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# In-situ remediation of chlorinated solvents in clayey till by enhanced reductive dechlorination

## Enhancement techniques for in-situ remediation and effect of ERD in clayey till matrix

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Contamination, constituting a threat to groundwater resources, often occurs in low permeability sediments such as clayey tills overlying aquifers. Clayey tills are mostly fractured, and contamination migrating through the fractures has spread to the low permeability matrix by diffusion. This results in a long term source of contamination due to back-diffusion. The challenge in remediation of the low permeability matrix by in-situ technologies is to bring the reactant (e.g. donor) and the contaminant and for biological methods the bacteria in contact. Remediation of the matrix is diffusion limited, as contaminants has to diffuse through the matrix to natural or induced fractures and/or sand stringers or lenses, where reactant and e.g. specific degraders can be supplied, or reactants has to diffuse into the matrix.

Enhancement techniques, such as environmental fracturing offers assistance to remediation efforts at contaminated, low-permeability sites via creation of active fracture networks, and hence, reduction of mass transport limitations set by diffusion in low-permeability matrices. Pilot studies of pneumatic fracturing, hydraulic fracturing and direct push injections, focusing on direct documentation of fracture propagation patterns and spacing and tracer distribution, were performed at a typical basal clay till site. The studies applied a novel package of documentation methods, including injection of 3-5 tracers with different characteristics (bromide, uvitex, fluorescein, rhodamine WT, and brilliant blue), subsequent tracer-filled fracture documentation via direct and indirect methods, and geological characterization of the fractured site. The direct documentation methods consisted of Geoprobe coring, augering, and excavation. Conceptual models and mass balances are being established for the distribution of the injected tracers in the subsurface.

Tracer from the pneumatic fracturing was distributed within 2 m of the fracturing well, mainly in existing fractures above the redox boundary (2 to 4 m bgs.; 5 to 10 cm spacing). Spacing of observed tracer-filled fractures was large (>1 m) at greater depths for the pneumatic fracturing test. Hydraulic fracturing was successful in creating a cm thick fracture at 3 m bgs with a radius of about 3 m. At 6-7 m bgs hydraulic fracturing was partially successful in creating sub-horisontal fractures and an attempt to create closely spaced (25 cm) hydraulic fractures failed. However, these hydraulic fractures were only encountered in few of the cored boreholes. At 9.5 m bgs hydraulic fracturing failed (fracture surfaced). Direct push injection in a single point and a 3 point cluster with 25 cm spaced injections at relevant depths (2.5-3.5, 6-7 and 8.5-9.5 m bgs) was successful in distributing tracer in natural and induced fractures at the same or shorter intervals at all relevant depths within about 1-1.25 m distance from the injection point. Individual tracer filled fractures were observed at > 2 m radial distance.

Stimulated reductive dechlorination (SRD) in some cases including bioaugmentation with specific degraders has proven to be very efficient for remediation of sandy aquifers, but knowledge regarding the effectiveness for remediation of low permeability sediments such as clayey till is lacking. SRD is currently being tested/applied at a number of Danish sites with chlorinated ethene contamination in clayey till, including the sites Rugårdsvej and Sortebrovej in/near Odense in Denmark. Model calculations reveal that the treatment period for significant reduction of the cis-dichloroethylene (c-DCE) level in the matrix at Rugårdsvej for a distance of 0.3 m to 1 m between fractures or sand stringers will likely be on the order of 10 to 100 years if biodegradation is restricted to the fractures or sand stringers.

At Rugårdsvej a slow release donor (Newman Zone) and specific degraders (KB-1 culture, by pulsed addition to donor solution being injected) was applied in two test-areas by injection in hydraulic induced sand-filled fractures and natural sand stringers, respectively. The biodegradation of c-DCE and vinylchloride (VC) was monitored in groundwater from the induced fractures and sand stringers. Complete degradation of c-DCE and VC to ethene in the induced fractures was obtained within 5 months, ethene was further reduced to ethane, the only degradation product remaining in the fracture after 18 months. In the sand stringer test site reduction of sulphate in the sand stringers was observed as the only clear evidence of an effect of the biostimulation. After 5 and 18 months the effect of the stimulated degradation in the clayey till matrix was documented through detailed profiles of sediment samples from intact cores of the clayey till. Profiles from the induced fracture test-area revealed a reaction zone in the clay in contact with the induced fracture, where biodegradation of c-DCE and VC in the low permeability matrix was documented. The degradation of the chlorinated ethenes occurred in the presence of relatively high concentrations of sulphate in the matrix.

At Sortebrovej full scale SRD of trichloroethylene (TCE) contamination in clayey till was initiated by injection of a slow release donor (EOS) and specific degraders (KB-1 culture, by pulsed addition to donor solution being injected) in natural sand stringers from a large number of wells each screened at 3 depth intervals covering the depth of the contaminated clayey till and with a spacing of 5 m. Monitoring of several wells (not used for injections) showed good progress of SRD with strongly reduced conditions, a high number of specific degraders and dechlorination of TCE via c-DCE and VC to ethene within about a year. However, detailed profiles of cores collected (12 cores in total) after 1.5 years revealed, that degradation was limited to sand stringer and other permeable features at variable spacing (cm to m) and a narrow reaction zone (few cm) surrounding them in the matrix. In/around most sand stringers/features only degradation of TCE to c-DCE was observed and specific degraders were not detected. Only in and around 2 sand stringers in one of the cores, complete dechlorination of TCE to ethene was observed and the presence of specific degraders documented. The study emphasise the importance of documentation of degradation in the matrix by soil/core-sampling, as monitoring of the aqueous phase may lead to overinterpretation of SRD effect.

The development of a reaction zone in the matrix greatly increases the potential for SRD in the clayey till. The impact on the timeframe for remediation of the clayey till will depend on the progress of the reaction zones over time. Enhancement techniques providing a sufficiently close spacing between reaction zones are essential for obtaining sufficient remediation of the clayey till matrix. Direct push injection was found promising with respect to enhancement of SRD in clayey till. The potential importance of the thickness of induced fractures or natural features, via which donor and specific degraders are applied, is yet unresolved.