolition of the plant in 2004, the upper clay layer of contaminated soil was exchanged (3.5 m thickness, 7,100 tons) and a drainage system was cut in the underlying claystone and sandstone. In total about 1,600 kg of CHC, 2,200 kg of copper, 40 kg of lead and 600 kg of mineral oil were removed.

A detailed site investigation estimated a total mass of between 10 and 100 tons of CHC in the underlying sandstone aquifers [1]. The concentration of CHC in the groundwater ranged from 1 mg/l in the saturated zone to up to 40 mg/l in the surface water drainage system (6 m bgs.). The content of CHC in the soil vapour was up to 4 g/m³ in the source zone and 200 mg/m³ in the surrounding area. The average CHC-content in the groundwater capturing wells GW10 - GW12 was 3,900 µg/L.

Below the exchanged soil from 3.5 m bgs on, a fractured claystone and sandstone (sot) of 6 m thickness forms the unsaturated zone, an underlying platy sandstone (sos) forms the upper aquifer, separated from the underlying lower aquifer (fractured siliceous sandstone, smk) by a thin layer of mudstone at approximately 22 m bgs. Underlying is bedrock granite at 37 m bgs. Both aquifers are confined. The water level in the wells is found at approx. 12 m bgs.

Tracer tests [2] with Uranine AP as fluorescent tracer indicated a porosity of 1% and an interaction of the confined aquifers. The natural seepage velocity ranges between 30 – 80 m/d. The hydraulic conductivity of the aquifer is approximately 3 x 10^{-5} m/s.

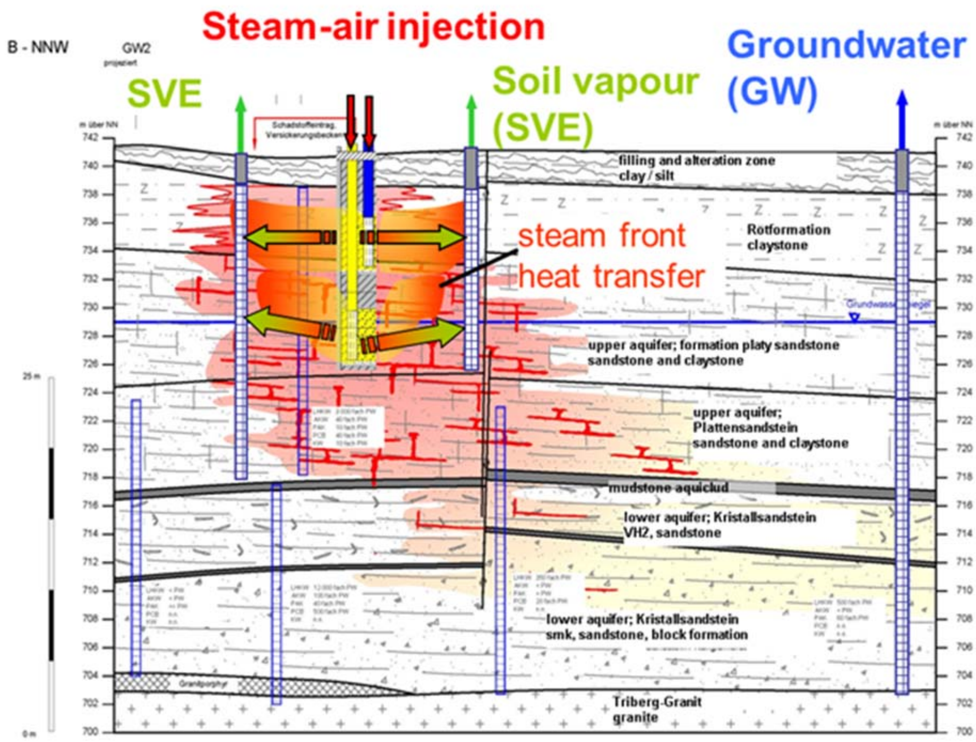

Figure 1: Soil profile and set up of injection and extraction wells

The steam-air mixture is injected on two different levels in order to control the required injection pressures: (1) in the platy sandstone (sos) below the groundwater level (12 – 15 m bgs) and (2) in the platy sandstone and claystone of the unsaturated zone (sot) (5 – 8 m bgs.) (Figure 1).

During the remediation process, the propagating steam heats the saturated and unsaturated zone by condensation. The air component acts as an inert carrier gas and transports the vaporized pollutants to the extraction wells. Considering the fractured sandstone aquifer and the claystone formation of the unsaturated zone with a low permeability there are two main processes related to heat transport. Steam-air will flush the fractured system, dewatering the fractures. Steam is condensing in the fractures, its surface and the porous sandstone matrix, thus transferring the heat by conduction to the sandstone matrix. Therefore as much steam as possible (maximal injection pressure) was injected into the fractured system in order to sustain the streaming steam propagation process as long as the sandstone matrix is being heated up.

The “azeotropic temperature”, the boiling point of the two-phase-system steam and contaminant (PCE and TCE), is 80°C in the upper unsaturated zone and 88°C in the saturated zone [5]. The procedure of the steam-enhanced remediation is to exceed this temperature in a heating phase lasting 6 weeks and to maintain the higher temperatures for additional eight weeks to evaporate the contaminants.

Remediation concept and procedure

The treated site extends to 15 m depth, 40 m width and 70 m length (43,000 m³ in volume), (Figure 2). In total, there are 32 two-level injection wells, 37 soil vapour extraction wells and three groundwater wells downstream of the contaminated area. In total, there are 89 temperature measurement tubes emplaced to monitor the heat propagation at 20 different vertical profiles.

The site was be treated stepwise in sections of approximately 4,000 m³ of fractured sandstone and claystone, each following the groundwater flow direction from north to south. Each of the in total nine sections is equipped with 4 – 5 dual injection wells surrounded by 10 - 12 SVE wells. The grid distance between the injection wells is 7 m; the distance between SVE-wells is 10 m.

The heating power should have ranged between 300 and 450 kW for operating 8 to 10 injection wells with injection pressures of 1.4 – 1.6 (top level) and 2.6 – 3.2 bar (sandstone). More than 600 m³/h of the soil vapour was to be extracted from the surrounding 10 SVE wells.

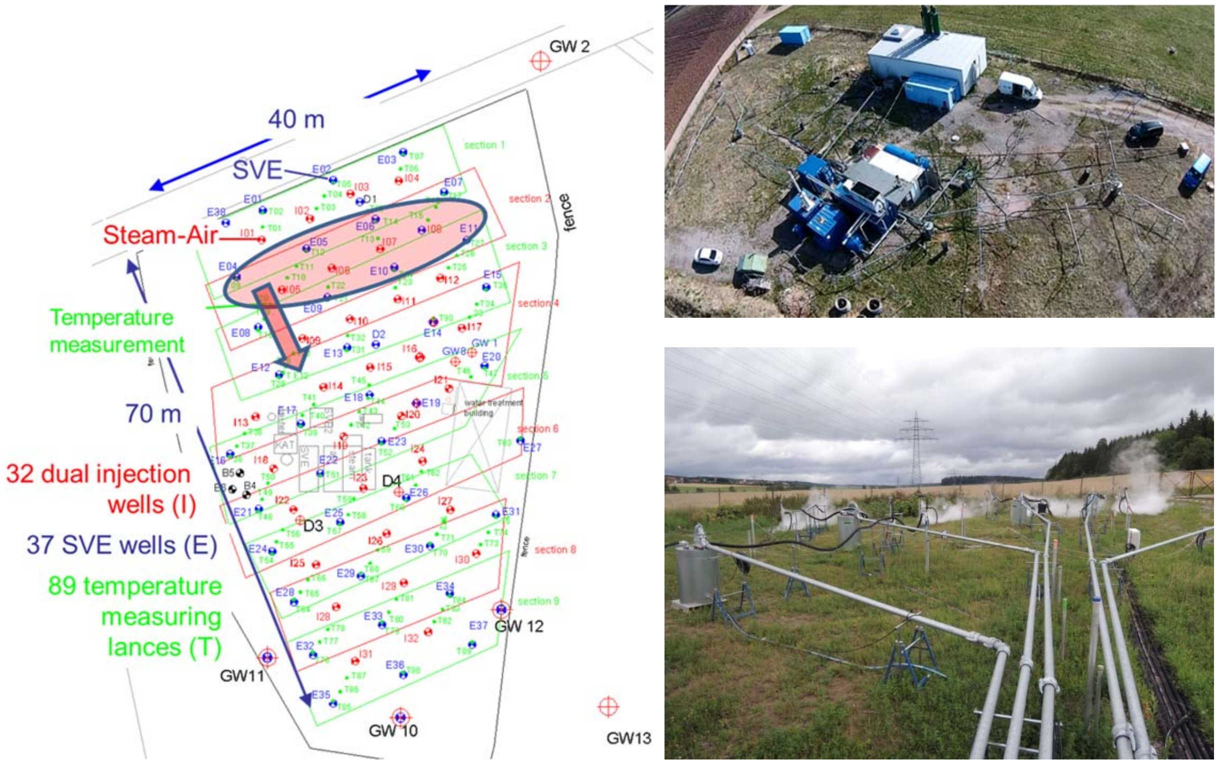

Figure 2: Map and perspectives of the completed remediation

The site is hydraulically contained downgradient of the treated area by two wells (GW11, GW12) operating in the upper aquifer (sos) and one well (GW10) in the lower crystalline sandstone aquifer (smk). The total discharge of this groundwater containment was to be 12 – 14 m³/h to capture the overall downgradient contaminant emission. A second line of observation wells (GW13 – GW16) is located 20 m downstream the pumping wells.

The process equipment includes two steam generators, an air compressor, groundwater discharge, storage and treatment units as well as a soil vapour extraction system (blower, condenser, cooler) to treat the contaminated hot soil vapour by catalytic oxidation including acid washer. The groundwater treatment unit using 2-stage strippers and activated carbon was already in use on the site. Cleaned groundwater was discharged into a nearby creek. It was also to be used as cooling water and to generate the steam.

All essential process data (pressure, temperature, flow rates of steam, air, soil vapour and water) were monitored online. Contaminant concentrations in the extracted soil vapour and purified off-gas were analyzed hourly using a process gas chromatograph and online monitoring.

In addition, SVE samples from all the wells were analyzed monthly. Groundwater samples were analyzed every second month.

Implementation

The thermally enhanced remediation started in July 2012 and was stopped in August 2016 after 4 years of operation. The operation of the SVE and groundwater containment continued until the end of March 2017 in order to cool down the field to an average temperature below 40°C. During a 12 months monitoring phase the operation of a low-flow SVE and the groundwater capturing will be operated to verify the remediation goal of less than 20 g/day of CHC-emission from the source zone.

In the beginning of the remediation procedure (section 1 and 2), the steam-air mixture was injected by means of four to five dual-screened wells and 300 – 500 kW to heat up the individual treatment sections. After passing the target temperatures and reaching a typical peak-shaped decrease in the contaminant mass removal (Figure 5) the next remediation section was to be selected.

During the treatment of section 2 an increase of contaminants in the soil vapour in the SVE wells of section 1 was observed. The observation of overpressure indicated a steam driven mobilization of CHC from south to north. In addition the long term heat storage in the sandstone layers might cause the evaporation and desorption process of CHC present in the matrix of an already treated section.

The steam and heat propagation was found to be wider (factor of 1.5 – 2) as predicted from the pilot study. So, the number of operating SVE wells was doubled (10 → 20) to capture the mobilized contaminants. During the following two months of operation the contaminant mass removal increased steadily indicating a long desorption time of the impregnated CHC from the bedrock. The process of desorption lasted for 5 months (see Figure 5, April - September 2013, section 2). This is 45% longer than expected.

The system was adopted since the wide heat propagation of 5 – 10 m in radius allowed the simultaneous treatment and steaming of two and in the end of three sections. The steam and heat flux (less than 500 kW) was not changed. This resulted in an energy-effective doubling of the treatment time for each section (7 months) and thus sufficient time for desorption of the contaminants. Overall, the time demand increased from 33 to 48 months of steam injection, the energy consumption increased less than 30%.

The SVE system was revised and the extraction rate was increased by 50% to cover 4 – 5 treatment sections for pneumatic control. The heat propagation was found to extend over 3 sections (Figure 4) due to heat storage and heat conductivity.

In the beginning of April 2015 the steam-air mixture (600 kg/h) was injected via 16 injection wells and approximately 600 m³/h (800 – 900 kg/h) of hot soil vapour was extracted by means of 45 wells. In total 13 m³/h of groundwater was extracted and treated.

In 2016 when reaching section 9 the number of injection wells was diminished to 8 wells according the SVE monitoring while the SVE-rate of 49 wells was optimized to obtain a maximum contaminant removal rate and ensure a pneumatic control.

Table 1: Comparison of design and effective remediation strategy

Design of remediation process (2010)	Applied remediation process (2013 – 2016)
• 9 sections • stepwise downgradient duration of 3 months for each section • 2 injection levels, 5 wells • 450 … 300 kW total power • Heating strategy: 6 weeks heating and steam propagation + 8 weeks desorption time	• 9 sections → transient simultaneously steaming of 2 – 3 sections • stepwise downgradient duration of 7 months for each section • 2 injection levels, 5 – 8 wells → 2 section • 500 …350 kW total power (maximum 600 kW) • Heating strategy: 5 weeks heating and steam propagation of claystone (sot), 200 kW + 11 – 13 weeks desorption time of claystone including heat propagation in sandstone (sos), 300 kW + 9 weeks desorption time (sos), 150 kW
→ 1 week cooling SVE phase each section	→ Seven months final cooling SVE time

After falling below the remediation goal of a maximum emission of 20 g/d of CHC via the groundwater and less than 20 mg/m³ in the soil vapour the steam injection was stopped by the end of August 2016.

During the cooling phase 26 SVE wells and 15 former injection wells in the sections 4 – 9 were operated. The SVE rate increased up to 850 kg/h. After 7 months of cooling to fall below an average temperature in the sandstone matrix of 40°C, the monitoring phase started in May 2017. The operation of the groundwater capturing is continued, the emissions are below 6 g/d CHC and the SVE will be operated at 12 wells in the “warm” sections 5 – 9 for at least 12 months.

Heat propagation

The steam propagation in each section was observed during the first 6 – 10 weeks after starting the injection (see Figure 3, section 1 and section 2) by a continuously increase of the temperature. The steam front arrived at the extraction wells four weeks after start-up.

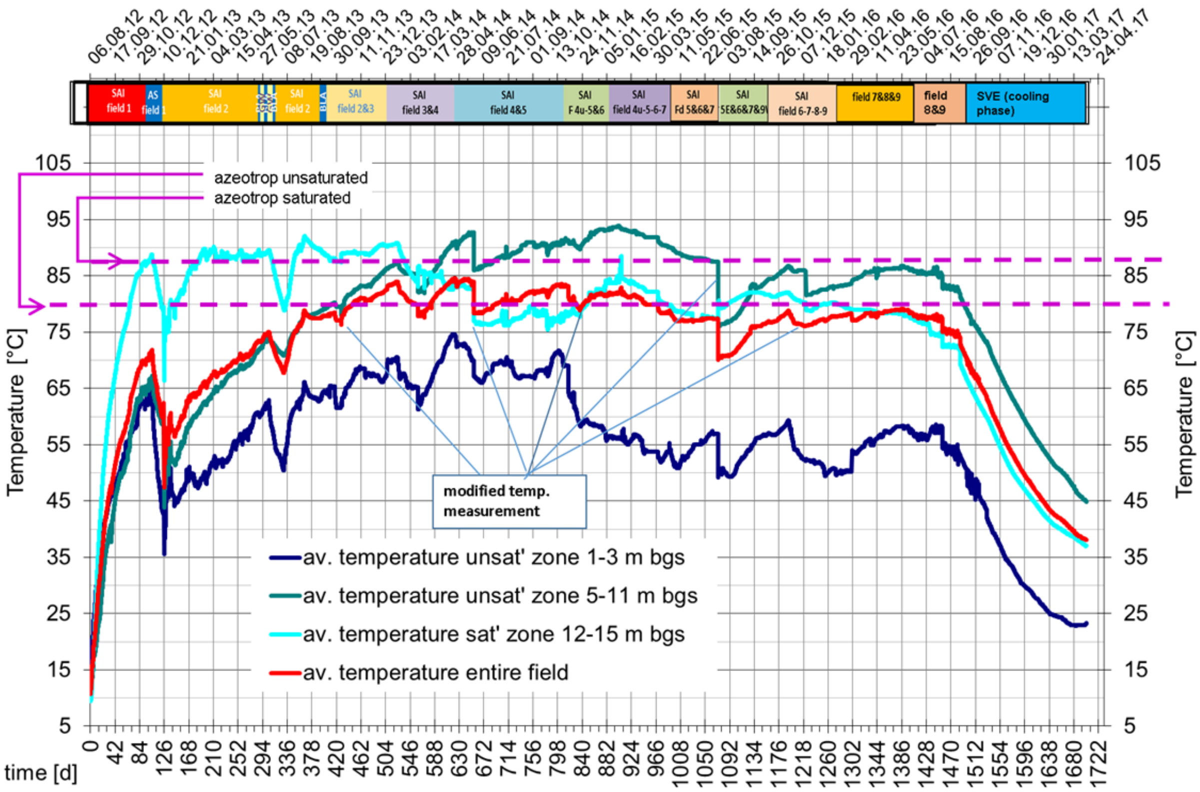

Figure 3: Temperature development during remediation process

The desired azeotropic temperature of 88 °C in the saturated zone was achieved after the heating phase. During the treatment of section 1 and 2 the average temperature in the unsaturated zone above 11 m bgs was below 65 °C. Thus the evaporation of the contaminants was slower as expected indicated by the long lasting increase of mass removal during the treatment of section 2 (Figure 5).

Changing the treatment concept led to meet the target values in both zones during the treatment of sections 2 and 3. Following the water displacement by steam and the operation of the groundwater capturing system the water table fell below 14 m bgs. The resulting target temperature of the azeotrope ranged between 80 to 82 °C for both zones. The temperature between 3 – 13 m bgs. was held above the target temperature as on 2014 the treatment of section 4 started (Figure 3, 630 days).

In August 2015 the temperature measurement system was changed to focus on the temperature distribution in all operated treatment sections. Solely the temperatures in the sandstone and claystone matrix were monitored. Thus lower temperatures were averaged and the aim of exceeding more than 80°C in both zones was realized from October 2015 until the end of steam-air injection (August 2016) while section 5 – 9 were decontaminated.

A coherent steam zone between 14 – 6 m bgs was created as visualized in 3-D heat images (Figure 4). As of 2014, the steam zone covered an area between 800 – 1,000 m² (two sections). For an estimated thickness of 7 – 8 m an extensive steam-filled cubature of 5,600 – 8,000 m³ of fractured bedrock was treated. The thermal range was extended in summer 2015, covering up to 1,500 m² meaning 12,000 – 15,000 m³ of fractured bedrock.

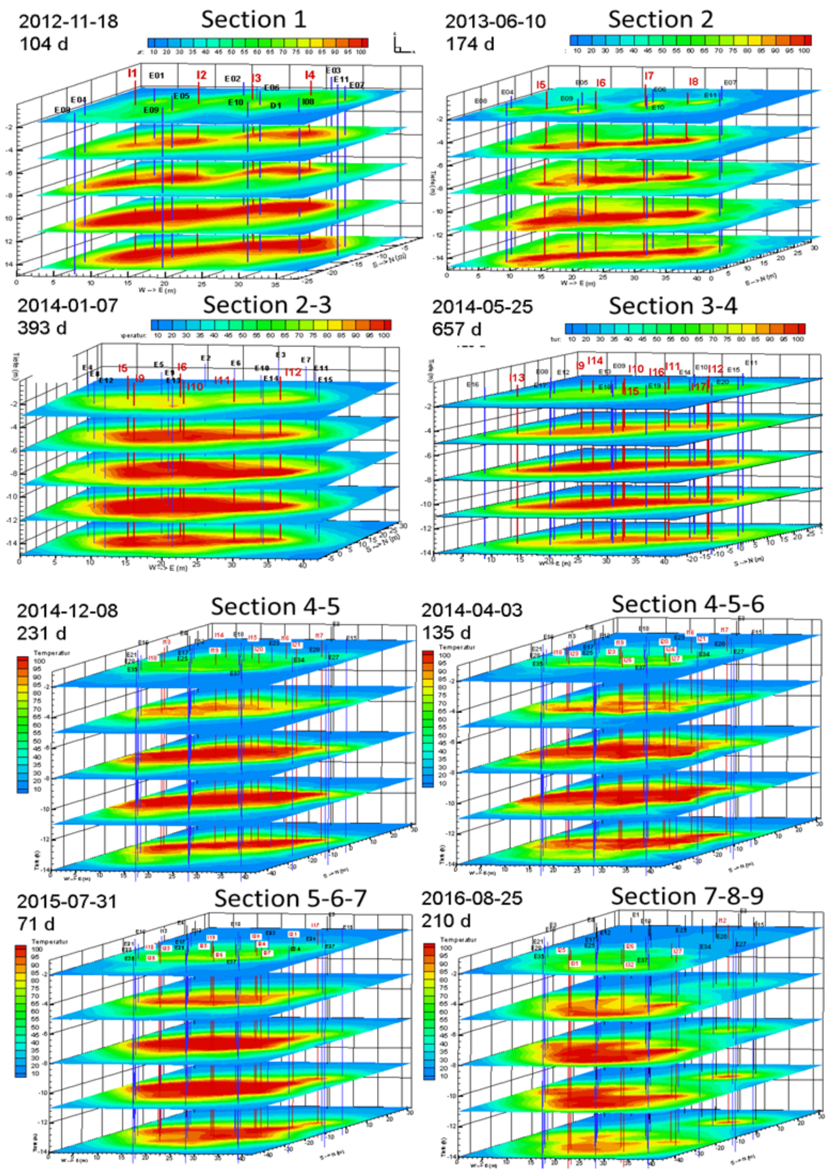

Figure 4: Heat propagation (48 months of steam injection)

Energy consumption

During 48 months of steam-air injection the power intake was 390 kW meaning an average steam-air rate of 520 kg/h. In average 700 kg/h of steam-containing hot soil vapour with an average temperature of 54°C was extracted. In total 13,800 MWh of energy were consumed. Energy losses in a dimension of 175 kW occurred due to the discharge of warm groundwater (10-13 m^3/h, maximal 22°C) and the extracted hot soil vapour after the steam breakthrough. Thus, in total 6,200 MWh of energy, or more than 44% of the energy input, were direct losses. The energy stored in the subsurface is estimated as 620 MWh for approx. 15,000 m^3 of heated soil (3 sections, area equipped with temperature sensors) with an average temperature of 80 °C as of April 2015.

Contaminant removal

The remediation process requires a steam propagation in the fractures and the direct heating of the highly contaminated surface of the sandstone close to the fractures and its fast thermal desorption. The first steam breakthrough at the extraction wells is typically accompanied by a high mass removal rate of contaminants (Figure 5). The effect is mainly and mostly visible after 1 - 2 weeks of steam-air injection in a new treatment section. During the first 8 – 10 weeks the bedrock is heated up while the contaminant mass removal increases (Figure 5, section 2, reddish area). After passing the peak of contaminant removal the desorption phase continues. The next treatment section was added to the steamed zone when the contaminant removal fell below 2 kg/d of CHC removal, meaning 100 – 150 mg CHC per m³ of soil vapour. Thus the general observed behavior was integrated in the optimized treatment concept.

As of summer 2015 the sections 5 – 9 were steamed. When the concentration in the observed wells in the northern section were falling short of 20 mg/m³ CHC in the soil vapour the correlated injection wells were stopped and the steam-air injection was focussed on the sections in the south of the source zone. In the beginning of 2016 only section 8 and 9 were steamed. To verify the remediation success the steam-air injection was operated periodically; on workingdays"on", over the weekend "off". Once the concentration at the start of the steam-air injection was below 20 mg/m³ CHC the steam–air injection was stopped in August 2016 (Figure 5).

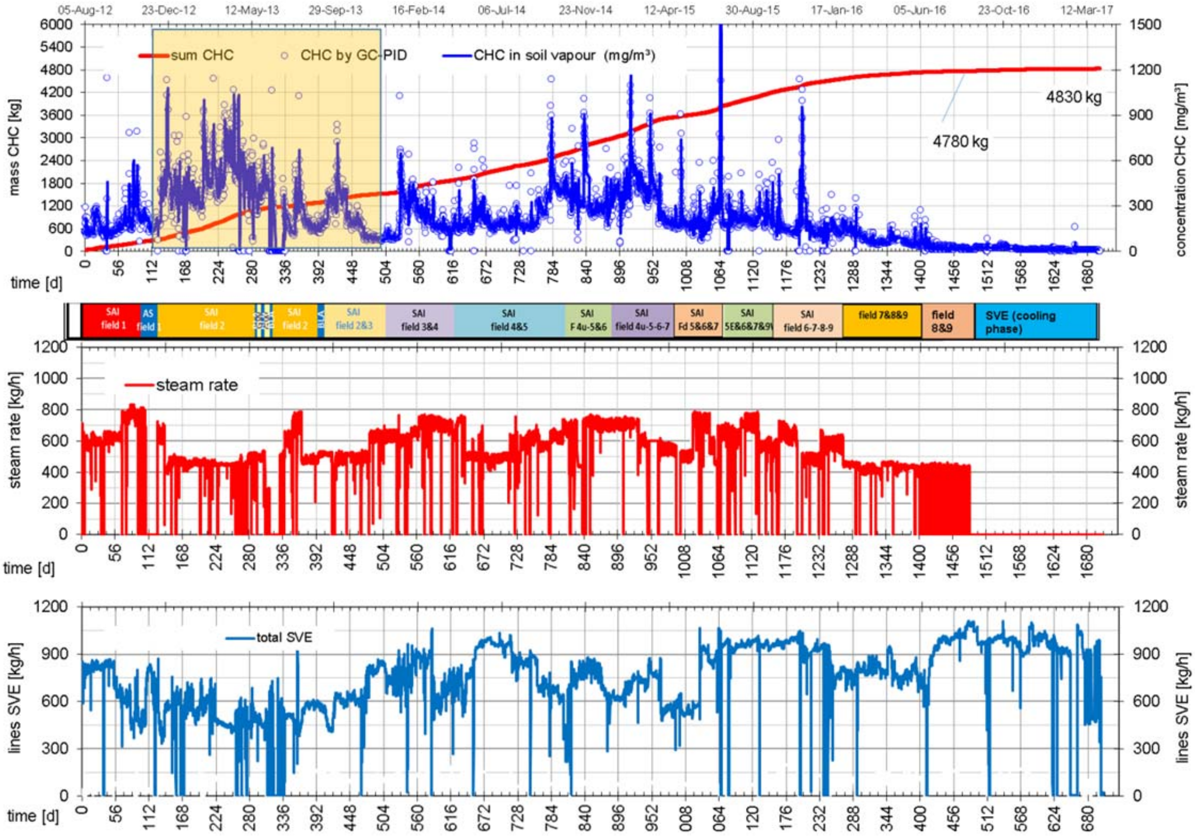

Figure 5: CHC removal rate, concentration in soil vapour and mass balance, mass flux of steam and soil vapour extraction

Until the end of August 2016 in total 4,970 kg of CHC were removed (Figure 5). Approximately 4,770 kg CHC were removed by SVE and 200 kg CHC by the groundwater containment. Including the pilot trial in 2009, in total more than 5,300 kg CHC were removed by the soil vapour extraction during the thermally enhanced remediation. The mean mass removal rate of the completed thermal remediation is approximately 3.2 kg of CHC per day. During the treatment of section 2 and section 6 the daily mass removal even exceeded 20 kg of CHC. At the end of the steam-air injection the mass removal was below 100 g/d, meaning a mass reduction by > 97%.

At the end of the cooling phase in April 2017 the concentration in the downstream plume ranges between 14 – 26 µg/L captured by the pumping wells GW10 – 13. This indicates a CHC mass flux of 5 - 6 g/d. The CHC content in the capturing wells GW 10 – 12 decreased by 99.4% from 3905 µg/L to 18 µg/L.

The goal of the remediation of less than 20 g/d of CHC emission was achieved since March 2016. The monitoring phase started in April 2017 since the fractured bedrock is still warm and a maximum of emission from the unsaturated zone of 40 – 70 g/day CHC was determined after a three weeks lasting shut down of the SVE in February 2017. The operation of the SVE and groundwater containment will last for at least another 12 months to substantially fall below the limit of 20 g/day of total CHC emission.

Contaminant distribution in the soil vapour

The remediation process was monitored by monthly analysis of the soil vapour from all SVE-wells on the site (Figure 3). The major criteria to conclude the treatment of a single section is to achieve a threshold level of less than 20 mg CHC per m³ of soil vapour. The temporal development of the spatial CHC distribution indicates the successful remediation (from red over green to blue, Figure 6) of the sections from north to south.

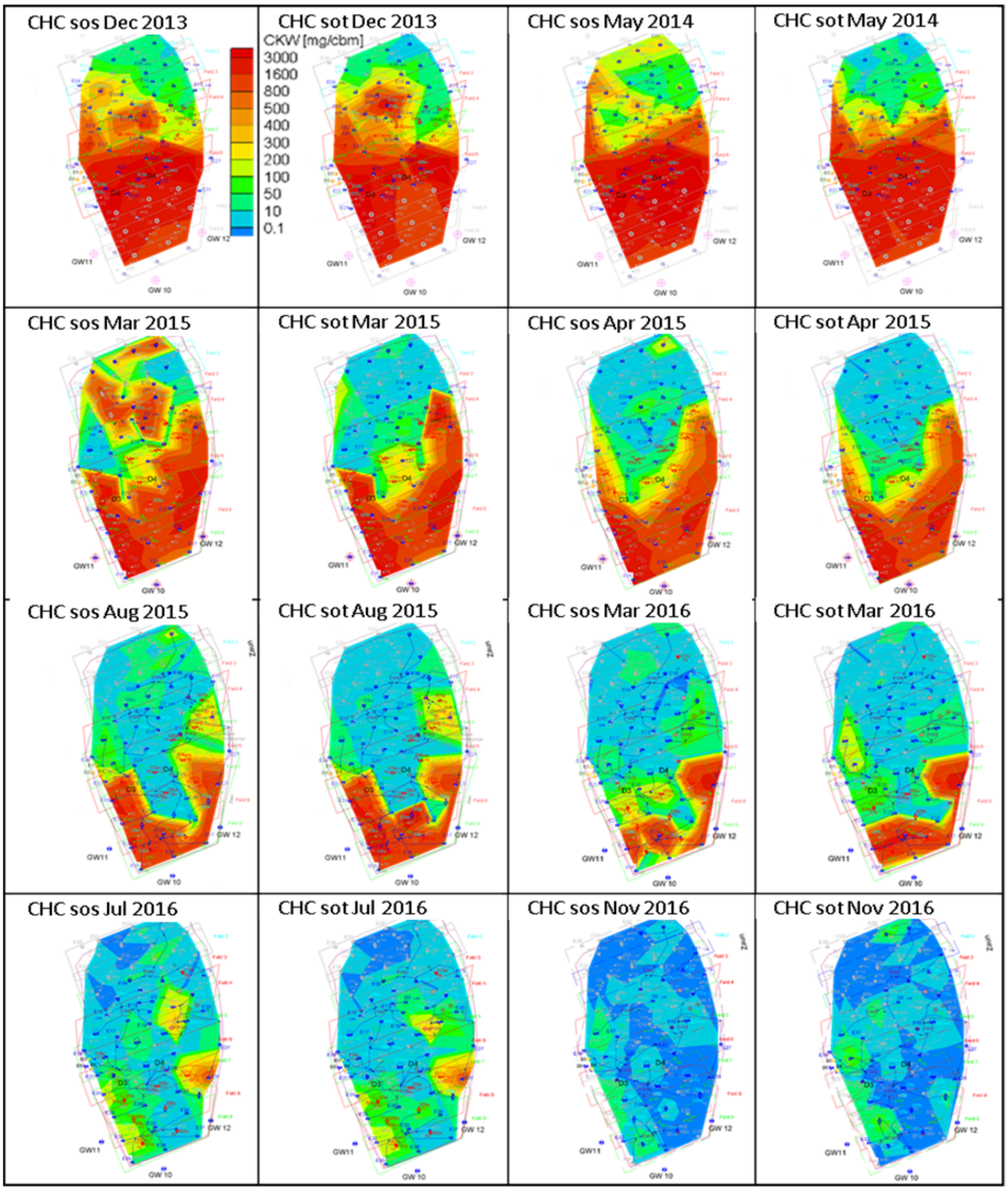

Figure 6: CHC distribution in the soil vapour since 2014 for the lower sandstone layers (sos) and the upper sand- and claystone layers (sot)

Green and blue marked areas indicate the target values of the contaminant concentration level after the treatment. The red coloured areas indicate the initial contaminant concentration of more than 800 mg/m³. During 2014 the sections 2 – 4 were treated and the remediation progress was clearly visible until the end of 2014 in both layers, the claystone and sandstone layer (sot) and the platy sandstone layer (sos), respectively. In March 2015 a significant increase in the soil vapour of the lower sandstone layers (sos) was observed in the northwest. The different concentration levels in sot and sos indicate a migration of contaminants from the lower part of the sandstone (sos) below 14 m bgs or a rebound from the compact bedrock. A slight overpressure of 20 – 50 mbar was found at some former injection wells in section 2 and 3 indicating a steam flow based mobilization of contaminants from section 6 and 7.

Thus the pneumatic control was enhanced in order to avoid any further contaminant mobilization. To capture CHC from the ongoing desorption processes the SVE system was extended to operate 40 SVE wells and increasing the SVE from the northern boundary of the treatment section.

To enhance the desorption in the southern part, the number of operated injection wells was increased to 20 wells while treating the sections 7 – 9. Nevertheless, the sections 4 – 7 had to be treated for more than 12 months bearing the most of contaminant mass. The less contaminated section 8 and 9 were remediated during 10 months of steam-air injection.

Remediation costs

The final remediation costs will be 4.4 Mio. EUR incl. VAT. The costs for the operation including the energy costs amounts to 60% (2.62 Mio EUR). About 22% (0.98 Mio EUR) were required for the remediation infrastructure like drilling and remediation process technology and 14% (0.6 Mio EUR) were used for engineering and analytical costs. The costs for the monitoring phase are estimated as 0.18 Mio EUR. In 2012 the remediation costs were estimated to be 3.5 Mio EUR. Due to the longer remediation time the costs for the thermal remediation increased by 28% from 3.3 to 4.2 Mio EUR. The costs for energy increased by 0.3 Mio EUR (19% of the planned budget of 1.9 Mio EUR). The consumables and personal costs to operate the remediation plant increased by 165.000 EUR to 1.265 Mio EUR. The costs for engineering and analytical cost increased by 110.000 EUR.

In comparison to a remediation by pump-and-treat the steam-air injection is significantly more cost-effective to remove the same amount of contaminant mass. Removing more than 5,000 kg of CHC takes 84 years using the existent pump-and-treat system. Assuming a duration of 80 years and regular yearly operational costs, the remediation costs are estimated as 7.6 Mio EUR. The thermal enhanced remediation costs 4.4 Mio EUR and lasted 5 years.

Summary

On the site of a former incineration plant for liquid organic waste (CHC, BTEX) in Villingen-Schwenningen, Germany, a long lasting contaminant leakage caused a plume covering several hectares. The source zone extended over 2,800 m². The affected fractured sandstone aquifers were contaminated down to 37 m bgs. The upper platy sandstone comprising the groundwater fluctuation zone and the unsaturated zone contained the majority of contaminant mass.

The application of a thermally enhanced remediation using steam-air injection was selected to remove chlorinated hydrocarbons from a fractured sandstone aquifer. A pilot application was conducted in 2009 to determine the effectiveness of the technology prior to designing the full scale thermally enhanced remediation scheme [5].

The full scale remediation started in 2012 to treat approximately 43,000 m³ of sandstone and claystone. The site was divided into nine treatment sections. The duration of the steam-air injection phase (steam injection power of 400 kW) was determined to last 33 months. A total of 32 two-level injection wells and 37 soil vapour extraction wells (SVE) were installed on site. The total costs of the four years running remediation was estimated as 3.5 million EUR incl. tax.

The thermal enhanced remediation process is divided in two steps. First, a heating phase when the steam-air mixture propagates in the fractures and heats up the bedrock by conduction. During this phase the easily accessible contaminant mass is evaporated. Second, a thermal desorption phase where impregnated contaminants are slowly desorbed from the sandstone and claystone matrix.

In 2013 the concept of a compartment-wise treatment of the bedrock had to be adapted. Both, the effective heating time and the duration of the desorption phase were significantly longer as indicated during the pilot trial. The time demand was increased by 45%, the energy consumption by 35%. The heat propagation of up to 10 m in radius allowed the simultaneous treatment of two sections and in 2016 that of three sections. The steam and heat flux was less than 500 kW. This resulted in an effective doubling of the treatment time of a single section to seven months duration which provides the required desorption time. In addition the SVE was revised and extended by 50% to cover 4 – 5 treatment sections. Instead of the initially intended operation of 10 SVE wells and 4 – 5 two-level injection wells up to 45 SVE wells and 10 two-level steam injection wells were under operation.

Due to the steam front propagation and the groundwater containment the initially saturated zone below 11 m bgs. was drained. Thus, the target temperature could be minimized from 88 °C to 80 °C. The temperatures in the bedrock ranged between 80 – 88 °C.

Until the end of August 2016 in total 4,970 kg of CHC were removed from the site. 4,770 kg of CHC were removed by SVE and 200 kg CHC by the groundwater containment. Including the mass removal during the pilot study (560 kg), in total more than 5,300 kg of CHC were removed by the soil vapour extraction during the thermally enhanced remediation. The mean daily mass removal was 3.2 kg of CHC with a maximum of 20 kg per day from the sections 2 and 6. At the end of the steam-injection period less than 10 mg/m³ CHC was measured in the soil vapour, down from initial values above 2,000 mg/m³.

After 48 months of steam-air injection the remediation was successfully finished. The CHC emissions in the groundwater were below 10 g/d. The latest groundwater analysis indicate a reduction of the CHC content by 99.4% from 3,905 µg/L CHC to 18 µg/L. The remediation target was an emission of 20 g/d.

After 7 months of cooling the average temperature was below 40°C, and the emission was 6 g/d. In May 2017 the monitoring phase started for at least 12 months by operating a low-flux SVE and a groundwater treatment to further cool the aquifer and control the low emission from the warm bedrock in the unsaturated zone.

Due to the irregular fractured system the duration of the desorption of the contaminants was prolonged by 35%. The remediation procedure was to be adopted and optimized several times. In total the state of Baden-Wuerttemberg and the town Villingen-Schwenningen will have spent 4.4 Mio EUR for the remediation. This is an increase of the estimated cost of 3.5 Mio EUR in 2012 by 26%.

Parties involved and responsibilities

The environmental agency of Baden-Württemberg (LUBW), the regional council (RP Freiburg) and the community of Villingen-Schwenningen support the application of a thermally enhanced remediation of the site by steam-air injection. The remediation is carried out with financial support of the community of Villingen-Schwenningen and the regional council and the State of Baden-Württemberg.

The local consultant GEOsens assisted the pilot study. GEOsens and is the leading consultant for the remediation. The scientific supervision for the pilot and the remediation is the responsibility of *VEGAS*. Bauer Umwelt GmbH is the executive remediation company.

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