MAGPlan
Management plan to prevent threats from point sources on the good chemical status of groundwater in urban areas

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1 Summary

Working with volatile chlorinated hydrocarbons (CHC) over several decades has caused a severe groundwater contamination as well as a contamination of the mineral springs and certified spas of Stuttgart. Despite the site-specific remediation measures which have started thirty years ago and removed about 25,000 kg CHC in the inner city of Stuttgart, the contamination of the deeper aquifers has not changed significantly. Since 1988 no sustained decline in CHC concentration in the mineral springs and spas could be observed. This indicates that the mechanisms of lateral and vertical CHC migration between the complex structured layers of the Keuper and the mineral water-bearing layer of the upper Muschelkalk were not fully understood.

This is where the MAGPlan project began. The MAGPlan project applies the concept of "integral management of contaminated sites". It focuses on the spatiotemporal analysis of CHC migration and the associated degradation and transformation processes between the contamination sources and the mineral springs and spas. Due to the complex hydrogeological conditions, new and advanced investigation strategies and methods are used, which in combination allow for multiple lines of evidence. The overarching principle is an integrative-adaptive approach with a gradual improvement of the level of knowledge. The hydrogeological conditions and the chemical mechanisms derived from this approach, as well as from extensive existing data sets, are combined in a conceptual (hydrogeological) model. This consists of two modules, the conceptual aquifer model describing the hydrogeological system and the conceptual contaminant model describing the chemical processes in the system. Important elements of the conceptual contaminant model are so-called “site profiles” with substance-specific information on 21 sites. These sites contribute significantly to the CHC contamination of the groundwater. The compound-specific carbon isotope analysis of CHC substances shows that degradation and transformation processes substantially influence the current contamination situation. In addition to reductive dechlorination, there is clear evidence for selective degradation of TCE under aerobic conditions. This is also confirmed by laboratory tests.

Based on the conceptual hydrogeological model, a numerical model with 17 layers was built to simulate the CHC transport in groundwater. The calibration of flow and transport was performed in an iterative process by varying hydrogeological parameters. After calibration, the numerical model was validated with data from environmental tracers (sulfur hexafluoride) and isotopes (tritium), and layer-specific transport parameters for the Gipskeuper, the lower Keuper, and the upper Muschelkalk were determined. The tracer test data shows that the double porosity behavior of the upper Muschelkalk only plays a significant role when simulating the results of tracer experiments, but is negligible for CHC transport.

The CHC migration was simulated in a multi-species model, accounting for the degradation and transformation processes. The model incorporates both reductive dechlorination and oxidative degradation and distinguishes between three zones:
strongly anaerobic conditions, moderately strong anaerobic (post-oxic) conditions and aerobic conditions. During calibration, the roughly outlined zones of the conceptual contaminant model were further differentiated and compound-specific degradation coefficients were estimated for each zone. Sensitivity studies showed that the sorption rates of CHCs must be significantly lower than expected from the measured organic carbon contents in the Muschelkalk.

The calibration was carried out for the period of 1960-2010 by comparing observed and calculated CHC concentration time series. The focus was on reproducing the order of magnitude of the concentrations and the characteristic temporal evolution of the individual substances and with the objective to predict correctly the impact of individual sites in the project area. The release rates of CHCs at the relevant sites were based on downstream monitoring wells and the CHC extraction by hydraulic remediation measures, such that only the effective load from the sites was considered in the model. The results of transport modeling shows that the identified „profile“ sites fully characterize the spatial distribution of the groundwater contamination. With regard to the vertical migration of CHCs into deeper aquifers, five „key locations“ can be identified. The plumes up to three kilometers long, which emanate from some sites in the southern part of the project area, do not reach beyond the city center around the main station within the Muschelkalk aquifer. PCE is first transformed to TCE under anaerobic conditions and then mineralized in the oxidative degradation zones. The contamination of mineral springs is mainly caused by three sites in the near vicinity of the springs. With this information, it is possible to reproduce the basic contamination patterns in the mineral springs and spas.

The mass balance of the CHC transport indicates a remaining PCE input of 60 g/d in 2010 and a TCE input of 20 g/d. Prior to the start of the remediation measures, the CHC discharge from the source zones was much higher. Within the emanating plumes, up to 2,000 kg PCE and 100 kg TCE were stored. The main fraction of the mass was stored in the Gipskeuper, as there the flow velocities are comparatively low. Since the Muschelkalk aquifer shows a high flow rate, the CHC concentrations are significantly diluted when flowing from the lower Keuper to the Muschelkalk. Thus, a PCE and TCE mass of 5 kg is stored, respectively. At the mineral springs and spas, about 20 g/d CHC are discharged in total. This corresponds to an average CHC concentration of 1 µg/l and a total outflow of 225 l/s.

The results of the integral investigation of the groundwater and contaminated sites and the measures required to ensure a good groundwater status are summarized in the groundwater management plan. In reports, maps, data bases, and with a computer-based visualization tool, it provides the basis for an optimal treatment of CHC contaminants in groundwater. The management plan sets priorities for contaminated site remediation. The numerical transport model provides the opportunity to analyze different remediation scenarios for individual sites and to make predictions of future contaminant migration. Sites with a particularly high remediation priority can be identified. Furthermore interactions between contamination sources and receptors in the project area can be included in the risk assessment and the
evaluation of the sites. The remediation feasibility study as part of the groundwater management plan describes the measures to be taken in order to achieve the remediation targets. It is designed for those sites, where the model predictions have indicated the need for additional investigation and remediation efforts. In order to verify the impact and success of the remediation concept, remediation measures have to be controlled in a monitoring program. The groundwater management plan becomes binding through the involvement of the responsible boards and agencies.

The strategy of contaminated site management is a project “from the practice to the practice” and, as a guideline, leads the way for the future groundwater protection in Stuttgart.

2 Integral management of contaminated sites in Stuttgart – basics, strategy and approach

2.1 Strategic approach and basics

The conventional single site management of contaminated sites is predominantly applied and considered state of the art in the management of contaminated sites. This approach has proven itself in the handling of small-scale singular contamination. It corresponds to the polluter pays principle. However, large-scale contamination of groundwater has occurred in many areas which were used intensively in the past for industry. If several adjacent or even overlapping contamination sources emit pollutants, they can – especially in the case of mobile volatile chlorinated hydrocarbons (CHC) – create complex pollution patterns downstream the source areas. Here the current practice of site specific procedures reaches its limits. The contribution of individual contaminated sites or contamination sources to the overall pollution pattern can hardly be assessed. This is especially the case, if multiple sites emit the same pollutant group (such as CHC), complex hydrogeological conditions determine the contaminant transport, and degradation and transformation processes modify the pollutant spectrum along the flow path. In such cases often no liable polluter can be identified. The situation described can only be resolved if all sites potentially affected are investigated simultaneously and groundwater situation apart from the contaminated sites is taken into account.

Another important aspect is to assess the impact of contaminated sites on the affected receptors. In Figure 2.1, the integral investigation strategy is illustrated, whereby the groundwater is investigated beginning at the receptor all the way back to the contamination sources. The integral strategy requires to gain an understanding of the whole hydrogeological system, which determines the processes between the contamination sources and the vulnerable groundwater or groundwater use. Building on this, prioritization can occur, setting priority treatment on those contaminated sites that significantly contaminate groundwater and the other receptors.
In Stuttgart the integral approach has been used since 1996 (Figure 2.2). It began in the project INCORE first with an investigation in the Neckar Valley [1, 2]. In this total area of 5.2 km², 129 industrial sites and 15 waste deposits, of which 51 were in overlapping areas, were investigated using the integral investigation approach. Based on 34 integral pumping tests at seven control planes, a two-dimensional numerical groundwater flow and transport model was built for the quaternary gravel aquifer in the Neckar valley and an area of 46 hectares. Under observation was the mass flux of petroleum hydrocarbons, polycyclic aromatic hydrocarbons (PAHs), aromatic hydrocarbons (BTEX), total CHC and ammonia. In the results, the key sources of the pollutants mentioned and the geometries of the pollutant plumes were determined.

The concept of integral management of contaminated sites was developed as part of two further projects: MAGIC and FOKS in Stuttgart-Feuerbach for an area of 316 hectares [5, 6, 3]. In the MAGIC project an integral investigation approach was processed with five control planes and 37 integral pumping tests in a three-dimensional numerical flow and transport model. The pollutant concentration trend
obtained by means of integral pumping tests played an essential role in the calibration of the model.

In the follow-up project, FOKS, the focus was on identifying the emissions of contamination sources and the prioritization of remediation needs. They found that of the 193 CHC suspected sites handled in Stuttgart-Feuerbach, four areas require priority action, because more than 90% of CHC input into the groundwater in Feuerbach [3] originates from them. Thus, the tools were provided to assess in a further step such a hydrogeological complex area of 26.6 km² with eight aquifers as the Stuttgart valley basin upstream of the mineral springs and certified spas.

A general aim of the MAGPlan project is to demonstrate the application of the integral management of contaminated sites in an urban area with complex hydrogeological conditions. The comprehensive summary of the results was published in [11]. In the project implementation new and continuously developed investigation strategies and methods are used. Some of them were developed in the projects mentioned above and are summarized in the LUBW guidelines [4].

The overarching principle is the integrative-adaptive approach, as apparent in Figure 2.3: various strategical and methodological approaches are compiled and ready to be selected depending on the task. An optimization in several turns (iteration) are gone through, in each of which improvement in the level of knowledge will be sought. In doing so, it should be always questioned what strategies and methods are specifically appropriate or need to be adjusted and thus lead to success in the simplest way (adaptation). A specific project objective is to create an overall picture for the 26.6 km² urban project area. To do this, spatial concepts of hydrogeology, groundwater flow, the environmental conditions and the mass transport, i.e. a system and process understanding, must be developed. The focus is – as opposed to single-site handling – the overall picture, i.e. the space and its effects, and the processes involved. The space is characterized by the structure of the geological system, the location of contaminated sites and the receptors, i.e. assets endangered or affected by the contaminants.

The size and complexity of the MAGPlan project area require a review of the procedures, applied successfully in previous projects, and their further development. The concept of determination of pollutant loads, based on integral pumping tests along control planes, is only applicable here to a limited extent. It is beyond any financial scope with regard to the number of control planes required in the different aquifers. However, integral pumping tests provide important results and facilitate mass flow rate calculations for individual areas. The extrapolation to the spatial dimension is calculated with the groundwater flow and transport model. It is equally used as an investigation, analysis and forecasting tool, thus making it the core feature of the project.
Both, the characteristics of the integral study and the build-up of the complex groundwater flow and transport model, require a step-wise approach, as practiced in MAGPlan. Based on the analysis of the set of extensive data already available, initial investigations are focussed on the groundwater flow as well as for further characterization of contamination sources and their pollutant emissions. Next, the transport conditions are explored in groundwater. Then follows the detection of microbial degradation and other attenuation processes. All findings are merged in a conceptual (hydrogeological) model, consisting of conceptual aquifer model and
conceptual contaminant model. Based on this, a numerical flow and transport model is developed. In the course of further project work the adjustment between hydrogeological model and numerical model is done iteratively. With the calibrated reactive transport model, the basis is laid for further work on remediation plans and pollution-progress forecasting as well as the preparation of monitoring plans.

The investigation strategy is based on the combination of different research methods. These include innovative methods alongside conventional standard methods. The combination of different methodological approaches for multiple lines of evidence proves to be effective in the project. Variance in interpretation of individual results or even uncertainties in the system and process understanding can be further narrowed down [7], in particular by the combination of hydraulic, chemical and forensic tests.

Overall, the investigations carried out in MAGPlan are primarily aimed at understanding the processes on the regional scale. For this, spatially integrating methods that cover a large volume in the investigation area or provide spatially averaged characteristic values are of great significance. In reality, however, this understanding is only reliable, if it is linked with findings from the local scales. Underground exploration activities must at least backup important steps in the laboratory or core scales, the local scales (site-based) and the regional scale (MAGPlan project area). Table 2.1 provides an overview of the methods used in the project and the scale of the related investigation areas.

Table 2.1 Methods used, focal points and scope of the investigations taking into account the scale. The laboratory and drilling core scale (E1 < 1 m), the local scale (for example contaminated site with a direct discharge (E2 from 1 m to < 500 m) and the regional scale (the project area, E3 > 500 m to < 50,000 m) are distinguished between [11]
2.2 Initial situation in Stuttgart

Stuttgart’s city center was characterized in the past by intensive commercial and industrial usage. Many small business sites distributed over the area were characteristic of the Nesenbach valley. The decades of dealing with highly volatile chlorinated hydrocarbons have resulted in a high hazard potential with respect to groundwater, mineral springs and certified spas. As at present the city sources its drinking-water not from the urban area, the efforts for groundwater protection are primarily focused on the mineral springs and certified spas. In Stuttgart the assessment of groundwater contamination was characterized from the beginning by the situation that this natural resource was affected or threatened. All measures taken had the objective to restore its natural purity.

Stuttgart is one of the few big cities in Europe with extensive mineral water resources. Groundwater recharge of the Stuttgart mineral springs and certified spas is taking place in the South of the city (Region of Sindelfingen) in the karstified limestones of the upper Muschelkalk. The mineral water flows from there NE under the Stuttgart basin through to the Cannstatter basin, where it is forced to rise and sluice. The water from the upper Muschelkalk discharged in the springs, in the Neckar valley sediments and into the Neckar itself amounts to about 525 l/s (see Figure 2.4).

Figure 2.4 Morphology and geology in the area of the Stuttgart basin – viewed from the southwest [11].
The CHC investigations of groundwater started in 1983 show contamination of many wells within the urban area. Even the low and highly mineralized Bad Cannstatter and Berger mineral waters are affected by volatile chlorinated hydrocarbons. Based on current information, CHC input takes place in the limestone aquifer in the Stuttgart urban area itself. Numerous groundwater wells and springs in the upstream areas are consistently free of pollutants.

In response to the detection of CHCs in the mineral springs and certified spas, Stuttgart started in 1984 to register those sites, which were suspected to be contaminated [8]. The systematic management of contaminated sites has been initiated with the widespread survey of potentially contaminated areas in the urban area done 1993 to 1995. 3,915 suspected contaminated sites were detected in Stuttgart [9]. The data collected still forms an important basis for the management of contaminated sites. The data is updated annually.

Figure 2.5 Relevant CHC suspected sites in the project area [11].

After detailed evaluation and selection, 182 sites in the project area remained relevant. Of the 182 sites, 21 sites showed high CHC contamination of the
subsurface in previously performed investigations (red dots in Figure 2.5). For 161 sites, which were not yet technically investigated, relevant CHC input cannot be excluded. The 161 sites are shown as yellow dots in Figure 2.5.

By combining directly adjacent and similar-used land, the 21 sites mentioned above were reduced to 19 sites, which are described in site profiles of the most serious CHC pollution in the project area.

Table 2.2 CHC removal by various remediation activities in the project area over the years 1985 to 2010 [11]

<table>
<thead>
<tr>
<th>No.</th>
<th>Site</th>
<th>SVE</th>
<th>GE</th>
<th>Excavation</th>
<th>Total</th>
</tr>
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<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Removed CHC mass by remediation until 2010 [t]</td>
</tr>
<tr>
<td>1</td>
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<td>-</td>
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<td>-</td>
<td>262</td>
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<tr>
<td>6</td>
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<td>161</td>
</tr>
<tr>
<td>9</td>
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<td>1.540</td>
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<tr>
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<td>79</td>
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<tr>
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<td>46</td>
<td>-</td>
<td>590</td>
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<td>155</td>
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<td>574</td>
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<td><strong>4.524</strong></td>
<td><strong>1.238</strong></td>
<td><strong>23.177</strong></td>
</tr>
</tbody>
</table>

No. = number of site profile, SVE = soil vapor extraction, GE = groundwater extraction

In the project area, remediation measures of the subsurface were undertaken at 30 sites. The first measures were started in 1986, most of them, however, were started during the period 1988-1995. At 14 sites the remediation continued throughout 2014. The CHC removal in the project area achieved until the year 2010 is summarized in Table 2.2. The information for the 19 site profiles is displayed in detail, the additional 11 sites are recorded summarily. It can be seen that more than 1,000 kg CHC was removed at four sites respectively, thus amounting to 89 % of the total quantity removed. The total amount of 23.18 tons is composed of
• 17.42 tons of CHC through soil vapor extraction from the unsaturated zone,
• 1.24 tons of CHC by excavation during construction or remediation measures,
• 4.52 tons of CHC from the saturated soil or groundwater.

In addition

• 2.25 tons from the water for industrial use since 1960.

On adding, it is clear that the remediation measures since 1983 have led to a removal of approximately 25,000 kg CHC in Stuttgart city center. Moreover the extensive investigations have also improved the data regarding hydrogeology, site history, contaminant origin and contaminant fate and transport [10].

However, the stagnation of CHC concentrations in the mineral springs and certified spas since 1988 makes clear, that the extensive site-related measures alone are not sufficient to understand the processes occurring in the subsurface to a sufficient extent. A sound remediation strategy can be defined only if the hydrogeology and the processes taking place in the underground of the Stuttgart Nesenbach valley can be comprehensively described and understood. The MAGPlan project was carried out to fill the existing gaps. It leads on from previous practice of a site-specific approach following the polluter-pays principle, towards a more integral and holistic investigation.

2.3 Literature Chapter 2


3 Methods for the identification and spatial characterization of degradation processes

Besides the assessment of the CHC input, degradation or transformation processes affecting CHC as well as their spreading in space and time constitute significant elements of the MAPlan project. Depending on the environmental conditions, microbial processes cause a transformation or degradation of chlorinated ethenes. Degradation in the sense of a truly natural attenuation process in the system takes place only at a complete dechlorination or mineralization of chloroethenes.

In the case of an anaerobic reductive dechlorination, each chlorine atom is gradually replaced by a hydrogen atom respectively. For chlorinated ethenes, this process proceeds stepwise from PCE via TCE to 1,2-Dichloroethene (DCE, usually cis-1,2-dichloroethylene cDCE, more seldom trans-1,2-Dichloroethene tDCE) and on to VC and Ethene. Ethene is eventually mineralized to water and CO2.

The aerobic-oxidative degradation occurs cometabolic in many cases, i.e. the bacteria do not use the pollutant, but an auxiliary substrate as a growth substrate. In contrast to anaerobic degradation, in recent years metabolic aerobic degradation, in which the pollutants are used as a growth substrate, has been described for cDCE and VC in several publications [6, 7]. Since 2014, also the aerobic-metabolic TCE degradation has been scientifically proven in field samples [2, 10].

In the case of the low-chlorinated ethenes cDCE and VC there is a further oxidative degradation process in slightly anaerobic environments: the so-called post oxic degradation. The exact degradation processes are not yet fully understood. It was considered as an anaerobic oxidation, where for example nitrate acts as an electron acceptor. More recently, it is presumed that here oxygen serves as electron acceptor too, which is however only present in trace amounts below the detection limit in these milieu conditions. In Figure 3.1 the basic microbial degradation pathways of chlorinated ethenes are shown.
In the following, the essential methods by which the MAGPlan project succeeded with the identification and characterization of CHC degradation processes will be discussed.

### 3.1 Synoptic milieu maps

In water-saturated areas, microorganisms consume the oxygen during the degradation of organic matter. Depending on the degradation activity, the supply of dissolved oxygen in the ground water of max. 10 mg/l is rapidly depleted so that aerobic processes gradually come to a standstill. Alternatively, other oxidizing agents are used, such as nitrate, iron and manganese oxides and sulphate as electron acceptor. The microbiological degradation processes can be simply described as redox reactions. Knowing the redox milieu is therefore an important basis for the assessment of possible environmental or degradation processes of solutes in the aquifer system, in particular for CHCs. The processes occurring in the groundwater can be determined from the measurement of the oxygen content, the redox potential, or from the detection of redox-sensitive materials, which serve quasi as biogeochemical tracers.

Nitrate is a good way to gain an initial overview of the redox conditions. This redox indicator was measured at many groundwater investigations in the Stuttgart urban area and is object to a more reliable measurement technology than, for example, the determination of the redox potential, the oxygen or even the methane concentrations. In addition, for many samples, nitrate measurements at different times are available, so that concentration trends over time can be considered.
The use of sulphate is irrelevant for the characterization of redox processes in Stuttgart, as there is a massive geogenic influence due to sulphate rock dissolution in the Gipskeuper.

In the synoptic milieu maps, zones are identified (Figure 3.2) where one of the degradation processes described (reductively-anaerobic degradation, oxidation-aerobic degradation, oxidation-anaerobic post oxic degradation) occurs or at least is possible. Distinction between differently anaerobic areas (for example nitrate or sulphate-reducing) is not possible on the current basis. The spatial representation of the redox conditions and thus a first approximation of the degradation conditions was carried out on the basis of nitrate distribution maps drawn up for the project area.

![Figure 3.2 Synoptic milieu map for the lower Keuper [8].](image)

### 3.1.1 Reductively-anaerobic degradation

Regions with nitrate concentrations of less than 1 mg/l are considered potentially suitable for anaerobic degradation by reductive dechlorination. Anaerobic oxidation is also not excluded. To distinguish between anaerobic degradation by reductive dechlorination and anaerobic oxidation, additional criteria are therefore needed. As an indicator of anaerobic degradation, the molar cDCE fraction (metabolite of the
reductive dechlorination) is accepted: this is divided into three levels (< 1 mol%, 1-30 mol% and > 30 mol%). This mapping is carried out selectively at the respective measuring points, but is considered in the attribution of the degradation zones. cDCE fractions of more than 30 mol% are considered as evidence of anaerobic degradation, even if high nitrate concentrations occur. The anaerobic degradation is then likely to occur on a small scale in the immediate upstream. In this case, the area is still marked as anaerobic.

3.1.2 Oxidative-aerobic degradation

As a minimum requirement for aerobic degradation, nitrate concentrations higher than 1 mg/l are assumed. The basic classification is deviated from this in areas where indications of anaerobic degradation exist despite high nitrate levels.

Furthermore, isotopic signatures (stable carbon isotopes) are also taken into account. The isotope signatures of individual substances TCE and cDCE as well as the CHC-sum (if determinable) are presented selectively. δ13C signatures of below -23 ‰ are taken as an indication of minor (red) degradation; from -23 to -20 ‰ for moderate (yellow) and about -20 ‰ for good (green) degradation (Figure 3.2).

Significant isotopic enrichment (green) of cDCE and/or TCE in an aerobic environment is considered as evidence of aerobic degradation. If these substances have a high fraction of the CHC sum, the overall signature is green. If the fraction is small, the overall signature can also be red. There is then a slight PCE-signature that affects the total signature at high PCE fractions because the aerobic degradation only concerns TCE and/or cDCE.

3.1.3 Oxidative-anaerobic (postoxidisation) degradation

Areas in which little or no cDCE can be detected despite nitrate concentrations below 1 mg/l are designated as areas with possible postoxidation reduction.

A concrete indication of postoxidation reduction in the upstream to the certified spas results from the composition and isotope signature of certified spas. Since the first detection of CHCs in the 1980s and 90s, TCE has dominated in all springs. Traces of PCE and cDCE are partially detectable, but are below the quantification limit. The δ13C signatures could only be measured for TCE. With values between -18 and -17 ‰, they demonstrate an isotopic enrichment, which proves TCE degradation.

TCE degradation without cDCE formation is only possible through aerobic degradation. This can, however, be ruled out due to the environment conditions in the area of certified spas. If the TCE degradation takes place in the vicinity of the certified spas, this degradation would have to occur by reductive dechlorination, due to the prevailing iron to sulphate-reducing conditions.

However, with reductive dechlorination, a significantly higher cDCE fraction would be expected. An accumulation of TCE as the first metabolite of the reductive
dechlorination of PCE would be exceptional, so far there is no evidence of this. It is therefore assumed that the degradation takes place essentially in the upstream to the certified spas. The evidence of cDCE shows that a reductive dechlorination occurs first. But since cDCE is only measurable in trace amounts in the mineral springs and certified spas, substantial elimination of this metabolite over the flow path to the mineral springs must be assumed. As a process for the selective cDCE degradation, postoxidation degradation can be considered.

3.2 Evaluation of CHC distribution patterns

To visualize the temporal development of the chlorinated ethenes, a bar chart was created for each piezometer in the project area, where the average annual values of CHC concentration and composition are displayed. The temporal CHC development in measuring points of the project area shows different patterns. For plain PCE pollution without accompanying contamination by petroleum products, usually no significant change over time in the composition can be ascertained. The concentrations stagnate or decline as a result of remediation measures (Figure 3.3).

![Figure 3.3 Typical temporal evolution of plain PCE pollution [8].](image)

However, a temporal CHC development as shown in Figure 3.4, occurs frequently. In the case of a heavy decline in CHC sum concentrations, there is a reduction in the TCE-fractions and a relative PCE-enrichment. One possible reason may be the
preferred dissolution of TCE compared to PCE (by about a factor of ten). In the case of advanced solutions of TCE from a mixed contamination, this would lead to the observed progression. Another possible cause could be a selective aerobic TCE degradation, which would lead to a progressive elimination of this substance and thus for mixed contaminations, to an increasing percentage of PCE. Isotope studies and microbiological tests show that the selective aerobic TCE degradation plays a significant role here.

Figure 3.4 Examples of a TCE decline or a PCE percentage increase [8].
### 3.3 CHC distribution in the total area

The CHC concentrations in the project area range within a framework of five orders of magnitude. The ranges of fluctuation in the various aquifers are summarized in a generalized way in Table 3.1. It is notable that the concentrations below the aquifer of Grenzdolomite decrease dramatically.

#### Table 3.1 Ranges of CHC concentrations in the different aquifers (see Figure 4.2) [8]

<table>
<thead>
<tr>
<th>Aquifer</th>
<th>Range of CHC concentrations [µg/l]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mittlerer Gipshorizont</td>
<td>0 – 10.000</td>
</tr>
<tr>
<td>Dunkelrote Mergel</td>
<td>0 – 40.000</td>
</tr>
<tr>
<td>Bochinger Horizont</td>
<td>0 – 20.000</td>
</tr>
<tr>
<td>Grundgipsschichten</td>
<td>0 – 60.000</td>
</tr>
<tr>
<td>Grenzdolomit</td>
<td>0 – 80.000</td>
</tr>
<tr>
<td>Lower Keuper</td>
<td>0 – 2.000</td>
</tr>
<tr>
<td>Upper Muschelkalk</td>
<td>0 - 60</td>
</tr>
</tbody>
</table>

In Figure 3.5, the spatial distribution of CHC in the Quaternary and Gipskeuper layers is shown. All the data in the Quaternary and Gipskeuper groundwater monitoring wells was used without further differentiation. The illustrated CHC distribution therefore approximately reflects the situation in the shallow groundwater. To create the figures, the averages of the chlorinated ethenes for the years 2005-2014 were used. The illustration of the CHC distribution is done with pie charts. The diameter is proportional to the concentration. The segments indicate the molar fractions of the compounds PCE, TCE, cDCE and VC. This form of presentation visualizes the distribution of the various CHC compounds and gives an initial impression of the occurrence of anaerobic degradation processes – recognizable by cDCE and VC.

In the southern part of the project area, PCE pollution dominates, indicated by the red color, mostly caused from chemical cleaning processes. Relevant TCE concentrations (blue) are only detectable in the area of a former metalworking company (located at site no. 1, see Figure 2.5). cDCE (green) as an indicator of anaerobic degradation only occurs there occasionally. The situation is different in the northern part of the project area. There, the CHC composition is significantly inhomogeneous. The more intensive industrial use, compared to the southern and central project area, can be seen from a higher TCE-fraction, which comes from metal degreasing and from various tank storage depots. In the vicinity of the tank storage, contamination by petroleum products also stimulates the anaerobic CHC degradation. This is recognizable from the widespread occurrence of cDCE and partly also of VC (yellow).

The CHC distribution in the lower layers, exemplary for the lower Keuper, can be seen in Figure 3.6.
Figure 3.5 Spatial distribution of CHC in the Quaternary and Gipskeuper [8].

Figure 3.6 CHC distribution in the lower Keuper [8].
3.4 Degradation experiments in microcosms

Degradation experiments in microcosms provide an opportunity to show the microbial processes occurring in the field and to investigate precisely in a controllable system [1]. This provides sound information on the degradable processes and the products that may arise. Stimulation possibilities of microbiological degradation, for example by the addition of auxiliary substrates and/or nutrients, may also be tested.

Microcosm tests are quite time-consuming, as after the groundwater sampling, a so-called "lag" phase ("phase delay") can occur. The micro-organisms responsible for the CHC degradation generally take a period of several weeks (up to three months) of adjustment until the degradation resumes. The simplest version of microcosm tests is groundwater incubation in bottles. At regular intervals, samples from each microcosm batch are taken. If more than one preparation per sample exists, the sampled bottle is then discarded. In each, CHC concentrations and redox-specific parameters are measured.

Due to the presumed importance of aerobic TCE degradation in the project area, groundwater samples were taken to carry out degradation experiments in microcosms from selected representative measuring points. Aerobic TCE degradation was evident in investigated groundwater from all five monitoring stations, chosen as representative examples, in different parts of the project area. The comparison with the sterile control with constant TCE levels shows that in the active experimental set-up, biodegradation of TCE occurred (Figure 3.7). The PCE preserved at the start of the experiment showed no degradation during the entire course of the experiment.
The groundwater used did not contain the possible auxiliary substrate methane [3] at all, and only low levels of ammonium [4]. Also, the DOC (dissolved organic carbon) as a measure of the organic matter contained and therefore for non-specific auxiliary substrates was low. No significant correlation of ammonium or decline of DOC with TCE degradation was observed over the duration test. The comparison with literature data for auxiliary substrate consumption during co-metabolic degradation [5] indicates that the measured DOC quantities are not sufficient, even with a complete transformation, for the observed TCE degradation.

Thus it can be assumed that the TCE degradation took place in groundwater microcosms without known auxiliary substrates, at low ammonium and DOC levels and in a defined mineral medium. This means that TCE was aerobically-productively degraded in the investigated groundwaters from the project area [2].

These test results and mappings have been incorporated into the numerical transport model.

### 3.5 Literature Chapter 3


4 Hydrogeological and numerical modelling for spatiotemporal understanding of CHC Transport

4.1 Conceptual (hydrogeological) model

The conceptual (hydrogeological) model divides into two modules: the "conceptual aquifer model" and the "conceptual contaminant model". Core elements of the aquifer model are the geological conditions (strata buildup, layer geometries, layer bedding). They serve as a basis for the hydrogeological system description (hydrogeological entities, hydrostratigraphy, and aquifer geometry). Moreover, basic concepts of regional groundwater flow and related hydraulic boundary conditions, assessments of water balance (groundwater recharge, boundary inflow, vertical hydraulic interaction between levels, management), range of the permeabilities and other hydraulic parameters are displayed [1, 3]. The core element of the contaminant model is the behavior of the CHC compounds in the system. It documents and describes the mechanisms of CHC-input, the spreading in space and time and ongoing CHC degradation or transformation processes depending on the environmental conditions. Based on data from field studies and taking into account the hydrogeological a priori knowledge a model concept is developed. It correlates to the understanding of the hydrogeological system, the behavior of CHC compounds and the major processes driving in the system.

In the project area, the strata buildup is determined by the Keuper, which extensively overlays the mineral-water-producing upper Muschelkalk aquifer over the entire model area (Stuttgart basin and Cannstatter Basin). The different sequences of limestone, mudstone, dolomite and sulphate rocks result in a hydrogeological strata structure with seven bedrock aquifers (Figure 4.2). Carbonate karstification, the subrosion of sulphate rocks and the strong tectonic stress of the layer sequence play an important role for the geometry of these entities and their hydrogeological function. The upper Muschelkalk – and in particular its top layer Trigonodus dolomite – forms a high yield karst aquifer, which extends along the lower Keuper far beyond the city limits of Stuttgart. In contrast, in Gipskeuper there are local aquifers with low yield restricted to the Stuttgart basin.
In the large 26 km² model area the data situation will inevitably remain incomplete or for certain sub-areas even fragmentary. This requires the abstraction of the natural system to its essential relationships and system-determining factors (Figure 4.1). The hydrogeological modelling process is dynamic because the quality of the conceptual models is continuously updated with the on-going investigation programs and their results within the project lifetime. The working hypotheses are tested, accepted or maybe also rejected with each new finding. In the MAGPlan project, the numerical flow and transport model, which were compiled alongside the hydrogeological model, involved the review process from the beginning. The constant interaction with the numerical model facilitates a mutual plausibility checking. In addition to its function for quality assurance, the hydrogeological model is a tool for communication between the project partners and thus the core feature in the project.

The dedicated geological and hydrogeological characterization of the system is only possible for the project area because data was documented and archived, over several decades in the context of construction activities and investigations of contaminated sites. The extensive hydrogeological exploration work for the railway tunnel project “Stuttgart 21”, as well as the hydrogeological investigations reaching beyond Stuttgart to the designation of the certified spas protection zone contributed significantly [2].

The compilation and evaluation of all available data at the project start has proved to be fundamental to the successful implementation of the project. Thus, access to reliable data sets was possible, which formed the basis for initial assessment and reviews, and in addition helped to identify data gaps. With a targeted approach, new field investigations for obtaining this data could be prepared. Thus, the limited resources for new investigations were optimally used. Measures could be well targeted to improve the knowledge of the natural system and on-going processes in key areas.
The technical data retrieved in the information systems was selected, professionally processed and displayed in the form of maps, cross sections, diagrams with respect to area and time [3]. The interpretation of the data requires the local experience of hydrogeologists. This is especially important for areas that are strongly influenced by tectonics and karstification, and thus very heterogeneous on a small-scale [4]. Incorporating the knowledge of structure-forming geological and hydrogeological processes (for example hydraulic function of linear structures, selective karstification) has decisively helped to a near-natural representation. Figure 4.2 demonstrates the procedure on the basis of a conceptual model cross-section of the site no. 9 (for location see Figure 2.5).

4.2 Numerical groundwater flow model

4.2.1 Model setup

The basis of the numerical groundwater flow model is the hydrogeological model, in which the system interrelationships of the complex aquifer system are summarized. Based on the conceptual aquifer model, model boundaries have been determined and boundary conditions assigned, that represent the natural hydrogeological conditions for each hydrogeologic unit. Since the numerical groundwater model is a mathematical description of the hydrogeological conditions, a further abstraction of the hydrogeological model is necessary. This concerns in particular to site-specific conditions, which are subject to a hydrogeological interpretation with high spatial resolution. Hydraulically active elements such as fault zones or discrete fracture elements can only be included abstractly in the numerical model. It is to be noted that these hydrogeological entities are fractured and karst aquifers, where the flow takes place mainly in the fracture system. Nevertheless, an exchange with the matrix is provided which primarily allows storage of dissolved
water ingredients being transported. This has been taken into account via the double porosity approach in the transport model.

To simulate the flow the MODFLOW model system in the 2005 version developed by the USGS was used [5]. The mass transport is simulated with the multi-species model MT3D99. The basis of the mass transfer model was complied with by [6] and further developed by [7] for multi-species models.

The permeability distribution of the hydrogeological model was considered as starting values at the beginning of the model calibration. It was modified in the course of calibration so that the best possible match between measured and calculated information (piezometers heads and transport data) was achieved. In addition, fault zones with big displacement value compared to the aquifer thickness were taken into account by a reduced horizontal permeability. This is based on the assumption that the groundwater flow is hindered by the displacement. The location of these fault zones is shown in Figure 4.3. The faults considered as hydraulically significant are the Birkenkopf fault and the Schloss fault.

Figure 4.3 Location of the faults with reduced horizontal permeability in the groundwater model [3].
Faults with large displacement can lead to flow around the fault zone as well as potentially allow a vertical connection between layers. This is justified by the fact that the bedrock nearby the fault can be shattered, leading to improved hydraulic interaction. The disrupted areas where this may be the case were determined during the calibration taking into consideration the piezometric heads or concentration distributions.

The calibration of the permeability was initially accompanied by comparing the measured and calculated piezometers heads. Since the permeability structures and in particular the vertical exchange rates between the hydrogeological entities influence the transport of dissolved water ingredients, it was necessary to adjust the permeability of structures in the course of the calibration of the CHC-transport model.

The quality of the model calibration can be derived from the comparison between measured and calculated piezometers heads. Plotting the observed compared to the calculated piezometric heads, displays the relationship shown in Figure 4.4. As the analysis shows, the values pairs are scattered around the bisecting line, so that it can be assumed there is no systematic difference between measured and calculated piezometers heads.

Figure 4.4 Comparison between measured and calculated piezometers heads in the model area [3].

Figure 4.5 Steady state water balance for the flow model, values in l/s [3].
With the help of the groundwater model, the steady state water balance was determined in the model area (Figure 4.5). Adding up all the inflows yields a water turnover of 1,044 l/s. This turnover corresponds to the sum of the discharge.

### 4.2.2 Review of groundwater flow with transport data

In the area of investigation, two tracer tests were done at the end of the 1990s, and another within the duration of the project. In the model, the tracer tests were simulated in order to determine the relevant transport parameters in the Muschelkalk. Since the Muschelkalk aquifer is a karst aquifer, fracture/matrix interactions have to be considered during the transport modelling. It has been found that breakthrough curves with long tailing typical for Karst aquifer could only be replicated by means of a double porosity approach. For this purpose, the transport system was divided into a mobile and immobile system and the differential equations according to [8] applied.

For the simulation of the plumes of the tracer tests and the tracer transit curves, the use of a longitudinal dispersivity of 25 m was necessary. The ratio between the horizontal longitudinal and transverse dispersion was set at 1:10. The vertical transverse dispersion was assumed to be smaller than the horizontal transverse dispersion by a factor of 10. These values refer to the limestone aquifer, where the tracer tests took place.

### 4.3 Numerical transport model

#### 4.3.1 Total CHCs and single compounds

In the groundwater model, initially, the sum of CHCs was considered. In doing so, the different compounds were converted into the PCE-equivalent concentrations taking into account their molar fractions. Based on this, the multi-species observation was conducted, in which, for example, the different compounds arising from reductive dechlorination were assumed, as follows:

\[ \text{PCE} \Rightarrow \text{TCE} \Rightarrow \text{cDCE} \Rightarrow \text{VC} \]

While PCE and TCE were used in different fractions in the contaminated sites, cDCE and VC are pure degradation products which are formed under anaerobic conditions. In the transport modelling it was taken into consideration that the single compounds are transported not only advectively and dispersively, but are also subject to sorptive processes.

The simulation of the CHC transport for the period from 1960 to 2010 was carried out on the basis of the steady flow model. The CHC input was resolved on an annual basis, whereby between 1960 and the 1980s, no variations were made. The temporal variations only begin in the 1980s because the remediation measures began and assumptions regarding the rate of change of pollutant input could be derived from this.
### 4.3.2 Input model of CHCs

To calculate the input rates, the input loads were determined based on the groundwater flow conditions in the control planes and the measured pollutant concentrations. The analysis of the sites in the conceptual contaminant model has shown that the most significant contamination of groundwater comes from six sites in the area of investigation. There are also three other sites whose contribution to the large scale pollution pattern cannot be excluded. For these sites the CHC input at the site, the extraction via decontamination and the residual downstream load were determined using the measured values for the PCE-equivalent of all species. The ratio of the CHC compounds was estimated according to the measured values of remediation wells.

In the model, the remediation measures were not explicitly taken into consideration as hydraulic boundary conditions but by reducing the input functions according to the respective CHC-removal rate. This results for all sites considered in total input rates of the CHC compounds, and summed up for the whole project area as shown in Figure 4.6. Before the start of extensive remediation measures in the 1980s, the PCE input amounted to 300 kg/year and the input of TCE and cDCE approximately 100 kg/year respectively. Thanks to the extensive remediation measures the inputs could be reduced by a factor of 3 to 4. It should be noted that, in reality, no direct input of cDCE and VC occurred, because these two CHC compounds were not directly used for commercial purposes. In case that the transformation process occurred on the contaminated site, the direct input of cDCE and/or PCE was applied in the numerical model.

![Figure 4.6 Summary input of CHC compounds for all sites in the model area between 1990 and 2010](image.png)

### 4.3.3 Advective and dispersive transport

The advective transport is determined by the flow field. This takes place in three dimensions in the area of investigation. The CHC compounds are horizontally transported within the hydrogeological entities and displaced via the vertical exchange between the layers. In the natural system, the vertical transport is oriented mainly to fracture pathways and, in the lower Keuper or Muschelkalk aquifer, also at fault zones. In the numerical model, in the case of known local vertical displacements, appropriate permeability structures were introduced. Additionally, an
areal vertical transport was applied, which may lead in the model to a spreading of plumes in case of depth displacement. This is particularly the case when the flow in an overlying layer runs transversely to the flow direction in the underlying aquifer.

The horizontal transport is also linked to the fracture system. Here, the transport takes place in the fractures in the lower Keuper or Muschelkalk with widths from a few centimeters to tens of centimeters. With the selected continuum approach the equivalent fracture system is described. Hence, it is assumed that each model cell provides sufficient gap structures. Due to the large size of the project area a horizontal resolution of 10 × 10 m was selected. This means that the model can only supply one integral value for a model cell. This must be considered when comparing model results with the measured values.

In addition to the advective transport, the dispersive transport is also taken into account. This is described by the dispersivity sensor. For the limestone the dispersivities from the evaluation of tracer tests were used ($\alpha_L = 25$ m, $\alpha_T = 2.5$ m).

4.3.4 Sorption

Generally, sorption processes lead to a storage in the aquifer and thus to a retardation of mass transfer. If sorption is irreversible, there is even a permanent removal of CHCs from groundwater. In the simplest case, the relationship between dissolved and adsorbed concentration is described by a linear isotherm, and expressed with the aid of a retardation factor $R$. The single CHC compounds differ strongly in their tendency to sorption. Their migration velocity is thus more or less strongly retarded.

4.3.5 Degradation and transformation – model approach

To simulate the reaction processes, in some parts of the model area the oxidative degradation (mineralization) and in others a transformation of CHC via reductive dechlorination is considered. To simulate the degradation or the transformation of CHC, reactions of the first order were used. For the degradation of CHCs, the degradation rate is proportional to the concentration. The reaction rate is also controlled by specifying a rate constant, the so-called half-life. To simulate the reductive dechlorination in the model, the procedure is similar. In addition to the degradation of the substance, the formation of the transformation product is also balanced. For example, using the stoichiometry factor in the transformation of PCE to TCE, from 1 g PCE only 0.79 g TCE is formed, as a result of elimination of a chlorine atom.

The establishment of the zones with aerobic CHC degradation or reductive dechlorination in the model area was based on the milieu map and the zoning, created in connection with the development of the conceptual contaminant model (see chapter 3).
4.3.6 Calibration by CHC-distribution curves and maps

First, in the downstream of contaminant source areas, CHC distribution curves were analyzed. With the assumptions made about the current input and the development over time, it was checked whether the measured ratios could be reproduced. Figure 4.7 shows an example of measured and calculated CHC concentrations in the downstream from two main contaminated sites. The numerical model was able to reproduce the concentration and the temporal development of the CHC.

The three-dimensional distribution of the CHC was evaluated and analyzed using the model. Figure 4.8 shows the calculated TCE distribution in the triogonodus dolomite for 2010. According to the measured information, the highest TCE concentrations can be found between the city center and Stuttgart-South. The low permeable Schloss fault (for location see Figure 4.3), identified in the course of the flow calibration, forms a barrier to the flow of groundwater. The remaining TCE contamination in this area flows around the Schloss fault, the TCE plume however terminates 500 m downstream at the level of the central station. This means that the increased TCE concentrations measured at the highly mineralized mineral springs and certified spas cannot originate from the inner city, but must be caused by contaminated sites located closer to the springs.

During the course of a sensitivity analysis, the site no. 9 (Figure 4.8) was identified as a possible source of the pollution of the highly mineralized certified spas. It has been shown that CHC must have reached the Trigonodus dolomite via fault zones close to the site. The CHC may still be present here as a free phase and represent a pollution source with sustained release of TCE. As part of the sensitivity study a release rate of 8.6 g/d TCE was identified. With this input, the TCE concentrations shown in Figure 4.9 can be explained.
While most of the highly concentrated mineral springs show an exclusive TCE contamination, PCE can be found in low-mineralized springs. In some of these springs, predominance of TCE has shifted to PCE since the 1990s. Using the model, the site no. 10 and site no. 19 (for location see Figure 2.5) were identified as a
potential sources of contamination of the low-mineralized springs. It can be assumed that site no. 10 was the main source of the CHC concentrations at the low-mineralized springs especially in the past. For site no. 19, so far, only results from the model suggest that the second source of contamination for the low concentration mineral springs can be found. Hence further specific investigations are necessary, triggered by the model results. The model-calculated (simulated) and measured (observed) concentrations of two low-mineralized springs are shown in Figure 4.10.

Figure 4.10 Calculated and measured CHC evolution curves at selected low-mineralized springs.

An important result of the modelling of CHC transport is the mass balance of each CHC compound. However, since the transport of dissolved matter is transient, a concluded mass balance can only be determined for the time-integrated mass.

The contaminant sources mainly release PCE. The input decreased from 280 to 60 g/d in 2010. The TCE input was 120 g/d in 1990 and then decreased to 20 g/d. The input of cDCE is of the same order of magnitude as that of TCE.

The main discharge of CHC masses occurs via the Gipskeuper and the Quaternary sediments into the Neckar [3]. The service water wells and building drainage systems in the Keuper and Muschelkalk also have a large proportion of the discharge of CHCs from the overall system. The CHC is discharged by up to 20% to the service water wells and up to 60% to the Neckar. Thereby, the fractions of TCE and PCE are approximately equal. The remaining 20% of the discharge take place via the mineral springs.

Due to the degradation model, both a transformation of CHC compounds as well as a mineralization takes place in the overall system. The temporal progression is shown in Figure 4.11 below. For PCE, there is only a transformation to TCE. In the whole model area, between 135 kg/year in 1990 and 80 kg/year in 2010 of PCE were converted to TCE. According to the stoichiometric ratio of TCE/PCE of 0.79, 105 to 60 kg/year of TCE are formed. From the total mass of TCE, in 2010 60 kg/year was converted to cDCE and 13 kg/year was completely mineralized by aerobic degradation. The TCE transformation results in 45 kg/year cDCE in 2010. Since cDCE is comparatively well aerobically degraded, those 45 kg/year in 2010 were completely mineralized. This mineralization rate in 1990 was 148 kg/year. Since the
major portion of cDCE is mineralized, only a small amount VC is formed. In 2010 it was still 5 kg/year. The same amount of VC was also mineralized in 2010.

Figure 4.11 Temporal progression of the transformation and the mineralization of CHC compounds in the model area [3].

The visualization tool, MAG IS (Management Information System) [3], developed during the project, enables the individual evaluation of the model results and visualization in combination with model input information. Various researchers and institutions can access the results through the internet and individually visualize and analyses the numerical groundwater modelling data, without any special knowledge thereof. This has proven to be an effective tool and communication aid.

4.4 Literature Chapter 4


The groundwater management plan for Stuttgart – priorities and concrete measures to be implemented by 2026

The structure of the groundwater management plan is derived from the Habitats Directive with three core elements: description of the current state, development goals and development measures. The description of the current state is in the form of an extensive study, which consists of reports, maps, databases, a hydrogeological model, and a numerical flow and transport model including a computer-based visualization tool that facilitates access to the data. It is clearly summarized in [10]. The derivation of the development goals and the development measures identified are presented in the following report.

5.1 Development Goals – priorities in contaminated site management

In general, the definition of investigation and remediation requirements is carried out in compliance with the legal framework and regulatory conditions applied to soil and water. Due to the outstanding importance of mineral springs and certified spas in Stuttgart, there is a particular focus on ensuring the quality of karst groundwater in the upper Muschelkalk. Thus, the requirements for protection of mineral springs and certified spas do also have an impact on the definition of priorities for municipal contaminated site management.

The prioritization serves to achieve the main objective of reducing CHC concentration in the mineral water aquifer of the upper Muschelkalk and the mineral springs and certified spas in the foreseeable future.

Within the context of contamination management, groundwater represents a protected resource and receptor. The prioritization for the groundwater management plan leads to a differentiation between three receptors:
MAGPlan
Clean Groundwater for Stuttgart

- certified spas receptor
- mineral springs and karst groundwater in the upper Muschelkalk receptor
- groundwater above the upper Muschelkalk (Quaternary to lower Keuper) receptor.

A particular urgency is attached to the protection of the certified spas, the mineral springs and the karst groundwater in the upper Muschelkalk. This leads to the definition of priorities, since not all necessary measures can be taken simultaneously. With the prioritization a temporal sequence of measures is proposed. High priority measures should be tackled before lower priority measures. However, the priority setting has no effect on measures that have already started. These continue to be carried out within the scope pertaining to the individual cases.

According to the importance of the mineral springs and certified spas, three different treatment priorities are differentiated. Specific evaluation criteria and remediation targets are assigned to each receptor, see Table 5.1. The result is a prioritization list of further measures. For each site, it is to be checked which treatment priority applies to the individual case.

Table 5.1 Specification of treatment priorities for the contaminated sites in the project area [10]

<table>
<thead>
<tr>
<th>Priority</th>
<th>Affected receptor</th>
<th>Criteria and decontamination goals</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Certified spas</td>
<td>Site affects receptor directly</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Decontamination goal is the complete elimination of a possible input into “Upper Muschelkalk”</td>
</tr>
<tr>
<td>2</td>
<td>Mineral springs and karst groundwater in „Upper Muschelkalk“</td>
<td>Concentration in receptor</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\sum \text{CHC} &lt; 5 \mu g/l$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Single species $&lt; 1 \mu g/l$</td>
</tr>
<tr>
<td>3</td>
<td>Groundwater above „Upper Muschelkalk“ (higher aquifers)</td>
<td>Emission and immission of contaminant sources</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\sum \text{CHC} &lt; 20 \text{ g/d}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\sum \text{CHC} &lt; 10 \mu g/l$</td>
</tr>
</tbody>
</table>

In the licensing and use of natural mineral water the following regulations are to be taken into account: the “Administrative regulations on the acknowledgement and usage permit of natural mineral water” [1] the "Mineral and table water regulation" [2], and the "Drinking Water Ordinance" [3]. In the Administrative regulation and the Drinking Water Ordinance, CHC is designated as parameter whose concentration is to be limited for the protection of human health.

The Drinking Water Ordinance defines a limit for this purpose of 0.010 mg/l (10 μg/l) for Tetrachloroethene (PCE) and Trichloroethene (TCE) as a "sum of proved and quantified individual substances". The administrative regulation refers to this requirement but adds "orientation values for contaminating substances in natural mineral water as a criterion for the original purity". As a criterion, "volatile organic
halogen compounds with 5 μg/l" (sum) are mentioned as the orientation value. Volatile chlorinated hydrocarbons with chlorinated ethenes PCE, TCE, DCE, and VC also fall into the group of volatile organic halogen compounds. In addition, Quentin, in [4] proposes, as a criterion for the natural purity of natural mineral water, 1 μg/l for each CHC component, i.e. for PCE, TCE, DCE, and VC respectively. In the "Quality standards for the nomination of spas, health resorts and mineral springs" the German Spa Association e. V. [5] recommends operators of certified spas to apply, "the laws on environmental protection...as minimum standards...".

From the above mentioned regulations and recommendations, the comparatively strict evaluation criteria and remediation targets in Table 5.1 are derived for the mineral and certified spas and karst groundwater in the upper Muschelkalk (priorities 1 and 2) for Stuttgart.

Priority 1 aims that no more CHCs reach into the certified spas. All sites that have an impact on their quality, i.e. contamination sources from which CHC emissions – independent of the concentration – lead to an impairment of the certified spas, must be decontaminated with the aim of completely suppressing CHC emissions into the upper Muschelkalk.

With priority 2, contaminant sources, which have an influence on the low concentrated mineral springs or the karst groundwater of the upper Muschelkalk, should be remediated. A requirement for treatment is fulfilled when the sum of the CHC exceeds 5 μg/l or the individual substances exceed 1 μg/l. If multiple sites are responsible for the exceeding of the aforementioned values, the emission at each contamination source is to be limited to the extent that the local input in the upper Muschelkalk is permanently below the concentration of 5 μg/l in total and 1 μg/l for each compound.

Priority 3 corresponds to the definition, applicable to all aquifers for each site in Baden-Württemberg, of the "case-by-case minimum requirements" [6]. These minimum requirements are used to establish the good quality of groundwater in each groundwater resource (waters) according to the Water Act or Groundwater regulations.

Thus, for the site-specific remediation targets at each site in need of remediation, of the three criteria, the one leading to the most stringent remediation requirements applies. Usually several aquifers are affected, for which specific remediation targets and measures are derived. The remediation plans are based on the decontamination of contamination sources. Technically, downstream remediation is impossible if the pollutants have penetrated into the karst aquifer of the upper Muschelkalk.

In all remediation target definitions, the principle of proportionality of resources applies. This has the consequence that the remediation plan is to be checked if the remediation targets cannot be achieved at reasonable cost [7].
5.2 Forecast of development of pollutant transport

The numerical transport model provides the opportunity to simulate different remediation scenarios and to make variant-specific forecasts for the future pollutant spreading. This observation can be performed for isolated sites. Each selected aquifer (receptor) can be represented by one or few groundwater wells. Analyzing the concentration trends in these wells, the effectiveness of remediation options is described for each scenario. Figure 5.1 shows an example of possible concentration trends of a groundwater well located downstream of a contamination source area for different remediation scenarios.

In Figure 5.1A, the concentration in the groundwater well and the representative receptor increases, if the remediation currently operated in the source area is ceased. The influence of the space between contamination sources and the groundwater well manifests itself in a delayed increase of the concentration.

If the concentration in the groundwater well remains approximately constant over the long-term despite an improvement of the remediation scheme (Figure 5.1B), thus the simulated remediation scenario is ineffective and the remediation plan is to be reconsidered. Figure 5.1C indicates a decrease of the concentration. However, it remains at a level above the remediation target. It cannot be expected that the remediation goal will be achieved in the foreseeable future. Therefore, there is a need for improvement also in this case.

The space between contamination sources and receptor can be of considerable significance if it acts as a pollutant storage. In this case, the concentration in the groundwater well reacts only very slowly to the source remediation (Figure 5.1D). An improvement of such remediation at the source is not effective because the majority of the pollutant, though desorption for example, enters the flow path between the sources and groundwater well.

An optimal and sustainable remediation is shown in Figure 5.1E. The remediation target is achieved quickly in this case; the space has no negative effects.

The numerical transport model was calibrated based on the observed trends in pollutant input and groundwater concentrations from 1960 to of 2010. With the support of this model and based on the worst-case assumption, that the pollutant emissions will remain constant in the 50 years forecast from 2010 to 2060, the remediation scenarios can be distinguished in Table 5.2 below.
Figure 5.1 Exemplary concentration trends of a groundwater well downstream in the simulation of different remediation scenarios for a contamination source [10].

To enable a simulation of different remediation options, four representative scenarios were defined:

**Scenario a)** No remediation: All ongoing remediation measures at the sites are terminated (worst-case scenario). The contamination sources consistently emit the maximum pollutant load.

**Scenario b)** Emission 2010: All current remediation measures (remediation 2010) will continue unchanged. Pollutants which are not covered by the remediation are emitted.

**Scenario c)** 50% of 2010 emissions: The ongoing remediation measures are optimized so that in future only 50% of the pollutant emissions are emitted, compared to b), from the contamination sources (optimization scenario).

**Scenario d)** zero emissions: The emission of pollutants is prevented completely (100% remediation of the pollution sources: an optimal scenario which is seldom achieved).

Applying this concept and its scenarios to the sites with high levels of contaminant input, the contaminant emissions (loads in grams per day) were simulated as compiled in Table 5.2.
For each prediction run, all four emissions scenarios a) to d) were modelled. In each case, representative downstream groundwater wells in effected aquifers, for example a mineral spring, a certified spa or one or several monitoring wells, were selected. The concentration trends clearly display how the respective remediation scenario will affect the groundwater wells and thus the receptors represented. All the site specific CHC substances were modelled. The results of the prediction runs are explained subsequently by the example of Berger North spring as a representative for the highly concentrated mineral springs (certified spas).

### 5.3 Forecast for the mineral springs

The certified spas in Stuttgart-Berg affected by the CHC contamination are represented by Berger North spring (Berger Nordquelle). This choice is justified by the fact that the concentration trend is approximately parallel to the mineral springs in this area and a distinctive CHC signal is measured at the Berger North spring. The CHC concentration detected varied between 0.3 to 2.9 μg/l in the past.

In Figure 5.2 the CHC concentration trends for the years 2010 to 2060 in the Berger North spring, for scenarios a) to d), in the site constellation ALL (all contamination sources are emitting in the project area) are shown for TCE, PCE and DCE. The CHC concentration in the Berger North spring (as in all highly concentrated mineral springs) is primarily caused by TCE, with a mean of 1.7 μg/l for the year 2010. All other individual substances, at concentrations of usually less than 0.1 μg/l, play a minor role.

Similarly, the CHC concentration trend for the site no. 9 is shown in figure 5.3, i.e. only this site emits, all others are switched off.

---

**Table 5.2 Emitted CHC loads for the relevant sites in the scenarios a) to d) [10]**

<table>
<thead>
<tr>
<th>No.</th>
<th>Site / Prediction Run</th>
<th>Scenario a) no remediation</th>
<th>Scenario b) emission 2010</th>
<th>Scenario c) 50 % of emission 2010</th>
<th>Scenario d) no emission</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Dornhaldenstraße 5</td>
<td>50</td>
<td>16</td>
<td>8</td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>Rotebühlstraße 171</td>
<td>160</td>
<td>30</td>
<td>15</td>
<td>0</td>
</tr>
<tr>
<td>3</td>
<td>Johannesstraße 60</td>
<td>53</td>
<td>23</td>
<td>11,5</td>
<td>0</td>
</tr>
<tr>
<td>6</td>
<td>Rotebühlplatz 19</td>
<td>16</td>
<td>16</td>
<td>8</td>
<td>0</td>
</tr>
<tr>
<td>7</td>
<td>Nesenbachstraße 48</td>
<td>412</td>
<td>5</td>
<td>2,5</td>
<td>0</td>
</tr>
<tr>
<td>8</td>
<td>Wolframstraße 36</td>
<td>15</td>
<td>2,4</td>
<td>1,2</td>
<td>0</td>
</tr>
<tr>
<td>9</td>
<td>Rümelinstraße 24 – 30</td>
<td>130</td>
<td>56</td>
<td>28</td>
<td>0</td>
</tr>
<tr>
<td>10</td>
<td>Mittnachtstraße 21 – 25</td>
<td>111</td>
<td>13</td>
<td>6,5</td>
<td>0</td>
</tr>
<tr>
<td>19</td>
<td>Prag-/Löwentorstraße</td>
<td>129</td>
<td>37</td>
<td>16,5</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>Sum total [g/d]</td>
<td>1,076</td>
<td>198</td>
<td>99</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>Sum total [%]</td>
<td>100</td>
<td>18</td>
<td>9</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>PCE-equivalent [g/d]</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Figure 5.2 Development of CHC concentrations in the Berger North spring for scenarios a) to d) due to the emissions of all sites (prediction run ALL) [10].

Figure 5.3 Development of CHC concentrations in the Berger North spring for scenarios a) to d) due to the emission of the site no. 9 (prediction run 9) [10].
A comparison of Figures 5.2 and 5.3 shows that the respective remediation scenarios lead to similar concentration trends. This underlines the assessment based on the overall evaluation of the investigation results that the TCE contamination in the mineral springs is mainly due to the site no. 9. Furthermore, it is clear that an effective remediation of the site no. 9 would achieve a very significant concentration decline in the mineral springs (Figures 5.2 c and d) and Figures 5.3 c and d).

A deep input in the upper Muschelkalk at the site no. 9 was identified as the essential reason for the CHC contamination in the Berger certified spas. This deep input was not affected by the remediation measures carried out so far. Remediation carried out so far in the Gipskeuper is apparently not primarily relevant for the concentration trend in the Berger certified spas. In conclusion, it is clear that the best possible reduction of the deep input of CHC at site no. 9 is necessary to prevent further CHC contamination of the Berger certified spas.

The evaluation of CHC concentration forecasting runs for the other relevant sites shows that such prediction runs of numerical transport modelling enable or effectively support the identification and characterization of sites with high remediation priority. In combination with the conceptual transport model, further action and respective targeted additional measures could be concretized for four sites of priority 1 and 2.

### 5.4 Stuttgart remediation feasibility study

The remediation feasibility study describes the measures that must be taken in order to achieve the described remediation targets of the priorities 1 and 2 sites. The remediation feasibility study complements the individual measures at these sites, it does not replace them, however. For this purpose, it is determined which technical options of an investigation or remediation exist. The individual options are evaluated non-monetarily with 23 assessment criteria and monetarily with cost estimates for alternative remediation methods [8]. On this basis, a recommendation for the next step is made. If additional investigation is required (for example if contamination sources zone delineation is required), appropriate investigative measures are outlined and investigation costs estimated. The results of the remediation feasibility study for the sites of priorities 1 and 2 in the MAGPlan project area are summarized in Table 5.3 below.
For site no. 9 it is proposed to investigate the use of microbiological in situ remediation. This is economically favorable compared to the pump and treat option and aims at decontamination of the sources. The remediation target is to reduce the emission of pollutants into the limestone, as far as technically possible. The numerical modelling shows that the direct entry of TCE in the upper Muschelkalk at the site no. 9 must be reduced by significantly more than 50% in order to achieve the remediation target of "good groundwater status" in the certified spas.

Since this site is extremely sensitive, the microbiological remediation may not allow any additional pollutant mobilization, as there is no further opportunity for prevention on its way to the upper Muschelkalk. Therefore, this remediation option can only be pursued after preliminary tests have been successfully carried out. Qualified statements on the applicability of the process, its risks and its chances of success can only be made on the basis of such pilot schemes.

The remediation feasibility study and the compilation in Table 5.3 show that near-surface excavation and pump & treat are not sufficient for sustainable remediation with regard to the mineral springs and certified spas. Rather, the application of in situ remediation techniques – despite all the limitations in karst aquifers – is essential. The suitability and permissibility of the remediation techniques proposed can only be decided based on pilot applications.
5.5 Monitoring Program

To verify the effectiveness of the remediation measures, it is necessary to monitor the impact and success by means of a monitoring program. The numerical transport model allows in prediction runs beforehand for many areas the determination of pollutant concentration trends at the selected groundwater monitoring wells.

If the simulated development according to the completed remediation measures is actually observed, thus it is likely that the hydrogeological system in this area has been understood and the measures taken are appropriate.

However, if the measured development deviates significantly from the calculated, either the remediation measures are not effective or the system-relevant circumstances are not yet understood sufficiently. In this case, it is to be considered what further measures to optimize the remediation or improve the understanding of the system should be taken.

The selection of monitoring wells occurs according to the following criteria:

- Pollutant emissions which affect the upper Muschelkalk aquifer have to be monitored as a priority.
- Every aquifer with non-permissible emissions should be recorded with at least one monitoring well.
- Existing groundwater monitoring wells are to be used predominantly. It is important that these groundwater monitoring wells record the effects of pollutants on the affected aquifers and provide quick response to changes in the pollutant input. If the existing groundwater monitoring wells do not meet this requirement, new groundwater monitoring wells are to be constructed.

The so-defined measurement network comprises 28 groundwater monitoring wells, including 11 new groundwater monitoring wells, which must be tapped in four different horizons. The selection of groundwater monitoring wells is taken with respect to the contamination sources. For each piezometer a sampling program is determined, see exemplified Table 5.4. Sampling methods and intervals for the groundwater monitoring well investigations are matched to the current monitoring program for the mineral springs and certified spas.

Table 5.4 Monitoring program exemplary for a groundwater monitoring well [10]

<table>
<thead>
<tr>
<th>Monitoring well</th>
<th>MAG 16N, downstream mo* site no. 9</th>
</tr>
</thead>
<tbody>
<tr>
<td>Task</td>
<td>Monitoring the evolution of CHC concentrations and the success of remediation for the site downstream in aquifer mo*</td>
</tr>
<tr>
<td>Sampling interval</td>
<td>Annually</td>
</tr>
<tr>
<td>Sampling depth</td>
<td>20 meters below ground level</td>
</tr>
<tr>
<td>Way of sampling</td>
<td>Short pumping test, extraction rate approx. 5 l/s, sampling at start, after 1 h, after 2 h, after 4 h and after 6 h.</td>
</tr>
<tr>
<td>Investigation parameters</td>
<td>Electrical conductivity, pH, O₂, Cl, SO₄, NO₃, CHC</td>
</tr>
</tbody>
</table>

* mo = upper Muschelkalk
5.6 Implementation

The success of the groundwater management plan is measured by the improvement in the quality of groundwater in Stuttgart. For this, it is crucial that the necessary measures, i.e. the investigation and remediation measures and the monitoring program be put into practice. To this end, a summary of the groundwater management plan, the measures and financing plan and the next steps were summarized in a document for the city council's information.

Measures for the remediation of the groundwater are defined and enforced by the competent authority in the context of its responsibility under the current legal framework. The city council has made a provision in the budget for the remediation of sites in urban responsibility. The implementation of the measures described in the groundwater management plan is thus secured.

5.7 Suitability of the "Groundwater Management Plan" instrument

The project shows that the MAGPlan strategy and the processes presented are particularly suitable when multiple pollution sources exist. Only by an integral study the contributions of individual sites to the overall situation can be identified and local remediation measures simulated with their spatial impact. The prognostic scenario modelling with the numerical model allows an investigation of the effects of remediation measures. This includes a thorough consideration of spatially effective pollution attenuation processes.

All measures that serve to achieve the quality objectives defined for the groundwater body are summarized in one document. Both the remediation feasibility study and the description of the measures are bundled in this document. This allows clearly defined decision-making processes, focused administrative procedures and reliable budget planning.

The initiative for the application of the instrument of the groundwater management plan will usually originate from the water and soil protection authorities in their efforts towards effective administrative action. Also local authorities will rely on it to support, for example, regeneration processes for urban quarters. In individual cases and usually on a smaller scale also land owners who are interested in effective remediation and resolution of liability issues with groundwater-contaminating neighbors may apply the methodology. Also on this scale, solutions achieved with the integral approach are effective.

The costs for the establishment of a groundwater management plan depend on the availability and quality of existing data, on the size and the hydrogeological conditions of the project area as well as the complexity of the pollutant situation. In the case of the city of Laupheim [9] for example, knowledge could be acquired at a cost of a few € 100,000, which allowed a reliable identification of trends relevant for the CHC-polluted sites.
In Stuttgart, although a comprehensive and high-quality data pool existed, this required a complex initial analysis. The project area is of high complexity because of its many small-scale sites and the hydrogeological conditions. In this respect, both the high financial expense of € 3.2 million as well as the long project duration of 6 years were necessary. Compared to the costs so far to investigate and remediate in the project area of around € 107 million, these expenses are well justified. Technically, there is no alternative to this approach in order to achieve the ultimate goal of CHC-free certified spas and mineral springs. Stuttgart has got much closer to this goal.

5.8 Literature Chapter 5


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