ThS 1C.27 Thermal remediation 2

STEAM-AIR INJECTION IN FRACTURED BEDROCK: RESULTS AND LESSONS LEARNED 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), developed and verified in several field applications by the Research Facility for Subsurface Remediation, *VEGAS,* are proposed for 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.

A novel application of steam-air injection was the removal of chlorinated hydrocarbons from a fractured sandstone aquifer. In 2009 *VEGAS* successfully completed a pilot trial of steam-air injection for the problem owner, the city of Villingen-Schwenningen in south-west Germany [6]. The test field extended over 2,000 m³ of fractured rock including the upper of two different 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. Contaminants that had penetrated into the sandstone matrix were thermally desorbed during the conductive heating of the bedrock due to 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.

Based on the results of the pilot, 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 size 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). The costs were estimated to 2.6 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 open lid storage and incineration basins covering a total area of about 800 m². Due to leaks in the basins liquid contaminants migrated into the underlying fractured sandstone formation. The extension of the contaminant source zone is estimated to extend over ca. 2,000 m².

During a first remediation 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 CHC per m³ in the source zone and 200 mg/m³ in the surrounding area.

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 bed rock granite at 37 m bgs. Both aquifers are confined. The pressure head in the confined aquifers is approx. 5 to 8 meters above the upper confinement, corresponding to a water level in the wells 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. A total discharge of more than 12 m³/h operating 3 groundwater wells is maintained for a complete capturing of the site's emissions.

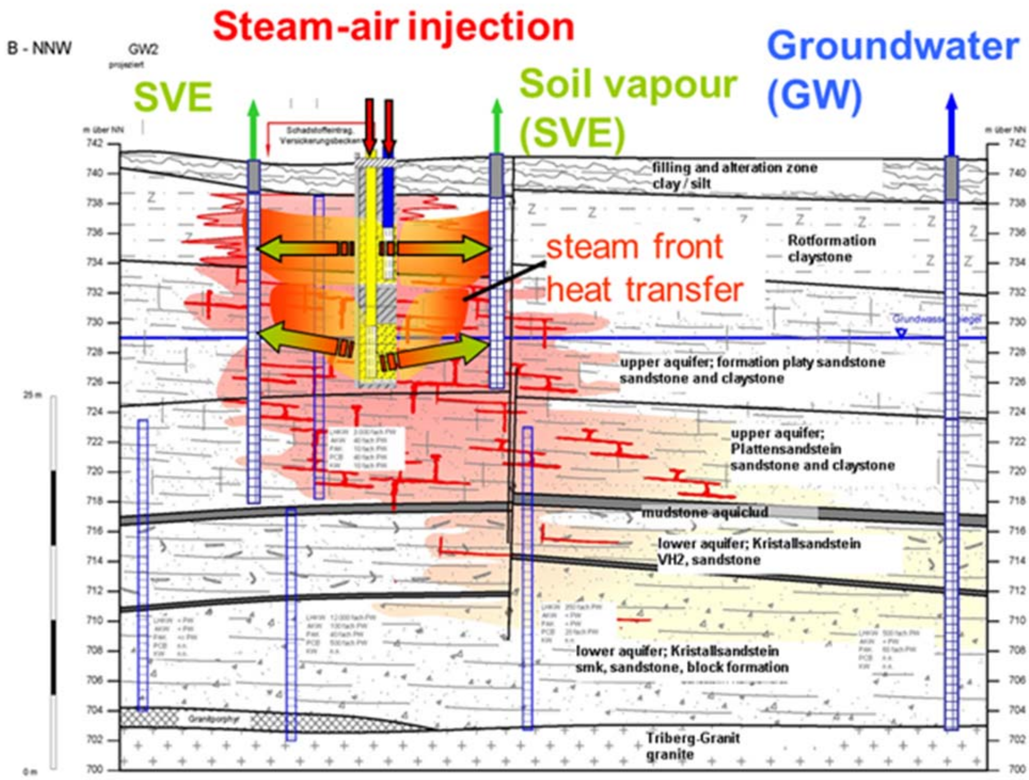


Figure 1: Soil profile and set up of injection and extraction wells

Following the heat propagation and the remediation success of the pilot study the steam-air mixture is injected on two different levels: (1) in the platy sandstone below the groundwater level (12 – 15 m bgs) and (2) in the platy sandstone and claystone of the unsaturated zone (5 – 8 m bgs.) (Figure 1). The injection levels were selected to differentiate between the depth-dependent injection pressures und temperatures.

In general the propagating steam heats the saturated and unsaturated zone by condensation, while the air component acts as an inert carrier gas and transports the vaporised 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 it was intended to inject as much steam as possible (maximal injection pressure) into the fractured system and 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 unsaturated zone. Dependent on the pressure of the water head and injection pressure it is 88°C in the saturated zone [6].

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), see fig. 2. In total there are 32 two-level injection wells, 37 soil vapour extraction wells and 3 groundwater wells downstream the contaminated area. 89 temperature measurement lances are used to monitor the heat propagation at 20 different vertical profiles simultaneously.

The initial design was to treat the site 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 total 9 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 is 10 m.

The heating power ranges between 300 and 450 kW when simultaneously operating 8 to 10 injection wells with injection pressures of 1.4 (top level) and 2.6 bar (sandstone). The soil vapour extraction rate of the corresponding 10 SVE wells operating at 150 mbar vacuum exceeds 600 m³/h.

The thermally enhanced remediation was design to last 30 to 33 months (April 2015).

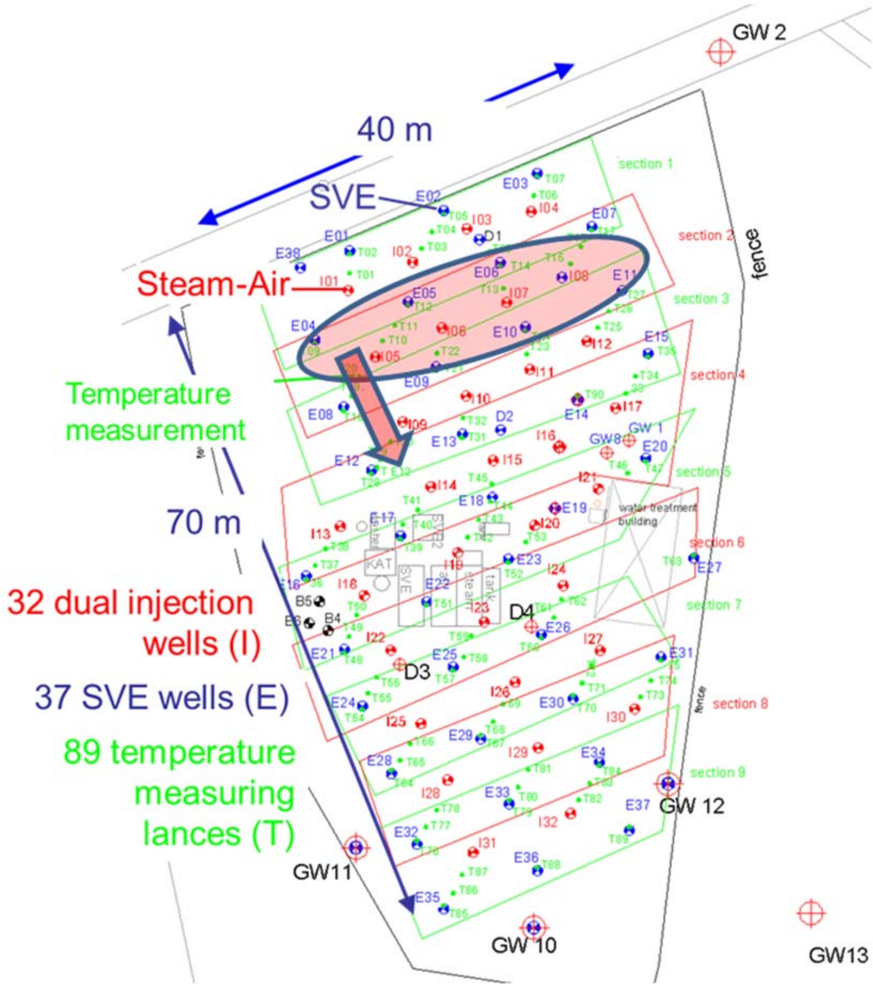


Figure 2: Map of the test site

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 is 12 – 14 m³/h to capture the overall downgradient contaminant emission.

To monitor the emission from the site a second line of observation wells (GW13 – GW16) is located 20 m downstream of the site. These wells might also be used for hydraulic containment.

Figure 3: Perspectives of the current remediation

The process equipment for the thermally enhanced remediation is delivered and operated by the remediation company Bauer Umwelt GmbH, Schrobenhausen, Germany. It includes two steam generators, 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. The groundwater is used as cooling water and process water to generate steam. Cleaned groundwater is discharged into a nearby creek.

A data acquisition system and ADSL modem for the online monitoring of essential process parameters (pressure, temperature, flow rates of steam, air, soil vapour and water etc.) is installed. Contaminant concentrations in the extracted soil vapour and purified off-gas are analysed hourly using a process gas-chromatograph.

To monitor the remediation process SVE samples from all the wells are analysed monthly. Groundwater samples are analysed every 2nd month.

Implementation

The thermally enhanced remediation started in July 2012. It is currently in full operation and the steam injection phase will last until January 2016. In the beginning of the remediation procedure (section 1 and section 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 obtaining a typical peak-shaped decrease in the contaminant mass removal (Figure 6) the next remediation section was to be selected for steam-air injection. The steam-air injection in the first section is stopped and the SVE will be maintained for at least one week. Then the next section will be treated by steam-air injection and SVE.

During the treatment of section 2 an increase of contaminants in the soil vapour in the SVE wells of section 1 which were during that time not in operation was observed. The observation of overpressure indicated steam driven mobilization of CHC from section 2 in direction of section 1. Since heat storage in the sandstone layers causes a slow cooling process (Δ T of 40 - 50 K takes 10 – 12 months) the evaporation and desorption process of CHC present in the matrix might also continue.

In addition the steam and heat propagation was found to be wider by a factor of 1.5 – 2 as predicted from the pilot study. In consequence the number of operating SVE wells was doubled 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 6, April - September 2013, section 2). In consequence the remedial operation had to be adopted.

The wide heat propagation of 5 – 10 m in radius allows the simultaneous treatment and steaming of two sections. Since the design steam and heat flux (less than 500 kW) was not changed this results in an energy-effective doubling of the treatment time for each section (now 7 months) and thus sufficient time for desorption of the contaminants. Overall this means that the time demand will be increased by 35%, the energy consumption by 25%.

In addition the SVE 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 (see Figure 5) due to heat storage and heat conductivity.

Table 1: Comparison of design and effective remediation strategy

Design of remediation process (2010)	Applied remediation process (2013 – 2015)
• 9 sections • stepwise downgradient duration 3 months 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 steaming of 2 sections simultaneously • stepwise downgradient duration 7 months each section • 2 injection levels, 5 – 8 wells → 2 section • 500 …350 kW total power • 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	→ Six months final cooling SVE time

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.

Heat propagation

The steam propagation in each section was observed and monitored during the first 6 – 8 weeks after starting the injection by a continuously increase of the temperature. The steam front arrived at the extraction wells four weeks after start-up.

The heating phase lasted 8 – 10 weeks (see Figure 4, section 1 and section 2) to reach constant temperatures above the desired azeotropic temperature of 88°C in the saturated zone. During the treatment of section 1 and 2 the average temperature in the unsaturated zone above 11 m bgs was below 65°C. Hence, the azeotropic temperature was not met. Therefore the evaporation of the contaminants was slower as indicated by the long lasting increase of mass removal during the treatment of section 2 (see Figure 6).

Changing the treatment concept led to meet the target values in both zones during the treatment of section 2&3. In consequence of the water displacement by steam and the continuous operation of the groundwater capturing system the water table fell below 14 m in the treatment zones whereas the pressure head was higher. Therefore the target temperature was defined to be between 80 to 82°C for both zones. In the beginning of 2014 the treatment of section 4 started. The average temperature between 13 – 3 m bgs. was maintained above the target temperature.

The average temperatures of the various distinct levels in the subsurface, sot and sos indicate lower temperatures due to the lower temperatures in the outer extraction wells extracting a mixture of steam and colder ambient air.

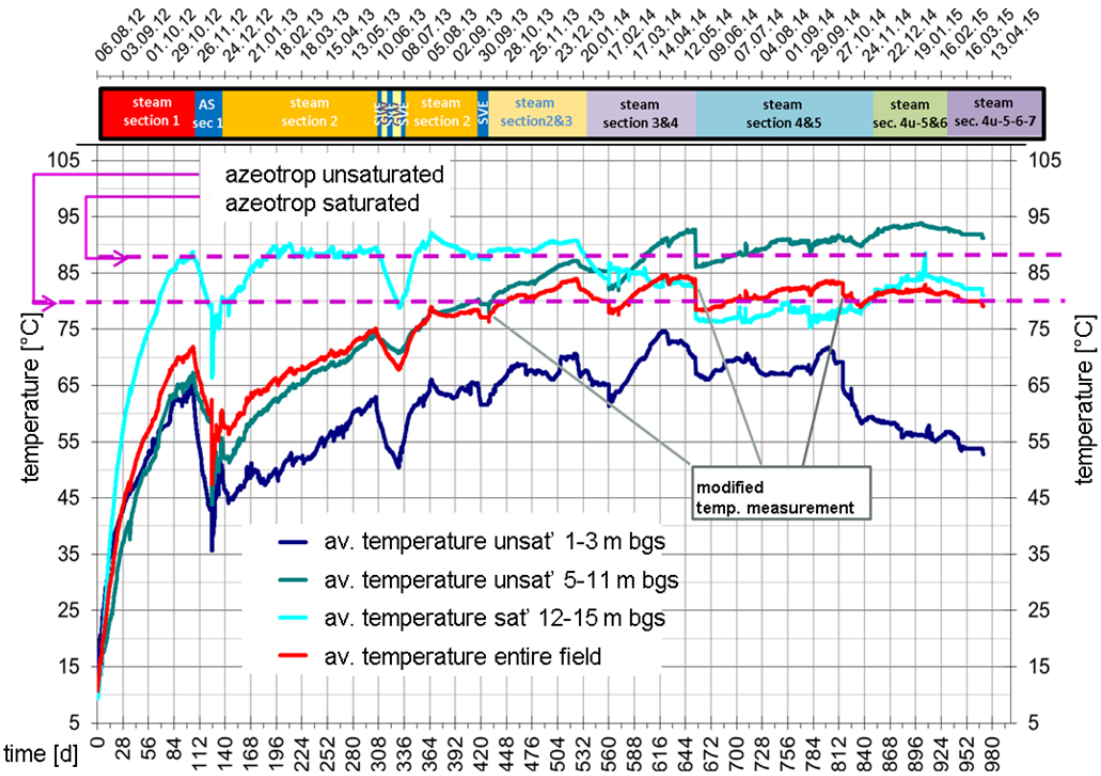


Figure 4: Temperature development during remediation process

The continuous steam zone ranging from 14 – 6 m bgs is visualized in detail in the 3-D heat images of the treated zones (Figure 5). As of 2014 the steam zone covered an area between 800 – 1,000 m² (2 - 3 sections). Its thickness was estimated to be 7 – 8 m resulting in an extensive steam-filled cubature of 5,600 – 8,000 m³ of fractured bedrock.

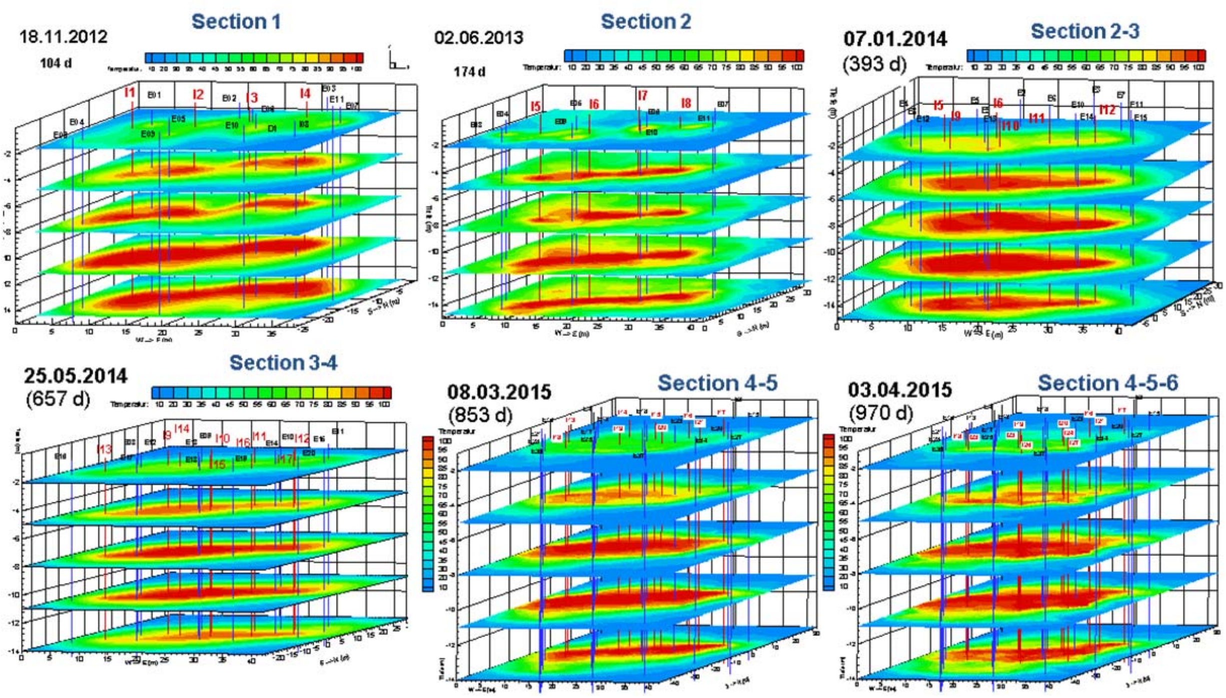


Figure 5: Heat propagation (30 months of steam injection)

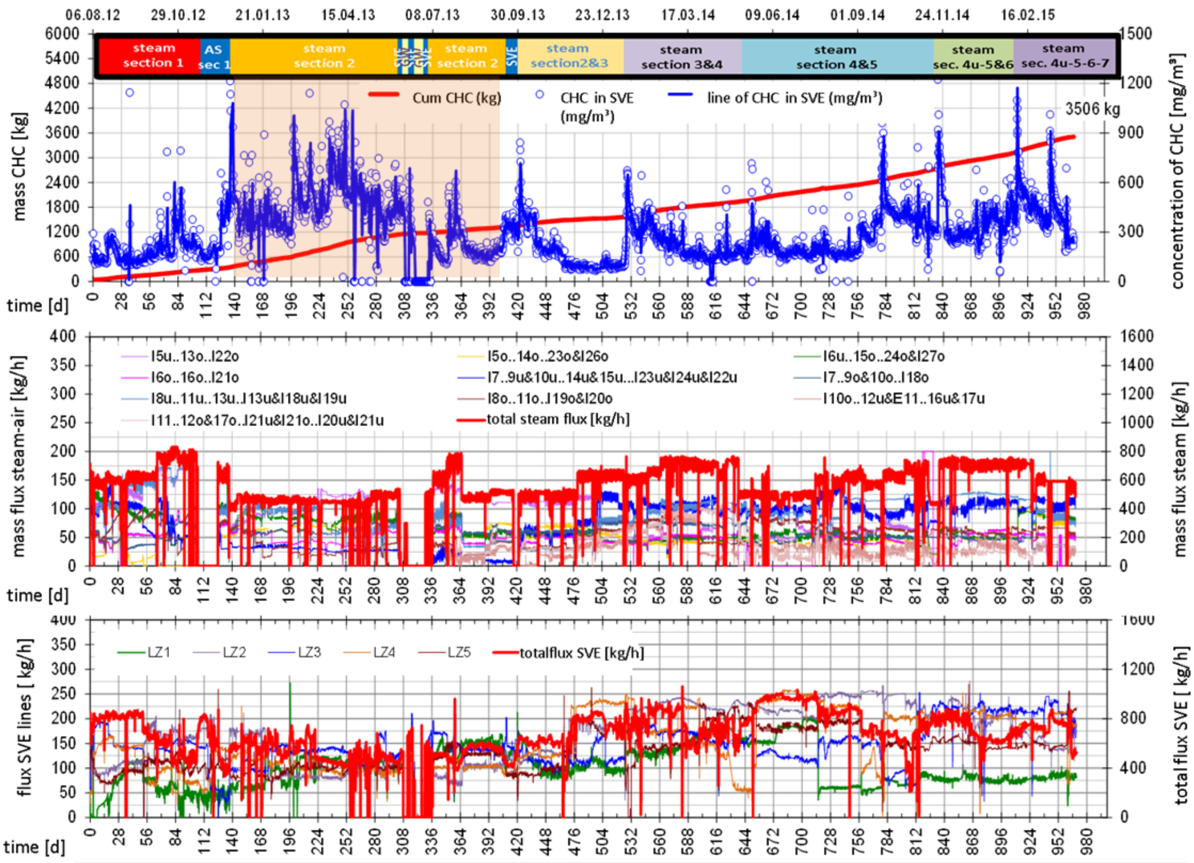
Energy consumption

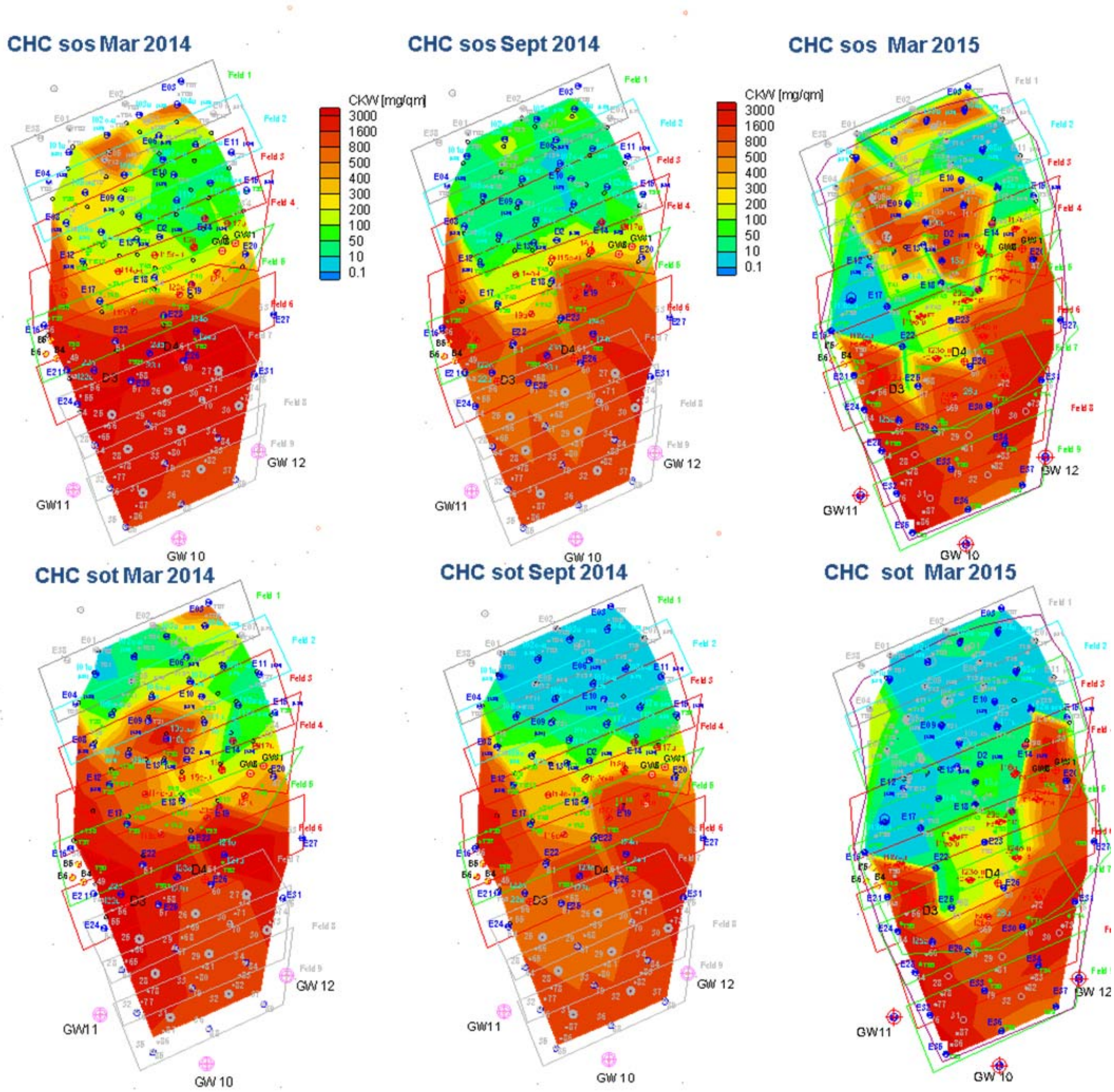
During the last 30 months of steam-air injection the power intake was 400 kW meaning an average steam-air rate of 540 kg/h. In average 670 kg/h of steam-containing hot soil vapour with an average temperature of 56°C was extracted, meaning 540 kg/h of hot air.

The steam-air mixture provided the energy to heat the subsurface (Figure 5). In total 9,300 MWh of energy were consumed. Energy losses in a dimension of 100 kW occurred due to the discharge of warm groundwater (GW10 – 13, maximal 22°C) and the extracted hot soil vapour after the steam breakthrough. Thus, in total 5,200 MWh of energy, or more than 55% of the energy input, were direct losses. The energy stored in the subsurface is estimated as 620 MWh for approx. 15,000 m³ of heated soil (3 sections, area equipped with temperature sensors) with an average temperature of 80°C in the beginning of April 2015.

Contaminant removal

The remediation process starts by the 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 6). 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 (





, section 2, redish area). Passing peak of contaminant removal the desorption phase continues. Once the contaminant extraction went below 2 kg/d of CHC removal, meaning 100 – 150 mg CHC per m³ of soil vapour the next treatment section was added to the steamed area.

Figure 6: CHC removal rate, concentration in soil vapour and mass balance, mass flux of steam, steam-air of single lines and soil vapour extraction, total and single lines

Until the end of March 2015 (simultaneous operation in sections 5-6-7) in total approximately 3,650 kg of CHC were removed (Figure 6). Approximately 3,500 kg CHC were removed by SVE and 150 kg CHC by the groundwater containment. Including the pilot trial in 2009, in total more than 4,000 kg CHC were removed by the soil vapour extraction during the thermally enhanced remediation. The mean mass removal rate of the current remediation is approximately 3.5 kg of CHC per day. During the treatment of section 2 and section 6 the daily mass removal even exceeded 20 kg of CHC.

The concentration in the downstream plume range between 100 – 300 µg/L captured by the pumping wells GW10 – 13. This indicates still a CHC mass flux of 40 - 80 g/d. The goal of the remediation is to decrease the maximum emission to less than 20 g CHC per day.

Contaminant distribution in the soil vapour

The remediation process is monitored by monthly analysis of the soil vapour from all SVE-wells on the site. 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 to to blue, Figure 7) of the sections from north to south

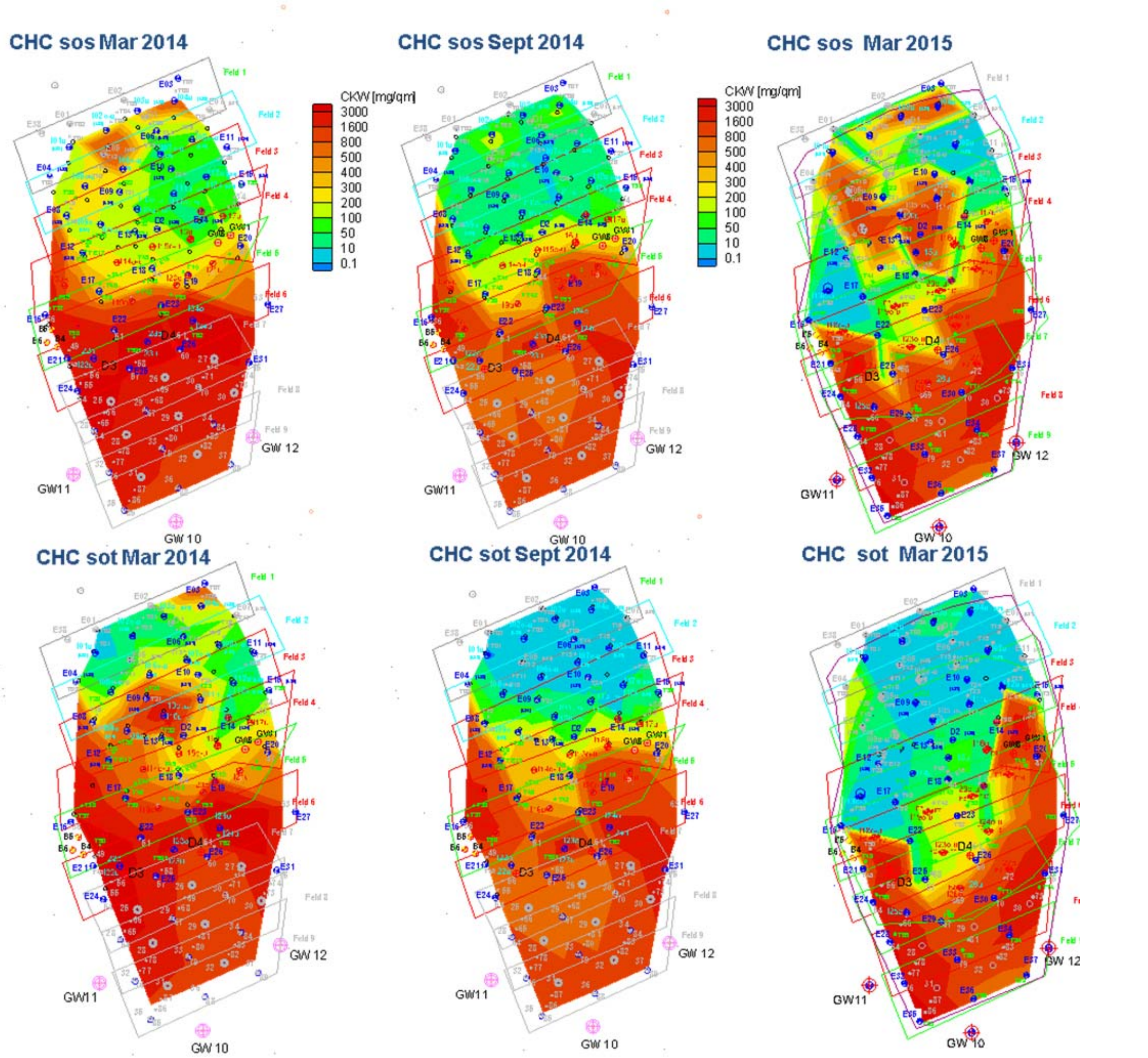


Figure 7: CHC distribution in the soil vapour since 2014 for the lower sandstone layers (sos) and the upper sand- and claystone layers (sot)

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 the last year of operation the sections 2 – 4 were treated and the steam-air injection stopped. To the eastern part of section 5 steam is still being applied. The remediation progress was clearly visible until end of 2014 in both layers, the claysone and sandstone layer (sot) and platy sandstone layer (sos), respectively.

In the beginning of 2015 during the thermal treatment of the sections 5 – 7 the CHC content in the spoil vapour of the lower sandstone layers (sos) increased by one or two orders of magnitude. This effect was not observed in the upper sand- and claystone layers. The different concentration level 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 couple of wells in section 3 still have temperatures above 70°C and elevated values of CHC are observed. In section 2 the temperatures are ranging between 40 – 50°C. A slight overpressure of 20 – 50 mbar was found at some former injection wells in section 2 and 3. So, ongoing evaporation processes and a steam flow enhanced mobilisation of contaminants from section 6 and 7 may cause the increase, too.

Currently, investigations are under way to find the reason of the rebound and migration effects during June 2015. For this a negative pressure barrier between section 3 and 2 will be created by turning off the steam injection for one week and monitoring and analysing the resulting CHC distribution.

To avoid further contaminant mobilisation and to capture CHC from ongoing desorption processes the SVE system will be extended to operate 40 SVE wells. To enhance the desorption in the southern part, in section 6-7-8-9 the number of operated injection wells will be increased to 20 wells.

Summary and outlook

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

The application of a thermally enhanced remediation using steam-air injection was a new approach 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 [6]. The steam and heat propagation extended to 5 m in radius from the injection well. During three months of operation approximately 560 kg of CHC were removed from about 2,900 m³ of fractured sandstone. A mass removal of more than 90% was indicated from the development of contaminant concentration in the soil vapour.

After a phase of planning, request for tenders and commissioning in summer 2012 the full scale remediation started to treat approximately 43,000 m³ of sandstone and claystone. The site is divided into nine treatment sections. The duration of the steam-air injection phase (steam injection power of 400 kW) was calculated 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 will be 4 million EUR incl. tax

The thermally 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.

During the remediation of the northern sections 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 are significantly longer as indicated during the pilot trial. The time demand was increased by 35%, the energy consumption by 25%. The heat propagation of up to 10 m in radius allows the simultaneous treatment of two sections at one time. The steam and heat flux is less than 500 kW. This results in an effective doubling of the treatment time of a single section to now 7 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 the initially intended operation of 10 SVE wells and 4 – 5 two-level injection wells currently 40 SVE wells and 10 two-level steam injection wells are under operation.

Due to the steam front propagation and the continuously operating groundwater containment the initially saturated zone below 11 m bgs. is being (partially) drained. Thus, the target temperature, the azeotropic temperature of the contaminant mixture and steam, could be reduced from 88°C to 80°C. The temperatures in the bedrock are controlled to range between 80 – 88°C. During the treatment of two sections approximately 15,000 m³ of sandstone are heated up to 80°C in average, storing 620 MWh of thermal energy. In total 9,300 MWh of energy were consumed during the so far 33 months of steam-air injection.

Until end of March 2015 in total approximately 3,650 kg of CHC were removed from the site after treatment of approximately 60 – 70% of the cubature. 3,500 kg of CHC were removed by SVE and 150 kg CHC by the groundwater containment. Including the mass removal during the pilot study, in total more than 4,000 kg of CHC were removed by the soil vapour extraction during the thermally enhanced remediation The mean daily mass removal is 3.5 kg of CHC. Maximum values of 20 kg per day were achieved during the heat and steam propagation phase in sections 2 and 6.

Transport and displacement of evaporated contaminants in the fractures was observed. The monthly depths-specific monitoring of the SVE-wells indicates the removal of the contaminants from the upper sandstone and the claystone layers between 3 – 8 m bgs. Currently CHC contents in the soil air are less than 20 mg/m³, down from initial values above 2,000 mg/m³. In some northern parts of the lower platy sandstone CHC contents of more than 600 mg/m³ were detected almost one year after the completion of the thermal treatment in these sections. In these areas the bedrock temperatures are still in a range of 40 – 70°C.

In order to decide whether this is due to a rebound from the bedrock matrix or a mobilisation of contaminants from the currently treated zones in the southern field, throughout the fracture a field a “vacuum fence” to differentiate between the different section is currently in operation.

After 33 months of steam-air injection the effectiveness and applicability of the technology to remediate a CHC contaminated fractured sandstone and claystone was proven. 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, affecting both, cost and time. It is foreseen to finish the steam-air injection in January 2016 after 42 months of operation.

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 pilot project was carried out with financial support of the Helmholtz Centre for Environmental Research, UFZ, Leipzig, the community of Villingen-Schwenningen and the regional council and the State of Baden-Württemberg.

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

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