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Haga Företagscenter - historical and preliminary technical investigation of a contaminated site in Anderstorp, Småland, Sweden

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Abstract

The *Haga Företagscenter* site in Anderstorp, Sweden has a long industrial history, which include the extensive use of chlorinated solvents. Trichloroethene (TCE) was used in large quantities here but the release to the environment has not been previously investigated. In this study, a historical investigation and preliminary technical investigation was carried out to identify the source zone and resulting groundwater plume, as well as to determine if contaminants are subject to degradation. Using depth-discrete groundwater sampling, TCE, dichloroethene (DCE), and at one location traces of vinyl chloride (VC) were detected at the site. The source zone of the contamination, having the highest concentrations of TCE and DCE, were identified below the point of a former TCE degreasing facility. Outside the source zone, TCE, DCE and VC concentrations were well below threshold values for drinking water, showing that TCE spreading is of a limited character. Stable carbon isotope data from compound-specific carbon isotope analysis (CSIA) revealed that TCE at the source zone is subject to degradation, producing the daughter compound cis-1,2-dichloroethene (cis-DCE). The low levels outside the source zone and the TCE degrading means that a minimally invasive remediation approach like monitored natural attenuation can be considered for the site.

Contents

Introduction

Context

Chlorinated solvents are some of the most common groundwater contaminants today. Due to their many industrial applications, especially in metal industries as degreasing and cleaning agents, they have been used extensively. Improper storage, handling, and disposal has resulted in chlorinated solvents being released to the environment. The physical nature of the substances makes them persistent sources of groundwater contamination, as they have the potential to readily migrate through the subsurface and dissolve over a long time to form a groundwater plume. (Pankow & Cherry, 1996). Trichloroethene (TCE), as is the focus of this study, has throughout the 20th century been the most used chlorinated solvent in Sweden, resulting in thousands of sites currently being potential sources of TCE contamination (Englöv et al., 2007). The *Haga Företagscenter* site in Anderstorp is one of many sites in Sweden where there has been a historical use of TCE, but the assumed release to the environment is yet to be confirmed and described by a technical investigation. This presents an opportunity to initiate an investigation in a first attempt to identify the presence, spatial distribution, and degradation status of the groundwater contaminants at this site.

Objective

The objective for this project is to determine the extent of a chlorinated solvent contamination at the *Haga Företagscenter* site in Anderstorp, Småland by conducting a historical investigation and preliminary technical investigation. The purpose of this is to a) identify the source of the contamination, b) estimate the extent of the resulting groundwater contaminant plume and c) to identify potential degradation activities. This information is of major importance for developing a remediation concept for the site and for the property owner to better estimate the value of the property.

Theoretical background and problem description

Chlorinated solvents as an environmental pollutant

In Sweden, contamination by chlorinated solvents is a relevant problem. According to the database of potentially contaminated sites, hosted by the County Administrative Board (Länsstyrelsen), there are currently 4741 objects (buildings, properties etc.) identified where the use of halogenated solvents has occurred (Länsstyrelsen, n.d.). The database does not distinguish between categories of halogenated solvents, consequently, what part chlorinated solvents make out. However, the same authority states that "thousands of sites" in Sweden have handled and used the chlorinated solvent trichloroethene (TCE) specifically, whereof workshops using TCE for degreasing being the most common site. In numbers, TCE-use peaked in the mid-1970s with over 11 000 tons/year, making out about half of the chlorinated solvent use in Sweden (Englöv et al., 2007).

Chlorinated solvents started being widely used in the 1950s and have since been released to the environment either by accidental spill or by what is now deemed improper storage, handling, and waste disposal. The solvents have been extensively used in industry, one example being as degreasing agents in surface treatment of metals. The dry-cleaning industry started using chlorinated solvents in the 1930s (Lohman, 2002). Although having a higher cost, there were benefits in their use for the excellent cleaning capabilities, as well as the non-flammable nature of chlorinated solvents, eventually replacing the petroleum products used since the early days of dry-cleaning (Doherty, 2000). Chlorinated solvents are common groundwater contaminants today due to their extensive use and that their occurrence in groundwater as well as their role as contaminants remained largely undiscovered for a long time (Pankow & Cherry, 1996).

Chlorinated solvents occur as dense non-aqueous phase liquids (DNAPL) which refers to their density being higher than that of water, and to their very low solubility in water. Owing to the low solubility (TCE: 1.1 g/L), and high density (TCE: 1.46 g/cm^3), when released in nature, the DNAPL will tend to sink through the subsurface, existing mainly in a separate phase from water. The DNAPL making up the source zone is commonly described to exist in two ways. In the first case, which is called residual DNAPL, some portion of the DNAPL remains in the sediment by occupying pore space as disconnected droplets or strings. In the second case, upon interception of a low-permeable layer in the aquifer, the DNAPL may come to rest on top of the layer in what is called *pooling*. This will occur if the capillary resistance of a sediment is sufficient to hold the mass of the DNAPL. Although the effect is greater the more fine-grained the material is, even different fractions of sand will allow for pooling. Over time, dissolution to the groundwater from the source zone leads to an aqueous phase plume forming and migrating in the direction of groundwater flow. Owing to their volatile nature, Chlorinated solvents may also exist in the unsaturated zone in the vapor phase. (Lerner et al., 2003).

A DNAPL's fate in an aquifer partially depends on the interactions with the material of the aquifer which it resides in. Pooling on top of a low permeable layer allows for the slow diffusion into and accumulation in the layer, and later the subsequent back-diffusion to the groundwater, once the contaminant concentration outside the low-permeable layer is low enough. This mechanism is one of the reasons for the long-term persistency of DNAPLs as groundwater contaminants (Pankow & Cherry, 1996).

Trichloroethene and degradation

TCE is a synthetic chemical belonging to the chlorinated ethenes. It was banned in Sweden in 1995, after which it could still be used under a licence (Englöv et al., 2007). Currently, it is listed in the EU regulatory framework (EC) No 1907/2006 *REACH* (2006) which states that after 2016, the sale and use of TCE within the EU (and consequently Sweden) is prohibited unless authorized. As of 2014, it is no longer possible to apply for authorization. The EU regulations, when put into effect, replaced the previous Swedish national regulations in this matter.

TCE may be subject to natural degradation given favourable conditions in the natural environment, and transformations facilitated by microorganisms play the greatest role. Highly chlorinated ethenes, such as TCE, have a high reduction potential and are thus likely to act as electron acceptors (Vogel et al., 1987). It is well established that under anaerobic conditions, TCE can partially or fully degrade through the process of reductive dechlorination. This was early on demonstrated in controlled settings (e.g. Bolesch et al., 1997; De Bruin et al., 1992; DeWeerd et al., 1998; Freedman & Gossett, 1989), as well as observed at contaminated field sites (e.g. 2005; Hunkeler et al., 2004; Lorah & Olsen, 1999).

TCE may undergo sequential reduction by hydrogenolysis, the stepwise removal of a chlorine atom, replaced by a hydrogen atom, making the daughter compounds less chlorinated in each step (figure 1). In this fashion, TCE transforms into one of three isomers of dichloroethylene (DCE): 1,1-DCE, cis-DCE or trans-DCE; which in turn transforms into vinyl chloride (VC); with the final step being ethene, which is fully dechlorinated (Vogel et al., 1987). Between the three isomers of DCE, cis-DCE is the most common and relevant in degradation, followed by the more rarely observed trans-DCE, and 1,1-DCE (Bradley, 2000). Although full reductive dechlorination is possible, partial dechlorination with DCE and/or VC accumulation as a result is more common due to their lower reduction potential compared to TCE. Other reasons for this stalling are non-ideal redox conditions, lack of halorespiring microbes, or limited access to compounds acting as electron donors. Another pathway that leads to the full breakdown of TCE is the initial anaerobic reduction to DCE and/or VC, followed by the anaerobic oxidation of DCE to VC or directly to CO2, and the oxidation of VC to CO2 (Bradley, 2000). The co-occurrence of the above-mentioned metabolites of TCE:

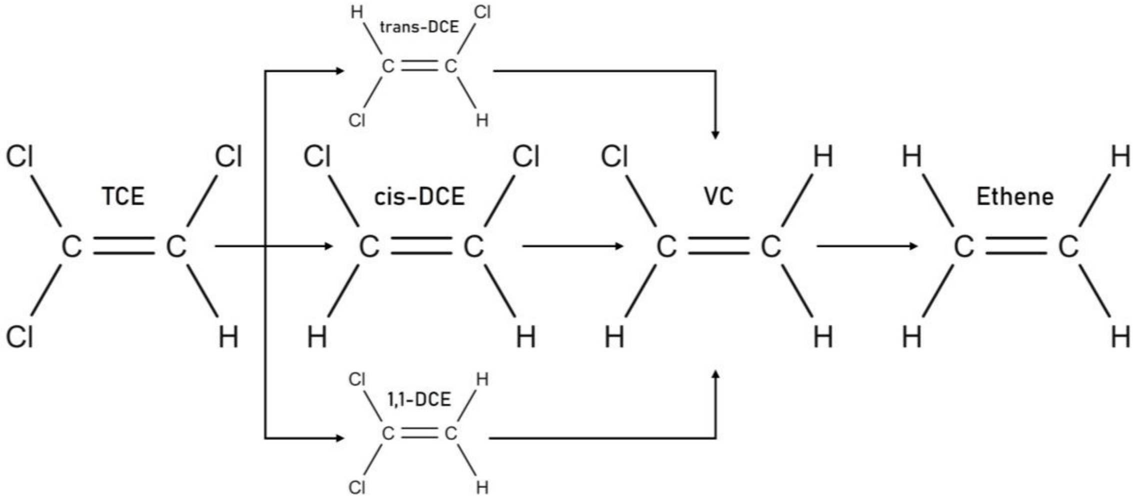

Figure 1. Sequential reductive dechlorination of the chlorinated solvent trichloroethene (TCE).

DCE and VC, is typically indicative of a site undergoing reductive dechlorination, although strictly speaking not a direct line of evidence since this would require proof that the chemicals were not directly disposed of at the site in the first place (Wiedemeier et al., 1999).

TCE and some of its daughter compounds formed during degradation are known to the European Union as carcinogens of varying degree and are listed in the regulation (EC) No 1272/2008 *Classification, labelling and packaging of substances and mixtures* (CLP) (2008) as follows: TCE (class 1B, *presumed carcinogen*); the less common dichloroethene isomer 1,1-dichloroethene (class 2, *suspected carcinogen*); VC (class 1A, *known carcinogen*). It is worth noting that the most common isomer of DCE formed during biodegradation, cis-DCE, is not a carcinogen according to the regulation. Swedish threshold values regulating maximum contaminant levels in drinking water exist for TCE as the sum of TCE and tetrachloroethene (PCE), and for VC. For DCE, there are no Swedish threshold values, so international guidelines may be used as a reference instead. Table 1 presents threshold and guideline values for the above-mentioned substances.

Although biodegradation plays the greatest role, abiotic degradation (not involving organisms) is also possible for TCE and other chlorinated hydrocarbons. In abiotic degradation, hydrogenolysis is an important degradation pathway for TCE, in which the sequential removal of chlorine atoms take place by replacement by hydrogen atoms, in the same stepwise order as shown in figure 1 (Tobiszewski & Namieśnik, 2012).

TCE degradation and its prerequisites are of interest when investigating the fate of the contaminant in the environment, for example when choosing a remediation strategy. Many strategies can be implemented in remediation to reduce contaminant levels, including physical or chemical removal of the contaminant. One strategy when dealing with a contaminant plume is monitored natural attenuation (MNA), where contaminant levels are allowed to reduce under observation within a reasonable time frame. Instead of human intervention, the method relies on natural processes such as dispersion and degradation to reduce contaminant levels. Alternatively, with the method of in situ bioremediation, the process is enhanced by technical intervention, by changing the natural conditions (bioaugmentation) as to allow for greater rates of

Table 1. Threshold and guideline values for drinking water, Swedish and international.

	Threshold values for drinking water in Sweden[1]	International guideline values for drinking water[2]
TCE (µg/L)	10 (PCE+TCE)	8
1,2-DCE (µg/L)	-	50
VC (µg/L)	0.5	0.3

[1]Livsmedelsverket, 2001. [2]WHO, 2022.

degradation (Kueper et al., 2014). The United States Environmental Protection Agency (USEPA) (1999) states that MNA is applicable when the contaminant only remains as a dissolved plume, whereas the contaminant source is better handled with other interventions to meet remediation goals.

Compound-specific isotope analysis for the assessment of degradation of chlorinated solvents

Compound-specific isotope analysis (CSIA) is an innovative tool that can provide unequivocal evidence for the degradation of a chemical compound. The method relies on the process of isotopic fractionation, which means that during degradation, a compound is being enriched in one isotope in relation to another, so that the ratio between the isotopes change. During both biological and abiotic degradation, fractionation occurs mainly due to the difference in bond dissociation energy, meaning that during chemical transformations, the breaking of bonds involving lighter isotopes is energetically preferred over that of heavier. Consequently, reaction rates will be faster for molecules with lighter isotopes, so that the remaining original compound attains a heavier isotopic signature from the (on average) heavier isotopes remaining. The product, or daughter compound, will consequently have a lighter isotopic signature (Sueker, 2001).

For chlorinated solvents, it is most common to investigate the stable carbon isotopes ^{12}C and ^{13}C. The natural abundance of these isotopes on earth are 98.93% for ^{12}C and 1.07% for ^{13}C (Laeter et al., 2003). When ratios of stable isotopes are close to that of the natural abundance, they are typically presented using the delta notation (δ) (Laeter et al., 2003). The isotopic ratio $^{13}C/^{12}C$ for the contaminant of interest is compared to that of a standard with a known isotopic ratio and expressed in $\delta^{13}C$, in permille (eq. 1). (Coplen, 2011). As a compound degrades, with the resulting enrichment of the heavier isotope, the $\delta^{13}C$ value will increase.

$$\delta^{13}C = \left(\frac{^{13}C/\,^{12}C_{Sample}}{^{13}C/\,^{12}C_{Standard}} - 1 \right) \cdot 1000\ ‰ \qquad (1)$$

The currently agreed upon threshold value to prove degradation is acknowledged by USEPA (2008) as 2‰ fractionation for carbon isotopes. Therefore, a change of 2‰ in the carbon isotope ratio is considered solid evidence for degradation. The threshold is based on studies that have determined that physical non-degradative processes such as dissolution, sorption and volatilization do not lead to significant carbon isotope fractionation for chlorinated solvents (Hunkeler et al., 2008).

Spatial variability of chlorinated solvent contamination

It is common to sample groundwater using conventional monitoring wells, which entails the sampling of an interval of the aquifer, using a long screen. However, two of the obvious downsides are the cost of installation and that the filter screens often cover such a long vertical length that a groundwater sample will show the average contaminant concentration for a large portion of the aquifer, giving a too generalized result. This can lead to over-interpretations of groundwater contaminant concentrations (Wiedemeier et al., 1999). Guilbeault et al. (2005) demonstrated the spatial variability of a contaminant plume that can exist even within homogenous aquifers. In a study, they found that contaminant concentrations varied orders of magnitude on as little as a 30 cm difference in sampling depth, highlighting the importance of depth-discrete aquifer sampling to better describe a contamination. As to what causes the spatial variability of a plume, logic asserts that there must be some heterogeneity in either physical, chemical, or biological properties of the system. For example, Parker et al (2008) showed that even very thin clay layers in an otherwise sandy aquifer can greatly affect the persistence of TCE, because of storage and back-diffusion from the clay, leaving only parts of the aquifer with elevated levels of TCE. In the case of (Guilbeault et al., 2005), where the aquifer had a uniform hydraulic conductivity, they attributed the spatial variability of contaminant concentrations to an uneven distribution of the DNAPL in the source zone. Due to the possible complex nature of a contaminant plume, with the possibility of spatial heterogeneity in composition, contaminant concentration and degree of degradation, a depth-discrete sampling approach should be considered the preferred way to delineate a contaminant plume. Using reusable groundwater samplers installed by hand with the direct-push method, as is used in this project, offers a cost-effective way to achieve depth-discrete sampling.

Study area

General

The study site for this project is in Anderstorp, which is a small town situated in the inland of the Swedish province Småland. This region is historically well known for its many small-scale industries, mainly in the metal, plastics, and leather industries (Nationalencyklopedin, 2021). The study site is a collection of properties in central Anderstorp with a long history of industrial activity. Currently, different companies occupy space in the buildings, and some of the warehouse space is leased for storage. While historical investigations in this project have revealed potential contaminant source zones, it is not obvious where a spill may have occurred during the industrial activity. Based on the industrial history of the study site, a single-component DNAPL plume mainly of TCE and possibly its breakdown products were expected to be present in the groundwater if a spill has occurred. An overview of the properties with respective property names is shown in figure 2.

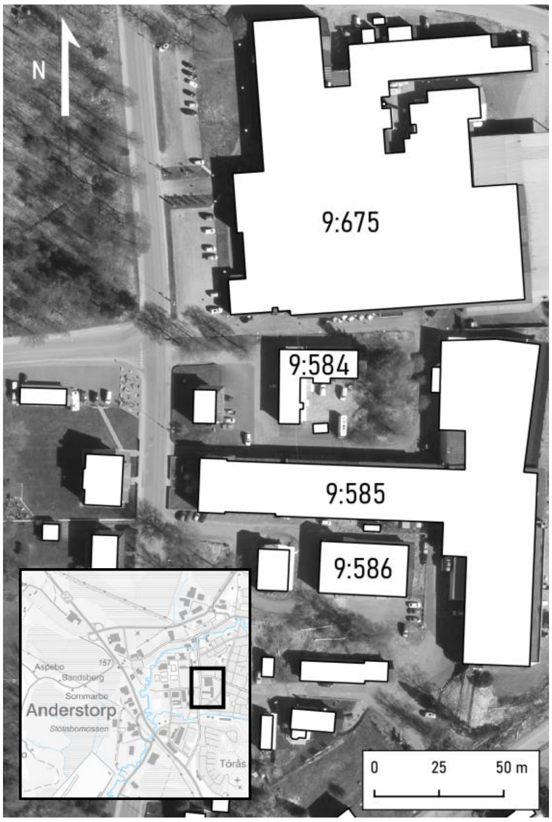

Figure 2. Study area with property name suffixes. ©Lantmäteriet.

Geology

Anderstorp is topographically confined within the Nissan River valley. Massive deposits of ice lake sediment and glaciofluvial material have filled the valley, being deposited by meltwater from retrieving glaciers when the ice sheet from the latest glaciation began retreating from the area between 13.5-14 ka ago. Ice lake sediment consisting of fine sand, silt, and clay is a general feature in the area below the topmost deposit of glaciofluvial sand. A typical feature directly on top of bedrock is sandy till, which drapes the bedrock topography with a thickness of 1-3 m (Sundevall, 2006). There are no available described geologic profiles in the vicinity of the study site, which means that apart from the geological information discovered in this investigation, the geology described for the larger area in general can be extrapolated to the local study site if needed.

Hydrogeology

To the south, and close to the site, Töråsbäcken flows westward into Anderstorpaån which in turn drains into the larger river Nissan, several kilometres downstream. The rivers are topographically confined within the same larger valley, surrounded by higher lying land with superficial bedrock. The theoretical groundwater gradient, inferred from the slope of the topography and the general direction of drainage in the area, is west to southwest.

Methods and materials

Historical investigation

A historical investigation was conducted to summarize the available information on the industrial history of the properties comprising the study site. This was mainly done to identify potential contaminant source zones, to support the development of a sampling plan for the technical investigation. The information on the industrial history of the properties is mainly derived from four separate MIFO phase 1 studies, one consultant report, correspondence with the property owner, as well as from conversations with people previously employed at the site. MIFO is a Swedish abbreviation translating to “method of surveying contaminated sites”, and it is the common way in Sweden to identify and evaluate contaminated areas in order assign a risk level for further action. Phase 1 is the initial study that gathers and summarizes information from existing data and conducted interviews to ultimately assign a risk class to a property (Naturvårdsverket, 1999).

Preliminary technical investigation

Field equipment

For groundwater sampling, the water sampling system *Water sampler DP 32/42* with an external filter, from the German company *Stitz* was used. These are temporary and reusable instalments offering the possibility for depth-discrete sampling at several locations reusing the same piece of equipment. The groundwater samplers were installed using a petrol-powered jackhammer (breaker) of the model *Atlas Copco Cobra TTe*. The jackhammer, originally designed for tamping ballast under railway sleepers, weighs 24 kg and is highly portable. With an adapter, the jackhammer is attached to the sections of the groundwater sampler which are then driven through the aquifer with a hammering action. For lifting of the groundwater sampler, a hydraulic lifting system, also from *Stitz*, was used. Soil core samplers, also installed with the use of the jackhammer and retrieved with the hydraulic system, were used to extract sediment cores to describe the geology. To sample the groundwater, a peristaltic pump was used. This is a suitable pump for environmental sampling since none of the possible contaminants are in physical contact with the pump’s mechanical parts, only with the tube that is installed in the pump, which limits cross contamination. A silicone tube was used for the extraction of water. Samples were taken in 40 ml glass vials suitable for VOA (Volatile Organic Analysis), with screw caps with PTFE coated silicone septa. To measure the chemical properties of the groundwater: oxidation reduction potential (ORP), pH, electric conductivity (EC) and temperature, a *HI-98194 multiparameter* from *Hanna Instruments* was used.

Installing the groundwater samplers

The groundwater sampler is installed with the direct-push method, which means that sections, in sequence, are driven through the aquifer to the desired depth of investigation. The process of installing the sampler is illustrated in figure 3. A jackhammer is used to drive down segments of the sampler. To expose the internal filter, the drive point, which is the tip of the sampler, is pushed out with an ejection rod inserted in the sampler, whilst lifting the whole sampler. This opens the system, allowing the internal filter, with the help of gravity and friction against the geologic material which collapses around it, to be extended and exposed to the aquifer. The drive point is lost during installation without the possibility of retrieval and is thus left in the ground. The equipment can be used for discrete-level sampling by pulling up the

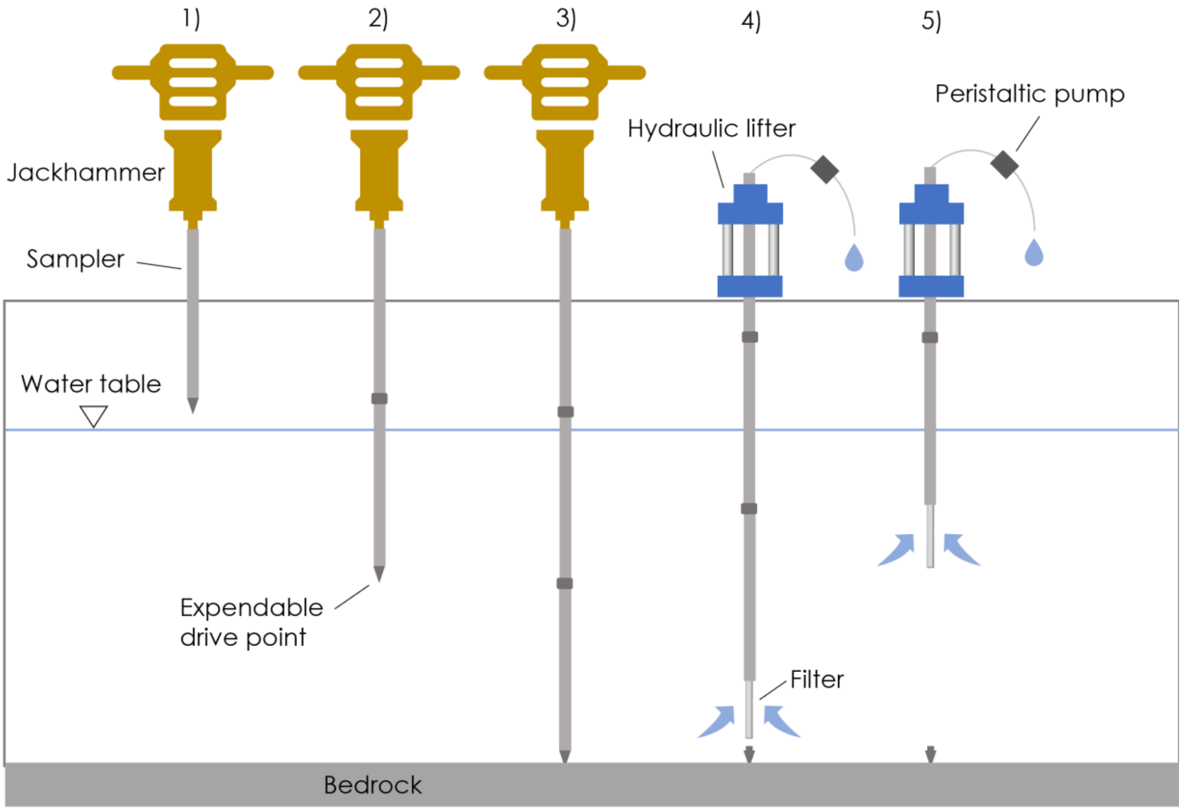

Figure 3. Installing the groundwater sampler and depth-discrete sampling. 1) The filter section is installed. 2) The sampler is extended by adding an additional section. 3) The sampler has reached bedrock and cannot go further. 4) The drive point is pushed out while the sampler is being lifted, allowing the filter to extend and sampling of the first level to be performed. 5) The sampler is lifted to sample a new level.

system and extracting groundwater at predetermined levels. The filter section was first installed in the ground, with extra care to drive it down vertically, as this establishes the direction for the following sections. Sections are driven down until ca 30 cm is protruding above ground, whereafter the next section is screwed on, tightened with wrenches, and equipped with an adapter before mounting the jackhammer again. Sections were installed successively in this fashion, until the sampler did not go down further, thus reaching what was assumed to be bedrock. At this point, the sampler was pulled up 5-10 cm, while the ejection rod was used to push out the drive point. After this, the sampler was lifted one filter length, exposing the length of the filter to the aquifer. The groundwater level was measured after being allowed to equilibrate, after which groundwater sampling could begin.

The speed at which the groundwater sampler is being driven into the ground can be considered a proxy for material properties in the ground mainly at the installation depth, such as grain size. This was not documented during installation of the samplers but could be used as information for when deciding on sampling levels, due to varying hydraulic conductivity related to grain size. It should be cautioned that since material is being pushed aside rather than removed when installing this type of groundwater sampler, the ground may become slightly raised at the surface due to the resulting upward pressure from the compacting material.

Determining the groundwater gradient

Due to an unexpected soil depth, several wells could not be installed at the same time for triangulation of the groundwater gradient. Instead, measured groundwater heads from three locations, measured on two consecutive days, were used. The three sampling points P1, P3 and W were used (figure 4). After determining groundwater heads relative to a fixed reference point, and the distance between wells, a conventional graphical solution for triangulation could be applied.

Groundwater sampling

To sample the groundwater, a tube was attached to the tape of a water level meter, which provided measurement of the depth to which the tube was lowered. The filter is 40 cm, and the sampling depth is considered the distance from the surface to the middle of the filter. The aim was to place the tube close to the filter. The tube was replaced between different sampling points, but due to practical reasons not between different sampling levels at the same sampling point. If the well was pumped dry, as indicated by air in the tube, sufficient time was given for the water to recover enough for sampling, or the well was raised in increments until reaching a more favourable level in the aquifer, with higher hydraulic conductivity. Samples were always taken with the aim of minimizing the introduction of air to the sample, samples were stored without any headspace in the vials.

Due to the small inner diameter of the groundwater sampler (~17 mm), effective purging of the sampler could be performed despite the relatively low flow rate provided by the peristaltic pump. The aim was to purge at least two times the volume of groundwater in the sampler, removing stagnant water to achieve a representative sample. As soon as the groundwater sampler is being lifted to a higher sampling level, the head of the water that remains in the sampler from the previous depth will need to equilibrate, forcing a flow into the aquifer from the filter end. If care is not taken, this could either introduce contaminants to an uncontaminated portion of the aquifer, resulting in a false positive result when sampling, or skew the concentration to appear higher, or lower, depending on the contents of the newly introduced water. Safeguarding from these possible errors are the purging procedure done at every sampling level, removing unwanted water from the sampler and the aquifer near the filter. Additionally, the sampler is raised relatively slowly, which means that water is released continuously on the way up, only insignificant amounts may be released at the new sampling level which can then be removed by purging.

Analyses

For concentrations of chlorinated hydrocarbons, samples were analysed at the commercial accredited laboratory *BEGAtec*, via the German company *Maul + Partner*

Ingenieurbüro GmbH. The method used was gas chromatography mass spectrometry with the static headspace sample preparation method (HS-GC-MS), according to the German standard DIN 38407-43. CSIA samples were analysed at *Isodetect GmbH.* The lab uses the method of gas chromatography combustion isotope ratio mass spectrometry (GC-C-IRMS). The method involves several steps to ultimately determine the ^{12}C- and ^{13}C-ratios for each analysed compound. First, the compounds are extracted from the sample by a purge-and-trap unit, then transported to a gas chromatograph for compound-specific separation. The compounds are then transformed to CO_2 during combustion, whereafter the $^{12}CO_2$ and $^{13}CO_2$ is separated and measured in an isotope ratio mass spectrometer (method description supplied by *Isodetect*).

Measuring of field parameters with a multiparameter meter

The probes were placed in a beaker with the electrodes at the bottom of the beaker, next to the tube. When pumping, the inflow of water generates an upward flow, passing the electrodes before escaping at the top of the beaker. The positive pressure in the beaker and resulting upwards flow ensures no contact with air and the method was thus deemed fit as an alternative to the commonly used flow-through cell not available at the time.

Results - historical investigation and sampling plan

Overview

In this section, the findings from the historical investigation are presented. Volumes of produced goods and amounts of chemicals used through the years are included when available to understand the scale of the production. Most important for this study are the locations where the use of TCE occurred, which is summarised below for each property for convenience.

Summary - TCE use

Anderstorp 9:675: TCE used 1950s-1990s, likely three TCE degreasing locations as confirmed by two sources.

Anderstorp 9:584: Conflicting information. No TCE use according to MIFO-investigation. TCE used according to accounts supplied by property owner.

Anderstorp 9:585 & 9:586: Degreasing has occurred, but no account of TCE use.

Anderstorp 9:675

This property is located on Brogatan 19 in central Anderstorp, and it contains the largest building on the site (figure 2). According to the MIFO investigation (Länsstyrelsen, 2002a), developments on the building have been performed several times, but since the last expansion in 1968 no new changes to the building have been made. The first industrial activity started in 1905 and at that time it was not more than a local smithy with a small-scale production. At the end of the 1920s the production increased, and some of the main products manufactured were balcony railings and gates. Industrialized production of e.g. rims for wheelbarrows started in the late 1930s. Other products manufactured were car parts, gas cans, galvanized valve grilles, etc. Not less than 80 tons of sheet metal goods were produced in the year 1972. Techniques used during production were cutting processes, machine pressing, tumbling, and coating. Surface treatment of metals (pickling) started in 1922. Galvanization likely occurred outdoors on a 3x5 meter cement plate, except during the 1950s, when the process was moved inside the building. The galvanization ended in 1969, but it is unknown when the process started. Degreasing with the use of TCE started in the 1950s and continued until the 1990s. The amount of TCE used between the years 1987 and 1991 was around 54 tons. Other chemicals used in the industry were cyanide, sodium hydroxide and sulphuric acid. 3000 L/yr. of these chemicals were used during the 1950s, and 550 kg in the year 1973 as the only specific examples. The amount of chemicals used during the years have a high uncertainty. TCE was used at different locations within the building, and they are presented in the sampling plan (Figure 4) The property owner has supplied approximate locations from the accounts of a carpenter who worked on the property for a long time. Locations are also provided from the MIFO-investigation which suggests three locations. Former employee L.E Larsson (discussion, December 8, 2021) provided a tour of the facility and identified three locations where he remembered TCE being used, they proved to be the exact same locations as supplied by the MIFO-investigations. During an inventory in 1990 it was documented that the building had three degreasing facilities of which only one was using TCE at the time. (Länsstyrelsen, 2002a).

Since 1995, the property is owned by *Haga Företagscenter AB* (org.nr. 556113-8552) and is used as a rental warehouse and rental offices. According to a liability investigation by COWI (2013), there is currently no property owner liable for contaminating activities.

Anderstorp 9:584

The property Anderstorp 9:584, is located on Brogatan 21 in Anderstorp. The present owner of the property is *Haga företagscenter AB*. According to the MIFO investigation (Länsstyrelsen 2002b), metal industries were active during the years 1935 to ca 1980 on the property. The processes that are deemed environmentally harmful started in 1935 and involved surface treatment of metal in the form of pickling as well as nickel, copper, and brass plating. Manufactured products include camping stoves, drum cymbals as well as various welded or forged products. After 1968, the industry was limited to pickling and a smaller facility for nickel plating which eventually ended in 1974. Regarding quantity of production, a quote from 1971 says surface treatment of metal was performed once a week, where a daily water usage for rinsing the metal was 300 l. At that time, 5000 kg of metal was surface treated per year. The building on the property has two levels and surface treatment of metals has occurred in various sections on the bottom floor. Chemicals listed that were used in the described processes above are: cyanide (and other unspecified acids), copper, nickel, and zinc. The only chemical which quantity is specified is nickel (200 kg salts of nickel per year). In addition to the chemicals noted in the MIFO phase 1 investigation, it is possible that TCE was used in the industry for degreasing, based on the information supplied by the property owner, shown in figure 4. These accounts differ from the information presented in the MIFO-investigation for this property, which, if true, implies that use of TCE at the property would have been missed in the MIFO phase 1 investigation.

There is no information regarding how disposal of process water from this property was handled before 1958, but between 1958-1972 it was led untreated via a storm drain to a creek (likely Töråsbäcken) which drains into Anderstorpaån. During this period, before 1968, cyanide was used in some processes. In 1972, a water treatment plant was installed on the property where treated wastewater was disposed of to the municipality’s sewerage system. (Länsstyrelsen, 2002b)

Anderstorp 9:585 & 9:586

At the site, there are other properties at which industrial activity has occurred. Two of these are Anderstorp 9:585 and 9:586. According to the MIFO phase 1 investigation (Länsstyrelsen, 2009), Anderstorp 9:585 was primarily used for plastic production but has also been owned by a company that performed surface treatment of metals for a short period. According to the MIFO phase 1 investigation (Länsstyrelsen, 2007), at Anderstorp 9:586, degreasing of metals is likely to have occurred, but the primary processes in production were galvanization and cutting of various materials such as steel and aluminium. Since surface treatment of metals have been performed at both properties, it is possible that TCE have been used at these properties, however, it is not specified if, or in which quantities.

Sampling plans

Sampling plan – 1st version

The sampling plan is based on the historical investigation. Previous studies, and secondhand accounts provided by the property owner have resulted in a hypothesis of two main sources of TCE contamination at the site, being the buildings on properties Anderstorp 9:675 and 9:584 (Figure 4). Based on this information, two sample points were planned in proximity of these possible source zones with three additional points to the west arranged in a profile, as this was in the direction of the assumed groundwater gradient. Sampling did not target properties 9:585 and 9:586 because they have not been suggested as sources of potential TCE contamination.

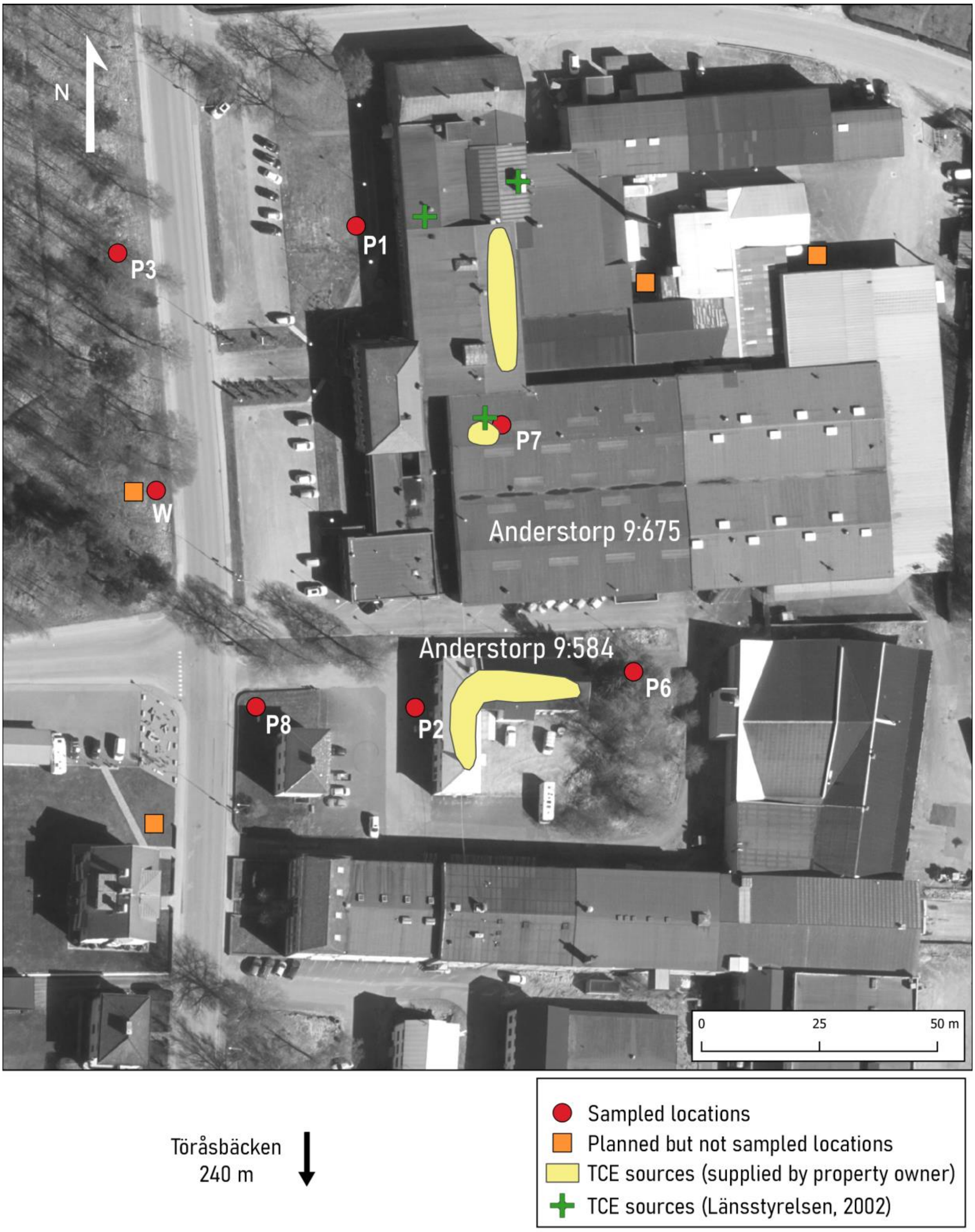

Figure 4. Sampling plan. Sampled as well as planned but not sampled points are included, with TCE sources retrieved from the historical investigation. ©Lantmäteriet.

The sampling plan include measuring the hydraulic gradient when three measuring points are established, to confirm or dismiss the estimated groundwater flow direction. This is important because it indicates in which direction a contaminant plume will travel. Once the groundwater flow direction has been confirmed, groundwater sampling is performed at five different locations within the investigated area. Two sampling points are located as close as possible to the supposed TCE source zones (one outside of each building at 9:675 and 9:584) to determine the concentration of TCE close to the assumed sources. The three remaining sample points are located along a transect 50-60 m west of the buildings and perpendicular to the estimated groundwater flow direction (figure 4). Sampling along a transect perpendicular to the assumed groundwater flow direction is performed to determine the extent of the plume. Requests for sampling south of Anderstorp 9:584 and 9:675 have been communicated by the environmental official due to concerns regarding a possible spread to a nearby housing area. This was considered but decided to not be included since the groundwater flow direction is estimated due west, meaning that pollutants would not reach south of Anderstorp 9:584 and 9:675. Additional support for this is near-surface bedrock at the housing area south of Anderstorp 9:584 and 9:675, which means that groundwater (and contaminants) is likely restricted from flowing south due to the upsloping topography of the bedrock.

Groundwater sampling is performed at different depths at each sampling point, starting from the bottom of the aquifer. The number of sampling depths is determined in field based on the aquifer thickness but should be 2-4. The number of samples are constrained largely by this project's budget. Furthermore, the exact location of the sampling points is also determined by practical limitations, such as below-ground gas and electric lines, as well as infrastructure.

Sampling plan 2nd version

The second version of the sampling plan is the result of initial findings effecting the suitability of the initial plan. The new sampling points (figure 4) were planned as to intercept a contaminant plume moving east according to the newly established groundwater gradient, which differs from the initially assumed direction. The purpose of P7 is to directly target one of the potential source zones. When arriving for a second sampling campaign, the groundwater levels in P2, left installed since the first campaign and W, contradicted the previously measured groundwater gradient. This suggests that the groundwater gradient at the study site is flat and fluctuates with time, barring any measuring errors, or that the gradient is a result from measuring in separate aquifer systems with different groundwater heads. This new information and the unforeseen discovery of a faint smell suspected to be TCE upon retrieval of the filter-end of the groundwater sampler at P2, lead to a final revision of this sampling plan. In the final revision, two points (P6 and P8) are placed on the eastern and western side of P2 respectively to make possible the interception of contaminants on either side. One sampling point is placed inside the main building at Anderstorp 9:675, directly on the location of a previous TCE degreasing station. Sampling directly on an assumed source zone is made to ensure the best chance of detecting contaminants to potentially identify a source zone and detect sufficiently high levels for CSIA analysis to be possible. The sampling of an assumed source zone was also requested by the environmental official at Gislaved municipality early in the project.

Notes on the sampling plans

Sampling points are labelled "P" for "point" followed by an identification number, except for the sampling point "W", a 2" pre-existing observation well, fortuitously discovered at a favourable location, from which samples could be taken. Samples are named after the sampling point followed by the sampling depth in cm below the surface at the sampling point (e.g. "P1-1780"). Two sampling plans have resulted in the final sampled locations, as shown in figure 4. The initial sampling plan was sent for revision and was later approved by an official at the Environmental Department at the Municipality of Gislaved.

Results – preliminary technical investigation

Field observations

The determined groundwater gradient was 86° east, which means that at the time it was roughly the opposite direction of what was assumed in the initial sampling plan. During the sampling campaigns, contaminants were only seemingly identified by smell in one location, P2, where upon extraction of the groundwater sampler, the filter smelled faintly of TCE. This was likely a misidentification, as indicated by the low contaminant levels found at the sampling point (figure 5). In some sampling points, the water had a distinct smell of hydrogen sulfide, indicative of an oxygen-poor environment with sulfate reduction (Appelo & Postma, 2005). Field parameters gathered with the multiparameter are presented in table 2. The redox conditions for all measurements are on the reductive side, with oxidation-reduction potentials (ORP) ranging between -277 and -33 mV. pH values show a normal, slightly acidic groundwater, between pH 6.17 and 6.97. Electric conductivity (EC) varies between 175 and 503 µS/cm, the variation in EC is directly related to dissolved ions, for example sulfate and chloride (SGU, 2013). The temperature has two distinct ranges depending on sampling point, close to 12.5°C for P7 and around 8°C for P2 and P6. Although the groundwater temperature is higher in P7, it should be noted that the field parameters were all measured during the same sampling campaign, in December 2021, but that groundwater extracted from P7 is beneath the large building at Anderstorp 9:675. P2 distinguishes itself from the rest having the most deviating values for almost all parameters (lowest ORP, highest pH, highest EC). Sediment cores were retrieved at P7 before the groundwater sampler was installed. The material and sediment retrieved from the cores is described based on visual inspection in the field and visualised as a profile (figure 5, inset). With the warehouse floor as a reference level, the profile consists of different materials and sediments with depth as follows: 0-20 cm: concrete; 20-90 cm: filling material (sand and gravel); 90-255 cm: medium sand; 255-315 cm: medium sand with silt; 315-370 cm: fine sand with silt.

Table 2. Physical and chemical field parameters.

Sample	ORP (±1 mV)	pH (±0.02)	EC (±1 µS/cm)	Temperature (±0.15°C)
P2-0809	-276.9	6.97	503	8.27
P6-0227	-74.5	6.28	320	8.11
P7-1307	-38.1	6.44	267	12.52
P7-0964	-37.7	6.28	218	12.19
P7-0621	-33.4	6.41	254	12.72
P7-0276	-50.1	6.17	175	12.61

Samples are named after the sampling point ID with the suffix indicating sampling depth below ground in centimetres.

Contaminant levels and spatial distribution

TCE and the compounds DCE and VC (at one location) were detected in the groundwater samples. Concentrations of these substances at respective sampling points and sampling depth are presented in figure 5. See the Appendix for a detailed report including all analytes. Swedish threshold values for drinking water are 10 µg/L for the sum of TCE+PCE (Livsmedelsverket, 2001), whereas international threshold values for DCE in drinking water are set at 40 µg/L (WHO, 2022). These threshold values are used as a reference to relate contaminant levels to a human health perspective. P7 is the only sampling point where concentrations for any contaminant exceed the threshold values. Here, the concentrations of TCE and DCE are highest at the topmost sampling depth and decrease significantly with depth. The highest concentration of TCE is 677 µg/L and the lowest is 6.73 µg/L, which is below threshold values for drinking water. For P7, the contaminant concentrations at the highest sampling level are considerably higher than that of the second highest sampling level, with 20 and 105 times higher levels of TCE and DCE respectively. For all other sampling points, the TCE levels range between 0.09 and 3.38 µg/L and the DCE levels range between 0.10 and 5.42 µg/L. VC is only detected in P3 at 0.15 µg/L, the Swedish threshold value for VC in drinking water is 0.5 µg/L (Livsmedelsverket, 2001).

CSIA results

The samples from sampling point P7, having the highest contaminant levels, were analysed with CSIA. All other samples had too low contaminant concentrations for CSIA to be possible. The results are presented in table 3 as well as in figure 5 as δ^{13}C with Vienna Pee Dee Belemnite (VPDB) as the standard, which is the conventional standard used in carbon isotope data. The sample ID is named after the sampling point and at which sampling depth in cm the sample is taken. What can be concluded from the results is that with depth, the values increase (become less negative), which means that TCE becomes more enriched in ^{13}C with depth. Sample P7-0276 has a δ^{13}C value of -23.8‰, and δ^{13}C for P7-0621 is -19.7‰. The isotope fractionation between these topmost sampling levels is 4.1‰. The two lowest sampling levels, P7-0964 and P7-1307,

Table 3. Isotope fractionation measured with CSIA.

		Sample ID	P7-1307	P7-0964	P7-0621	P7-0276
Trichloroethene (TCE)	δ^{13}C ‰ VPDB	Vial 1	-19.2	-18.1	-20.0	-23.9
		Vial 2	-14.5	-17.4	-19.3	-23.8
		Mean	**-16.9****	**17.8***	**-19.7**	**-23.8**
			(3.3)	(0.5)	(0.5)	(0.1)
1,2-cis-Dichloroethene (cis-DCE)	δ^{13}C ‰ VPDB	Vial 1	ND	ND	ND	-46.2
		Vial 2				-46.9
		Mean				**-46.5** (0.5)

* Measuring signal slightly below limit of detection, accuracy still typically within ±1‰.

** The values from vials 1 and 2 does not satisfy the criterion of reproducibility, although the mean is presented, the values are discussed separately.

have δ^{13}C values of -17.8‰ and -16.9‰ respectively. However, for P-1307, the values were not reproducible between the two vials that were analysed, and although the mean is presented, any interpretation should be made with caution.

Discussion

Chlorinated solvent concentration data

The higher contaminant levels in P7 combined with its proximity to the historical use of TCE, makes it a reasonable assumption that this point is a source zone for the contamination. Although the concentrations of TCE in P7 can be considered high from a health perspective, exceeding drinking water threshold values, the concentrations are still considered low in relation to the solubility of the substance (1.1 g/L). This finding is interesting as it relates to the nature of the contamination at the site. As a rule of thumb, a concentration exceeding 1% of the solubility is indicative of the presence of TCE in its DNAPL phase (Pankow & Cherry, 1996). The highest concentration at the site only amounts to 0.062% of the solubility, making the presence of free phase TCE unlikely close to the sampling point. If sampling point P7 is the source zone of the contamination, it is likely that TCE only exists here in the dissolved phase. In support of this, the actual release of TCE, as discovered in the historical investigation, must have occurred a long time ago, within the time frame of approximately 30 to 70 years ago.

DNAPLs are expected to readily move downwards, yet the highest concentration of TCE is found in the uppermost sampling level at P7, at a depth of 2.76 m below the surface. The most likely explanation is that the geologic material in the aquifer, which

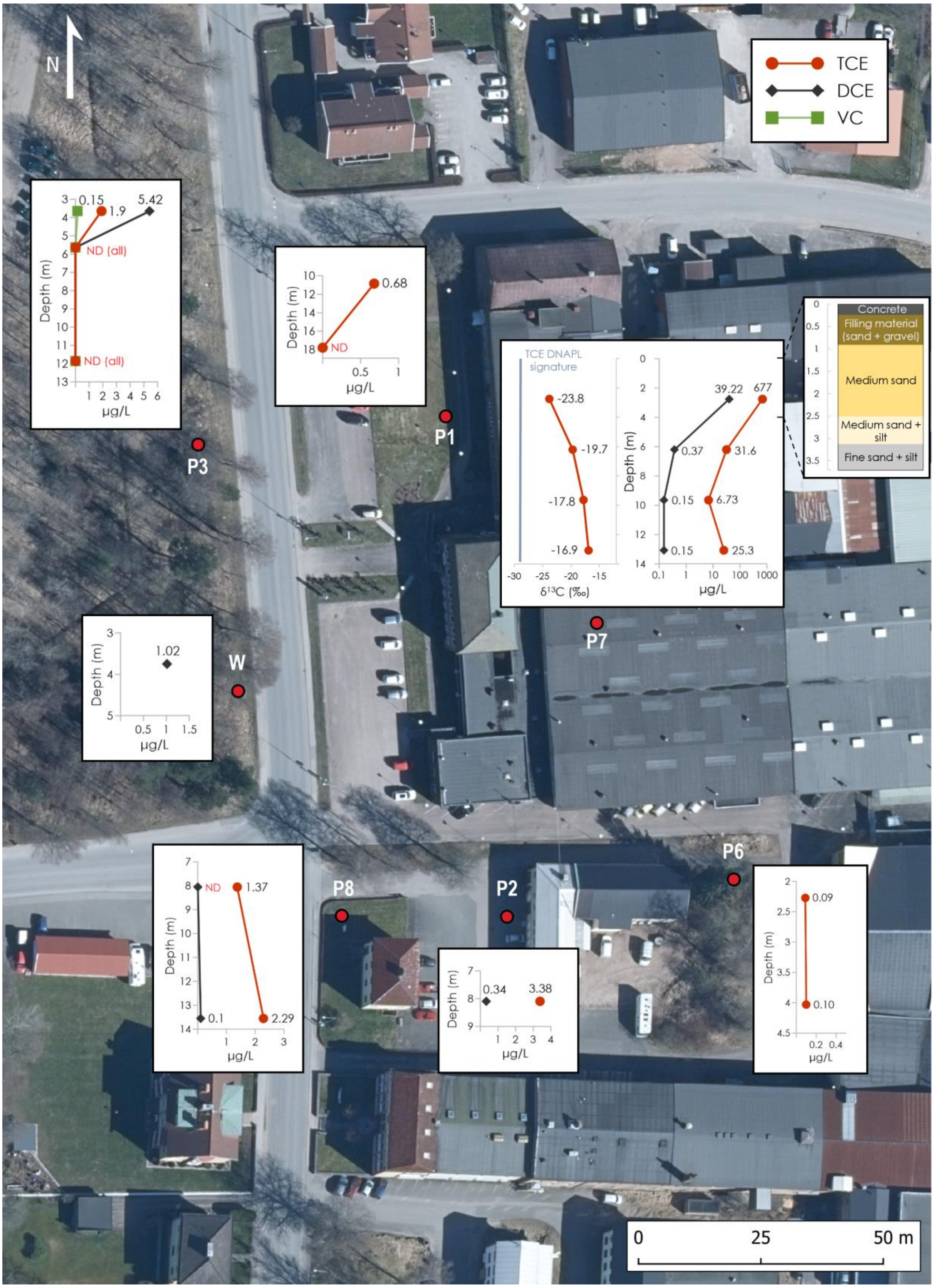

Figure 5. Contaminant levels, carbon isotope values, and geology. Contaminant concentrations in µg/L with depth for trichloroethene (TCE), the sum of all three dichloroethane (DCE) isomers, and vinyl chloride (VC). CSIA results with isotopic signatures for TCE in sampling point P7 The inset shows the profile derived from sediment cores taken at sampling point P7. Concentrations at P7 are displayed in semi-log. ©Lantmäteriet.

is fine-grained, has limited the downward transport of the DNAPL, leading to a resulting accumulation shallow in the aquifer. The sediment consisting mainly of fine sand and silt is closely situated below the highest contaminant levels (figure 5). As previously explained, the effect of "pooling" on top of fine-grained sediment is a well-known feature in DNAPL transport in the subsurface (Lerner et al., 2003). Another explanation that would account for the lower contaminant levels is that the degradation rate could be higher deeper in the aquifer.

Although the use of TCE may have occurred in the building at Anderstorp 9:584, as was noted by one source, only insignificant amounts of TCE were detected close to this building (sampling points P2 and P6). However, it is possible to have missed potential higher levels of TCE due to unsuccessful sampling at shallower levels at P2. It should be noted that it is possible for an industry to use TCE and not release it to the environment, which is one additional explanation for the low levels of TCE at these sampling points.

The low contaminant levels in all but one sampling point (P7) give credibility to the measured groundwater gradient being due east, since higher contaminant levels would otherwise be expected in the other sampling points as well, surrounding P7. As mentioned, a second groundwater measurement using only two measuring points still contradicted this gradient. This suggests that although the gradient could predominantly be to the east, the gradient is likely flat and fluctuates with time. Furthermore, the higher groundwater temperature at P7 compared to other sampling points can be explained by the large building providing insulation to a slow-moving groundwater, never allowing the temperature to reach the naturally cold groundwater temperatures for the sampling time (winter). This higher groundwater temperature, as it relates to degradation activities, is considered beneficial since bacterial metabolism generally increases with temperature (Wiedemeier et al., 1999).

Chlorinated solvent isotope data and degradation

From the CSIA results, it can be concluded that TCE is subject to degradation, as the presence of daughter compounds had indicated. The samples that filled the requirements for reproducibility show an isotope fractionation which exceeds the threshold value of 2‰ set by the USEPA (2008), which means that degradation of TCE is confirmed. Degradation rates cannot be inferred from the results but would appear to increase with depth given the diminishing contaminant levels. At first glance, this would seem to be supported from the CSIA results, which show that ^{13}C is being enriched with increasing depth. However, lower concentrations would, even given the same degradation rate, lead to higher fractionation, simply because the fractionated amount of TCE appears higher relative to the already low levels of TCE (Aelion et al., 2009). Comparing the isotope ratio ($\delta^{13}C$) of TCE found at the site with that of TCE from different manufacturers, literature values range from -33.5‰ to -24.5‰ (mean -

29.3‰, n=13). The δ^{13}C range at the site falls outside of this (-23.8‰ at lowest), which is expected due to TCE degradation. The DCE found at the site also has an isotope ratio different from values of manufactured DCE, supplied in literature, which range from -29.3‰ to -22.2‰ (mean -25.9‰, n=6), δ^{13}C at the site being -46.5‰, which is due to DCE being a product of degradation (Aelion et al., 2009). The pathway of degradation is evidently the breakdown of TCE to form DCE, but by which specific mechanism, including whether the degradation is of biological or abiotic nature, cannot be inferred from the results, and is beyond the scope of the study. The absence of VC in P7, which contains the highest contaminant levels, indicates that degradation terminates at DCE, which is a common outcome (Bradley, 2000). However, the possibility of DCE further degrading to form a product other than what is analysed for cannot be excluded, as that would require further studies. The field parameters of the groundwater support an anaerobic pathway of degradation, since the groundwater has a redox potential indicating reducing conditions. Although the smell of hydrogen sulfide would suggest sulfate reducing conditions, the redox measurements are not enough to categorize the redox processes (e.g. sulfate reduction, Fe(III) reduction etc.). The observed accumulation of DCE is preferred from a human health perspective to that of VC, which is a more harmful compound. No predictions are made concerning future levels of DCE, however, at this moment, levels of DCE do not exceed the international threshold values for drinking water for any sample.

The co-occurrence of TCE, DCE and VC in one sampling point (P3), if indeed originating from the source at the site, can indicate a chemical difference between the source and a more distant part of the plume. This means that while the source zone has characteristics restricting degradation to proceed past the formation of DCE, distant parts of the plume may allow for further degradation to at least VC.

Recommendations

Based on the contaminant levels at the site, it can be concluded that spreading from the source is of a limited character and are only causing low levels of contamination outside the source zone, well below threshold values for drinking water. However, this can currently only be said about parts of the site in directions emanating from the source zone that were sampled. For this reason, further sampling is recommended, mainly on the eastern side of the large building at the Anderstorp 9:675 property, as was originally planned but ultimately not performed in this study (figure 4, sampling plan). The TCE has been confirmed to be subject to degradation, which means that TCE concentrations have reduced since the release and should be expected to further reduce with time. Because of this, the need for extensive remediation efforts is likely not required and monitored natural attenuation should be considered as a potential remediation approach. At this moment there are no apparent exposure pathways which poses a risk to human health or the environment. Parallel with this investigation, the property owner arranged passive sampling of the indoor air over a period of 21 days, where no chlorinated solvent was detected, even next to sampling point P7 (correspondence, Volker Kelm, May 12, 2022). This means that even the highest measured concentrations, which are below the building, do not lead to vapor intrusion.

Conclusions

After conducting a historical investigation, depth-discrete groundwater sampling and compound-specific isotope analysis have been used to identify and describe a TCE contamination at the *Haga Företagscenter* site. The following conclusions are made:

- The historical use of TCE at sampling point P7 coincides with the highest concentrations of TCE and DCE being found at the site, which suggests that this is a source zone for the contamination. TCE concentrations are well below 1% of the solubility, indicating that while likely being released as a DNAPL, TCE may now exist only in the dissolved phase at the source zone.
- The vertical distribution of TCE at the source zone is heterogenous, where disproportionally higher concentrations of TCE and DCE are found at the topmost sampling level. Fine-grained sediment consisting of fine sand and silt is believed to have limited the downward transport of TCE, leading to the accumulation shallow in the aquifer.
- All sampling points outside the source zone have low levels of contaminants, below threshold values for drinking water, which means that the degree of TCE spreading is of a limited extent. Additional sampling is recommended on the east side of the source zone to gain a more comprehensive picture of the contamination, as this was not performed in this study.
- The CSIA results prove that TCE at the site is subject to degradation, where the pathway is the transformation of TCE to form DCE in the form of cis-1,2-DCE. The mechanism of degradation is not determined in this study.
- Due to TCE being subject to degradation and the spreading of contaminants being of a limited nature, monitored natural attenuation, which is less expensive than conventional methods, should be considered as a remediation approach.

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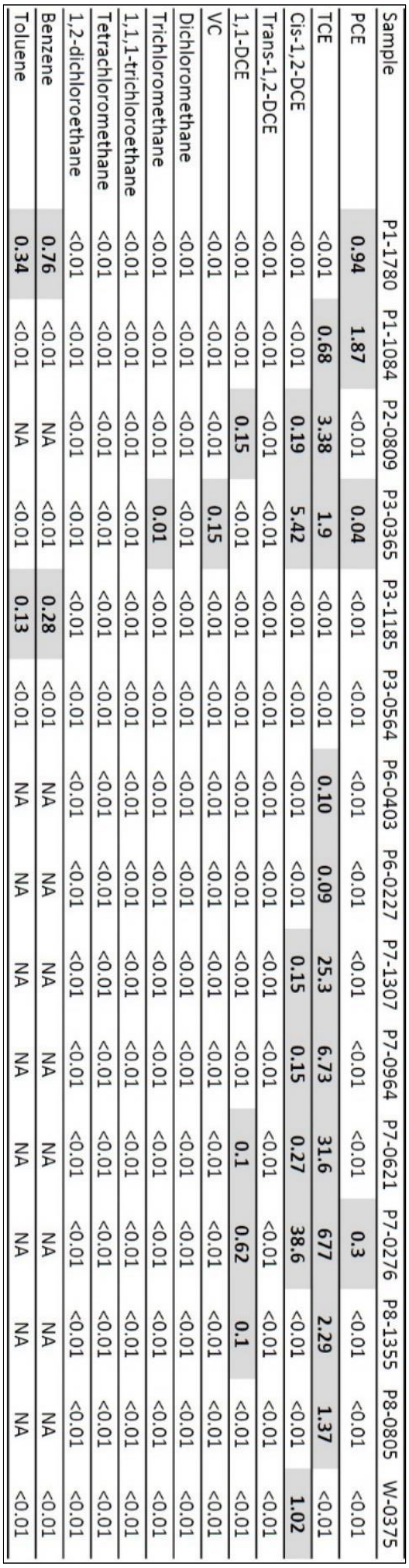
Appendix. Substance concentrations for all samples

All substance concentrations are in µg/L.



Sample	P1-1780	P1-1084	P2-0809	P3-0365	P3-1185	P3-0564	P6-0403	P6-0227	P7-1307	P7-0964	P7-0621	P7-0276	P8-1355	P8-0805	W-0375
PCE	**0.94**	**1.87**	<0.01	**0.04**	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	**0.3**	<0.01	<0.01	<0.01
TCE	<0.01	**0.68**	**3.38**	**1.9**	<0.01	<0.01	**0.10**	**0.09**	**25.3**	**6.73**	**31.6**	**677**	**2.29**	**1.37**	<0.01
Cis-1,2-DCE	<0.01	<0.01	**0.19**	**5.42**	<0.01	<0.01	<0.01	<0.01	**0.15**	**0.15**	**0.27**	**38.6**	<0.01	<0.01	**1.02**
Trans-1,2-DCE	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
1,1-DCE	<0.01	<0.01	**0.15**	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	**0.1**	**0.62**	**0.1**	<0.01	<0.01
VC	<0.01	<0.01	<0.01	**0.15**	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Dichloromethane	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Trichloromethane	<0.01	<0.01	<0.01	**0.01**	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
1,1,1-trichloroethane	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Tetrachloromethane	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
1,2-dichloroethane	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Benzene	**0.76**	<0.01	NA	<0.01	**0.28**	<0.01	NA	NA	NA	NA	NA	NA	NA	NA	<0.01
Toluene	**0.34**	<0.01	NA	<0.01	**0.13**	<0.01	NA	NA	NA	NA	NA	NA	NA	NA	<0.01