Centre of Competence for Soil, Groundwater and Site Revitalisation - TASK



Guidelines

In situ thermal treatment (ISTT) for source zone remediation of soil and groundwater



Funded by:

Federal Ministry of Education and Research

Helmholtz Centre for Environmental Research - UFZ





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Last edited: May 2013









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- Pilot application: Steam-air injection into a former benzene plant (after soil excavation) in the saturated and unsaturated zone
- Removing CHC contamination from the unsaturated zone using thermal wells (THERIS method)
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- Pilot application: RF treatment to support SVE at a former hydrogenation plant in Zeitz (Saxony)
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Abbreviations

AR	aromatic hydrocarbons
BTEX	benzene, toluene, ethylbenzene, xylene
CatOx	catalytic oxidation
CCl ₄	tetrachloromethane
CHC	chlorinated hydrocarbons
DCA	dichloroethane
DCB	dichlorobenzene
DCE	dichloroethene
DNAPL	dense non-aqueous phase liquid (density > 1000 kg/m³)
GWM	groundwater measuring point
HF	high frequency
ISTT	in situ thermal treatment
LEL	lower explosion limit
LNAPL	light non-aqueous phase liquid (density < 1000 kg/m³)
m bgs	meter below ground surface
NAPL	non-aqueous phase liquid
PAH	polycyclic aromatic hydrocarbons
PCA, PCEa	tetrachloroethane
PCE	tetrachloroethene = perchloroethene = perchloroethylene
PLF	power line frequency
PRO	petroleum range organics
RF	radio frequency
SVE	soil vapour extraction
SATP	standard ambient temperature and pressure
SZ	saturated zone (groundwater or aquitard)
TCA	trichloroethane
TCE	trichloroethene = trichloroethylene
TUBA [®] -method	in situ thermal remediation using steam(-air) injection
THERIS [®] -method	in situ thermal remediation using thermal wells
UEL	upper explosion limit
UZ	unsaturated zone
VC	vinyl chloride
VP	vacuum pump

Nomenclature

Symbol	Meaning	Unit
C _{gas}	substance concentration in air	[g/m ³ or vol. %]
Cw	substance concentration in water	[µg/l]
i	groundwater gradient	[-]
k _f	hydraulic conductivity	[m/s]
р	partial pressure (air)	[mbar]
p _d	steam pressure	[mbar]
V _f	Darcy velocity	[m/d]
ρ	density	[kg/m³]

Foreword

Since 1995, the German Federal Ministry of Education and Research (BMBF) has been promoting numerous practice-based joint and single projects in the areas of contaminated site management, conservation of soil and groundwater, and sustainable land revitalisation. Despite their high quality, practical suitability and potential for innovation, many of the developed methods have not been able to establish themselves on the market for various reasons. Potential users often lack the knowledge to sufficiently appreciate the advantages of these innovative products.

In August 2007, the "Centre of Competence for Soil, Groundwater and Site Revitalisation Leipzig" (TASK) was founded with the aim of dealing with such problems. The objective of the project, funded by the BMBF and the Helmholtz-Centre for Environmental Research – UFZ, is to promote and spread innovations in the area of investigation and remediation of contaminated sites. TASK is supported by a network of experts from engineering technology and administrative bodies. Associated measures have been developed and implemented at TASK to improve the transfer of technology and knowledge from research into practice.

The present guidelines "*In situ* thermal treatment (ISTT) for source zone remediation of soil and groundwater" funded by TASK, follow on from this idea. The guidelines are intended to act as a support to successfully perform *in situ* thermal treatments. The specific planning and implementation steps in the different project phases are illustrated for users, principals and authorities in an easy-to-understand, practice-based manner. The guidelines are also intended to assist in estimating and evaluating the site-specific remediation success of ISTT right from an early stage of planning. Furthermore, possibilities to compensate for investigatory and forecast insecurities by adjusting the overall design are demonstrated, and advice is given for the monitoring of the remediation operations and the evaluation of the remediation success.

The team of authors aims to provide a booklet of general guidelines which is intended to make the application of ISTT as easy as possible and thus promote this technique on an international level. Thus, the present guidelines have a special focus on Latin American specific site conditions illustrated by the example of Brazil since ISTT might represent a line of technology that is interesting for the Latin American market.

Prof. Dr. Georg Teutsch Scientific Managing Director of the Helmholtz Centre for Environmental Research – UFZ

Acknowledgements

We would like to thank Prof. Thilo Hofmann, University of Vienna, Department of Environmental Geosciences, Dr. Thomas Held, ARCADIS Deutschland GmbH, Michael Woisnitza, HIM GmbH, Dr. Karsten Menschner, CDM Consult GmbH, Quentin Hulm, Cornelsen Ltd. as well as altlastenforum Baden-Württemberg e. V. for their support and helpful comments.

1. Introduction, techniques, objectives

The remediation of source zones in soil and groundwater attains a high level of importance for the brownfield re-development of contaminated industrial sites. *In situ* remediation can make an important contribution to construction in existing contexts, insofar as it is integrated into the overall project planning with reliable deadlines and cost figures. Irrespective of the distribution of contaminants¹ at a site, schematically illustrated in Figure 1.1, *in situ* remediation must frequently lead to remediation success in source zone remediation within a given timeframe – often a period of only a few months.



Figure 1.1: Schematic representation of a possible distribution of contaminants in the sub-surface. *In situ* thermal treatments (ISTT) are focussed on the removal of highly contaminated source zones rather than on the removal of low concentrations in the outer fringe or groundwater plume.

Conventional *in situ* remediation techniques for organic contaminants such as conventional "cold" soil vapour extraction (SVE) or hydraulic groundwater control like "pump & treat" often require several years or decades to clean-up a plot of land due to the

¹ The processes of contaminant migration of LNAPL and DNAPL are described in detail in the literature. Therefore, these guidelines will not focus on these processes. Figure 1.1 illustrates the contaminant migration for two scenarios schematically.

diffusion-limited release of contaminants. *In situ* thermal treatment (ISTT) can provide effective solutions as it allows for the fast decontamination of source zones. Limitations arising from the characteristics of volatile contaminants (e. g. diffusion limitation) and substrates (e. g. inhomogeneity in the soil structure) can be overcome with ISTT by heating the entire remediation volume. Thus, the contaminants vaporise significantly faster, even in areas of low permeability.

After the first scientifically accompanied pilot projects, an increasing number of projects and implementations was carried out during the last decade as part of source zone remediation and brownfield redevelopment projects both in open spaces and beneath buildings [HIESTER & SCHRENK 2008].

The following text describes three types of *in situ* thermal treatments for an efficient source zone remediation: steam-air injection (TUBA® method), thermal wells (THERIS[®] method) and radio frequency (RF) energy. The described ISTT methods focus on the removal of source zone contaminations by vaporising the contaminants (NAPL = non-aqueous phase liquid) in the source zone through sub-surface heating and the subsequent extraction of the gas mixture from the sub-surface by soil vapour extraction. It is normally necessary to heat the sub-surface to 50 to 100°C (Figure 1.2). Contaminants with a density both smaller and higher than water (LNAPL², DNAPL³) can be remediated. Organic contaminants such as chlorinated or halogenated hydrocarbons (CHC, CVOC), benzene, toluene, ethylbenzene and xylene (BTEX) or petroleum-derived hydrocarbons up to a boiling point of approx. 200°C can be removed by ISTT [HIESTER 2009]. The recovered, contaminated soil vapour is treated by air purification systems. Standard air purification systems are, for instance, activated carbon filters or catalytic oxidation (CatOx). The described source-zone remediation projects have been executed in coarse-grained unconsolidated soil, cohesive or heterogeneous soil layers or fractured bedrock. Successful remediation has been achieved in the unsaturated as well as in the saturated zone (aquifer and aquitard).

At sub-surface temperatures above 120°C, processes take effect enabling contaminants to be chemically transformed. Examples are gasification and hydrolysis (above approx. 120°C), steam cracking in the case of benzene derivatives and linear alkanes (above approx. 180°C), torrefaction (in the case of high carbon content) and anaerobic pyrolysis (above approx. 250°C), aerobic cracking or pyrolysis (above approx. 500°C) or sintering (above approx. 1,000°C). Since such temperature ranges are not achieved by the application of ISTT which is described herein, or since the contaminants described vaporise at lower temperatures, these processes are not relevant for a successful application of ISTT.

² light non-aqueous phase liquid ($\rho_{pollutant} < \rho_{water}$)

³ dense non-aqueous phase liquid ($\rho_{\text{pollutant}} > \rho_{\text{water}}$)



Figure 1.2: Application ranges of sub-surface heating for remediation and fields of application of these guidelines (significant process temperatures between 50°C and 100°C at atmospheric pressure)

The fields of application of sub-surface heating, including its use in combination with other remediation methods, is constantly expanding. Besides the ISTT methods presented herein, there are further techniques for sub-surface heating. These techniques include, for example, the injection of warm or hot air (e. g. ProAir technique, ThermoAir technique), the injection of warm or hot water, electrical resistance heating (ERH), sintering at temperatures of 1,600–2,000°C), the Thermopile[©] technique or thermally enhanced liquid-phase recovery. These applications are only mentioned in these guidelines, since the scope of these guidelines would otherwise be exceeded significantly.

2. Background

2.1. Sub-surface heat input

2.1.1. Convective heat input: steam (-air) injection (TUBA[®] method)

To heat up the sub-surface, preferably a mixture of steam and air is injected into the unsaturated or saturated zone of the contamination source or in its fringe. Less common is the injection of saturated steam (dried saturated water vapour). The convective heat input is dominated by the flow and expansion of the steam portion, while the air portion accelerates and ensures the contaminant recovery through soil vapour extraction.

In the unsaturated zone with unconsolidated soil and finely fractured bedrock with good to moderate permeability (k_f -values) in the range of 10^{-2} to 5 x 10^{-5} m/s (gravel, sand, coarse silty soils), the injected steam condenses in the cold soil matrix, and releases its energy (enthalpy of vaporisation) to the soil matrix. On account of the on-going condensation process (until the soil has reached the temperature of the steam), the steam propagates from the point of injection to the steam front. According to the amount of injected steam, a smaller or larger vertical heat front is formed which ideally (at homogeneous sub-surface conditions) expands from the point of injection in a horizontal way and in radial symmetry. In the heated area, the (often) residual, volatile to moderately volatile organic contaminants are vaporised. The air portion supports the contaminant transport towards the extraction wells of the thermally enhanced soil vapour extraction.



Figure 2.1: Steam-air injection into the unsaturated and saturated zone

When the steam-air mixture is injected into unconsolidated soil aquifers (i. e. in the saturated zone) with a hydraulic permeability in the range of 5×10^{-4} to 5×10^{-5} m/s (gravelly sands to silty sand), the steam expands around the injection point (ideally, in radial symmetry) forming a steam-saturated zone. The size of this zone depends on the rate of injected steam and the permeability of the sub-surface. The existing groundwater is

partially displaced in this process. The area to be remediated is heated by the ongoing condensation process. Although the steam is usually injected through vertical wells laterally into the aquifer, the steam also expands vertically due to its lower density compared to water. The frictional forces at high injection rates and the anisotropy due to the natural stratification in the porous aquifer (the vertical permeability is mostly lower than the horizontal permeability) act against the buoyant force. Accordingly, a more horizontal steam expansion is achieved and thus the achievable radius of influence is augmented. After the penetration of steam into the unsaturated zone, there is no more significant horizontal expansion in the saturated zone. The air injected with the steam flows as a carrier gas from the point of injection to the steam front, integrates the gaseous contaminants, and transports these contaminants from the aquifer upwards into the unsaturated zone. A relatively high proportion of air (10–20 mass %) should be considered in order to generate a secure transport of the vaporised contaminants from the steam-saturated zone through the saturated zone towards the soil vapour extraction wells. Then, the gaseous contaminants are removed by soil vapour extraction.

2.1.2. Conductive heat input: thermal wells (THERIS method)

Thermal wells in the shape of electrically operated heating wells can be operated at temperatures of several hundred degrees Celsius and enable the heating of soil stratifications of low permeability by thermal conduction and heat-induced circulation processes (heatpipe, see chap. 2.2.2). The conductive heat input does not rely on a heat-carrying medium, in contrast to steam injection. The heterogeneity of a sub-surface is only of minor importance for the conductive heating of a sub-surface since the thermal conductivity of different soil stratifications only varies moderately [HIESTER 2009]. Nevertheless, the vaporised contaminants must always be intercepted by soil vapour extraction. Depending on the site-specific location of the source zone as well as the (hydro-) geological situation, it may be appropriate and particularly efficient to arrange the heating elements and soil vapour extraction wells at different depths in the subsurface. [HIESTER 2009].

The positioning and operating conditions of heating wells and soil air extraction wells may vary significantly depending on the site conditions and remediation goals. For alternating strata of cohesive and non-cohesive stratifications in the unsaturated zone, it has been demonstrated that it may suffice to heat the cohesive stratifications only and to apply soil vapour extraction solely to the non-cohesive stratifications in order to achieve an efficient remediation of the entire soil profile [HIESTER 2009]. Also in the case of cohesive stratification with vacuum pumps can be efficient [HIESTER ET AL. 2004, HIESTER 2009]. When installing heating elements in the saturated zone, sealing measures for protecting the electrical installations against water infiltration must be implemented.

In principle, thermal wells enable the sub-surface to be dried completely and be heated to temperatures > 100°C. However, in the fields of applications described in these guidelines, this effect is reached by the THERIS method only after a long heating period and only close to the heating wells (approx. one to two decimetres around the thermal wells). The scope of these guidelines does not cover special applications in which the soil between the thermal wells is completely dried and heated to temperatures significantly exceeding 100°C.



Figure 2.2: Schematic diagram of the THERIS method (heating with thermal wells)

2.1.3. Dielectric heat input: radio frequency (RF) energy

With radio frequency (RF) energy, the sub-surface (unsaturated and / or saturated zone) is heated by electromagnetic waves, similar to the operating principle of a microwave oven for heating food. Low MHz frequencies (3 to 50 MHz, RF range) are normally used. Dielectric heating on a volume basis does not rely on the availability of water. In comparison to microwaves, the penetration depths of the electromagnetic waves into the medium to be heated are significantly greater and within the technically relevant range of a few metres. Various soils (dry or wet, sandy or silty) can be heated by RF soil heating. In the soil, radio frequency (RF) energy is normally transformed into heat with an efficiency of more than 90 % [ROLAND ET AL. 2007a; ROLAND ET AL. 2008a].

The RF energy is generated in an RF generator and transferred by an electronic matchbox to the electrode system in the soil (Figure 2.3). The electronic adjustment maximises the energy input due to the fact that the electrical output returning to the generator is offset to zero. Rod electrodes, plate electrodes or gauze electrodes are used as electrode systems. Rod electrodes that can be simultaneously used as soil vapour extraction wells have been established for thermally enhanced *in situ* remediation projects [ROLAND ET AL. 2007a; ROLAND ET AL. 2008c; ROLAND ET AL. 2007b]. A specific design enables the energy to be transferred to selected depths in order to remediate the contamination in a defined manner (Figure 2.3). Parallel plate or gauze electrodes are suitable for improving microbiological processes as these types of electrodes enable a moderate increase in temperature at low gradients. In principle, other types of antenna geometry are also available if a very selective heating is desired [ROLAND ET AL. 2007b; KASEVICH, R.S., 1998]. In this case, the RF energy is introduced into the soil with the aid of a coaxial cable via the cable's unshielded area.

As it is the case with steam-air injection or the operation of thermal wells, extracted, polluted soil vapour can be purified either by activated carbon adsorption, or by thermal or thermal-catalytic oxidation. *In situ* oxidation using catalysts directly placed within the electrode (acting simultaneously as extraction well) is also possible as part of the RF method under certain conditions (sufficiently high concentrations of hydrocarbons in waste air) [utility model DE 202007014507.1, 2007].



Figure 2.3: Schematic diagram of the RF sub-surface heating

2.2. Influence of important processes and site conditions on remediation operations

2.2.1. Phase transitions (liquid – gas – liquid)

In the case of *in situ* thermal treatment, the processes of phase transition between the liquid and gas phase are in many cases more important than the substance properties of the liquid contaminant. Substances with a vapour pressure of > 70 Pa at standard ambient temperature and pressure (SATP) are considered to be volatile [US-EPA 1991]. In an ideal two-phase liquid-air system, the phase transition can be described by the vapour pressure p_v which increases exponentially with the temperature T. For example, vapour pressure can be calculated for pure substances by using the ANTOINE or WAGNER equations. The substance-specific coefficients (A, B, C) can be found in REID ET AL. [1987], for example. When a contaminated, wet soil is heated, the NAPL is vaporised together with water (steam distillation). The vapour pressures (p_v) of the two immiscible phases "water" (index W) and "contaminant" (index NAPL) are combined in the process [Equation 1] to form the co-boiling vapour pressure p_d . The co-boiling vapour pressure always exceeds the vapour pressure of the low boiling phase (often water). The co-boiling

temperature (azeotropic temperature) therefore represents the optimum of the substance transfer from the NAPL to the gas phase.

Equation 1: $p_v = \exp(A_w - B_w / (C_w + T)) + \exp(A_{NAPL} - B_{NAPL} / (C_{NAPL} + T))$

During the steam distillation of the components water and contaminant, the pressuredependent boiling temperature of the mixture in the open system (in this case: soilstructure pores) is always lower than the boiling temperatures of the individual substances. As an example, the vapour pressure curves of PCE (tetrachloroethene) and water, in addition to the vapour pressure curve for co-boiling (PCE with water), are illustrated in Figure 2.4.



Figure 2.4: Vapour pressure curves of water and PCE a) Vapour pressure curve of the single substances (H20, PCE) and the mixture (H20 + PCE) at co-boiling (azeotrop)

b) Determination of the co-boiling temperature of PCE and water according to the method of Badger-McCabe [Betz 1998]

To illustrate this process, it is possible to express the relationship of the vapour pressure of the water-contaminant mixture to the water-vapour pressure at normal pressure and on a temperature-dependent basis in form of the NAPL pressure coefficient (Table 1) [HIESTER 2009]. For contaminants with a boiling point of approx. 200°C, the NAPL pressure coefficient is approx. 1 since the co-boiling only leads to slight decreases of the water-vapour pressure. Consequently, the process of steam distillation for contaminants with boiling points > 200°C is of minor importance.

Within the radius of influence of a soil vapour extraction operated with several hundred millibars of vacuum, the co-boiling temperature can be reduced for CHC or BTEX contaminations by approx. 5–8°K.

In the saturated zone, a steam-saturated zone must be formed initially in order to vaporise contaminants. For this purpose, the energy losses arising through ISTT in the saturated zone by groundwater intake or groundwater pumping must be considered. Additionally, the hydrostatic pressure must be considered. The latter increases the boiling temperature of water and contaminant proportionally to the depth underneath the groundwater surface. This increase in temperature can amount to several degrees Celsius (Kelvin) compared to atmospheric conditions.

Table 1: Boiling temperatures and co-boiling temperatures of selected contaminants at normal pressure [derived from REID ET AL. 1987], NAPL-pressure-coefficients [HIESTER 2009]

Contaminant	Boiling temperature of the contaminant	Co-boiling temperature with water (azeotrop)	NAPL-pressure- coefficient
	[°C]	[°C]	[-]
1,2-cis-dichloroethene	60	55	6,0–10,0
trichloroethene	87	73	2,8–3,9
benzene	80	69	3,4
toluene	111	84	1,8
tetrachloroethene	121	88	1,6–1,8
(m-) xylene	139	93	1,3
mesitylene, trimethylbenzene	165	97	1,1
dichlorobenzene	180	98	1,08
naphthalene	218	99	1,0

For mixtures consisting of several individual contaminants, the partial vapour pressure can be calculated according to RAOULT from the sum of the products of the mole fraction and the individual substance vapour pressure [ATKINS 1988]. A simplified estimation of the boiling temperature of the mixture can be made on the basis of the vapour pressures of the remediation-relevant contaminant with the highest boiling temperature and water.

The transition of the contaminant dissolved in the pore water or groundwater into the gas phase (soil vapour) can be calculated by using the HENRY coefficient. HENRY's law is defined either as the ratio between the partial pressure of the substance in the gas phase and the concentration in the aqueous phase, or as the dimensionless ratio of the equilibrium concentrations in the gas and liquid phase [LFU BW 1995]. Accordingly, the volatility of a substance is a function of the HENRY coefficient (contaminants dissolved in water), the solubility in water and/or the vapour pressure (contaminant phase). This function depends on the pressure, the temperature and other water constituents like salts. An increase in the vapour pressure increases the solubility of the contaminants in the pore

water or groundwater. A comprehensive collection of substance property data for calculating the temperature-dependent HENRY coefficient was compiled by SANDER [1999].

2.2.2. Drying behaviour, relative permeability, transport processes

As a consequence of the sub-surface heating and the water vaporisation, a humid soil tends to dry during an ISTT. In this process, the proportion of air-filled pores increases compared to the proportion of liquid-filled pores. During the steam injection, this effect is mostly compensated by supplying water vapour. Local drying effects can occur if thermal wells or RF applications are used. As a consequence of the drying process, the proportion of vapour filled pores increases, as well as the relative permeability of the gaseous phase and the overall diffusion of the contaminants solved in pore water into the gas phase. In consequence, the contaminant recovery by soil vapour extraction is improved. This effect is more significant in cohesive soil types due to their high water-binding capacity compared to, e. g. unsaturated sandy soil types.

Complete drying processes as part of an ISTT normally occur only during the remediation process and in the close proximity of a few decimetres surrounding the thermal wells or rod-shaped RF probes. A complete soil drying is not necessary for an efficient remediation due to the steam distillation effect (cf. chapter 2.2.1). Even if ISTT is applied to the saturated zone (aquifer and aquitard), the formation of a steam-saturated zone does not necessarily imply a complete drying process [BAKER & HIESTER 2009].

Particularly in cohesive soils, a non-isothermal circulating air-water flow is induced (heatpipe effect) [UDELL & FITCH 1985]. This flow of water and vapour is generated by a temperature gradient and the capillary forces in the soil. According to the temperature gradient, the gases flow from the heat source to colder areas. The gases cool down with an increasing distance to the heat source and previously vaporised liquid condensates (as soon as the saturation concentration is exceeded in the gas phase), which strengthens the gradient (induced by the vaporisation) in the liquid phase. This gradient can be neutralized by capillary forces to enable pore water to be transported in the opposite direction and against the temperature gradient to the hot area.

When ISTT is applied to sandy soils, the heat-pipe effect can be neglected due to the low capillary forces. In contrast, the heat-pipe effect for ISTT in soils with low permeability accelerates the expansion of heat since a convective heat transfer is induced in the direct proximity of the heating wells in addition to the conductive heat transport. Simultaneously, water that has already been heated is fed back into the hot area surrounding the heating elements, causing a significant time delay of a complete soil drying [HIESTER 2009].

2.2.3. Geology, hydrogeology, anisotropy, settlement

The hydraulic conductivity of the sub-surface has a significant influence on the operating window of ISTT. While the advective or convective method of steam-air injection requires soils of good to moderate permeability, thermal wells or radio frequency heating can also be used in soils with low permeability.

A source remediation in aquifers containing structures of high conductivity or / and high hydraulic gradients using ISTT is usually less effective due to extensive heat losses. If the Darcy velocity $v_f (k_f x i)$ in unconsolidated soil aquifers exceeds the critical value of 0.5 m/d, a thermo-technical investigation is recommended.

The geological structure of the sub-surface is mostly characterised by sedimentation and erosion processes. The thickness of stratified layers can be up to several metres and thus influences both a contaminant migration and the selected thermal treatment method. Thin, alternating strata can lead to a dispersed contaminant migration, while thick, homogeneous strata of cohesive soils might cause a concentration of liquid contaminant phase (pools). Cohesive soil strata are heated by thermal wells using the effects of heat conduction or by introducing RF energy in order to desorb the contaminants through thermally enhanced diffusion. An external conductive heating of cohesive strata by a steam flow around the layer can be economically viable up to a strata thickness of approx. 1 m [FÄRBER & HIESTER 2006, TRÖTSCHLER ET AL. 2006]. Stratifications of unconsolidated soil (sands, silty sands, and gravel) above or below cohesive soil formations and layers can be used for extracting the vaporised contaminants [HIESTER 2009].

Sedimentation and erosion processes generally lead to an anisotropic permeability distribution in the sub-surface. Due to the sedimentation processes, the vertical permeability is often one order of magnitude smaller than the horizontal permeability. In the saturated zone, this always results in an enhanced radius of influence of the steam-air injection going along with a reduced amount of injection and extraction wells. Conductive heating methods are less to rarely ever influenced by anisotropy. Due to the low variance in the heat conductivity, different soil types have only a minor effect on the speed of conductive heating processes.

The load-bearing capacity of the soil in the unsaturated, heated zone is normally less impacted by changes in the water content [HIESTER 2009]. In the saturated zone, the steam partially displaces the pore water and takes the physical function of the water in the soil. Organic soil components such as peat can be degraded more quickly at higher temperatures causing a reduction of the volume. In extreme cases and at high loads, for example of buildings, such volume reductions may lead to settlements. Although recent laboratory studies have been attempting to simulate the monoaxial volume reduction of peat as a consequence of ISTT on soil samples [TRÖTSCHLER ET AL. 2011], the transfer of these test results to triaxial stress states during remediation operations is still a current research topic. However, especially in soils with high organic content, the boundary conditions and limitations for the application of ISTT must be thoroughly assessed and considered in the early planning stage.

Experience has shown, however, that settlement measurements were only carried out on approx. a quarter of ISTT during the last few years (Figure 2.5). The volume modifications caused by boreholes and modifications of the water contents amounted to a few millimetres. However, volume modifications of strongly organic soils (organic content > 10 %) can amount to several centimetres in extreme cases, depending on the thickness of the stratification and the loads. If ISTT are applied to soils of such conditions beneath

buildings or in their immediate proximity, the effects of heating on the foundation of the building must therefore be estimated in advance (planning stage) and monitored during the remediation process.



Figure 2.5: Number of ISTT performed with settlement measurements, differentiated according to remediation beneath or next to buildings as well as in open spaces

Heating in general influences the carbonate balance and solubility of water constituents (e. g. iron or manganese). In contrast to the increasing water solubility of contaminants with rising temperatures, lime increasingly precipitates at higher temperatures. In aquifers with high lime contents, a local precipitation of calcium carbonate on a pore scale is to be expected. A project-specific analysis is needed to assess whether such spatially limited effects could have a significant effect on the remediation process. Such precipitation effects are usually taken into account for an appropriate selection and design of the water treatment techniques.

2.2.4. Focus: geology and hydrology of tropical soils by the example of Brazil

Tropical soils were mainly formed during the Tertiary but can also be of Jurassic age. Besides terrain characteristics and the course of time, prevailing climate conditions have the most important influence on the soil genesis. Increased temperatures and a high moisture content enabled intensive chemical weathering processes and microbiological activities which reached depths of 20 to 30 m bgs. Due to long-term weathering, e. g. in very old and flat land territories in Africa or Southern America (old continental shields), these soils are often low in nutrients. In contrast to soils in the temperate zone, subtropical soils as well as tropical soils show a moderate to low horizontal stratification.

The processes of ferralisation and desilication can be summarised by the term "lateralisation" [SCHELLMANN 1966]. High precipitation rates in tropical areas in combination with constant formation processes led to the eluviation of highly soluble compounds such as sodium, calcium and manganese from the ferralisation process as well as silicium in the form of silica resulting from desilication. This way, less soluble elements e. g. aluminium and iron accumulated in the surface soil, and the formation of sesquioxides such as adamantine spar (Al₂O₃) or ferric oxide (haematite, Fe₂O₃) and hydroxides such as goethite (FE₃+O(OH)) or gibbsite (Al(OH)₃) is facilitated. The typical clay mineral contained in these soils is kaolinite (Al₄[(OH)₈|Si₄O₁₀]). Kaolinite is moderately active compared to other clay minerals in terms of its contaminant adsorption capacity.

One of the most common tropical soils is ferrasol which is also known as latosol or oxisol. The ferrasol soil type can be found up to depths of 30 to 60 m bgs and commonly has a pH-value of 4.5 to 5.5. A "ferralitic Bu-horizon" of several metres of thickness is often followed by an alteration zone (saprolite) of varying thickness.

Due to the weathering of clay minerals, vertical eluviation processes as well as clay illuviation take place and also the presence of acrisol horizons is characteristic. This soil type is characterised by its acid milieu.

Both ferrasol and acrisol become plastic with increasing water contents. In contrast, when soils dry up, contained oxides harden and form a solid crust. This process is irreversible, even if the water content increases again. When applying *in situ* thermal treatment methods, crusting processes can be expected, e. g. locally around heater wells.

Furthermore, soilification is a structure building process [FIEDLER 2006-2007]. As a result of partial SiO_2 -removal and silicate destruction, a stable cementation structure is formed with irregular subpolyhedrons. Small soil aggregates develop due to the adhesion of primer quartz and secondary clay minerals and the relocation of iron oxides, and form a solid texture (pseudo-sand) with a low bulk density.

Thus, ferrasol and acrisol reach a high soil aggregate stability by comprising macro pores between the aggregates. Depending on the pore diameter, these macro pores constitute preferential flow paths for multi-phase flow processes of different liquids (water, oil, dissolvent etc.). In case of small pore size characteristics (high capillary pressure and unsaturated conditions), NAPL infiltration lab experiments in aggregated soils showed a more homogeneous NAPL movement due to capillary forces [HIESTER 2009]. However, in case of the long term presence of a NAPL contamination, the contamination front will migrate into the soil matrix due to diffusive transport processes. The recovery process of contaminants during *in situ* remediation highly depends on the pore scales of the local soil: the contaminant extraction from micro pores is more complex than that from macro pores. The inhomogeneous distribution of pore scales ranging from micro to meso and macro pores makes it almost impossible to quantify the limitations of the overall remediation processes will overcome the pore scale effects. However, it can be expected that *in situ* heating processes will overcome the pore scale limitations for contaminant recovery due to the higher mobility and vaporisation of the contaminants.

2.3. Substance parameters and substance sizes of important contaminant groups

2.3.1. Volatile halogenated hydrocarbons

Volatile halogenated hydrocarbons are low-boiling organic compounds that besides carbon and hydrogen also contain halogen atoms (fluorine, chlorine, bromine, iodine). They have been used as cleaning agents, solvents (e. g. PCE or TCE), refrigerants, propellants, and fire-extinguishing agents (e. g. halones). Due to a density greater than water, an immiscibility with water (NAPL), and a low viscosity, most of the remediation-relevant halogenated hydrocarbons form pools that may sink to the bottom of the aquifer, i. e. to the aquitard, by gravity.

The contaminant phase distributes dropwise along its flow path in the unsaturated and in the saturated zone. This normally leads to a massive, remediation-relevant groundwater pollution. Most of the volatile halogenated hydrocarbons are classified as harmful to humans (including carcinogenic or mutagen) and harmful or toxic to aquatic organisms [GESTIS-DATABASE ON HAZARDOUS SUBSTANCES 2011].

Over longer periods of time, halogenated hydrocarbons can be biologically degraded to ethylene or CO_2 (aerobically or anaerobically) under certain environmental conditions. During the degradation process, intermediates such as cis-DCE or VC can be significantly more hazardous to health than the original substances.

Due to their comparatively high vapour pressures, halogenated hydrocarbons source zones in the unsaturated zone can be completely remediated at temperatures of approx. 75–95°C within a few months dependent on the contaminant composition (Table 1 and Table 2).

Substance	Boiling point	Vapour pressure 20°C 50°C 80°C	Water solubility (at 20°C)	Density at 0°C, 1013 mbar
	[°C]	[mbar]	[mg/l]	[kg/m³]
vinyl chloride	-13.4	>1,013 >1,013 >1,013	1,100	2.86
dichloromethane	40	470 >1,013 >1,013	20,000	1,330
1,2-cis-dichloroethene	60	216 704 >.1013	600–800	1,280
trichloroethene	87	78 284 812	1,000	1,460
tetrachloroethene	121	19 82 748	160	1,620

Table 2: Physical properties of selected volatile halogenated hydrocarbons [GESTISDATABASE 2011 or adapted from REID ET AL 1987]

2.3.2. Aromatic hydrocarbons, BTEX, petroleum-derived hydrocarbons

Aromatic hydrocarbons such as benzene, toluene, ethylbenzene, and xylene are organic compounds that are derived from benzene. The highly volatile and chemically stable BTEX compounds accumulate in the soil vapour and are highly mobile. Water solubility values can be found in Table 3. Benzene, toluene, and their derivatives are important solvents. Since the density of BTEX is lower than that of water, the BTEX phase should not migrate to the bottom of an aquifer. Nevertheless, an oil phase, for example, can also be

located below the groundwater surface. This effect occurs more frequently in the case of significantly fluctuating groundwater levels or large contaminant inputs of mixed contaminants. Many aromatic hydrocarbons are classified as harmful to humans (including carcinogenic or mutagen) and as harmful to toxic to aquatic organisms [GESTIS-DATABASE ON HAZARDOUS SUBSTANCES 2011].

Compared to BTEX, petroleum-derived hydrocarbons are less soluble in water and show a stronger adsorption on hydrophobic or organic soil constituents. Different types of oil show different distributions of the organic phase. Oils of high viscosity undergo a more lateral expansion (e.g. crude oil) while low viscosities tend to expand in a vertical direction (e.g. kerosene) [BLUME 1990].

Due to the low vapour pressures of ethylbenzene and xylene compared to tetrachloroethene and trichloroethene, target temperatures of 90–99°C (azeotrop co-boiling) are mostly applied during ISTT of BTEX source zones in the unsaturated zone (Table 1 and Table 3). Due to the increased temperatures during ISTT, there is an increased risk that explosive gas-air mixtures will be formed during the BTEX vaporisation. The lower (LEL) and upper (UEL) explosion limits and the ignition point help to estimate the reaction of the gas-air mixtures (Table 4).

Substance	Boiling point	Vapour pressure at 20°C 50°C 80°C	Water solubility (at 20°C)	Density at 0°C, 1013 mbar
	[°C]	[mbar]	[mg/l]	[kg/m³]
benzene	80	100 361 1,008	1,800	880
toluene	111	29 122 387	470	870
ethylenebenzene	136	10 47 167	140	870
m-xylene	139	8 41 151	200	860
trimethylbenzene, mesitylene	165	3 15 60	20	860

 Table 3: Physical properties of selected aromatic hydrocarbons [GESTIS DATABASE

 2011 or adapted from REID ET AL 1987]

An explosive gas-air mixture exists if the substance concentration of an explosive gas in the (soil) vapour is in the range between the lower and upper explosion limits (LEL $\leq c_{gas} \leq$ UEL). To cause an explosion, an ignition source (e. g. heat, sparks) and a certain minimum volume of the gas-air mixture are needed [ATEX 2008, BGR 104 2005]. In process engineering, this volume is undercut due to the mesh size of a flame arrester in pipes. The pore space between grains of sand and smaller particles provides a similar function. This way, numerous aromatic and petroleum-derived hydrocarbon contaminations, in some cases including floating NAPL, have been successfully treated by *in situ* thermal remediation methods [BARBIAN ET AL. 2009, KOSCHITZKY ET AL. 2007], even though explosive gas mixtures were present in the pore space.

Substance	Lower explosion limit (LEL)	Upper explosion limit (UEL)	Saturation concentration 20°C 50°C 80°C	Ignition temperature
	[g/m³ Vol. %]	[g/m³ Vol. %]	[g/m³]	[°C]
benzene	39 1.2	280 8.6	321 1,049 2,682	555
toluene	42 1.1	300 7.8	110 420 1,215	535
ethylbenzene	43 1.0	340 7.8	41 185 605	430
m-xylene	48 1.1	310 7.0	36 164 544	540
mesitylene	40 0.8	365 7.0	13 65 247	550
phenol	51 1.3	352 9.0	1.5 73 177	595

Table 4:LEL/UEL, saturation concentration and ignition temperature of selected aromatic hydrocarbons (BTEX) [GESTIS DATABASE 2011]

The safety rules for handling explosive gas-air mixtures must be complied with as specified in the safety precautions [ATEX 2008]. In particular, the input of ignition energy (e. g. as part of pile driving or chiselling) must be avoided during dry drilling as part of well drilling. In principle, the conditions in the soil vapour extraction wells used during ISTT are comparable to those in multi-phase extraction wells with floating NAPL (e. g. as in the case of kerosene contamination). In extraction wells, there is always an explosive gas-air mixture to be found above an LNAPL. In terms of process engineering, ATEX-approved pumps and blowers are to be used in addition to flame arresters.



Figure 2.6: Explosive atmosphere in an extraction well (schematic representation)

During ISTT, the composition of the gas-air mixture can change quickly in contrast to SVE. Hence, the use of automatic monitoring and regulation systems (explosion prevention and protection, fresh air valve, emergency stop) is necessary. When the SVE switches off or in case of any other operating state associated with a risk of explosion, the steam-air injection must shut down automatically. When applying heating elements or RF

energy, the necessity of an automatic shut down is to be considered in the scope of the site-specific safety analysis.

2.3.3. Polycyclic aromatic hydrocarbons (PAH)

Polycyclic aromatic hydrocarbons consist of two or more condensed aromatic rings that can be arranged in various manners. PAH are contained in crude oil and coal, but can also be formed from incomplete combustion processes (pyrolysis). In soils, they are mainly adsorbed by hydrophobic mechanisms. Despite a strong sorption to the soil, PAH can migrate into the groundwater and, e. g. as creosote (DNAPL), even to the aquiclude. Some PAH such as naphthalene, pyrene, chrysene, or benzo[a]pyrene are harmful and / or carcinogenic to humans.

Substance	Boiling point	Vapour pressure at 20°C 50°C 80°C	Water solubility (at 20°C)	Density (at 0°C, 1013 mbar)
	[°C]	[mbar]	[mg/l]	[kg/m³]
naphthalene	218	0.04 1.6 9.1	32	1,140
fluorene	295	13 (at 146°C)	1.8	1,200

Table 5: Properties of PAH	[GESTIS DATABASE 2011	or adapted from RE	EID ET AL 1987]
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An efficient steam distillation and consequently a complete contaminant removal of PAH will not be achieved due to the high boiling points of these substances. Nevertheless, enhanced removal rates in the temperature range of approx. 90°C–100°C have been achieved in laboratory tests, particularly with PAH with lower boiling points [ROLAND ET AL 2010]. The present guidelines do not deal with the use of thermal decomposition processes such as pyrolysis and hydrolysis in the case of PAH with temperatures >> 150°C.

2.3.4. Further substances

In general, ISTT can be used for the remediation of a broad spectrum of contaminants, mainly for organic contaminants with boiling temperatures below 200°C. A selection of substances is listed in Table 6.

In the unsaturated zone, even substances with boiling temperatures > 200°C can be thermally treated by a combination or extension of the thermal methods described, e. g. steam-air injection followed by the injection of hot air or in combination with thermal wells. On a laboratory scale, the potential remediation of mercury contaminations (boiling point 356°C) by ISTT at soil temperatures of 250°C has been proven. Complementary sitespecific investigations are necessary to approve and design such special applications. Furthermore, contaminant destruction processes occur at these temperatures, for example pyrolysis. However, these guidelines do not focus on such specific applications. For more information, please refer to the respective publications.

Contaminant (molecular formula)	Boiling temperature [°C]	Density [kg/m³]		
carbon disulfide	46	1,260		
methyl-tert-butyl ether (MTBE)	55	740		
chloroform (trichloromethane)	61	1,480		
hexane	69	660		
cyclohexane	81	780		
Halons				
dibromochloromethane (CHClBr ₂)	120	2,450		
1,2-dibromomethane (CH ₂ Br ₂)	131	2,180		
tribromomethane (CHBr ₃)	150	2,900		
Aromatic hydrocarbons				
chlorobenzene (C ₆ H ₅ Cl)	132	1,100		
dichlorobenzene (C ₆ H ₄ Cl ₂)	174–180	1,300		
phenol (C ₆ H ₆ O)	182	1,070		
cresols (C ₆ H ₈ O)	190–200	1,020		
Inorganic substances				
mercury	356	13,534		

 Table 6:
 Selection of further substances for ISTT

3. Application range and limitations of *in situ* thermal treatment techniques

3.1. Overview of application areas which have proved successful

After comprehensive research activities since approx. the mid 1990s and the following initial ISTT pilot projects during the subsequent 10 years, ISTT has been used more regularly in the last few years. Since remediation processes in the saturated zone are more complex compared to applications in the unsaturated zone, the application of ISTT often focuses on the unsaturated zone. But during the recent years ISTT is increasingly applied to the saturated zone (aquifer and aquitard) as well [KOSCHITZKY & TRÖTSCHLER 2010].

ISTT has been successfully applied to sands, silt, clay, and fractured bedrock, both in the unsaturated and saturated zone. More than 50 % of the groundwater pollution can be attributed to CHC, almost one out of three groundwater pollutions is caused by petroleumderived hydrocarbons, and one out of five by BTEX [BAVARIAN STATE OFFICE FOR WATER ECONOMY 2004]. Therefore, there are more referenced ISTT projects of remediated CHC contaminations. Other substances (e. g. PAH) have mostly been remediated as accompanying substances along with CHC or BTEX contaminations.

In the case of ISTT in the unsaturated zone and within a low vertical distance to the groundwater as well as in the case of ISTT in groundwater, the operation of a hydraulic barrier is often required since the contaminant solubility in water is increased by temperature and the risk of a potential mobilisation of the contaminants via groundwater exists. With the operation of more and more ISTT, this risk may be deemed less significant as so far only low and sporadic increases in the contaminant transport have been observed during groundwater extraction in cases of some specific ISTT. It is typically for the ISTT described herein that usually more than 90 % of the recovered contaminant mass is extracted by soil vapour extraction, even in case of a treatment in the saturated zone. If the vertical distance between the lower level of the contaminant source zone and the groundwater is more than three metres, and considering an adequate operation of the SVE, an additional contaminant transport into the groundwater is not to be expected due to the ISTT. In specific cases, it is recommended to deploy SVE wells in the transitional area between source zone and groundwater. In these cases, groundwater pumping is usually not needed to prevent an uncontrolled contaminant transport. Information and explanations on boundary conditions that, in specific cases, may cause a migration of contaminants when ISTT is applied are listed in BETZ 1998, SCHMIDT 2001, HIESTER 2009, BAKER & HIESTER 2009.

ISTT beneath buildings or in their direct vicinity were successfully realised on many occasions, often while these buildings were used for commercial purposes and private residence during the remediation. Examples can be found in HIESTER & SCHRENK 2008, HIESTER & MÜLLER 2010, KOSCHITZKY ET AL. 2011.

3.2. Major plant-specific and spatial conditions

3.2.1. Sub-surface heating system

Steam-air injection (TUBA method) requires a steam generator and a compressor for generating and injecting the steam-air mixture. Smaller steam generators in form of single units are typically used in buildings, for example. Larger steam generators usually come as container solutions. Before operating a steam generator, the relevant regulations have to be considered (TRD001-802). Depending on the steam output, the steam generators must be certified and / or its operation monitored by the respective technical inspection agency. An operator is usually necessary if the steam generator is classified as group IV (boiler volume > 50 I, operating pressure > 1 bar, product of boiler volume [I] and maximum operating pressure [bar] > 1,000). Dependent on the steam production and operating pressure, the presence of an operator may be required 24 hours a day. For this reason, thermal treatment systems are usually designed in such a way that a safe operation is possible without the presence of a boiler operator (limitation of the steam production or installation of several smaller units).

Steam generators usually operate on electricity, gas or diesel. The size of the fuel tank depends on the fuel consumption of the system and the filling intervals of the fuel tank. Regulations concerning the regular operation of tank systems have to be considered (including the Federal Water Act, state regulations on systems operated with water-hazardous materials, Ordinance on Industrial Safety and Health, Technical Regulations for Combustible Liquids). In particular cases, industrial process heat networks or existing steam supply networks were used for the steam generation. Compressors are usually operated by electricity.

When the THERIS method is used, electrically operated heating elements are installed directly into the sub-surface. The energy output of the heating elements must be adjustable for a proper operation. The regulation units can be centralised or decentralised.

A high frequency generator, a matchbox, and an electrode system are necessary for RF soil heating. The generator and the probe heads including its connections to the matchbox must be specifically shielded to protect the system from electromagnetic disturbances and to avoid electro-smog.

Particularly when these electrically operated methods are used, an infiltration of water into the electrical circuit must be inhibited. In the case of larger systems, it is economically viable to install a separate power supply. Systems with a special power supply of several hundred kilowatts have already been implemented. When electricity is purchased, it is recommended to choose special tariffs in the base-load range. This enables significant savings to be made compared to tariffs for private users. Tariffs offered especially for building sites cannot be recommended as they are not economically viable.

3.2.2. Soil vapour extraction, hydraulic groundwater control, vapour and water treatment

Compressors and pumps must be chosen according to the site-specific geology and hydrogeology, the arrangement of the well, and the length of the pipelines to ensure that the designed flow rates are achieved. It is recommended to mount the soil vapour extraction units inside containers. This helps to reduce noise emissions, for example.

During ISTT, the recovered soil vapour has a humidity of 100 % during most of the operating time. Since the gas flow cools down between the SVE wells and the system container, the formed condensate must be separated prior to the vapour treatment. It is also necessary to dry the hot, humid soil vapour by way of condensation in a heat exchanger, and to separate the condensate from the air flow to guarantee an effective treatment and waste gas purification (particularly in the case of air activated carbon). Therefore, it is always necessary to implement a purification stage for the aqueous phase (e. g. water activated carbon) even when SVE is performed.

If a hydraulic groundwater control is applied (e. g. by pump & treat), the groundwater and the condensate can be merged in a storage tank. The application of phase separators to separate organic liquids from the aqueous phase and a separate waste disposal of the separated NAPL are only in exceptional cases economical, for example if a multi-phase recovery from groundwater wells is likely to be needed. The treated groundwater can be used on a commercially viable basis to cool the air flow. The cooling water can be discharged into a drainage canal or (after cleaning) alternatively reinjected into the aquifer via wells if the permitted "input" temperatures are met.

3.2.3. Remediation-accompanying measuring systems

ISTT process installations are more complex and more expensive in terms of the daily operating costs compared to conventional treatment installations. A more comprehensive process engineering is required during ISTT compared to conventional systems. The higher daily operating costs result from a continual and committed project and remediation management including regular optimisation of the system's operation. The remediation processes as part of ISTT are significantly faster than those of common soil vapour extraction since the contaminant removal and extraction of a standard SVE can be increased by ISTT by a factor of 10 to 50. However, this is always dependent on the site conditions. Therefore, ISTT require the regularly and prompt evaluation of the measured data. Depending on the dynamics of the remediation processes (heat propagation, contaminant recovery, etc.), the boundary conditions and the requirements (e.g. explosion protection), it may be necessary to analyse the measured data on a daily basis in order to control the system and the remediation process.

For monitoring and optimising the system, it is rather common to quantify the heat input, and to compile the main measure and control values in the system as a whole, for example, flow, pressures and contaminant mass flux. A temperature monitoring in the ISTT field is necessary to monitor the heat propagation, to analyse the remediation progress and to control the heating process. The measuring sections and intervals are to

be chosen in such a way that all remediation-relevant processes can be interpreted in the field under an entire range of operating conditions. Automated compilation systems with remote data transfer are common. Systems for remote data transfer and alarm messages from the system and process control have also become state of the art.

Depending on the project, it may be necessary to expand the monitoring concepts to remediation-accompanying tasks. Typical interfaces are the protection of underground cable to prevent that temperature limits are exceeded, indoor air monitoring in buildings, or stability and settlement monitoring of buildings (e. g. sub-surface settlement measuring, fracture monitoring). These accompanying tasks and boundary conditions may arise during the planning and operating phase and may result in restrictions to the remediation process. The compliance of such restrictions is to be monitored by the remediation management.

3.3. Potentials and limitations of steam (-air) injection (TUBA method)

3.3.1. Unsaturated zone

The field of application includes remediating source zones in consolidated soil and finely fractured bedrock with good to moderate permeability (gravel, sand, silty soil) and a hydraulic permeability (k_f value) in the range of 5 x 10⁻⁵ to 1 x 10⁻² m/s. The condensation and accumulation of contaminants at the steam front (in case of a solely injection of steam instead of a steam-air injection) might – under unfavourable conditions – increase the potential risk of a NAPL downward migration to the groundwater. In the unsaturated zone (UZ), a solely steam injection should be restricted to LNAPL remediation. An additional air injection accelerates the contaminant recovery and thus reduces the remediation duration as well as the risk of LNAPL condensation. In the case of a solely steam injection, the remediation design should imply the transport of NAPL by way of high extraction rates or even multi phase extraction and treatment options. As previously described, groundwater pumping is often appropriate. In the case of DNAPL remediation (e. g. CHC), a steam-air mixture should be injected to prevent a potential accumulation of contaminants. The co-injected air acts as a carrier gas. It penetrates the condensation front and removes the contaminants in the gaseous phase with the soil vapour.

Even silt or clay lenses with a low permeability and up to a thickness of approx. 1 m [FÄRBER & HIESTER 2006] can be heated with steam from the exterior of the lenses by extensive conduction. For this purpose, the steam preferably flows in areas of higher permeabilities below or above the cohesive stratifications or lenses. The vaporised contaminants are able to reach areas of better permeability where they can be removed by SVE. In the case of cohesive layers of great thickness, it is recommended to heat the stratifications from the interior by operating thermal wells or by introducing electromagnetic waves.

3.3.2. Saturated zone

The field of application includes porous aquifers (fine sand to silt) with a hydraulic permeability in the range of $k_f \approx 5 \times 10^{-5}$ to 5×10^{-4} m/s, in addition to appropriate fractured rock aquifers with a comparable permeability (uniform fractures, no dominant wide fractures). Experience gained from pilot and full-scale applications in porous aquifers suggests the following rule of thumb: For anisotropic stratifications in the above mentioned permeability range, a lateral horizontal steam expansion of a radius of 3–5 m is to be expected at an injection rate of approx. 150 kg/h of saturated steam (per metre of injection filter length), a ratio of air of approx. 10 mass %, and a water level above the injection filter of 4–5 m [OCHS, S.O. 2006, TRÖTSCHLER ET. AL. 2006].

The steam expands laterally and vertically in the aquifer and is not intercepted by the SVE until it reaches the UZ. Prior experience suggests so far that a distance between the injection depth and the UZ of up to 10 m is feasible. Depending on its permeability $(5 \times 10^{-5}-1 \times 10^{-2} \text{ m/s})$, the UZ should be between 3 and 5 m thick. An additional steam-air injection in the area of the UZ can be economically viable if the UZ exceeds approx. 3 m in thickness. The remediation planning may include pilot tests to determine the range of influence of the steam expansion in the saturated zone with extraction wells arranged appropriately, in addition to the temperature and contaminant monitoring. Site-specific pilot tests may help to clarify open issues for applications in heterogeneous structures or beyond the application areas stated above.

In the case of intermediate stratifications of low thickness < 1.5 m, for example silt or clay lenses, the steam flows underneath the areas with poor hydraulic permeability and is able to heat the soil located above through extensive conduction ("steam-override") within several weeks [FÄRBER & HIESTER 2006, KLEINKNECHT 2011]. The contaminants in the steamed-saturated zone are vaporised and transported to the areas located above due to the temperature gradient. The contaminants can now be captured and removed by SVE from the unsaturated zone. Any possibly condensed contaminants are vaporised again by propagating heat. In the case of cohesive layers of great thickness, the heating should be initiated from the interior of the structures. The operation of thermal wells (chapter 3.4.1) or the input of electromagnetic waves (chapter 3.5) is recommended.

3.3.3. Dimensioning guidelines

The procedure and design of a steam-air injection in the unsaturated zone are based on calculations of the steam expansion [SCHMID ET. AL. 2000, SCHMIDT 2001] which are based in turn on analytical solutions for combining thermodynamic conditions and flow conditions [FÄRBER 1997].

The expansion of a steam front in the groundwater can be estimated on the basis of type curves [OCHS 2006, KOSCHITZKY & TRÖTSCHLER 2008]. For this purpose, it is necessary to thoroughly investigate the geological and hydrogeological properties of an aquifer. A

software tool for calculating steam-air injections⁴ was published in 2012 within the framework of TASK (<u>http://www.task-leipzig.de</u>).

ISTT beneath buildings is possible in both the unsaturated and the saturated zones while maintaining the use of the buildings during the remediation activities. A technical inspection of the affected buildings including the documentation of the state of the buildings is required before the remediation and at regular intervals during the remediation process.



Figure 3.1: Example of the estimation of the radius of thermal influence using the "DLI-Tool" software



Figure 3.2: Steam-air injection at the "Biswurm" demonstration site left: container system, right: field of wells with steam-air injection, soil-vapour extraction (SVE) and groundwater pumping (GW)

⁴ The user interface and the manual of the "DLI-Tool" software are available in German only.

Parameter	Effect on	Dimensions
Hydraulic conductivity		
Unsaturated zone 10 ⁻² – 5 x 10 ⁻⁵ m/s	Steam rate SVE extraction rate Time required	0.1–0.5 kW/m³ soil 0.1–0.5 Nm³/(m³ soil x h) 0.03–0.06 d/m³ soil
Saturated zone 5 x 10^{-4} – 5 x 10^{-5} m/s	Radius of thermal influence Steam rate SVE extraction rate Time required	3–5 m radius (see also text above) 0.3–0.5 kW/m ³ soil 0.1–0.5 Nm ³ /(m ³ soil x h) 0.06–0.12 d/m ³ soil
Source zone	Economic viability Remediation duration	> 500 mg contaminant / kg soil 0.5–2 a
Dimension of the contamination (cubage)	System size → section-by-section remediation	200–1000 kW injection power (>1 MW e. g. [4. BIмSснV 1997])
Contaminants	-	-
Boiling temperature < 100°C (Benzene, DCE, TCE, DCA, TCA, CCl ₄)	Contaminant recovery rate SVE concentration level	> 95 % achievable > 10 g/m³ soil air
Boiling temperature 100– 150°C (PCE, PCA, BTEX, petrol, chlorobenzene)	Contaminant recovery rate SVE concentration level	> 90 % achievable 1–10 g/m ³ soil air
Boiling temperature 150 - 180°C (diesel, (decane), phenol, DCB, naphthalene, trimethylbenzene)	SVE concentration level	 > 70 % achievable > 1 g/m³ soil air
Remediation zone with/without built-up	Technical effort, costs for site preparation and safety mea- sures, area required for the installation of the system, structural inspection, indoor air monitoring, safe pneumatic capturing or cooling of sen- sitive areas by air injection	Distance injection well / extraction well UZ: 10–15 m SZ: 4–8 m
Surface sealing	Polyethylene tarpaulin covering, concrete, asphalt, mineral sealing	Necessary if the surface $k_f > 10^{-5}$ m/s or if the remediation area is not covered sufficiently
Thickness of UZ	Pneumatic capture Ratio: number of extraction wells to number of injection	Minimum 3–10 m Mass flow extraction / injection > 1.5 Minimum 2–6
	wells Maximum injection pressure	Soil superimposed load x security (0.3–0.7)
Thickness of SZ	Applicability Injection capacity Maximum injection pressure	Currently to be designed up to approx. 8 m (see above) Soil superimposed load x factor of safety (0.3–0.7) + water column above injection level

Table 7: Dimensions for steam-air injection (TUBA method)

3.4. Potentials and limitations of thermal wells (THERIS method)

3.4.1. Unsaturated zone

Cohesive or non-homogeneous soil layers are generally considered little or non-suitable for soil vapour extraction. However, source zone remediation can also be efficiently implemented in these soils by using conductive heat input and transfer. In contrast to a convective heat input (steam-air injection), the permeability of the sub-surface is only of minor importance for the energy input.

Nevertheless, the soil properties affect the radius of influence of soil vapour extractions. Source zone remediation activities with the THERIS method have been successfully completed in soils with hydraulic conductivities of less than $k_f \approx 10^{-9}$ m/s. A pre-requisite are high-performance extraction units (vacuum pumps) for generating a high vacuum.

This method has been used for many sites in urban areas thanks to its simple operation, the good installation options and the noiseless operation of thermal wells. Irrespective of this, noise pollution arising from conventional system components such as pumps and compressors must be reduced.

3.4.2. Saturated zone

Thermal wells can be used efficiently for source zone remediations in aquitards. Scientific studies have shown a tendency that moderate to low hydraulic conductivities ($k_f < 1 \times 10^{-7}$ m/s) have a positive influence on the efficiency [BAKER & HIESTER 2009]. To ensure that the contaminant recovery predominantly takes place through the gaseous phase, a steam-saturated zone must be initially created by vaporising water, i. e. the subsurface is heated to the boiling temperature of water. A low hydraulic conductivity reduces the lateral infiltration of water into the remediation area and supports the heat to accumulate there. The contaminants vaporised in the saturated zone are transported into the unsaturated zone as a consequence of a heat-induced soil air flow where they are collected by soil vapour extraction.

To enable a sufficient remediation success, the steam front generated in the saturated zone must completely cover the entire source zone. On a technical scale, a moderate lateral transport of contaminants has been observed as side effect in cases of small distances of a few decimetres between the steam front and the source zone. It is caused by the condensation at the steam front [BAKER & HIESTER 2009]. This process is similar to a potential contaminant accumulation when saturated steam is injected. Hence, a complete source zone investigation is important for field applications. However, in the case of field applications, the side effect described above is of minor importance since the area to be thermally remediated is less affected by side effects than on the technical scale. First THERIS remediation projects in aquitards have already been implemented. For example, a simultaneous source zone remediation of two aquifers with the TUBA method and the aquiclude with the THERIS method was classified at the BROWNFIELD BRIEFING AWARDS 2011 as "HIGHLY COMMENDED" in the category of "Best In-situ Treatment" (see project example G).

3.4.3. Dimensioning guidelines

The energy input mainly depends on the contaminant inventory, the remediation goals, the soil moisture, and the intended duration of the source zone remediation. The infiltration of stratum water in the unsaturated zone should be known or is to be estimated during the planning phase since it must be taken into account when designing the system and the wells.

Site-specific issues can be clarified during the respective pilot tests (Figure 3.3). In general, large remediation fields show an improved economic viability due to reduced energy losses via the boundary. In the case that the electric power available on-site is not sufficient to remediate the complete source zone in one go, the installation of additional electrical power lines is usually more economical than dividing the remediation area into several sections. Both options have already been implemented.



Figure 3.3: Site-specific THERIS[®] pilot test. The radius of thermal influence at the surface becomes obvious by the melting of the 20 cm thick snow cover (project of the HIM GmbH⁵).

For the SVE wells, a well spacing of few metres is recommended in clay soils [VDI 1997]. The distance between the heating elements amounts to several metres as well and varies according to the cubage of the source zone, the contaminants and the intended remediation duration. Soil air extraction tests as described in the ITVA guidelines [2002] are not convincing since they do not cover the processes in the heated state. The

⁵ For further information see the publication in German: WOISNITZA ET AL.: *Thermische In-situ-Sanierung (Pilotversuch) am Beispiel "ehemalige Lederfabrik Berninger" in Idstein.*

dominating remediation processes are particularly temperature dependent in cohesive soils since the increased vapour pressures of the fluids interfere with changes in the water balance and with thermo-hydraulic processes such as the heat-pipe effect. The latter affect the mobility of the contaminants significantly.

Remediations are carried out frequently beneath buildings or in their direct proximity in both the unsaturated or saturated zone. A constructional inspection and the documentation of the building structure have to be carried out before commencing a remediation as well as in regular intervals during the remediation process. The protection of temperature-sensitive components in the sub-surface such as cables can be implemented in an easy way when using the THERIS-method by adopting design and operating methods to site-specific characteristics. To prevent damages, e. g. information is required on existing underground cables and their sensitivity against heat and varying temperatures which can be obtained from the system operator. Moreover, a comprehensive documentation (temperature measuring) and project coordination is needed.

3.5. Potentials and limitations of radio frequency (RF) soil heating

3.5.1. Unsaturated zone

In contrary to low-frequency heating (power-line-frequency [PLF] heating, six-phase heating) [BILSHAW-BIDDLE 2000; CKY INCORPORATED ENVIRONMENTAL SERVICES 1995], RF soil heating does not require a certain soil water content [ROLAND ET AL. 2007a; ROLAND ET AL. 2008a]. Even dry soils can be heated to temperatures far above 100°C with high efficiency which makes the RF method ideally suited for the application in the unsaturated zone.



Figure 3.4: Options of electrode arrangements for the RF-supported soil vapour extraction at different sites (right: protection of the RF electrodes)

Specific rod electrodes [ROLAND ET AL. 2008b, ROLAND ET AL. 2007b] or antennas [ROLAND ET AL. 2001] can be used to cover large depths or remediation areas beneath buildings. Rod electrodes can also be installed in offset bore holes (Figure 3.4). When combining RF soil heating with SVE, the air flow in the soil can be used as heat transfer medium in order to homogenise the temperature profile. The electrode spacing can then amount to several metres (mostly 3 to 5 m) [HOLZER ET AL. 2010].

3.5.2. Saturated zone

RF soil heating can basically be used in the saturated zone as well. However, lowfrequency heating methods (PLF heating or six-phase heating) are suited much better due to the high electrical conductivity of this zone [BILSHAW-BIDDLE 2000]. Energy losses due to the transformation of network energy into HF energy are omitted (approx. 45 % to less than 20 % for newer generators) [VOLKMAR & WRONA 2010]. In many cases, the contamination extends, however, beyond the capillary fringe at the boundary between the saturated and unsaturated zones (e. g. LNAPL). In order to prevent condensation and readsorption, large temperature gradients are to be avoided in this area during RF soil heating.



Figure 3.5: Coupling of several frequencies for the homogeneous heating of the capillary fringe

For these cases, a combination of resistive (low-frequency) and dielectric (high-frequency) heating has been developed. The method was successfully tested at a model site (former storage facility for solvent tanks) [ROLAND ET AL. 2008b; ROLAND ET AL. 2007b; ROLAND 2008]. An electrode system is used to introduce energy with two frequencies into the soil (frequency coupling similar to "internet from the socket"; Figure 3.5).

Appropriate coupling devices ensure that the two sources (e.g. 50 Hz and 13.56 MHz) can be controlled independently. As a result, the boundary area between the saturated

and the unsaturated zones can be heated homogeneously (Figure 3.6) which could not be achieved with a single frequency alone [ROLAND 2008; ROLAND ET AL. 2008a].



Figure 3.6: Homogeneous heating of the border area between saturated and unsaturated zone (at 1 and 3 m depth, groundwater level at 2 m bgs, soil type: sandy soil, in part filling material)

3.5.3. Dimensioning guidelines

The final temperatures which can be achieved depend on the power density related to the soil volume, the availability of water in the soil to be heated and the heat transport into the environment including the thermal insulation on the surface. There is almost no restriction in terms of soil material or soil type.

The spacing between the single RF rod electrodes or between the electrodes and the extraction wells during SVE should be 3-5 m as the RF method heats up a radius of 4 and 5 m. Reference values for the upper limit of a soil cubage are 300 m^3 for a 15 kW and 500 m³ for a 30 kW RF container system.

In combination with SVE, the requirements of the latter mainly influence the overall design of the process. In principle, the method is based on a counter current principle. Soil vapour is extracted via the RF electrode(s) which leads to a transport of soil air from cold to hot soil areas. The convective heat transport is directed opposite to the dielectric heating and the thermal conduction. This procedure would be appropriate for spatially limited contaminations concentrated around the RF electrode(s). In addition, it helps to prevent a contaminant transport from warmer to colder areas.

It is also possible to arrange the extraction wells around the RF electrode(s). Due to the direction of current (direct current principle), the soil air flow supports the expansion of the heat front in the soil by convective heat transport. This extraction process allows for the heating of large areas of contaminated soil. This procedure should be preferred in the case of high contaminant concentrations above the lower explosion limit in the extracted soil vapour.

3.6. Combining methods – When is it appropriate?

3.6.1. Combined application of various ISTT methods

Even though a conductive heating of cohesive surfaces can be achieved by applying convective heating methods (steam injection), and even though it is possible to heat sandy-gravel layers with thermal wells, such applications are only appropriate for layers of a few decimetres of thicknesses. In the case of alternating strata of non-cohesive and cohesive soils of layers of several metres of thickness, a combination of the described ISTT methods is common to efficiently remediate sites with complex geological and hydrogeological conditions as well.

Several TUBA-THERIS combinations have already been implemented since both methods stand out particularly by their application in various geological structures. Both a lateral arrangement of remediation fields for convective and conductive heat input as well as "sandwich" concepts, i. e. a vertically overlapping arrangement of the various ISTT methods, have been used. An example for a TUBA-THERIS-TUBA combination is illustrated in the project data sheet G. In this project, the source zone was remediated down to a depth of 18 m bgs in the first and second aquifer (TUBA in each case) including the clay aquiclude (THERIS). Such complex systems require separate control, pump and monitoring systems for the heating and extraction operations of each remediation horizon to meet the boundary conditions in the respective remediation areas in the best possible way. An intensified project management is essential in order to quickly detect interactions between the individual remediation areas and to control the remediation process in real time by exploiting synergy effects.

For a simultaneous purification of unsaturated and saturated zones using RF, a method of combining resistive (low-frequency) and dielectric (high-frequency) heating has been developed and successfully tested at a model site. Details can be found in chapter 3.5.2. In addition, it would also be possible to combine RF soil heating with *in situ* catalytic oxidation [UTILITY MODEL DE 202007014507.1, 2007]. This particularly applies to high concentrations of non-chlorine organic contaminants in the soil air.

So far, RF soil heating has not been combined with other ISTT methods (TUBA and THERIS). Similar to the TUBA-THERIS combination, a combination of different heating strategies (RF heating and TUBA) is plausible. The RF technology would heat the cohesive soil layers predominantly in vertical direction, while TUBA would lead to the heating and contaminant output in lateral direction due to a convective mass and energy transport.

3.6.2. Combination of ISTT with other remediation & precautionary measures

ISTT can be combined with other remediation and precautionary measures. When ISTT is performed in the groundwater, the operation of a hydraulic barrier is recommended as already described. This way, a potential contaminant transport in the groundwater can be prevented at moderate costs. Experience shows that there may be a short-term increase in the downstream contaminant transport during some of the ISTT applications. As the

contaminant removal via the groundwater is low compared to the removal via SVE, the contaminants transported in the groundwater during ISTT hardly contribute to the mass balance.

In order to accelerate ISTT In the saturated zone, hydraulic measures can be used to create a temporarily unsaturated zone. Since the thermodynamic requirements for vaporising large volumes of water in order to create the necessary gas and steam volume and in order to compensate for heat losses in the groundwater can be significant, accompanying hydraulic measures might be of economic benefit.

The remediation of source zones in the groundwater to achieve drinking water quality by using only one remediation method may be possible from a technical point of view in some cases. However, it is not always efficient and often disproportionately cost-intensive. Already in the planning phase, it is possible to combine ISTT with other remediation and precautionary measures to be applied in a chronological and spatial order ("treatment train"). Then, ISTT enables the removal of contaminants from source zones in the surrounding plume in a fast and secure way. These residual contaminants can then be treated more economically with a temporarily limited, downstream hydraulic operation (operating duration approx. 2 to max. 5 years) or in combination with measures in the contaminant plume (e. g. stimulation of microbiological activity). Corresponding concepts have already been successfully implemented and have been classified at the Brownfield Briefing Awards 2011 as "highly commended" in the "Best In-situ Treatment" category (see appendix: project example G).

The kind of combination of various measures in the source zone and in the plume can affect the operational control and the switch-off criteria of single measures, including those of an ISTT. Publications concerning implemented projects with combined methods are currently being prepared.

3.7. Current developments in the field of remediation methods

3.7.1. Fractured bedrock remediation

Even in case of steam-air injection in fractured bedrock the previously described ISTT remediation processes are based on contaminant vaporisation as well. In contrast to unconsolidated soil aquifers, the fractures in the fractured rock determine the available routes for water, steam and air. The 'radius of thermal influence' of the steam-saturated zone in fractured aquifers cannot be calculated for stochastic reasons. However, it can be mathematically estimated in anology to porous aquifers.

As part of a pilot test in a fractured sandstone aquifer, the bedrock matrix was conductively heated by injecting a steam-air mixture [TRÖTSCHLER & KOSCHITZKY 2009, KOSCHITZKY ET AL. 2009]. During the process, the contaminants impregnated in the sandstone were desorbed and vaporised by the steam propagation in the fractures.

The presence of an effective pneumatic connection between the fractures in the saturated and unsaturated zones and the soil vapour extraction wells has been proven as part of an additional site investigation. A gas tracer test (CO_2) was conducted in advance of the intended thermal *in situ* remediation. A high portion of air (approx. 10 mass %) was added to the steam flow and this way, a gas-based transport of contaminants and their subsequent removal were achieved.



Figure 3.7: Remediation scheme showing the site lithology and injection and extraction wells in the pilot field

A radius of thermal influence (steam expansion) of more than 10 m in diameter was achieved in the upper aquifer and in the unsaturated zone. In total, more than 510 kg of CHC were removed during the 7 months of operation. It was decided to implement the ISTT for the entire site based on the thermally induced increase in remediation efficiency in the upper aquifer area and the UZ, in addition to reduced emissions from 210 g CHC/d to 37 g CHC/d [KOSCHITZKY ET. AL., 2009]. The thermal remediation started in September 2012 and will last until end of 2014.

3.7.2. Improved multi-phase extraction

A further development of ISTT is the thermal improvement of flow properties of NAPL to increase the liquid-phase extraction. Initial investigations with oil phases in the laboratory and in the field yielded good to very good results in terms of an increased contaminant output. This applied to creosote, for example. Related publications are still to be expected.

3.8. Legal aspects and approval requirements

The handling of contaminated sites in Germany, including the application of *in situ* remediation techniques, is regulated by the Federal Soil Act [BBodSchG 1999] and the Federal Soil Protection and Contaminated Sites Ordinance [BBodSchV 1999] in conjunction with the respective state-specific legal regulations.

In situ remediation techniques in the unsaturated zone or in the groundwater require an approval in accordance with the water regulations. Therefore, the current version of the Federal Water Act (WHG)⁶ often applies for ISTT – even when it is used in the UZ. In compliance with § 8, section 1 WHG, the use of a body of water (including groundwater bodies) requires approval or permission, unless provided otherwise in this Act or the regulations decreed by the Act. The application of physical, in this case thermal, *in situ* treatment techniques constitutes the elements of usage according to § 9, sections 1 and 2 WHG. In compliance with § 10 section 1 WHG, the approval authorizes the entity in question to use a body of water for a specific purpose in a certain manner depending on the type and extent of the situation. Permissions according to water regulations are issued by the districts and district-free towns in their function as subordinate water authorities.

In addition, further areas of regulation – predominantly the legal areas of immission control, building regulations, waste disposal and nature conservation – may apply and the respective permissions may be required.

The planning documents must comply with the requirements of the relevant laws and ordinances of the German Federation and the federal states, with technical regulations, occupational safety and health regulations, regulations of professional associations, guidelines, safety rules, principles, technical bulletins and standards issued by the German Institute for Standardization (DIN) in their respective valid versions. Information on the requirements for approval and the types of required permissions are to be obtained from the responsible authorities. The installation and operation of ISTT systems has to comply with the legal requirements for safety and occupational health.

3.8.1. Legal aspects and approval requirements at the example of Brazil

Even though the application of ISTT methods might be of interest to several countries, it is the local legal framework as well as the national authorisation and regulatory authorities which decide on the implementation of ISTT methods. Factors influencing the authorisation process are, e. g. the effective legal provisions for groundwater, hazardous waste and waste disposal or time requirements and limitations of the remediation process. For example, in some countries, groundwater is considered private property that belongs to the land surface owner, in others it is mainly regarded as public good. However, the legal situation determines the possible decisions and actions taken by local authorities concerning source zone remediations and the conditions for the sale of real estates.

⁶ The current version dates from 01/03/2010.

In general, country-specific information can be obtained from the local chambers of commerce. To give an example, the legal regulations for Brazil are presented in the following. For Brazil, the UK TRADE & INVESTMENT (2010) recently published the officially recorded number of about 15,000 contaminated sites. However, the number of unknown contaminated sites might be significantly higher. In 2009. CETESB (http://www.cetesb.sp.gov.br, São Paulo State Environment Agency) counted more than 2,500 contaminated sites of which 77 % were related to petrol stations. Thus, the state of São Paulo passed a law in July 2009 (13.577/09) stating that site owners, leaseholders and waste producers are to assume responsibility for the protection of the soil quality and the funding of ongoing remediation actions. Spatial limitations and the economic growth of São Paulo stimulate brownfield redevelopment for domestic, trade and industrial purposes. Further information about the current legal situation can be obtained, e.g. from the Brazilian environmental agency (Ministério do Meio Ambiente, dos Recursos Hídricos e da Amazônia Legal – MMA (<u>http://www.mma.gov.br</u>)) or the institute for environment and renewable natural resources (Instituto Brasileiro do Meio Ambiente e dos Recursos Naturais Renováveis – IBAMA, http://www.ibama.gov.br).

The Brazilian association for technical norms, the Associação Brasileira de Normas Técnicas (ABNT, <u>http://www.abnt.org.br/</u>), is responsible for the release of standards for many industrial products. Some of these products such as cables, pipelines or ATEX-equipment require a further certification by an INMETRO accredited organisation. Furthermore, international standards like ISO/IEC-norms are established.

3.9. Ecological and economic considerations

3.9.1. Life cycle assessments, energetic analysis

Life cycle assessments [DIN 14040] can be used as an "*instrument to (...) quantify environmental effects*" [SCHRENK 2005] of brownfield remediation methods. These assessments help to investigate the impacts of a technical system (in this case: soil and groundwater remediation) on the environment (e. g. emissions, waste, consumption of resources). In the final analysis, it is distinguished between the different ecological impacts, i. e. between consumption (e. g. energy and water consumption), material and energy flows (analysis of tangibles: e. g. material manufacture, creation of waste, transport) and effects (e. g. from carbon dioxide as a greenhouse gas). For exemplary purposes, "cold" SVE and the subsequent ISTT measures were analysed and evaluated for various sites. Field data were processed using a specific software tool to determine the environmental footprint of brownfield remediations [LFU BW 1999; HIESTER & SCHRENK 2005].

For ISTT methods, the available data of remediation activities showed a reduction in the specific energy consumption compared to "cold" SVE. This applied irrespectively of the site and was a consequence of the short ISTT operating durations. Comparisons of ISTT with other "conventional" remediation methods showed similar tendencies [HIESTER ET AL 2003, SCHRENK 2005].

Since most of the energy during ISTT is expended for sub-surface heating and contaminant extraction, conventional extractions and ISTT can be evaluated in a simplified way based on the contaminant-specific operating energy for extracting one kilogram of contaminant. For exemplary purposes, Figure 3.8 illustrates the contaminant removal and the associated energy consumption of a THERIS remediation in the UZ via SVE and paralleloperated hydraulic control (P&T). In addition, Figure 3.9 illustrates the development of the soil vapour concentration during this ISTT in order to demonstrate the remediation progress (evaluation of > 20 SVE wells).

The North Rhine-Westphalia State Environment Agency [LUA NRW 2001] provides reference values for the energy efficiency analysis of an extraction measure. For exemplary purposes, specific operating energies of ISTT sites have been compiled (Table 8). The lowest power consumption was found in the case of highly contaminated source zones and soils with high permeability (Figure 3.10).



Figure 3.8: Project example for the specific energy consumption during a THERIS remediation. Upper diagram: cumulative contaminant discharge and energy consumption, divided into soil vapour extraction, subsoil heating and P&T, lower diagram: specific energy consumption for ISTT and P&T.



Figure 3.9: Complementary to Figure 3.8: The course of the CHC-pollution at all SVE-wells (range of variation (max-min values) and average values (points))



Figure 3.10: Specific energy consumptions of soil remediation methods (SVE and ISTT) depending on the contaminant loads and the soil classification (project examples from Table 8)

			"C	Cold" SV	Έ		ISTT	
Project	ISTT- method	UZ/SZ (decisive soil type)	Contami- nant discharge	Energy *	specific energy**	Contami- nant discharge	Energy*	Specific energy ***
			[kg]	[MWh]	[kWh/kg]	[kg]	[MWh]	[kWh/kg]
Biswurm: pilot test (500 m ³ fractured rock)	TUBA Steam-air injection (SAI)	UZ + fractured GW	117	13	111	394	315	797
		SAI in SZ				15.4	27	1,753
		GW- extraction	12.5	21	592	22	61	2,795
Zeitz pilot test	TUBA	UZ	4,050	21	5	2,521	275	109
(1.500 m ³ granular S soil) i	Steam-air	SAI in SZ				300	150	499
	(SAI)	GW- extraction	16	3	156	56	10	180
Hamburg II a	THERIS	UZ (loam)	5.05	3	657	1,525	326	214
UK-NH	THERIS	capillary fringe+ SZ (clayey sand)	1.03	3	2,906	110	65	591
Idstein	THERIS	UZ (loam with layer groundwat er)	0.06	0.7	>11,500	19	21	1,100
Landshut	THERIS	UZ (Quaternar y, swamp loam)	19	18	956	527	572	1,084
Manston (Ecologia)	RF	UZ (limestone)	32	2	68	913 ¹ 951 ²	56 27	61 29
Zeitz	RF	UZ (mixed)	600	2.5	4	660 ¹	55	83

Table 8: (Operating) Energy consumption of selected ISTT projects

* The energy consumption values refer to thermal energy (steam production) and electrical energy (remediation system, heating elements, match-box)

** "Cold" SVE: specific energy consumption during commissioning = best value because of the increase of the specific energy consumption due to the declining recoveries and constant energy consumption during the remediation process

*** ISTT specific energy consumption for the whole remediation including SVE, groundwater protection, and air and water treatment

¹ The mass balance study to assess the extent of the removed contaminant was based on organic carbon due to the presence of a contaminant mixture.

² During the remediation, an alternating operation was tested in addition to the parallel operation of the RF heating and SVE. From the discharge rates measured for this option, the presented key figures were calculated for a similar total recovery of contaminants.

3.9.2. Definition of remediation objectives for ISTT

The basis of an ISTT design are the delineation of the source zone, its position and its composition. Furthermore, the design includes an integral site analysis including the definition of desired remediation goals and site-specific boundary conditions (e.g. available space, building use, desired remediation duration).

The remediation objectives for a site are normally defined in the scope of the remediation plan and have to be approved by the local government. In this context, ISTT can enable a sustainable remediation of the source zone. Therefore, it is often decided to remove a high amount of the present contaminants. On numerous sites, the ISTT of the source zone is followed by further measures in the plume (e. g. temporary P&T downstream). In these cases, it is usually more economical to formulate operating and shut-down criteria of an ISTT on the basis of moderately higher residual contaminant concentrations in the source zone and further measures in the plume, irrespectively of the technical capabilities of an ISTT to remove even smallest residual contaminations. The limits of a long-term remediation objective like drinking water quality may not be applicable for a source zone application in the context of a treatment train concept.

On the other hand, appropriate shut-down criteria for ISTT can support economic and ecological solutions. In this respect, the development of contaminant removal rates via SVE and the groundwater (emission evaluation) and the specific energy consumption per kilogram of contaminant might serve as shut-down criteria, among others. Depending on the site it is a pre-requisite to achieve a defined target temperature (normally the coboiling temperature) and to maintain this temperature for a defined period of time taking into account the development and / or reduction of the contaminant output. The long-term remediation objectives (desired "residual contaminant concentration" or the remaining residual contaminant concentration) are also to be taken into account.

3.9.3. Ecological and economic considerations for semi-tropical and tropical regions

When applying ISTT in semi-tropical to tropical areas, it can be expected that less energy is needed for the sub-surface heat-up process due to usually higher sub-surface temperatures compared to the temperate zone. However, it is not clear whether moderately more energy is needed to treat an aggregated soil structure as opposed to a non-aggregated soil.

Even conventional SVE or P&T might be more efficient in semi-tropical to tropical areas than in the temperate zone. The contaminant recovery processes are limited due to diffusion even at temperatures of 20 to 25°C. As shown for several sites in the temperate zone by life cycle assessment balances, ISTT causes less environmental impacts and uses less energy than conventional methods like SVE or P&T [HIESTER & SCHRENK 2005]. Remediation costs are always site-specific due to site-specific boundary conditions like the location of the contamination source, the site's infrastructure or the remediation objectives. Hence, the estimation of the overall budget contains high uncertainties.

Nevertheless, data from one (European) site with different remediation scenarios are shown in Figure 3.11.

As shown, the specific remediation costs per m³ decrease with an increasing total remediation volume. Based on this information, some site and scenario specific cost factors like human resources and energy vary along the specific costs, others like measurement devices seem to be mainly independent.

If the presented site was located in Brazil, the local prices could be calculated roughly by using the site cost structure and trade information like [ASSUNTO.DE 2006; DEINTERNATIONAL 2010; GERMANY TRADE & INVEST 2011]. As an overall result, the remediation costs of a similar site in Brazil would approx amount to 75 % of the European costs, as shown in Figure 3.11.



Figure 3.11: Relation between remediation volume and specific remediation costs and impact of volume on cost items [reconsite GmbH 2012]

3.9.4. Economic viability of ISTT

The results of life cycle assessments cannot be financially quantified in an appropriate manner. An economic evaluation can include an increase in remediation costs due to other project-specific, relevant (additional) positions. For instance, building monitoring, protective devices and monitoring measures for the proximity or traversable remediation installations may result in costs without significantly influencing the life cycle assessment.

Usually, it is economically viable to use ISTT for remediating source zones. The remediation of large ISTT fields generally reduces the specific remediation costs per unit volume (per m³ soil]. Site-specific cost estimates are necessary to consider the multitude of parameters influencing a site remediation assessment.

4. Summary

In situ thermal treatment (ISTT) by means of steam-air injection, thermal wells or radiofrequency energy enables a fast, reliable and economically viable remediation of source zones. Organic contaminants can be removed from highly permeable, non-cohesive soils, cohesive soils with low permeability and fractured bedrock within a few months.

ISTT have proved successful in remediation projects in urban areas, below and in the vicinity of buildings in use, and at active and former industrial sites. Due to its efficiency, ISTT is used for an increasing number of brownfield redevelopment projects in order to combine fast source-zone remediation and project development.

These guidelines provide an overview of the various application options as well as on the limitations of the individual methods (see Table 9). It is feasible to combine different ISTT methods and ISTT with conventional remediation methods. Considering the importance of a fast remediation, such combinations can be appropriate in the case of complex tasks.

Pre-requisites for planning and implementing economically viable remediation concepts are a thorough assessment of the geological and hydro-geological conditions and a reliable delineation of the source zone. This applies particularly to remediation measures in the saturated zone. The remediation target values and monitoring methods must be determined on a consensual, site-specific basis as part of a risk assessment within the regulatory frameworks. This includes the (future) use of the area, of the soil and of the groundwater. Consensus must be reached with the authorities at an early stage of the planning approval, taking into account the valid legal regulations and the proportionality of the measures (cf. e. g. the pathways of effects of the BBODSCHV 1999). Information on boundary limits and evaluation methods including emissions can be found in the Federal and State Soil Acts, in instructions and guidance documents from the federal states, and the recommendations of the Federal States Working Groups on Soil (LABO) and Water (LAWA) in their respective valid versions. For example, in the case of point sources, a successful source zone ISTT may lead to a modification of post-closure groundwater protection measures [LABO / LAWA 2006].

Primarily fields of application		Steam-air injection (TUBA)	Thermal wells (THERIS)	Radio- frequency- energy (RF)
UNS	SATURATED ZONE			
Soil	type	1	1	1
. ≥	gravel	++	0	+
ION HES	sand	++	0	++
2 00	silty sand, sandy silt	+	++	++
IVE	silt	0	++	++
IESI	loam, marl	-	++	+
CO	clay	-	++ to +	+
Con	taminants			•
CHO	2	++	++	++
BTE	X	++	++	++
PRC)	0	+ to ○	+ to ○
PAF	4	-	∘ to -	0
S A T	TURATED ZONE			
Soil	type	1	1	1
ËR	gravel	+ to 0	-	○ to -
NF	sand	++	-	○ to -
AG	silty sand, sandy silt	+	+ to \circ	+
RD	silt	-	+	++ to +
JIT 4	loam, marl	-	++ to +	+
AQI	clay	-	++ to +	+
Contaminant				
CHC		++ to+	++ to +	++ to +
BTE	X	++ to +	++ to +	++ to +
PRC		0	+ to 0	+ to 0
РАН		0	0	0
EQU	JIPMENT COMPONENTS			
Energy supply for heating		gas, diesel, (electrical)	electrical	electrical
SVE – extraction unit		blower	vacuum pump (VP)	blower, VP
Cooling of extracted soil vapour and		imperatively	mostly	mostly
condensate separation			recommended	recommended
Extracted air treatment		e. g. activa	ited carbon, catalytic	oxidation
OTHER FIELDS OF APPLICATION		00.0/	00.0/	00.0/
		> 90 %	> 90 %	> 90 %
Remediation on freely accessible areas				
Remediation beneath and next to		++	++	++
build	dings	++		77
Mai	ntaining the building use	++ to +	++ to +	++ to +
during a thermal <i>in situ</i> remediation				
 partly possible / individual examination of boundary conditions 				

Table 9: Primarily fields of application and boundary conditions of ISTT

partly possible / individual examination of boundary conditions inappropriate boundary conditions for an economic application, individual examination necessary

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RECONSITE GMBH cover picture, Figure 1.1 – 2.5. 3.3, 3.8 – 3.11

Research Facility for Subsurface Remediation (VEGAS), university of Stuttgart Figure 3.1 – 3.2, 3.7

ECOLOGIA ENVIRONMENTAL SOLUTIONS LTD Figure 3.4 right

HELMHOLTZ CENTRE FOR ENVIRONMENTAL RESEARCH (UFZ) Figure 3.4 left and 3.5 – 3.6

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Exemplary fields of application and projects



Project description:

The centre of a CHC contamination with perchloroethylene of the unsaturated and saturated zones comprised an area underneath the building (approx. 200 m²) and courtyard (approx. 100 m²) of a former dry cleaners. Based on soil samples (850-3,800 mg CHC per kg soil), the vertical expansion of the contamination centre was restricted to 5 m below ground surface (bgs). Nowadays, the ground floor of the historical building is used as an artist's workshop and sales studio, whereas the upper floors are used for private residency. During a three-month lasting pilot test (2005), the radius of steam expansion in the saturated zone was determined to 4 m. Approximately 450 kg of PCE were removed from the upper aquifer and from a silty layer forming the transition zone to the unsaturated zone. Based on the thermal radius of 4 m and the injection depth of 7-8 m, the entire site was divided into four treatment sections to be treated sequentially during the full scale remediation (2010). Two injection wells were installed in each treatment section. An overall of eight steam injection wells, a surrounding ring of soil vapour and groundwater wells, and a horizontal soil vapour drainage underneath the building were installed. During the five-month steam injection period, starting from early summer 2010, the saturated zone above 5 m bgs was heated to the required azeotrop of 92°C. The contaminated groundwater fluctuation zone and the silt layer were heated to 85°C and 75°C respectively. During the ten-month lasting overall remediation period, 50 kg of CHC were removed from the silt area (2.5-3.5 m bgs) and the upper aquifer (3-5 m bgs). Reaching the target values of 10 mg CHC per m³ of soil air and 10 µg CHC/l in the groundwater, respectively, the steam injection was ceased at the end of 2010. After the completion of the subsequent two-month cooling phase, the target values were confirmed (13 µg/l in the groundwater and 1.1 mg/m³ in the soil vapour). During the remediation control period of two years, all downstream values and in the former source zone were below the target values indicating drinking water quality.



Project description:

As part of the SAFIRA II research project, a steam-air injection was carried out at the former hydrogeneration plant in Zeitz, Germany. The pilot remediation focussed on the area of the former distillation unit. The soil was heated to the boiling temperature of benzene (85°C) as the site is extensively contaminated with this hydrocarbon. A mobile module-based thermal *in-situ* remediation plant (MOSAM), developed for the UFZ, was used for remediation. This system was extended to be compliant with ATEX standards (benzene). The remediation field was thermally treated using three injection wells each equipped with two injection depths (saturated and unsaturated zone). Moreover, six soil vapour extraction wells surrounded the pilot field.

The objective of the pilot application was to prove a removal of contaminants from the saturated and unsaturated zone in excess of 99 % of the initial contaminant mass. The contaminant inventory was estimated at 1,700 kg benzene before the ten-month application. During the first phase with "cold" soil vapour extraction and air sparging, 4,050 kg benzene were removed. Initially, the unsaturated zone was thermally remediated for comparative purposes (2,280 kg benzene). The contaminant removal from the saturated zone was comparatively low (300 kg benzene). During the subsequent treatment of the capillary fringe and the treatment of a one meter thick silty layer by steam over- and underride, another 240 kg benzene were removed from above the groundwater level.

The remediation success was proven by testing the soil vapour and calculating the content in terms of the contaminant masses adsorbed and remaining in the soil. The average content after thermal remediation was 0.16 mg benzene per kg soil. This was verified 6 months after the remediation completion by way of soil samples (0.1 mg/kg in the unsaturated zone and 0.5 mg/kg in the saturated zone).

Due to the high benzene content in the unsaturated zone and the high recovery rates using cold soil vapour extraction, a conventional treatment represents an economically viable option. Since it is not likely to meet the remediation target values by the conventional SVE method a combined remediation of steam-air injection and SVE was proposed to remediate the site.

C) Removing CHC contamination from the unsaturated zone using thermal wells (THERIS method)



Relevant contaminants: Initial contamination (Σ CHC):	PCE; secondarily TCE and cis-DCE Soil air: > 1,000 mg/m ³ Soil: > 1,000 mg/kg (max. 1,985 mg/kg) Groundwater: > 400 µg/l (downstream up to	recon
Remediation goal:	1,000 µg/I) Source zone remediation in the unsaturated zone: 10 mg/m ³ during 4-hour extract at-	
Removed contaminant mass:	tempts per soil air level Approx. 353 kg Σ CHC, of which approx. 5 kg via groundwater	
	Veer 2010	



Before remediation start / recovery 0 kg CHC



Year: 2010

Remediation-relevant cubage: Area: 540 m² Depth: from 3 to 6 (7) m bgs Volume: approx. 1,600 m³

Remediation operation: 290 d (incl. cold SVE: 14 d)

Contaminant recovery: via soil air: 348 kg CHC via groundwater: 5 kg CHC

Specific energy consumption: soil air: approx. 1,500 kWh/kg groundwater: approx. 1,890 kWh/kg



5 months of remediation / recovery 316 kg CHC After remediation end / recovery 350 kg CHC voc Lager 1000 100 50 25 10 25 10 Wer We /erkaufsbe /erl Parkplatz Parknlatz Ausstellun Ausstellur

Project description:

The total area was split into four sections due to the boundary conditions. 2 and 3-metre long heating elements were used in the unsaturated zone. Soil vapour was extracted close to the surface in order to prevent contaminants from migrating into the ambient air. The primary contaminant recovery took place from the deeper located contaminant area between 4 and 6 m bgs. Waste air was cleaned using air active carbon. In addition, groundwater was pumped through a central well.

The four diagrams (above) illustrate the soil vapour contamination upon remediation commencement, after two and five months, and upon remediation completion (after release). After just four months, the soil vapour contamination had reduced compared to the initial contamination by approximately one order of magnitude. This is equivalent to a contaminant recovery of approximately 90 % of the initial inventory.

CHC pumping via groundwater was broadly constant during the remediation process and made a contribution of 1.5 % to the total recovery. CHC concentrations in the area of the former source zone were significantly reduced in the groundwater upon remediation completion. Two years after the completion of the source zone remediation, the groundwater values in the immediate proximity were between 100 and < 10 μ g/l.



Project description:

Conventional in-situ remediation (SVE, air sparging) was applied to remediate the contaminated area of a petrol station from October 2004 to October 2007. Although contaminants in excess of 2 tonnes were removed, a residual inventory remained in the subsurface.

During a subsequent THERIS pilot test, two more groups of heating elements of 3 m length were installed around two higher contaminated areas with already installed wells. 12 further soil vapour wells were installed in the periphery to support the SVE. The SVE was equipped with an emergency- shut-down to be able to immediately stop the operation in case of the formation of explosive gas-air mixtures (e. g. benzene). The waste air was cleaned using active carbon. The diagrams illustrate the transport of contaminants in form of benzene equivalent (BE, left) from the pilot field and the surrounding area, in addition to the average BE transport per well (right) in the course of the remediation.

Initially, a "cold" SVE was applied for two weeks. Then it was transformed into "warm" SVE with a moderate heat input (35 days). In the course of the "warm" SVE, the BE recovery from the entire field increased from approx. 70 g BE/day to approx. 100 g BE/day. In the subsequent THERIS operation (70 days) with high heat performance, the recovery increased by 2-3 magnitudes to up to 19 kg BE/day. The maximum recovery levels achieved on the days 49 and 91 resulted from the steam penetration in the smaller and larger partial areas.

After approx. 98 days, the contaminant transport in the pilot area reduced by two magnitudes within two weeks. Due to heat expansion, high recovery from the surrounding area was still achieved at the end of the pilot test (approx. 300 g BE/day). In total, approx. 60 % of the contaminant recovery was accounted for by the 2 wells in the THERIS field; approx. 40 % by the 12 SVE wells in the surrounding area.

E) Pilot application: RF treatment to support SVE at a former hydrogenation plant in Zeitz (Saxony)

Relevant pollutants: Initial concentrations:

BTEX (mainly benzene) Soil vapour: 60 g/m³ org. C (max. 90 g/m³) Soil: 100 mg/kg org. C (max. 250 mg/kg) A HELMHOLTZ

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Remediation goal: Demonstration of source removal in the unsaturated zone of a sandy to silty soil in depths between 3 and 8 m bgs, groundwater at approx. 8.5 m bgs

Removed contaminant mass: approx. 660 kg BTEX as org. carbon equivalent (org. C)



Project description:

Supplying an average RF power of 13 kW (15 kW RF system) to a specially modified, single RF electrode (E1) for 71 days, a soil cubature of approx. 500 m³ in a depth between 1 and 7 m bgs was heated up to an average temperature of 54°C (figures in the middle). The radius of influence of the RF heating was approx. 4 m. The installation of three extraction wells (E2-E4) positioned in a triangular array around the central RF electrode (Fig. top) supported the heat transport within the soil due to a co-current flow direction of soil vapour from the RF electrode to the extraction wells and therefore led to more homogeneous temperature profiles in radial direction.

The extracted soil vapour was treated using a catalytic combustion unit. RF soil heating increased the pollutant concentrations in the soil vapour measured at several soil vapour extraction points by a factor of 4 to 8 (figure bottom, left). Due to exceeding 20 % of the lower explosion limit of benzene in the extracted soil vapour, a dilution of the soil vapour with air had to be ensured during the entire remediation period. A rapid recontamination of previously cleaned soil compartments was observed, which was caused by a mobile organic phase floating in the ground water aquifer (figure bottom, right).

F) RF treatment to support SVE at a former petrol station, Manston, Kent, UK

Relevant contaminants: Initial contamination: BTEX and TPH Soil air: > 10,000 ppmv (max. 43,000 ppmv) Soil BTEX: 9,300 mg/kg (max. 66,500 mg/kg) Soil TPH: 6,400 mg/kg (max. 23,000 mg/kg)

Remediation goal: Source zone remediation in the unsaturated zone of chalk soil in depths between 2 and 7 m bgs, groundwater at 47 m bgs **Removed contaminant mass:** approx. 1,100 kg TPH/BTEX





Year: 2008

Treated cubature: Area: 132 m² Depth: between 2 and 7 m bgs Volume: approx. 660 m³

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Remediation period: 117 d (incl. cold SVE: 25 d)

Specific energy consumption: Approx. 61 kWh/kg

Figures:

L.: Setup of the field site R.: Temperature distributions



Project description:

Highest pollutant concentrations were detected in depths between 2 and 3 m bgs (middle figure). Supplying an average RF power of 22 kW (30 kW RF system) to the RF electrodes, which were arranged in a triangular array with a horizontal-spacing of 3 m for 27 days, soil cubatures of approx. 120 m³ each were heated up to mean temperatures of 49.1, 31.7 und 18.7°C in depths of 3.5, 5.5 and 7.5 m bgs, respectively (top figure).

The extracted soil vapour was treated using a granular activated carbon filter. After obtaining an average soil temperature of approx. 40°C (in 3.5 m bgs), the pollutant extraction rates increased by a factor of 10 in comparison to a cold SVE.

Based on the analysis of soil samples, the pollutant concentrations were reduced by more than 95 % for BTEX and light TPH fractions (< C_{10}) and by approx. 80 % for the TPH fraction C_{10} to C_{16} (bottom figure). Toluene und xylenes were removed by more than 99 %. In addition to the extracted mass of 945 kg of TPH/BTEX, about 172 kg of organic phase (mostly BTEX) along with 1,300 L aqueous condensate were recovered by the gas-liquid separator of the SVE.

G) Combined TUBA-THERIS remediation of the source zone at an industrial site

Relevant contaminants:

Initial contamination: **Remediation goal:**

Removed contaminant mass:

Contaminant cocktail, e. g. CHC, BTEX phase, aliphate, PAH Up to 140,000 μ g/l Σ AR in groundwater Removal of source zones in unsaturated and saturated zone: TUBA 1st aguifer -THERIS aquiclude - TUBA 2nd aquifer, downstream plume remediation Approx. 250 kg via SVE, approx. 31 kg via GW, phase pumping and condensation



TAMDOWN

REMEDIATION & ENABLING

steam-airinjection (TUBA) conductive heating (THERIS) steam-airinjection (TUBA)

Year: 2010



Project description:

The site was split into three stratifications, consisting of the 1st and 2nd aguifers and the aguiclude located between them. The two aguifers were heated using the TUBA method; the aguiclude was heated using the THERIS method. Steam-injection wells, and heating elements of 3 and 6 m length were used. The contaminants were recovered by twophase extraction wells (aquifer) or soil vapour extraction wells (aquiclude). Waste air was cleaned using air active carbon. Prior to this, the liquid phase was deposited, filtered, and cleaned using water active carbon.

In total, approx. 107 kg of contaminant were recovered from both aquifers and approx. 145 kg from the aquiclude. The four diagrams (above) illustrate the temperatures and soil vapour contamination in the 2nd aquifer after 50 and 80 days. A reduction in the contaminant concentration (top right) as a consequence of increased temperatures (bottom right) can be seen.

The project was classified at the Brownfield Briefing Awards 2011 in the Best In-situ Treatment category as highly commended.

Brownfield	
Awards	Nighly Commended
ARDS 2011	



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