Performance assessment of liquid activated carbon enhanced bioremediation of a TCE plume by use of isotopic and molecular biology techniques

Chlorinated solvent plumes continue to pose challenge. Enhanced reductive dechlorination (ERD) by donor addition and bioaugmentation constitute a potential sustainable technology. A novel technology combining ERD with liquid activated carbon (LAC) amendment has the potential to introduce a bioactive zone with increased degradation. Documentation and quantification of biodegradation is challenged by the combined sorption and degradation. At a TCE plume undergoing LAC amended bioremediation, compound specific isotope analysis (CSIA) for documentation and rate determination, and molecular biology tools (MBT) for determination of specific chloroethenes degraders and their genes are applied to water and sediment. A laboratory treatability experiment is carried out to support the field data evaluation. The challenges in evaluation of the new remediation technology and in applying CSIA and MBT and the lessons learned will be discussed.

The attendees will benefit by research insights in the new technology and CSIA and MBT assessment.

Conceptualization of flow and transport in a limestone aquifer by multiple dedicated hydraulic and tracer tests

Limestone aquifers are of great interest as a drinking water resource in many countries. They often have a complex crushed and fractured geology, which makes the analysis and description of flow and transport processes in such aquifers a challenging task. In this study, the solute transport behavior including fracture-matrix interaction in hydrogeological units of a limestone aquifer in eastern Denmark was characterized by designing, conducting and interpreting six depth-specific tracer tests involving natural- and forced-gradient conditions with multiple tracers representing different diffusion properties. To determine flow parameters, the tracer tests were complemented by a comprehensive set of depth-specific borehole and hydraulic tests.

Based on the tests, a new and stronger conceptual understanding was developed for the different aquifer units. The investigated limestone aquifer is composed of a glacially crushed unit and two fractured units, with calcarenitic and bryozoan limestone of similar hydraulic properties. Hydraulic tests revealed, that the crushed unit has a lower hydraulic conductivity than the fractured limestone units, likely due to the crushed conditions with small limestone clusters and small-aperture fractures potentially filled with fine material.

In the fractured limestone units, a distinct preferential flow and primary transport along major horizontal fractures was inferred from the tracer tests under forced-gradient conditions. The dominant horizontal fractures were identified on impeller flow logs and appear connected between wells, having an extent of up to several hundred meters. Connectivity between the aquifer units was investigated with a long-term pumping test and tracer tests, revealing restricted vertical flow and transport. A very pronounced hydraulic conductivity contrast between major fractures and matrix could also be inferred from the borehole and hydraulic tests, which is consistent with the findings from the tracer tests. However, the difference in the matrix diffusion behavior of the simultaneously injected tracers and a long tailing in the breakthrough curves revealed that matrix diffusion has a strong influence on the solute transport in the fractured limestone.
Conceptualization of contamination using depth discrete monitoring of dynamic PCE concentration changes during pumping

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Conceptualization of residual contamination using depth discrete monitoring of dynamic PCE concentration changes during and after remedial pumping and pumping test

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Conceptualization of residual contamination using depth discrete monitoring of dynamic PCE concentration changes during and after remedial pumping and pumping test

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Forced-gradient tracer tests in a fractured limestone aquifer designed and interpreted by modeling

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Integrity of clay till aquitards to DNAPL migration: Assessment using current and emerging characterization tools

Field investigations were carried out to determine the occurrence of tetrachloroethene (PCE) dense non-aqueous phase liquid (DNAPL), the source zone architecture and the aquitard integrity at a 30-50 year old DNAPL release site. The DNAPL source zone is located in the clay till unit overlying a limestone aquifer. The DNAPL source zone architecture was investigated through a multiple-lines-of-evidence approach using various characterization tools; the most favorable combination of tools for the DNAPL characterization was geophysical investigations, Membrane Interface Probe (MIP), core subsampling with quantification of chlorinated solvents, hydrophobic dye test with Sudan IV and Flexible Liner Underground Technologies (FLUTE) NAPL liners with activated carbon felt (FACT). While the occurrence of DNAPL was best determined by quantification of chlorinated solvents in soil samples supported by the hydrophobic dye tests (Sudan IV and NAPL FLUTE), the conceptual understanding of source zone architecture was greatly assisted by the indirect continuous characterization tools. Although mobile or high residual DNAPL (St >1%) only occurred in 11% of the source zone samples (intact cores), they comprised 86% of the total PCE mass. The data set, and associated data analysis, supported vertical migration of DNAPL through fractures in the upper part of the clay till, horizontal migration along high permeability features around the redox boundary in the clay till, and to some extent vertical migration through the fractures in the reduced part of the clay till aquitard to the underlying limestone aquifer. The aquitard integrity to DNAPL migration was found to be compromised at a thickness of reduced clay till of less than 2 m.

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Modeling of flow and transport processes in a fractured limestone aquifer

Characterization of chlorinated solvent contamination in limestone using innovative FLUTE® technologies in combination with other methods in a line of evidence approach

Characterization of dense non-aqueous phase liquid (DNAPL) source zones in limestone aquifers/bedrock is essential to develop accurate site-specific conceptual models and perform risk assessment. Here innovative field methods were combined to improve determination of source zone architecture, hydrogeology and contaminant distribution. The FACT™ is a new technology and it was applied and tested at a contaminated site with a limestone aquifer, together with a number of existing methods including wire-line coring with core subsampling, FLUTE® transmissivity profiling and multilevel water sampling. Laboratory sorption studies were combined with a model of contaminant uptake on the FACT™ for data interpretation. Limestone aquifers were found particularly difficult to sample with existing methods because of core loss, particularly from soft zones in contact with chert beds. Water FLUTE™ multilevel groundwater sampling (under two flow conditions) and FACT™ sampling and analysis combined with FLUTE® transmissivity profiling and modeling were used to provide a line of evidence for the presence of DNAPL, dissolved and sorbed phase contamination in the limestone fractures and matrix. The combined methods were able to provide detailed vertical profiles of DNAPL and contaminant distributions, water flows and fracture zones in the aquifer and are therefore a powerful tool for site investigation. For the limestone aquifer the results indicate horizontal spreading in the upper crushed zone, vertical migration through fractures in the bryozoan limestone down to about 16–18 m depth with some horizontal migrations along horizontal fractures within the limestone. Documentation of the DNAPL source in the limestone aquifer was significantly improved by the use of FACT™ and Water FLUTE™ data.
Modeling contaminant plumes in fractured limestone aquifers

Determining the fate and transport of contaminant plumes from contaminated sites in limestone aquifers is important because they are a major drinking water resource. This is challenging because they are often heavily fractured and contain chert layers and nodules, resulting in a complex transport behavior. Improved conceptual models are needed for this type of site. Here conceptual models are developed by combining numerical models with field data. Several types of fracture flow and transport models are available for the modeling of contaminant transport in fractured media. These include the established approaches of the equivalent porous medium, discrete fracture and dual continuum models. However, these modeling concepts are not well tested for contaminant plume migration in limestone geologies. Our goal was to develop and evaluate approaches for modeling the transport of dissolved contaminant plumes in fractured limestone aquifers in 3D and to test methods for determining the required flow and transport parameters.

The models were compared for a contaminated site in Denmark, where a plume of dissolved PCE has migrated through a fractured limestone aquifer. Numerical modeling was used in the planning of field tests and to update the conceptual model in an iterative process. Field data includes information on spill history, distribution of the contaminant (multilevel sampling), geology and hydrogeology. To describe the geology and fracture system, data from borehole logs, packer tests, optical televiewers and cores was combined with an analysis of local heterogeneities and data from analogous sites. A combined pump and tracer test was performed at the site with simultaneous contaminant sampling to determine flow and transport parameters of the fractures and matrix, and to quantify the contaminant distribution in the aquifer. Different models were used for the planning and interpretation of the pump and tracer test.

The models were evaluated by examining their ability to describe collected field data. The comparison with data showed that the models have substantially different representations of the contaminant behavior, with different consequences for evaluation of contaminant risk and potential remediation strategies. For instance, the fractured aquifer means that tracer tests result in fast breakthroughs, while larger scale plume transport is much slower. On the plume scale, the equivalent porous medium model and the dual-porosity model can reproduce the main features of the plume at a given time. However, small-scale fracture-matrix interactions such as diffusion of contaminant into the matrix result in non-linear plume speeds, and these cannot be represented with an equivalent-porous medium model. The paper concludes with recommendations on how to identify and employ suitable models to advance the conceptual understanding and as decision support tools for risk assessment and the planning of remedial actions.
Modeling contaminant plumes in fractured limestone in 3-D: comparison of modeling approaches

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Modeling contaminant transport in limestone aquifers

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Modeling contaminant transport in limestone aquifers

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Evaluation of modeling approaches to simulate contaminant transport in a fractured limestone aquifer

Understanding the fate and transport of contaminants in limestone aquifers is important because they are a major drinking water resource. This is challenging because they are often heavily fractured and contain chert layers and nodules. Several modeling approaches have been developed to describe contaminant transport in fractured media, such as discrete fracture, equivalent porous media, and dual continuum models. However, these modeling concepts are not well tested for real limestone geologies. Our goal is therefore to develop, evaluate and compare approaches for modeling transport of contaminants in fractured limestone aquifers.

The model comparison is conducted for a contaminated site in Denmark, where a plume of dissolved PCE has migrated through a fractured limestone aquifer. Field data includes information on spill history, distribution of the contaminant (multilevel sampling), geology and hydrogeology. To describe the geology and fracture system, data from borehole logs and cores was combined with an analysis of heterogeneities and fractures from a nearby excavation and pump test data.

We present how field data is integrated into the different model concepts. A challenge in the use of field data is the determination of relevant hydraulic properties and interpretation of aqueous and solid phase contaminant concentration sampling data. Traditional water sampling has a bias towards fracture sampling; however, concentrations in the limestone matrix are needed for assessing contaminant rebound and remediation strategies.

Each model is compared with field data, considering both model fit and model suitability. Results show a considerable difference between the approaches, and that it is important to select the right one for the actual modeling purpose. The comparison with data showed how much information is required to discriminate between models, and recommendations on how to identify the best modeling approach are made.

High-resolution characterization of DNAPL source zone architecture in clay till
Method of tracing engineered nanoparticles

The present application discloses a population of non-aggregated polymer-coated nanoparticles having a mean particle size (diameter) in the range of 1-100 nm, said population comprising (i) a first subpopulation of (re)active particles coated with a first polymer, and (ii) a second subpopulation of non-(re)active tracer particles coated with a second polymer, wherein the weight ratio between said first subpopulation and said second subpopulation is from between 85:15 to 99.99:0.01, as well as a composition comprising such a population. The population of particles is particularly useful for tracking of particles in methods for remediation, including in-situ remediation, of contaminated ground water and for water treatment.
Characterization of chlorinated solvents in fractured limestone aquifers is essential for proper development of site specific conceptual models and subsequent risk assessment and remediation. High resolution characterization is challenged by the difficulties involved in collection of intact core samples as water flushing during drilling often results in extensive core losses, especially from zones with soft limestone in contact with flint beds. Field investigations with alternative characterization techniques have been carried out at two Danish sites with tetrachloroethene (PCE) contaminated fractured limestone aquifers. The two sites represent different scales (source and plume) and contaminant levels (DNAPL and dissolved). The scope of the investigations was to evaluate different techniques for characterization of the contaminant distribution in the limestone aquifers and to obtain an improved conceptual understanding of contaminant transport.

At both sites limestone cores were collected with significant core losses. The discrete quantification of chlorinated solvents in the retrieved limestone cores was compared to different FLUTe technologies at the DNAPL site and passive and active multilevel groundwater sampling at the plume scale site. Important information regarding contaminant distribution and potential presence of DNAPL was provided by FACT (FLUTe activated carbon technique) and Water-FLUTe multilevel sampling. The data was used to validate a model based tool for interpretation of the FACT field measurements, which allows the conversion of discrete activated carbon concentrations to aqueous concentrations at given hydraulic parameters and FACT parameters. The passive groundwater sampling with snap samplers resulted in significantly different concentration levels and concentration profiles over depth compared to the active sampling by separation pumping with heat pulse technology. The differences between the two techniques decreased with distance to the source area. Overall, the borehole characterization techniques provided an improved conceptual understanding of the contaminant distribution compared to the data obtained by quantification of chlorinated solvents in the limestone cores.
Assessing the risks posed by contaminated sites to water resources

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Challenges in subsurface in situ remediation of chlorinated solvents
Chlorinated solvent source zones in the subsurface pose a continuous threat to groundwater quality at many sites worldwide. In situ remediation of these sites is particularly challenging in heterogeneous fractured media and where the solvents are present as DNAPL. In situ remediation by chemical as well as biological degradation of chlorinated solvents is a contact sport and requires direct contact between the contaminant and the reactants and/or degrading microorganisms. In fractured geologic media, where contaminants have spread to the low permeability matrix by diffusion, the contact between contaminant and reactant is limited by slow back diffusion of contaminant and in-diffusion of reactant if the only access for the reactant is via the high permeability fractures/conduits. Where DNAPL is present the mass distribution is very heterogeneous and the reactive degradation is often limited by dissolution of the DNAPL. Most recent research has been aimed at overcoming these challenges by enhanced and targeted reactant delivery methods. These include a wide range of very diverse technologies such as: enhanced injection methods, including fracturing; electrokinetic enhancement of delivery; ZVI-clay mixing for contact; hydrophobic and/or mobile nano-reactants targeting DNAPL. The complexity of the technologies varies greatly and the current level of implementation ranges from multiple full scale applications to bench scale testing. However, the basic degradation reaction involved is usually well established. Enhanced injection with fracturing increases the access to contaminants in clay/clayey media matrixes by shortening the diffusive distance and with ZVI-clay technology by physically mixing the reactant with the contaminated clay/clayey media. The efficiency of the injection technologies has been very variable and rather unpredictable in heterogeneous geologic media, hence, further developments are needed. The novel techniques involving electrokinetics induce migration of primarily ionic species/reactants independent of hydraulic permeability differences, hence transporting the reactant into the contaminated matrix and may be applicable for limestone/bedrock as well as clayey media. Only laboratory studies of electrokinetic enhancement have yet been published, and there is a need for thorough pilot scale studies and supporting laboratory studies. Injectable nano-particles with an affinity for DNAPL surfaces (or phases) may overcome dissolution limitations and provide direct contact with contaminant, limiting reactions with other reactive sites in the subsurface. Challenges lie in obtaining stability and mobility in water, affinity for DNAPL and at the same time maintain reactivity with contaminants. Upscaling to production for pilot studies without loss of efficiency is not trivial. In conclusion there continues to be a need for research and development and in particular for well documented pilot/full scale field studies.

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Integrated Characterization of DNAPL Source Zone Architecture in Clay Till and Limestone Bedrock

Background/Objectives. Characterization of dense non-aqueous phase liquid (DNAPL) source zone architecture is essential to develop accurate site specific conceptual models, delineate and quantify contaminant mass, perform risk assessment, and select and design remediation alternatives. The activities of a distribution facility for perchloroethene (PCE) and trichloroethene (TCE) at the Naverland site near Copenhagen, Denmark, has resulted in PCE and TCE DNAPL impacts to a fractured clay till and an underlying fractured limestone aquifer/bedrock. The scope of the investigations was to evaluate innovative investigation methods and characterize the source zone hydrogeology and contamination to obtain an improved conceptual understanding of DNAPL source zone architecture in clay till and bryozoan limestone bedrock.

Approach/Activities. A wide range of innovative and current site investigative tools for direct and indirect documentation and/or evaluation of DNAPL presence were combined in a multiple lines of evidence approach.

Results/Lessons Learned. Though no single technique was sufficient for characterization of DNAPL source zone architecture, the combined use of membrane interphase probing (MIP); coring with quantitative subsample analysis, SudanIV test, and PID; and NAPL FACT FLUTe gave good insight in the source zone architecture in the clayey till. Surface geophysics with ground penetrating radar (GPR) and seismic reflection and refraction combined with geologic information supplemented the conceptual understanding of transport and distribution of DNAPL in the fill and clayey till and the interface to the limestone. Core loss in the limestone, particularly from soft zones in contact with flint beds, was caused by the water flush applied during drilling and likely also resulted in loss of DNAPL from high permeability features. Hence, coring and subsampling for quantitative analysis and SudanIV tests continues to be an unresolved challenge in limestone. The coring may also have impacted DNAPL in high permeability zones near the borehole, thereby, potentially affecting the use of the NAPL FLUTe. Water-FLUTe multilevel groundwater monitoring and sampling (under two flow conditions) and FACT-FLUTe sampling and analysis provided important information regarding potential presence of DNAPL versus dissolved and sorbed phase contamination in the limestone matrix. These combined methods provided an improved conceptual understanding of DNAPL source zone architecture in fractured limestone. The DNAPL source zone architecture in the clay till was consistent with conceptual expectations. In contrast the documentation for and quantification of DNAPL in the limestone aquifer was limited and demands refinement of techniques and further characterization.

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Polymer coated gold nanoparticles for tracing the mobility of engineered nanoparticles in the subsurface

Nanoparticles (NPs) are manufactured for their specific properties providing possibilities for new and improved products and applications. The use of engineered nanoparticles (ENPs) has therefore brought significant innovation and advances to society, including benefits for human health and the environment. At the same time, little is known about the potential risk associated with the inevitable release of these new materials to the environment, and their new properties are poorly understood. Suspensions of ENPs are not very stable, as they tend to aggregate thereby losing their properties as single particles. Coatings, including a large variety of natural and synthetic polymers, are used to enhance the colloid stability in high concentrations. However, increasing the stability of these materials may lead to unintended effects, such as enhancing their mobility in surface water and groundwater leading to inadvertent impacts on aquatic ecosystems and human health. Detection of ENPs in natural water systems, however, has proved very challenging. Hence, there is a need for tracing of ENP behaviour in the environment. We suggest a possibility of introducing inert gold NPs with the same mobility as the reactive NPs, as tracer particles. Colloidal gold has been of great interest for centuries due to its vibrant colors produced by the interaction with visible light. The unusual optical-electronic properties, high chemical stability and relatively low toxicity have made them the model system of choice in this context. Also, the natural occurrence of these particles in the proposed environment is very rare. Laboratory based experiments conducted in sand columns show that stable aqueous suspensions of gold NPs coated with amphiphilic block co polymers (PVP-VA and PVA-COOH) are extremely mobile (retardation factors of 1.0-1.2) with high recovery values (50-95%). The specific retardation and recovery depends on the coating type, concentration and grafting method. The NPs also show significant partitioning to organic phases such as tetrachloroethylene (PCE) and trichloroethylene (TCE), which are considered as potential ground water pollutants accumulated in the subsurface as DNAPLs (dense non-aqueous phase liquids). Being a noble metal, nanogold is to be detected by nondestructive optical methods at a concentration of at least 1000 fold lower than ENPs. Using conventional spectrophotometric technique equipped with liquid waveguide capillary cell (LWCC), nanogold is detected at very low concentration range (1 ppm – 62.5 ppb). Compared to uncoated particles, surface modified nanogold with polymers retains the plasmonic peaks at 520 nm when diluted with artificial ground water.

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Polymer coated nanogold for tracing mobility of engineered nanoparticles in subsurface water
Gold nanoparticles coated with amphiphilic block co-polymer PVP-VA are found to be extremely mobile in sand columns in laboratory based experiments. The ultra-stability obtained by such surface modification is also shown by diluting down to a concentration of 62.5 ppb in groundwater having other ions. Such a low level detection of particles is achieved by non-destructive simple UV-VIs spectroscopic technique equipped with liquid wave guide capillary cell (LWCC) units. These results propose the possibility of using surface engineered gold nanoparticles as model system for tracing the mobility of other engineered nanoparticles in the subsurface.

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Risikovurdering af overfladevand, som er påvirket af punktkildeforurenet grundvand

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Risk assessment of contaminated sites and diffuse sources to water resources

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Site characterization and in-situ remediation in fractured geologic media contaminated by chlorinated solvents

General information
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Towards Coated Nano-Gold Particles as Non-Reactive Tracers in Coated nZVI for In-Situ Remediation

Background/Objectives. Chlorinated solvent (e.g. trichloroethene and tetrachloroethene) source zones in the subsurface pose a continuous threat to groundwater quality at many sites worldwide. Remediation of these contaminated sites is especially challenging in the presence of Dense Non-Aqueous Phase Liquids (DNAPLs) and the development of innovative remediation technologies is still needed. It has been proved, in laboratory scale as well as in field tests, that nanoparticles of zero-valent iron (nZVI) are efficient in degrading chlorinated solvents in aqueous solution, due to their large surface area and reducing ability. Stability of nZVI in aqueous emulsions and mobility of nZVI in the subsurface has been obtained through coatings. However, lack of knowledge of the fate of nZVI once they are released to the environment, including their transport, reactivity, mobility, and affinity towards various environmental phases, hinders the implementation of an effective environmental remediation process. Also, from a risk assessment perspective, it is of great interest to know the dispersion of such reactive nanoparticles in the environment. A possibility is the introduction of non-reactive tracer particles with the same mobility and partitioning properties as the reactive particles. In this study, the use of gold nanoparticles (nAu) mixed in tracer amounts to track the fate of nanoparticles in porous media is investigated, initially at the lab scale.

Approach/Activities. Colloidal nAu have been of great interest for centuries due to their vibrant colors produced by the interaction with visible light. These unique optical – electronics properties along with their very low natural abundance, high chemical stability and relative low toxicity have made them useful in such tracing applications. In this project, 30 nm sized citrate stabilized nAu were and coated with amphiphilic block copolymers such as PVP-VA and PVA-COOH. The coated nAu were found to be stable in water and in a number of organic solvents as well. The partitioning of the nAu from an aqueous solution (5 mg/L) into two different DNAPLs (tri- and tetrachloroethene) was tested. The nAu mobility was investigated in sand packed columns and the breakthrough was monitored using spectrophotometry on the effluent.

Results/Lessons Learned. The results showed nanoparticles with great mobility and a significant affinity for the DNAPL. The breakthrough curve from the mobility study showed that only minimal retardation and tailing occurred at both the front and back of the nAu plume. These results obtained by conducting model experiments (lab scale aerobic) using nAu coated with amphiphilic block copolymers like PVP-VA and PVA-COOH, hold great promise for the co-injection and DNAPL affinity of both reactive (nZVI) and tracer nanoparticles. DNAPL affinity of the coated nZVI is expected to target treatment of DNAPL source zones.
Characterization of DNAPL Source Zone Architecture in Clay Till and Limestone Bedrock by Integrated Site Investigations with Innovative and Current Techniques

Characterization of dense non-aqueous phase liquid (DNAPL) source zone architecture is essential to develop accurate site specific conceptual models, delineate and quantify contaminant mass, perform risk assessment, and select and design remediation alternatives. The activities of a distribution facility for perchloroethene (PCE) and trichloroethene (TCE) at the Naverland site near Copenhagen, Denmark, has resulted in PCE and TCE DNAPL impacts to a fractured clay till and an underlying fractured limestone aquifer/bedrock. A range of innovative and current site investigative tools for direct and indirect documentation and/or evaluation of DNAPL presence were combined in a multiple lines of evidence approach. The scope of the investigations was to evaluate innovative investigation methods and characterize the source zone hydrogeology and contamination to obtain an improved conceptual understanding of DNAPL source zone architecture in clay till and bryozoan limestone bedrock.

Surface investigations included Ground Penetrating Radar (GPR) and seismic reflection and refraction. Investigations in the clay till included membrane interface probing (MIP) with FID, ECD and GC-MS analysis; coring with discrete subsampling for quantitative analysis, SudanIV hydrophobic colour test, colour spray test, PID and geologic descriptions; NAPL and FACT FLUTe exposure and discrete FACT subsampling and analysis; liquid sampling from boreholes; and a radon and PCE/TCE soil gas survey. Investigations in the limestone aquifer included coring with discrete subsampling for quantitative analysis, SudanIV hydrophobic colour test, PID and geologic descriptions; NAPL and FACT FLUTe exposure and discrete FACT subsampling and analysis; FLUTe liner hydraulic conductivity profiling; Water-FLUTe installation and multilevel groundwater sampling for quantitative analysis under two flow conditions.

Though no single technique was sufficient for characterization of DNAPL source zone architecture, the combined use of MIP; coring with quantitative subsample analysis, SudanIV test, and PID; and NAPL FACT FLUTe gave good insight in the source zone architecture in the clayey till. Surface geophysics with GPR and seismic combined with geologic information supplemented the conceptual understanding of transport and distribution of DNAPL in the fill and clayey till and the interface to the limestone. Core loss in the limestone, particulary from soft zones in contact with flint beds, was caused by the water flush applied during drilling and likely also resulted in loss of DNAPL from high permeability features. Hence, coring and subsampling for quantitative analysis and SudanIV tests continues to be an unresolved challenge in limestone. The coring may also have impacted DNAPL in high permeability zones near the borehole, thereby, potentially affecting the use of the NAPL FLUTe. Water-FLUTe multilevel groundwater monitoring and sampling (under two flow conditions) and FACT-FLUTe sampling and analysis provided important information regarding potential presence of DNAPL versus dissolved and sorbed phase contamination in the limestone matrix. These combined methods provided an improved conceptual understanding of DNAPL source zone architecture in fractured limestone. It is expected that down-hole logs, cross-borehole geophysical methods, and flow and solute transport modeling can supplement the conceptual understanding of DNAPL distribution in limestone aquifers.

The DNAPL source zone characterization showed DNAPL at the fill-clay till interface, vertical migration through fractures in the upper part of the clay till, horizontal migration along fractures and/or other high permeability features around the redox transition zone in the clay till, and then continued vertical migration through fractures to the underlying sand and limestone. This is consistent with conceptual expectations based on contaminant distribution at other clay till sites. For the limestone aquifer the results indicate horizontal spreading in the upper crushed zone, vertical migration through fractures in the bryozoan limestone down to about 16-18 depth with some horizontal migration along horizontal fractures within the limestone. In conclusion, the documentation for and quantification of DNAPL in the limestone aquifer is limited and demands refinement of current and innovative techniques and further characterization.

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Research output: Contribution to conference › Conference abstract for conference – Annual report year: 2013 › Research › peer-review

DNAPL i moræneler og kalk - vurdering af undersøgelsesmetoder og konceptuel modeludvikling: Naverland 26AB, Albertslund
Field methods for determining point source pollution impacts in rivers: A case study of the Grindsted stream

Well-known organic contaminants such as chlorinated solvents, as well as new classes of compounds or emerging micropollutants (e.g., pharmaceuticals) are extensively produced, utilized and then discarded in society and subsequently released to streams from multiple sources. To address this, the EU Water Framework Directive requires member states to evaluate all types of contamination sources within a watershed in order to assess their direct impact on water quality. Understanding and accurately characterizing groundwater-surface water interactions (GSI) and groundwater discharge is...
thus becoming an increasingly important activity for the hydrogeological investigations of rivers and streams. In cases where groundwater contaminant plumes are discharging to streams, determination of flow paths and groundwater fluxes are essential for evaluating the transport, fate and potential impact of the plume. This implies that investigators have the tools to easily and accurately evaluate the governing parameters, including an appreciation of the scale of variability, as well as conceptual models that incorporate the various mechanisms affecting flow.

An in-depth field investigation of the Grindsted stream was carried out in 2012, to develop the theoretical basis for conducting risk assessments for contaminated sites impacting surface waters. Grindsted stream was chosen, as groundwater flow is known to comprise an important part of the total water supply to the stream. It is also a well-studied site, affected by two major polluting point sources, Grindsted factory and Grindsted landfill, representing two of the 43 large-scale contaminated sites in Denmark. Our overall aim was therefore to (i) test the applicability of different methods for mapping groundwater pollution as it enters streams at a complex site, and (ii) perform a risk assessment of the stream’s chemical status, including documentation of emerging contaminants. A secondary aim was to identify and ideally separate the entry point for the two plumes to Grindsted stream.

We successfully detected six significant local-scale GSI "contact" zones along a 5 km stream stretch, which were not visible at the regional scale, using systematic temperature measurements. We then correlated the two highly contaminated contact zones, using piezometers placed where streambed temperature measurements were <10°C, to concentrations in downstream surface waters. Transects placed perpendicular to stream flow in the contact zones allowed us to effectively localize the Grindsted factory plume using samples containing a unique compositional footprint consisting of chlorinated solvents, barbiturates, sulfonamides, sulfanilic acid and bromide specific for the contaminated site. Notably, the highly volatile and toxic compound vinyl chloride was found to exceed the surface water quality criterion (0.05 µg/L) for a ca. 5 km stretch, to our knowledge the only documented study of its kind. Additionally, the sum of sulfonamides was also shown to exceed the recommended criterion (4.6 µg/L) twice along the same stream stretch. Further investigations will be necessary to finalize the location of the Grindsted landfill plume where it enters the stream; specifically we recommend that two-dimensional cross-sectional characterization be conducted to identify the direction and width of this groundwater plume.
Metoder til undersøgelse af jordforurenings påvirkning af vandløb

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Hele abstract samlingen

Abstract

Udvikling af konceptuel forståelse af DNAPL udbredelse i moræneler og kalk

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Udvikling af konceptuel forståelse af DNAPL udbredelse i moræneler og kalk ved integreret anvendelse af direkte og indirekte karakteriseringsmetoder

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Research output: Chapter in Book/Report/Conference proceeding › Conference abstract in proceedings – Annual report year: 2013 › Research › peer-review

Zero valent iron reduces toxicity and concentrations of organophosphate pesticides in contaminated groundwater
The potential of zero valent iron (ZVI) for remediation of contaminated groundwater from an abandoned chemical disposal site was examined through batch and column experiments. The key contaminants were organophosphate pesticides but the chemical analysis also comprised additional 22 compounds including synthesis intermediates and degradation products of organophosphates. The ZVI treatment showed that all the contaminants were degraded with the exception of two diesters (phosphorothioates). The most rapid reduction was found for methyl parathion, ethyl parathion and malathion, which had first-order degradation rate constants on the order of 10−3 min−1. In the study, acute toxicity towards freshwater crustaceans (Daphnia magna) was included to evaluate the overall efficiency of ZVI treatment of the complex mixture. The acute toxicity tests with D. magna showed that the untreated groundwater was highly toxic. Thus, 50% of the daphnids were unable to swim upon 24h exposure to groundwater diluted 770 times. ZVI facilitated degradation resulted in a complete toxicity removal for the first four pore volumes, where after a three times dilution caused 50% inhibition of the mobility of the daphnids. The rapid degradation of the highly toxic organophosphates combined with the significant decrease in the ecotoxicological potential shows a promising potential for site remediation of organophosphates with ZVI technologies.

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Organisations: Water Resources Engineering, Department of Environmental Engineering, Environmental Chemistry, Residual Resource Engineering, Technical University of Denmark
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Source zone remediation by ZVI-clay soil-mixing: Reduction of tetrachloroethene mass and mass discharge at a Danish DNAPL site

The presence of chlorinated solvent source zones in the subsurface pose a continuous threat to groundwater quality. The remediation of Dense Non-Aqueous Phase Liquid (DNAPL) sites is especially challenging and the development of innovative remediation technologies is needed. Zero-valent iron (ZVI) technologies have proven effective for remediation of chlorinated compounds. ZVI-Clay soil-mixing is a new remediation technology, which combines abiotic degradation (via ZVI addition) and immobilization (via soil-mixing and clay addition), whereby a great potential for reduction of both contaminant mass and mass discharge is obtained.
The technology was tested at a Danish DNAPL site, where the secondary aquifer was heavily contaminated by tetrachloroethene (PCE). ZVI-Clay soil-mixing was tested at a small source zone (~200 m³) with soil concentrations ranging up to 12,000 mg/kg. The objective of the field test was to document in situ destruction of the contaminant mass and the down-gradient response in contaminant mass discharge.

The field sampling consisted of baseline measurements and a 19-month monitoring program (7 sampling campaigns) subsequent to the implementation of ZVI-Clay soil mixing. The concentrations of chlorinated ethenes were monitored via soil sampling at the source zone and groundwater sampling at a control plane with multilevel samplers covering the entire contaminated plume down-gradient (3 m) of the source zone.

The results showed a significant mass depletion of PCE (2-3 orders in magnitude) with ethene as the main degradation product. The down-gradient reduction of contaminant mass discharge occurred more slowly; after 19 months a mass discharge reduction of 76 % was obtained for PCE. However, due to a temporary increase in cis-DCE, the overall down-gradient reduction of all the chlorinated ethenes was limited to 21 %. Long-term modeling (Comsol Multiphysics) was used to predict that a contaminant mass discharge reduction of 2-3 orders in magnitude will take 3-5 years.

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Source: dtu
Research output: Chapter in Book/Report/Conference proceeding
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Research year: 2012
Research type: Conference abstract in proceedings – Annual report

**Source Zone Remediation by ZVI-Clay Soil-Mixing: Reduction of Tetrachloroethene Mass and Mass Discharge at a Danish DNAPL Site**
ZVI-Clay soil-mixing is a relatively new in situ remediation technology for remediation of chlorinated dense non-aqueous phase liquids (DNAPLs). The technology combines abiotic degradation (via zero-valent iron, ZVI, addition) and immobilization (via soil mixing and clay addition), whereby both contaminant mass and contaminant mass discharge can be reduced. The technology was tested at a Danish tetrachloroethene (PCE) site. The field sampling consisted of baseline measurements and a 19-month monitoring program (7 sampling campaigns) subsequent to the implementation of ZVI-Clay soil-mixing. The concentrations of chlorinated ethenes were monitored via soil sampling at the source zone and groundwater sampling at a downgradient control plane. The results showed that within one year ZVI-Clay soil-mixing resulted in significant mass depletion of PCE (2-3 orders in magnitude) with ethene as the main degradation product. The down-gradient reduction of contaminant mass discharge was slower; after 19 months a mass discharge reduction of 76 % was obtained for the parent compound PCE, while the overall mass discharge reduction of chlorinated ethenes was 21 %.

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Temporal Changes to the Mass Discharge of Tetrachloroethene Subsequent to ZVI-Clay Soil Mixing at a DNAPL Source Zone

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Place of publication: Columbus
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Research output: Chapter in Book/Report/Conference proceeding \ Conference abstract in proceedings – Annual report year: 2012 \ Research \ peer-review

ZVI-Clay remediation of a chlorinated solvent source zone, Skuldelev, Denmark: 1. Site description and contaminant source mass reduction
Field investigations on the effects of ZVI-Clay soil mixing were conducted at a small DNAPL source zone with PCE as the parent compound. In a one-year monitoring program, soil samples were collected at three horizontal sampling planes (2.5, 5.0 and 7.5m bgs.). PCE was found to have a pseudo first-order degradation half-life of 47 days resulting in more than 99% depletion of the source mass after one year. The main degradation product was ethene, while only low concentrations of the primarily biotic sequential degradation products (cDCE, VC) were detected. The soil mixing resulted in more homogeneous vertical conditions, while the horizontal homogenization was very limited. Iron was delivered in the full targeted depth with an average iron enrichment of 3.1%, and an average decline in the oxidation-reduction potential of more than 500mV. Due to the applied top-down addition of ZVI, the iron content decreased from 4.6% to 2.1% on average over a depth of 5m; hence, there is a potential for optimization of the delivery method. Most in situ technologies are limited by subsurface heterogeneities, whereby the successful dispersion of geological units and contaminants holds great promise for remediation of DNAPL source zones with ZVI-Clay soil mixing.
ZVI-Clay remediation of a chlorinated solvent source zone, Skuldelev, Denmark: 2. Groundwater contaminant mass discharge reduction

The impact of source mass depletion on the down-gradient contaminant mass discharge was monitored for a 19-month period as a part of a field demonstration of the ZVI-Clay soil mixing remediation technology. Groundwater samples were collected from conventional monitoring wells (120 samples) and a dense network of multilevel samplers (640 samples). The hydraulic gradient and conductivity were determined. Depletion of the contaminant source is described in the companion paper (Fjordbøge et al., 2012). Field data showed four distinct phases for PCE mass discharge: (1) baseline conditions, (2) initial rapid reduction, (3) temporary increase, and (4) slow long-term reduction. Numerical modeling was utilized to develop a conceptual understanding of the four phases and to identify the governing processes. The initial rapid reduction of mass discharge was a result of the changed hydraulic properties in the source zone after soil mixing. The subsequent phases depended on the changed accessibility of the contaminant mass after mixing, the rate of source depletion, and the concentration gradient at the boundaries of the mixed source zone. Overall, ZVI-Clay soil mixing resulted in a significant down-gradient contaminant mass discharge reduction (76%) for the parent compound (PCE), while the overall reduction of chlorinated ethenes was smaller (21%).

Source zone remediation by zero valent iron technologies

Chlorinated solvents have been widely used as industrial solvents. Decades of extensive use have resulted in the contamination of an estimated 800,000 sites in the western world alone. A medium to high risk of problems related to dense non-aqueous phase liquids (DNAPLs) has been assessed to exist at a fifth of these contaminated sites. These source zones pose a serious threat to soil and groundwater quality. Remediation of the heterogeneous source zones is challenging due to irregular downwards migration patterns in the subsurface, low aqueous solubility and matrix diffusion. To protect the soil and groundwater resources from long-term deterioration, the development of in situ technologies suitable for remediation of DNAPL is warranted. Currently, an array of aggressive in situ remediation technologies remediation exists. These technologies may be suitable under various site specific conditions; however, most of them are limited by subsurface heterogeneities and/or the risk of inadvertent DNAPL displacement during field application. This thesis presents the results of an investigation of the potential for remediation of chlorinated solvent source zones by emerging zero valent iron (ZVI) based technologies. These in situ remediation technologies include the injection of nanoscale ZVI (nZVI) particles and the addition of ZVI and clay through soil mixing (ZVI-Clay soil mixing). The investigation of the nZVI technology is based on an elaborate literature review, while the investigation of the ZVI-Clay soil mixing technology is based on experimental data from bench-scale studies and field applications. Generally, both technologies are efficient at degrading chlorinated solvents with only a minor production of persistent chlorinated intermediates. To date, the nZVI technology has been applied at around 40 contaminated sites, while the ZVI-Clay soil mixing technology has been applied in full-scale at 9 contaminated sites. Scarce performance data exist for most of these sites; this limits the assessment of the potential for source mass depletion and risk mitigation at down-gradient receptors by the ZVI technologies. When investigating the nZVI technology specifically, this technology utilizes the decreased particle size (}

Case Study on ZVI-Clay Remediation with Mass and Flux Reduction of PCE

Environmental benefits and risks of zero-valent iron nanoparticles (nZVI) for in situ remediation: risk mitigation or trade-off?
Flux and Mass Reduction Resulting from ZVIClay Remediation of a PCE DNAPL Source Zone

In December 2008 the first Danish field implementation of the ZVIClay soil-mixing remediation technology was initiated on a PCE DNAPL contaminated site. The technology ideally combines source zone degradation via zero-valent iron with a reduction of the saturated hydraulic conductivity via addition of bentonite clay. The degradation of PCE in the treated source area and the development in the downstream flux of chlorinated compounds have been monitored in six sampling campaigns. A PCE half-life of 50 days and a reduction of the average concentration of PCE of more than 99% were found during the first year. The main degradation product was ethene. The flux of chlorinated compounds through the downstream transect showed dechlorination. A 56% decrease in the flux of PCE and a corresponding increase in the flux of the degradation product cis-DCE were observed.

Full scale test of DNAPL remediation with ZVI-clay/soil mixing, monitoring of flux, remediation of level, hydraulic properties and geotechnical properties

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Afprøvning af ZVI Clay metoden, Område V, Skuldelev: Beskrivelse og dokumentation af installation

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Effekt af oprensning med ZVI-clay teknologien: Simulerings af massefjernelse og masseflux fra en DNAPL kilde

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Source-ID: 240187
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Environmental benefits and risks of zero-valent iron nanoparticles (nZVI): risk mitigation or trade-off?
The use of nanoscaled particles in environmental remediation is gaining increasing amounts of attention in recent years, including the use of zero-valent iron nanoparticles (nZVI) for soil and groundwater remediation. The main advantages of its use include high degrees of reactivity towards a wide range of contaminants, enhanced mobility of the often coated particles, and its cost-effective in situ applications. Numerous studies have shown that compared to larger sized iron particles nZVI may have some superior properties, due to high surface areas and small sizes associated with nanoscale dimensions. While the use and further development of nZVI is understandably heralded as an environmentally-beneficial technology, the potentials risks of introducing these nanoparticles into the environment also needs to be considered. To date most research has focused on the potential benefits of nZVI and very little research has investigated its potential health and environmental risks. Nonetheless, some recent studies have documented adverse effects from its exposure including the generation of reactive oxygen species (ROS), oxidative stress, bactericidal effects, DNA damage, and inflammatory responses. Moreover, field site injections often involve the use of large quantities of nZVI (10-50 g/L) which may be directly injected into groundwater flow. Combined with the pursuit of designing more mobile and reactive particles, this may potentially lead to risks related to environmental exposures of substantial concentrations. In this study, we provide a brief synopsis of the expected environmental benefits and potential risks of nZVI, particularly focusing on its environmental fate and behavior and potential role as contaminant carrier. These are some areas of primary concern for risk assessors. Furthermore, we estimate and compare the span between probable environmental concentrations from its use in the field and concentrations which have been shown to cause adverse effects in laboratory settings. This is in light of the challenges that quantitative risk assessments face for nZVI and other nanoparticles, in part due to extensive and fundamental uncertainties. These data may provide a starting point to more thoroughly investigate the potential risks of nZVI and ultimately help scientists, engineers, and decision makers make better informed decisions regarding the use of nZVI for environmental remediation.

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Fuld-skala test af ZVI-clay-metoden til oprensning af DNAPL

General information
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year: 2009 › Research › peer-review

Afprøvning af ZVI-Clay metoden til oprensning af DNAPL på lokaliteten Vestergade 5, Skuldelev: Fase 1: Litteratur studie og treatability forsøg

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Organisations: Department of Environmental Engineering, Water Resources Engineering, Residual Resource Engineering, NIRAS A/S
Contributors: Fjordbøge, A. S., Kjeldsen, P., Christensen, A. G., Riis, C. E., Larsen, M. B.
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Anvendelse af ZVI-clay teknologien til oprensning af DNAPL-forurening

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Assessment of zero-valent iron technologies for source remediation of organophosphorous insecticides

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Contributors: Fjordbøge, A. S., Kjeldsen, P.
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Oprensning af forureningen på depotet ved Høfde 42 ved hjælp af nul-valent jern

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Evaluering af afværgeteknologier med nul-valent jern til oprænsning af høfde 42 depotet

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