INTRINSIC AND ENHANCED BIOREMEDIATION IN AQUIFERS CONTAMINATED WITH CHLORINATED AND AROMATIC HYDROCARBONS IN THE NETHERLANDS

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SUMMARY

The feasibility of intrinsic and enhanced bioremediation approaches for 16 contaminated sites in the Netherlands are discussed. At at least five out of 10 chlorinated solvent sites, natural attenuation can be used as one of the tools to prevent further dispersion of the plume. At two sites stimulation of the intrinsic dechlorination processes in a bioactived zone is required, and pilot field tests are currently under way. For three sulphate-reducing/methanogenic aquifers contaminated with aromatic compounds, microcosm studies demonstrated that natural anaerobic degradation of the risk-determining benzene does not to occur spontaneously. Benzene biodegradation could be initiated by feeding small amounts of oxygen (in all samples) or nitrate (only in one sample), and these techniques are currently tested in field-pilot studies. Redox-microbiological characterisation of two locations contaminated with hexachlorocyclohexanes indicated significant intrinsic biodegradation of HCH's in methanogenic/sulphate-reducing parts of the contaminant plumes, and of the HCH-degradation products chlorophenol and benzene under sulphate/iron reducing conditons. Biostimulated zone's remain required to complete the degradation of HCH and the degradation product monochlorobenzene. Risk-based guideline's for assessing the feasibility of natural attenuation approaches for various contaminant situations are currently under development, making use of already existing protocols (USA) and taking European and National specific conditions, knowledge and regulatory constraints into account.

Contaminated Soil '98, Thomas Telford, London, 1998, 109–112

Introduction

The ideas concerning best approaches for remedation of polluted soil and groundwater are rapidly changing. A few years ago many engineers and regulators considered contaminated aquifers as biologically inactive systems which could only be remediated by very drastic and expensive methods like soil vapour extraction or large scale pump-and-treat. Nowadays, one has become aware that the natural microbial population in soil and groundwater react actively when confronted with pollution, and thus offer new and better ways to protect groundwater resources at acceptable costs.

Often, natural biodegradation processes can convert large amounts of contaminants without any external stimulus. Sometimes, the natural processes can completely degrade or immobilise pollutants during their down-gradient transport. When sufficient time and space is available, an intrinsic remediation approach can in principal be completely protective for man and surrounding ecosystems. At other sites, the natural biodegradation processes need to be enhanced in order to obtain a sufficient risk reduction and groundwater protection. Such a stimulation can be performed in a bioactivated zone or bioscreen. In such a zone, the autochtonous bacteria are supplied with the appropriate electron donors, acceptors or nutrients. At some sites, the microbial population is not yet fully adapted to the most optimal biodegradation process. In such a case bioaugmentation of the bioscreen may be a solution. Intrinsic remediation, when necessary combined with bioscreens, is becoming the new approach to deal with large and complex contaminated sites.

Natural and enhanced attenuation as a risk based solution for contaminated soil and aquifers is currently being investigated at various sites in the Netherlands. A number of 16 cases investigated by TNO will be discussed below.

Chlorinated solvents

At the Rademarkt site (Groningen, The Netherlands) contaminated with perchloroethylene (PCE) and trichlorethylene (TCE), mixed redox conditions control the intrinsic biodegradation processes. In a methanogenic/sulfate reducing zone a complete reductive dechlorination reaction via vinylchloride to ethene and ethane occurs. However, the natural transformation rates of vinylchloride observed in the field (and in the laboratory) are too slow to prevent migration of this hazardous compound to areas to be protected. The low amounts of DOC (< 10 mg/l) indicate a lack of sufficient amounts of electon donor naturally present. Laboratory experiments identified a special mixture of electron-donors to be the most suitable cost-effective product to enhance the in situ reductive dechlorination. An in situ pilot test with an anaerobic activated zone designed for complete reductive dechlorination is planned this fall. In another flow direction in the field, the redox condition changes from methanogenic/sulfate reducing to oxic conditions. In the reduced part, PCE and TCE are transformed to cis-1,2-dichloroethylene (DCE) and vinylchloride (VC), which both disappear after entering the oxic zone. Probably, these compounds are removed by intrinsic oxidation. Hence, in this flow direction a complete sequential degradation of the chlorinated compounds is achieved.

At another PCE/TCE site in Maassluis, the contaminated aquifer contains high concentrations of organic carbon (up to 700 mg/l DOC). Here, complete reductive dechlorination is observed. Most likely, this is a result of the high amounts of intrinsic electron-donor and a well adapted autochtonous microbial population.

The results of the characterisation of redox conditions and intrinsic biodegradation at 10 chlorinated solvent sites are as follows. For at least five of these sites, intrinsic bioremediation is an important part in an approach to effectively control the risks. At at least two sites, in-situ biodegradation needs to be stimulated in in-situ activated zones to protect down stream area's.

Intrinsic chlorinated solvent remediation is further investigated by testing electron donors and measuring in-situ hydrogen pressures in the field and in laboratory microcoms. Thus a further insight into mechanisms involved will be obtained.

At present new technologies are being developed by TNO for application in bioscreens and remediating chlorinated solvent hot-spots and contaminated soils and subsurface layers with low permeability. Combination of electroreclamation techniques and biological methods appear to bring new solutions in the near future.

Aromatic and oil-related hydrocarbons

Aromatic compounds are often the risk controlling compounds at oil and gas production sites, and at sites of coating and nutrition industries. At three sites in the north part of the Netherlands, deep anaerobic aquifers contaminated with Benzene, Toluene, Ethylbenzene or Xylenes (BTEX) have been investigated. Under the existing sulfate-reducing conditions, the intrinsic biodegradation of toluene and ethylbenzene could be demonstrated in the field and in microcosm studies. Benzene biodegradation could not be evidenced with the field data. Laboratory microcosm studies with five different sediment samples were performed. For each sample a series of comparable triplicate microcosms were incubated; in total about 300 microcosms were used. The results demonstrate thus far (200 days of incubation) that spontaneous intrinsic anaerobic benzene biodegradation does not occur in these sediment and groundwater samples.

Microcosm method development studies indicated that special care is required to prevent artefacts: "apparent intrinsic anaerobic" benzene biodegradation could be demonstrated to originate from low (often not measurable) amounts of oxygen introduced into the systems, when inappropriate materials and techniques were used.

Microcoms were also used to investigate possibilities to stimulate biodegradation of benzene and TEX compounds. Especially, addition of nitrate and low amounts of oxygen to the anaerobic samples was studied. In one out of the five series of sediment-microcosms, nitrate-reducing benzene biodegradation appeared to occur after lag-times greater than 80 days. In addition, anaerobic benzene degradation could be initiated in all samples by spiking low amounts of oxygen. Further investigations are underway to elucidate mechanisms and quantify remediation process parameters. The results are used for designing pilot demonstration tests to be performed this spring/summer.

At an oil refinery site in the Rotterdam Harbour area, an aerobic reactive-trench bioscreen is tested for managing a plume of the dissolved fraction of a mineral oil contamination (80% of the compounds belong to the C6 - C12 fraction). Bench scale experiments are currently performed to establish i) optimal grain-size and packing density for the porous media used in the trench, ii) optimal oxygen supply rates to sufficiently initiate aliphatic hydrocarbon biodegradation and to minimise clogging with iron(III)oxides.

Chlorinated pesticides

Hexachlorocyclohexane (HCH) isomers are important pollutants introduced by the production of lindane (gamma HCH). At two sites, anaerobic intrinsic biodegradation of HCH and corresponding degradation-intermediates (benzene and monochlorobenzene) is currently being investigated. At one site, natural biodegradation processes appear to completely degrade all compounds except the monochlorobenzene; possibly some biostimulation may be required at the downstream end of the plume. At the other site, interception of the HCH/Chlorobenzene/benzene plume is required. A bioactivated zone as an alternative to conventional large scale pump-and-treat is currently being investigated. At present, laboratory process research aimed at developing a combination of anaerobic-microaerophilic in-situ stimulation in such a bioactivated zone is being performed. The results indicate good prospects of such an approach, and further development and pilot scale demonstration is being planned.

Natural attenuation guidelines

Dutch (NOBIS) and Nicole-supported guidelines for assessing the feasibility of natural attenuation approaches for various contaminant situations are currently under development, making use of already existing protocols (USA) and taking Dutch/European specific conditions, knowledge and regulatory constraints into account. These guidelines are to become tiered decision tools. In the first steps a minimum of data is required for a first feasibility screaning, thus saving costs. At higher tiers, data-requirements are designed to address natural degradation under various redox-situations (including sequential redox conditions) that often occur in European sedimentary regions.

Conclusion

Intrinisic and enhanced in-situ bioremediation approaches become more and more accepted as appropriate cost-effective solutions for aquifers and soils contaminated with organic chemicals. These techniques still need to be further developed and demonstrated and could also be expanded to control and remediate mixtures of organic and inorganic (heavy metal) pollutants.

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