

Source Remediation vs. Plume Management: Critical Factors Affecting Cost-efficiency

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Abstract

This summary paper reviews just some of the extensive scientific literature from the past 20 years on the various aspects of contaminant source remediation and plume management. Some of the major findings of the numerous research projects are presented. The first section outlines the controlling mass transfer mechanisms which need to be considered for any in situ source remediation technology. It then outlines the innovative technologies, such as the application of surfactants, use of co-solvents, thermal technologies or bio-enhancement, which try to overcome some of the physico-chemical difficulties for in situ source remediation.

In the second part of the paper, various plume management approaches are discussed as an alternative to in situ source remediation. Due to the longevity of the contaminant emission, the long-term cost-effectiveness of the various plume management approaches becomes the key issue. Therefore, a combination of physico-chemical and economical modelling is needed to provide a quantitative basis for decision-making. An example of a combined modelling approach for a reactive barrier is given. Finally, the various factors in the determination of whether source remediation or plume management is likely to be a preferable, i.e. more cost-effective, option, are summarised.

Key words: source remediation, plume management, cost efficiency

INTRODUCTION

One of the biggest challenges for many environmental scientists during the past 20 years has been the development of efficient tools for locating contaminants in soil and groundwater, and the design of cost-effective measures for the remediation of the subsurface environment. Various strategies have been developed to either excavate, isolate or *in situ* flush (to decontaminate) the *contaminant source zones*, i.e. the locations where the contaminants have penetrated into the subsurface. However, so far all of the source decontamination technologies available have proved to be very costly when applied to either larger sites or greater depths. Lacking any alternative, simple pump-and-treat concepts have been commonly employed: i.e. the *contaminant plume*

rather than the source, is treated through downstream pumping wells. However, pump-and-treat itself might be very cost-inefficient if realistic operation periods of sometimes tens of years are considered. These findings are supported by site statistics data, such as those published by the US NRC (1994).

Consequently, there has been increasing interest in concepts which extend beyond simple pump-and-treat but still focus on the control of the groundwater plume rather than the elimination of the source. *Natural attenuation* can be seen as one such approach for the control of problematic compounds which are sufficiently biodegradable. *Reactive barriers* might be used if the use of natural attenuation alone would lead to an unacceptable length of the contaminant plume.

It should be noted that in the context of this paper, source remediation is understood in terms of an *in situ* remediation. As this paper focuses on cost-efficiency issues at large industrial sites, remediation options such as *in situ* containment or excavation are not considered

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here because these techniques are in general too expensive.

NATURAL SOURCE ZONE DEVELOPMENT

As a consequence of the long history of intensive industrial activity in Europe, numerous areas now face severe soil and groundwater pollution problems. These may, on the national scale, lead to remediation bills in the multi-billion dollar range. From a drinking water quality perspective, point-source groundwater contaminants frequently cause problems, in particular a particular group of manufactured organic compounds. These are the chlorinated solvents used in metal manufacturing and the electronic industry, petroleum products stored at refineries, gas stations and in underground storage tanks, as well as tar oil and distillates used at former gas production plants.

Many of these compounds are highly persistent in the subsurface environment, i.e. they are still present in high concentrations many decades after the contamination occurred, and they cannot be removed from the subsurface within a reasonable time period by pump-and-treat. This persistence may be caused by slow *dissolution kinetics* of the compounds from non-aqueous phase liquids (NAPLs), slow *diffusion* of the contaminants from low permeability zones (which have accumulated the pollutants over decades) or resistant *desorption* of the contaminants by the aquifer material. All these processes reduce the bioavailability of the organic compounds, leading to long remediation times, even for easily biodegradable compounds.

NAPL dissolution

The dissolution of the organic compounds present in the organic phase is the major contaminant release mechanism. In the saturated zone, recharge water transports the dissolved contaminants into groundwater. The organic phase is in direct contact with the mobile groundwater so that a much larger volume of water may be affected.

Residual NAPLs are commonly very complex organic mixtures of compounds with a broad range of physico-chemical properties. The maximum (equilibrium) concentration $c_{sat,i}$ [M L⁻³] for dissolution out of a mixture is given by Raoult's law (Prausnitz *et al.* 1986; Atkins 1986; Loyek 1998):

$$c_{sat,i} = c_i \cdot g_i \cdot S_{w,i} \quad (1)$$

where c_i denotes the mole fraction of the component i in the mixture [-], g_i the activity coefficient of the component i in the organic mixture [-] and $S_{w,i}$ the water solubility of the pure compound i [M L⁻³]. According

to Raoult's law, the more soluble components of a mixture are depleted first. The saturation concentrations are, however, variable over time, due to changing molar fractions as dissolution advances.

In porous media NAPLs can be distributed as small droplets ('blobs') or ganglia entrapped in the pore space at high residual saturation; they may also form lateral coherent bodies ('pools') on low permeability layers/lenses. The overall mass flux due to dissolution depends on various physical and chemical parameters of the mobile phase (water) and the immobile phase (NAPL); on grain diameter; on the porosity of the porous medium; and on the specific interfacial area available for mass transfer.

For larger NAPL-water interfacial areas (NAPL distributed as blobs or ganglia) the mass transfer zone is relatively short, and so equilibrium concentrations are reached after short distances. In a homogeneous porous media the length of the mass transfer zone (saturation distance) is on the order of centimetres for 'blobs' (Fitzer *et al.* 1997; Geller and Hunt 1993; Miller *et al.* 1990; Powers *et al.* 1994). However, under natural field conditions, the saturation distance will be longer, in the range of metres, owing to aquifer heterogeneity and the heterogeneous distribution of the organic phase within the subsurface, leading to 'dissolution fingering' (Mayer and Miller 1996; Imhoff and Miller 1996; Imhoff *et al.* 1996). Consequently, close to the source, the contaminant concentration will generally be close to the compound specific saturation, according to Raoult's law. This equilibrium dissolution assumption has been confirmed in dissolution experiments conducted in the laboratory at different scales – columns (Eberhardt 1995; Weiß 1998), tanks (Loyek 1998; Eberhardt *et al.* 1998), as well as in the field (Frind *et al.* 1998; Zamfirescu and Grathwohl 1998; Zamfirescu 2000). The mass flux for equilibrium dissolution is directly proportional to the groundwater flow velocity, i.e. faster velocities lead to a faster dissolution of the residual NAPL.

The duration for complete equilibrium dissolution of NAPL 'blobs' depends on the aqueous solubility of individual compounds, as well as on the ratio between the saturation distance and the length of the domain containing residual NAPL. 'Pools' dissolve even slower than 'blobs', because there is a much smaller interfacial area between the NAPL and the circulating groundwater. Typical timescales for complete dissolution range from weeks to hundreds of years for 'blobs', and from months to thousands of years for 'pools'. It should be noted that for most realistic spill scenarios at industrial sites, it is quite unlikely that the NAPLs will be present in the form of 'blobs' only, i.e. more or less evenly distributed within the source zone.

For illustration, Figure 1 shows the computer simulation of a DNAPL (dense non-aqueous phase liquids) spill (TCE) on top of a heterogeneous sand and gravel aquifer (Figure 1a) (Whittaker *et al.* 1998). Based on a

realistic hydraulic conductivity distribution from outcrop data (Klingbeil *et al.* 1999), the spill is simulated (shown in Figure 1b) using a multi-phase numerical simulator (Unger *et al.* 1995). As seen in Figure 1c, the

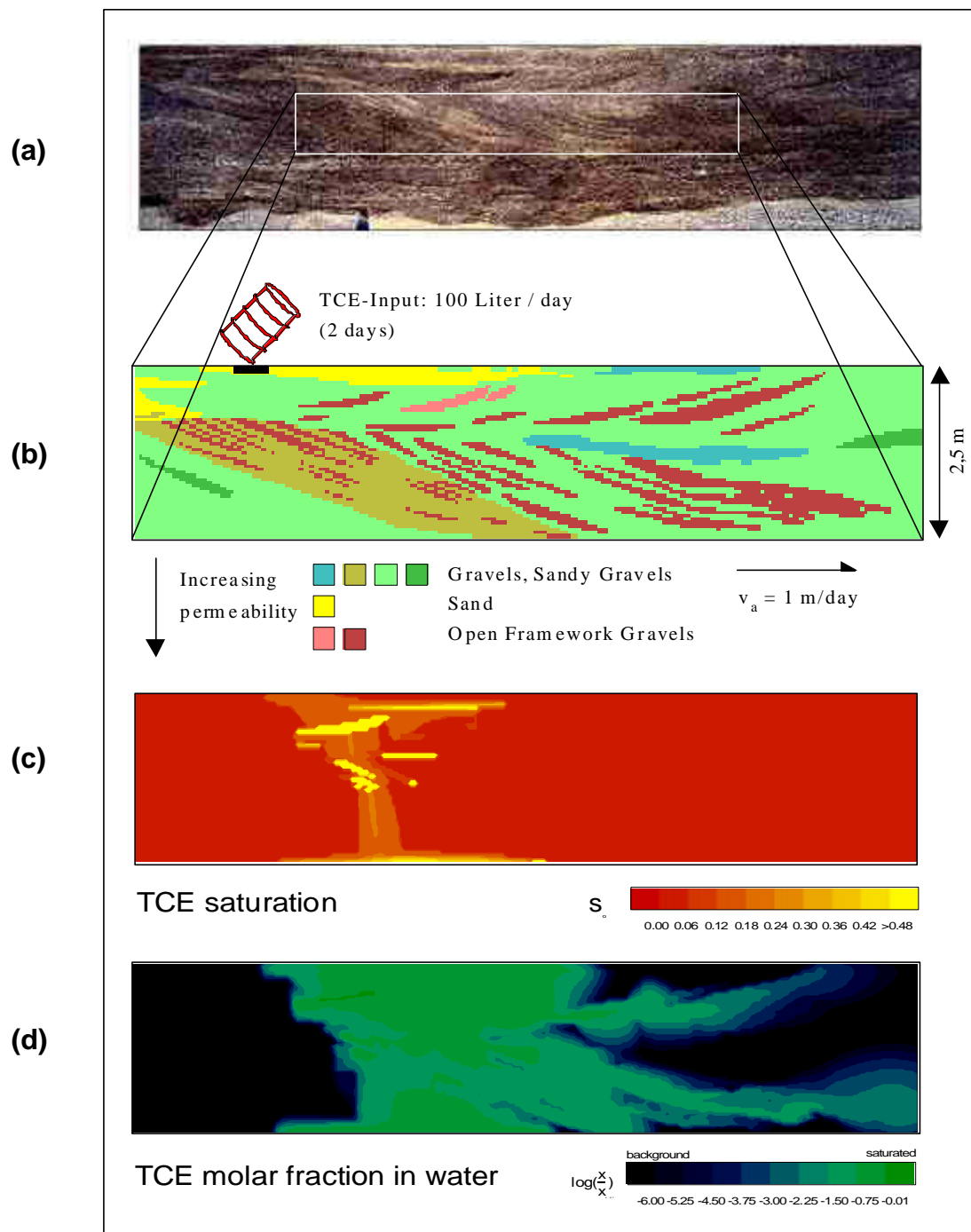


Figure 1. Computer simulation of a DNAPL spill in a heterogeneous aquifer (Whittaker *et al.* 1998) with (a) outcrop photograph, (b) hydraulic conductivity distribution, (c) DNAPL distribution and (d) dissolved contaminants in the groundwater flowing from left to right. A TCE spill of 200 litres leads to a very complex distribution of both the contaminants in the source zone (DNAPL –distribution) as well as in the contaminant plume.

distribution of the DNAPL in the subsurface is strongly controlled by the subsurface heterogeneity. 'Pools' develop at the interfaces between high and low conductivity layers (e.g. the base of gravel lenses). 'Blobs' develop between the 'pools', where the DNAPL cascades down towards the bottom of the aquifer. The size and spatial distribution of 'pools' and 'blobs', and consequently the estimated remediation times, strongly depend on the local scale heterogeneity. However, because of the relatively short mass transfer zone, the contaminant concentrations in the groundwater immediately downstream of the source zone will tend to be close to saturation throughout most of the entire cross-section (Figure 1d). This indicates that the dissolution of the DNAPLs in the source zone is simply limited by the equilibrium dissolution of the DNAPL in the water. Even though the concentrations downstream of the source zone are high compared to drinking water standards, the contaminant emission (mass flux) is still small compared to the total mass stored in the source zone. The consequence is a very long remediation time. Furthermore, further downstream of the source, the contaminant concentrations might be significantly lower than saturation concentrations, depending upon where groundwater samples are taken. This is because the contaminant plume will follow highly conductive flow paths, and uncontaminated or less contaminated groundwater will then mix into the sample cross-section. The result will be apparent dilution.

Desorption

Desorption is the second major process affecting the removal of contaminants present in the subsurface. Especially with older contamination, a significant proportion of the dissolved contaminant mass is not stored in the mobile water but diffused into the porous particles forming the aquifer sediments.

In the case of desorption, the equilibrium concentration of a compound is determined by the distribution coefficient, K_d , between solids and water. Unlike dissolution out of the residual phase, desorption is in most cases a non-equilibrium process (Grathwohl 1998). Since desorption is a transient process, the length of the mass transfer zone becomes a function of time. The desorptive flux decreases first with the square root of time and later exponentially with time. Unlike the mass flux for dissolution out of the residual phase, it is independent of the groundwater flow velocity.

Desorption from coarse grains and removal of highly sorptive compounds such as the polycyclic aromatic hydrocarbons (PAH) requires a timescale of decades to hundreds of years (Kleineidam *et al.* 1999; Rügner *et al.* 1999). The same holds true for desorption out of the matrix of fractured rocks, or large confining layers, even for compounds with lower sorption capac-

ities than PAHs such as chlorinated solvents. Long time periods have to be expected for the decontamination, even when sorption equilibrium has not been reached during the period of contamination (Grathwohl 1998).

ENHANCED (*IN SITU*) SOURCE ZONE REMEDIATION

The *in situ* remediation of NAPLs will generally take a considerable period of time. Therefore, various technologies are being developed which try to overcome some of the rate limiting processes described above. These are outlined below.

Surfactants and co-solvents

One possible way of overcoming the low solubilities of most organic compounds during *in situ* remediation is to use surfactants or co-solvents. In both cases, the idea is to increase the mass removal by moving the NAPL either as a dissolved or separate phase in a short time towards a remediation well. Surfactants may work by either solubilising (micellar-enhanced aqueous concentrations) or mobilising (ultra-low interfacial tensions) the NAPL phase. If a co-solvent is present (usually an alcohol) the solubility enhancement is determined by the fraction of the co-solvent in the aqueous solution and also by the co-solvent strength which depends on both the co-solvent and the solute (Pinal *et al.* 1990). However, a significant solubility enhancement is expected only if the fraction of co-solvent is greater than 10 % (Grathwohl 1998).

A site-specific surfactant or co-solvent mixture is usually prepared above ground and applied to the subsurface through injection wells placed within the source area. A second set of wells is used to abstract the groundwater with the contaminants and to control the flow. In general, a controlled flow zone needs to be established in order to ensure that no contaminants will move beyond the remediation area. In addition, economic considerations may require the recovery and re-injection of the flushing fluid. The maximum size of the controlled flow zone depends on various hydraulic factors, including the natural groundwater flow velocity at the site and the geology of the site.

Some initial field experiments under controlled conditions (usually within sheet-piles) have recently been conducted with LNAPLs (light non-aqueous phase liquids) and DNAPLs. The results described (e.g. in Sabatini *et al.* 2000) are generally quite promising and are expected eventually to lead to a more frequent application where source access is restricted. However, it seems likely that it will be difficult to obtain regulatory permission for the application of surfactants or co-solvents to DNAPL problems. There is a considera-

ble possibility that parts of the mobilised DNAPL will be re-distributed into deeper aquifer zones instead of reaching the abstraction wells.

The cost of enhanced NAPL-recovery with surfactants or co-solvents is difficult to assess so far, because it is very dependent upon the specific conditions at any one site and the remediation target. The complexity of the hydraulic control, the requirements for above ground water treatment and the complex design will dictate that these technologies will tend to incur large costs. However, where other alternatives are not available and the size of the domain is not too large, there is likely to be a market for surfactant and co-solvent flushing.

Thermal technologies

Thermal technologies include direct heating of the subsurface by thermal conduction or by the injection of heated fluids, mostly hot air, hot water or steam. Steam injection is believed to be the most effective owing to the high-energy injection rates and the ability of steam to displace pore fluids from saturated soils (Udell 1998).

So far, the applications of thermal technologies have been quite successful in laboratory experiments, but only limited experience exists at field scale. Practical applications are seen mostly for LNAPL spills. The application of thermal technologies for spills of volatile DNAPLs, even though quite effective, is likely to be restricted owing to fear of re-mobilising the contaminants.

In a heterogeneous environment the remediation efficiency required to reach drinking water standards is seldom achievable by the use of thermal technologies. In this case steam injection, similar to all fluid-based flushing technologies, relies on the efficient sweep of the contaminant source zone. It shows only limited success in removing NAPLs located in lower conductivity or difficult to access parts of the aquifer system. However, thermal technologies do represent a valid remediation option for removing a significant part of a LNAPL contamination within a limited area.

Bio-enhancement

Bioremediation is a process in which indigenous or inoculated microorganisms (i.e. fungi, bacteria and other microbes) degrade (metabolise) organic contaminants found in soil and/or groundwater (US EPA 1997). In general, naturally occurring microorganisms are able to readily degrade released soluble contaminants under aerobic conditions. The microbial degradation leads to depletion of oxygen within the contaminated zone. Therefore aerobic processes are limited to the fringes of a contaminant plume where oxygen is supplied by diffusion from the surrounding non-contami-

nated area. In the centre of the plume anaerobic processes dominate, i.e. degradation that relies upon electron acceptors other than oxygen. Degradation activities under anaerobic conditions are in general slow (ASTM, E-1943-98).

In practice, enhancement of biodegradation processes is achieved mainly by increasing the concentration of electron acceptors and nutrients in soil and leachate. Oxygen is the main electron acceptor for aerobic degradation, while nitrate and sulphate serve as an alternative electron acceptor under anoxic conditions. To ensure aerobic conditions in a biodegradation process, air or hydrogen peroxide may be injected into the subsurface (FRTR 1995). Similarly, nutrients used for microbial cell growth may be added (WDNR 1999).

Probably the most significant limitation for bio-enhancement is the limited ability to mix the contaminants present in the groundwater plume with the injected electron acceptors and nutrients in the subsurface. If the contaminant moves without being retarded, then any injected fluid will just displace but barely mix with the contaminant.

High concentrations of heavy metals, highly chlorinated organics, long chain hydrocarbons, or inorganic salts are likely to be toxic to microorganisms, leading to reduced turnover rates.

Soil vapour extraction and air sparging

Soil vapour extraction (SVE), also known as 'soil venting' is widely used as an *in situ* remedial technology in the unsaturated zone (US EPA OUST 1995). This technology has proved to be most effective in reducing concentrations of volatile organic compounds such as gasoline and chlorinated solvents. However, concentration reductions greater than about 90% are usually difficult to achieve (Di Giulio *et al.* 1998). Diesel fuel, heating oils, and kerosene, which are less volatile than gasoline, are not as easily removed by SVE. The technique may be best applied to sites where the unsaturated zone is at least a few metres deep. The limitations for this technology are in general subsurface environments with low gas conductivity.

Air sparging (AS), also known as '*in situ* air stripping' is an *in situ* remedial technology that reduces concentrations of volatile constituents that are adsorbed to soils and dissolved in groundwater (US EPA OUST 1995). It involves the injection of contaminant-free air into the saturated zone, enabling a phase transfer of hydrocarbons from a dissolved state to a vapour phase. This air is then vented through the unsaturated zone. When used appropriately, air sparging has been found to be effective in reducing concentrations of volatile organic compounds (e.g. chlorinated solvents; BTEX) but tends to be generally less success-

ful for compounds which have vapour pressures < 5 mm Hg (Gierke *et al.* 1998).

Air sparging may stimulate biodegradation processes but the portion of contaminant mass reduction by biodegradation is usually small (e.g. Johnston *et al.* 1998). In general, air sparging faces the problem of strong fingering (narrow, singular paths where air passes through the heterogeneous subsurface environment) resulting in a limited sweep efficiency with respect to the total contaminated soil volume.

PLUME MANAGEMENT APPROACHES

Owing to the limitations of the *in situ* source remediation options discussed above, but also due to its relatively low investment cost, pump-and-treat has become the standard remediation technology for many larger sites. However, this plume control technique does not shorten significantly the source lifetime. Numerous practical experiences during the past 20 years have shown that in most cases the remediation time estimates have been far too optimistic and anticipated operation costs far too low. The US NRC report (1994) states that almost all of the large-scale groundwater remediation projects have actually been failures with respect to attaining the remediation targets. However, insufficient or inappropriate monitoring has in many cases prevented this from becoming obvious during the lifetime of the projects.

Consequently, concepts and technologies which focus on long-term groundwater plume control rather than 'aggressive' source remediation have received increasing attention during recent years. For compounds which are sufficiently biodegradable, *natural attenuation* or *enhanced natural attenuation* can be an

option (Wiedemeier *et al.* 1994; Rifai *et al.* 1995; US EPA OSWER 1999), whereas engineered *reactive barriers* (Gillham and Hannesin 1992; Teutsch *et al.* 1997a) represent an alternative to *pump-and-treat* in situations where the compounds are more persistent.

Figure 2 shows four plume management approaches. These are, from left to right: pump-and-treat, reactive barriers (here a funnel-and-gate system), natural attenuation, and enhanced natural attenuation.

All four approaches have in common that the contaminant source remains (partly) in the ground and continues to emit for potentially a very long time. Any decision for or against a plume management approach has to consider the ecological, technical and economic aspects throughout the lifetime of the contaminant source. A thorough analysis is required for all three aspects. This paper focuses on the economic aspects only, leaving out site-specific ecological, logistical or technical issues.

COST-MODELS FOR PLUME MANAGEMENT

Pump-and-treat

Plume management costs are relatively simple to calculate for pump-and-treat systems, if the hydraulic conditions are well understood, and the position of the pumping wells is known. The only uncertain parameter remaining is the anticipated operation time, i.e. the lifetime of the source. Good practice is to calculate or estimate from site data the mass flux leaving the source area (emission) under natural flow conditions and so estimate the minimum amount of contaminant mass stored in the source zone. This approach is likely to be less error prone than trying to extrapolate concentration time-series into the future. For many of the larger sites,

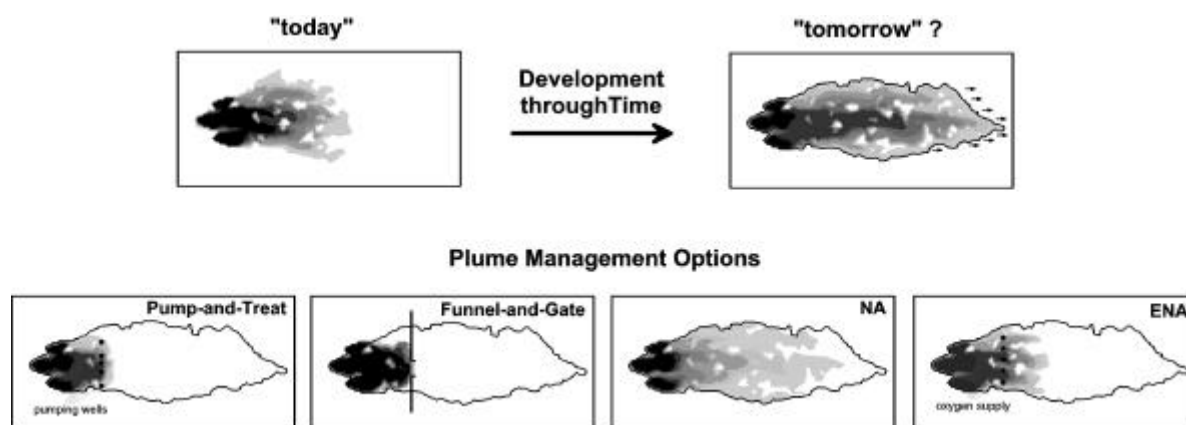


Figure 2. Plume management approaches

operation times will probably be in the range of at least tens of years, reaching hundreds or even thousands of years for very hydrophobic compounds like some of the PAHs. However, because future costs are discounted, i.e. converted to net present value, the impact of any payment in the future will decrease with increasing operation time (Ross *et al.* 1995; US OMB 1992).

For more complex conditions, pump-and-treat system design requires optimisation. Typically, this is done in practice by trial and error. This is not an optimal approach. Given the large number of factors encountered for a complex site – the number and position of the pump-and-treat wells, the heterogeneous distribution of the contaminants in the subsurface, the various above ground treatment options etc. – it becomes obvious that an optimal solution can hardly be identified by simple trial and error. Numerous more rigorous optimisation schemes have been proposed for pump-and-treat operations (e.g. Ahlfeld and Heidari 1994; McKinney and Lin 1996; Aly and Peralta 1999). Bayer (2000) went a step further and added an above ground water treatment simulator to the cost model which already included a numerical flow and transport module. He showed, for example, that passive flow barriers placed upstream of the contaminant source zone could be a cost-efficient solution to reduce the volume of contaminated groundwater passing through the source, and consequently reduce the required pump rate at the downstream plume control well.

Reactive barriers

Reactive barriers are an alternative to pump-and-treat where the operation time for pump-and-treat is expected to be long. The idea is that the higher initial investment costs for constructing a reactive barrier will be compensated in the long run by the lower operation costs of the passive technology. However, this may not always be true and depends on a number of factors such as: the type of contamination and availability of adequate 'passive' technologies, the contaminant concentrations, the width and depth of the contamination, the hydraulic conditions at the site.

A comprehensive example of how the total cost of plume management can be evaluated for a reactive barrier system has been demonstrated by Teutsch *et al.* (1997b) in their presentation of the application of a coupled hydraulic-geochemical-economical simulation model for the design of *in situ* reactive barrier systems. More specifically, the model is used for cost-optimisation and sensitivity analysis of a conceptual funnel-and-gate system with an *in situ* sorption reactor. The groundwater flow and advective transport are simulated under steady-state conditions using a numerical model. This model is coupled to an analytical solution describing the sorption kinetics of hydro-

phobic organic compounds within the reactor (gate). The third part of the model system is an economic model which calculates (a) the investment costs for the funnel-and-gate construction and (b) the operation cost based on the number of reactor refills, which depends on the breakthrough time for a given contaminant and the anticipated total operation time. For practical applications, a simplified approximation of the cost-function is derived and discussed.

Figure 3 shows the example cost functions for a given parameter set describing: the anticipated operation time (50y), the aquifer conditions (aquifer thickness, hydraulic conductivity, hydraulic gradient), the type of construction technique (trench cutter), the type of *in situ* reactor treatment (sorption parameters) and the prescribed contaminant degradation factor (C_0/C_m , which is the ratio between inflow and outflow concentrations at the *in situ* reactor).

The left-hand figure displays the total investment costs for different contaminant plume widths (dotted lines) in relation to the funnel (y-axis) and the gate length (x-axis). For example, for a plume width of 50 m, the cost-optimal funnel-and-gate system would have a funnel length of 130 m and a gate length of 7.5 m. The investment cost would be about DM 550,000 (280,000 Euro). In the right-hand figure, total costs are displayed, which include investment as well as operating costs for a 50-year time period. The total cost functions have a 'jagged' form because over the total operation time of 50 years, a discrete number of *in situ* reactor refills (activated carbon) is needed. The total cost for the above-mentioned funnel-and-gate system is estimated to be about DM 625,000 (320,000 Euro), including the replacement of the activated carbon during the 50 years of operation.

(Enhanced) natural attenuation

Interest in enhanced natural attenuation has increased during the past five years as more reliable field data has become available on the specific mobility and persistence of the most frequently observed (organic) groundwater contaminants. For example, a chlorinated hydrocarbon source may produce an extended contaminant plume which will move almost without being retarded. Conversely, a petroleum hydrocarbon source will lead to a plume with a limited length due to the relative ease of biodegradation of petroleum products. Natural attenuation (NA) of hydrocarbons is now considered a valid remediation strategy in the USA as well as in some European countries (Rice *et al.* 1995; Nyer and Duffin 1997; Wiedemeier *et al.* 1999; US NRC 2000). In effect, NA is a reactive zone which represents that part of the aquifer downstream of the source zone, which is needed for the attenuation of the contaminants down to background levels.

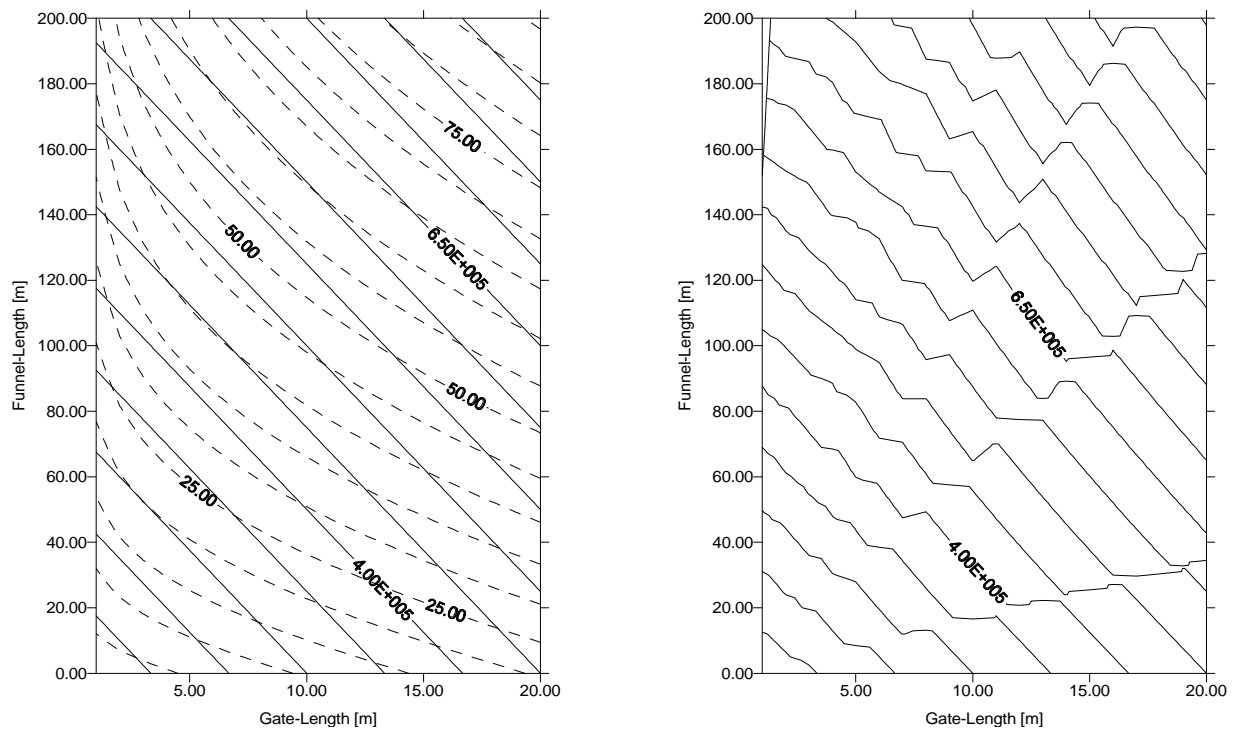


Figure 3. Funnel-and-gate investment and total cost (construction technique: hydraulic trench cutter, $t_{op} = 50$ y, $C_0/C_m = 10^3$, $T_{Ga} = 0.5$ m, $K_{Aq} = 10^{-3}$ m/s, $grad\ h = 10^{-3}$, $K_d = 10^4$ cm³/s, $D_a = 10^{-13}$ m²/s, $m = 10$ m) (Teutsch et al. 1997b)

Enhanced natural attenuation (similar to the reactive barriers technology) requires some kind of a reactant (e.g. electron acceptor or donor) to be supplied to the subsurface. This supports the biodegradation of the dissolved contaminants on their downstream path. The resulting reactive zones are intended to be shorter than is the case for natural attenuation.

Cost models for natural attenuation or enhanced natural attenuation need to include not only the extra subsurface characterisation, modelling and monitoring costs which are needed to quantify (enhanced) natural attenuation at a site, but also the possibility of costs for the lower land value in the reactive zone downstream of the source. However, these values may sometimes be hard to quantify because the change in value may depend on the future land and water use (US EPA OSWER 1995; Di Mauro *et al.* 2000). In addition, environmental costs resulting from soil and groundwater contamination in the reactive zone may add to the bill for natural attenuation. (US EPA OPPTS 1995; Bardos *et al.* 2000).

At present, there is not yet a generally accepted cost model for natural attenuation projects. Present research focuses on the development of tools which allow the comparison between different plume management and source remediation options. Appropriate decision support tools (e.g. NOBIS 1998) will need to be extended

to handle the technical, economic, ecological and social factors of plume management.

SUMMARY AND CONCLUSIONS

Source remediation

As can be seen from the above case studies, *in situ* remediation of contaminant source zones is a formidable task which in many cases will not be achievable within one generation. Furthermore, a significant reduction of the contaminant emission (concentrations and mass flux) occurs only if more than 90% of the source mass is removed (Grathwohl 1998; Eberhardt *et al.* 1998). Therefore, the effect of source zone mass removal will certainly affect the lifetime of a source, but not necessarily the contaminant concentrations observed downstream of the source during that lifetime.

Source removal through excavation or containment might be a valid and cost-effective option where the position of the source(s) is known and its extent is limited. However, at large industrial sites, the ratio between the volume sampled from the subsurface through cores or groundwater and the volume of the potentially contaminated area is so small that the result-

ing levels of uncertainty are sometimes too high for a multi-million dollar decision (Liedl and Teutsch 1998).

All technologies which try to overcome the mass-transfer limitations through use of surfactants, co-solvents or thermal methods may work well if the contaminants are accessible to the flushing fluid, and if the flow control works. In these cases, the extent of the treatment zone will be relatively limited, and particularly limited for DNAPLs. An unresolved question relates to the degree of remediation which can be achieved under practical conditions. Total remediation costs will be very sensitive to this issue.

Bio-enhancement relies on the availability of the contaminants and the availability of electron acceptors and nutrients in the mobile water. The availability of contaminants is mass-transfer limited, i.e. controlled by the physico-chemical parameters at the specific site. The ability to deliver electron acceptors and nutrients for biodegradation is limited by the ability to mix two fluids in a porous media under laminar flow conditions, i.e. controlled by the hydro-mechanic parameters at the specific site.

Plume management

Plume management approaches like pump-and-treat, reactive barriers, natural attenuation and enhanced natural attenuation are valid options for site remediation where the position of the source(s) is not known exactly or the source is inaccessible.

Pump-and-treat is a proven technology in an operational sense, with relatively low investment costs but significant operation costs. Many former applications of this technology might have been less attractive in terms of total costs, if a more realistic, i.e. longer, operation period had been used in the calculation.

Reactive barriers are a new approach which requires higher initial investment, but has lower operation costs. Several reactor materials are available, the most common being (a) granular iron for the degradation of chlorinated aliphatic compounds like TCE or PCE, or (b) activated carbon for sorption of hydrophobic compounds like PAHs. Numerous other reactor types are presently under development. With respect to long-term total costs, reactive barriers should be less costly than most pump-and-treat technologies. However, existing field experience is less than ten years so far; the longevity of the reactor performance will be a key factor of the total costs.

Enhanced natural attenuation is another new plume management approach which tries to combine the advantages of reactive barriers with natural attenuation. If applied to easily biodegradable compounds, enhanced natural attenuation can be very cost-effective. Because it is not a passive technology, operation

costs are critical. Cost models have to include the environmental costs for the reactive zone. Enhanced natural attenuation is a low to medium investment technology with low to medium operation costs.

Natural attenuation became popular when it was accepted as a valid remediation option for fuel spills. However, there is increasing field evidence that some degradation products, as well as lesser known by-products, may be more mobile and less degradable than the original compounds. The best known case is that of MTBE, an additive to common gasoline, which in 1996 caused the closure of half of the water supply wells of the city of Santa Monica, California (Keller *et al.* 1998; Schirmer and Barker 1998). Other, more toxic by-products have recently been found downstream of former gasworks sites such that the environmental risk will have to be reconsidered in future cases (Zamfirescu 2000).

Natural attenuation is of course very attractive from a cost perspective since the investment costs and operation costs are believed to be low. However, it should be noted that natural attenuation would always require a more detailed site (plume) investigation as well as continuous monitoring throughout the lifetime of the source. Furthermore, the reactive zone, which will be larger than for the enhanced natural attenuation option, will need to be evaluated with respect to its stationarity in time (growing or shrinking plumes). Similar to the enhanced natural attenuation case, no generally accepted cost models yet exist.

One should finally bear in mind that especially at large industrial sites, where variable types of sources of different extent and levels of accessibility exist, cost-optimal and eco-efficient groundwater protection measures are likely to be a combination of various source and plume management concepts.

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