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Groundwater contamination: Pump-and-treat remediation

Second of a five-part series

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Almost all remediation of groundwater at contaminated sites is based on groundwater extraction by wells or drains, usually accompanied by treatment of the extracted water prior to disposal. This often causes an initial decrease in contaminant concentrations in the extracted water, followed by a leveling of concentration, and sometimes a gradual decline that is generally expected to continue over decades. In such cases, the goal of reaching stringent health-based cleanup standards is very remote and the ultimate cost of cleanup very high (1).

The purpose of this paper is to explore reasons for the observed difficulty of groundwater cleanup and note some implications that become clear during this process. Our discussion is limited to organic contaminants because they are the most common health-threaten-

ing chemicals detected in groundwater and because the greatest difficulties in groundwater remediation have been encountered at organic contamination sites.

Organic contaminant plumes

Prior to the passage of the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) and revised Resource Conservation and Recovery Act (RCRA) legislation in 1980 and 1984, respectively, detailed monitoring of groundwater at industrial and waste disposal sites was rare, particularly for trace organic contaminants. Now, as the end of the decade approaches, knowledge of the nature of organic contamination of groundwater has advanced considerably because of the expenditure of more than a billion dollars on site investigations and cleanup activities. Hundreds of plumes of organic contaminants have now been delineated by networks of monitoring wells.

Several examples of organic plumes in the United States and Canada are given in Table 1. Table 1 pertains to plumes in sand and gravel aquifers; although there are many plumes in fractured rock, summary data such as we

present in this table are not readily available because the outer boundaries of such plumes and the fracture porosity are very difficult to determine. Plumes such as those in Table 1, which extend 0.5–10 km from the source and which generally have formed over decades, are common in North America and Europe. Each of the sites listed in the table represents a major plume in the sense that millions of dollars have been spent on plume characterization, feasibility studies for alternative remedial actions, and attempts at remediation. Cleanup programs are now planned or underway for some of the plumes in the table at ultimate costs for each plume estimated at tens of millions of dollars or more. The CERCLA and RCRA programs address hundreds of plumes such as those in the table. There are probably thousands of other such plumes in North America that are not addressed by these programs.

Of the various organic contaminants found in groundwater, the widely used industrial solvents and aromatic hydro-

carbons from petroleum products are most common (2–4). Much if not most of groundwater contamination of this type is caused by leakage, spillage, or disposal of organic liquids immiscible with water (nonaqueous-phase liquids [NAPLs]) into the ground. Dissolution of the NAPL and subsequent transport of the dissolved constituents by groundwater is thought to generate many plumes (5), although there is mounting evidence that migration within the vapor phase of the unsaturated zone with subsequent transfer of vapor-phase contamination to soil water and groundwater may also cause formation of groundwater plumes (6, 7).

The plumes in Table 1 are listed in decreasing order of the mass estimated to be present in the dissolved form (expressed as equivalent volume of NAPL in the right column). Note that there are documented examples of plumes that encompass much larger volumes of groundwater and greater masses of dissolved contaminants than listed in this short table. Also, not all types of con-

TABLE 1
Relatively well-documented organic contaminant plumes in sand–gravel aquifers^a

Site location and plume map	Presumed sources	Predominant contaminants ^b	Plume volume (liters) ^c	Contaminant mass dissolved in plume (as equivalent NAPL volume in liters or 55-gal drums) ^c
0 5 km Flow →				
Ocean City, NJ	chemical plant	TCE TCA PER	5,700,000,000	15,000 (72 drums)
Mountain View, CA	electronics plants	TCE TCA	6,000,000,000	9800 (47 drums)
Cape Cod, MA	sewage infiltration beds	TCE PER Detergents	40,000,000,000	1500 (7 drums) ^d
Traverse City, MI	aviation fuel storage	Toluene Xylene Benzene	400,000,000	1000 (5 drums)
Gloucester, ON Canada	special waste landfill	1, 4 Dioxane Freon 113 DEE, THF	102,000,000	190 (0.9 drum)
San Jose, CA	electronics plant	TCA Freon 113 1, 1 DCE	5,000,000,000	130 (0.6 drum)
Denver, CO	trainyard, airport	TCE TCA DBCP	4,500,000,000	80 (0.4 drum)

^a Readers aware of other well-documented cases for which reliable estimates of contaminant mass distribution and organic carbon content (f_{oc}) of the aquifer solids are available are encouraged to contact the authors, who plan to expand this compendium.

^b TCE = trichloroethylene; TCA = 1, 1, 1 trichloroethane; PER = per-, i.e., tetrachloroethylene; 1, 1DCE = 1, 1 dichloroethylene; CHCl₃ = chloroform; DEE = diethyl ether; THF = tetrahydrofuran; DBCP = dibromo-chloropropane.

^c Approximate estimates derived from plume length, groundwater velocity, contaminant concentration distributions, etc., provided for illustrative purposes only. Estimated contaminant mass accounts only for the dissolved phase (i.e., does not account for contaminant sorbed to the aquifer media throughout the plume or for NAPL contaminant, if any, from the sources). Most of basic data is from unpublished sources; data on three plumes are published (13, 27, 28, 29).

^d This mass estimate is for the halogenated contaminants only (i.e., detergents are excluded).

taminants encountered in groundwater plumes are represented, though most of those listed are quite commonly detected. Nevertheless, the table is instructive. It is clear, for example, that the contaminant load in an aquifer cannot be judged from the magnitude of the contaminated area; two of the apparently larger plumes have the least contaminant mass dissolved in them. This, of course, reflects the differences in the concentrations within the various plumes as well as their total volume. All but two of the plumes in the table contain more than a billion gallons of contaminated water. Yet the two smallest plumes contain relatively large amounts of contaminants, in part the result of their relatively high solubility compared with the halogenated compounds in the other plumes.

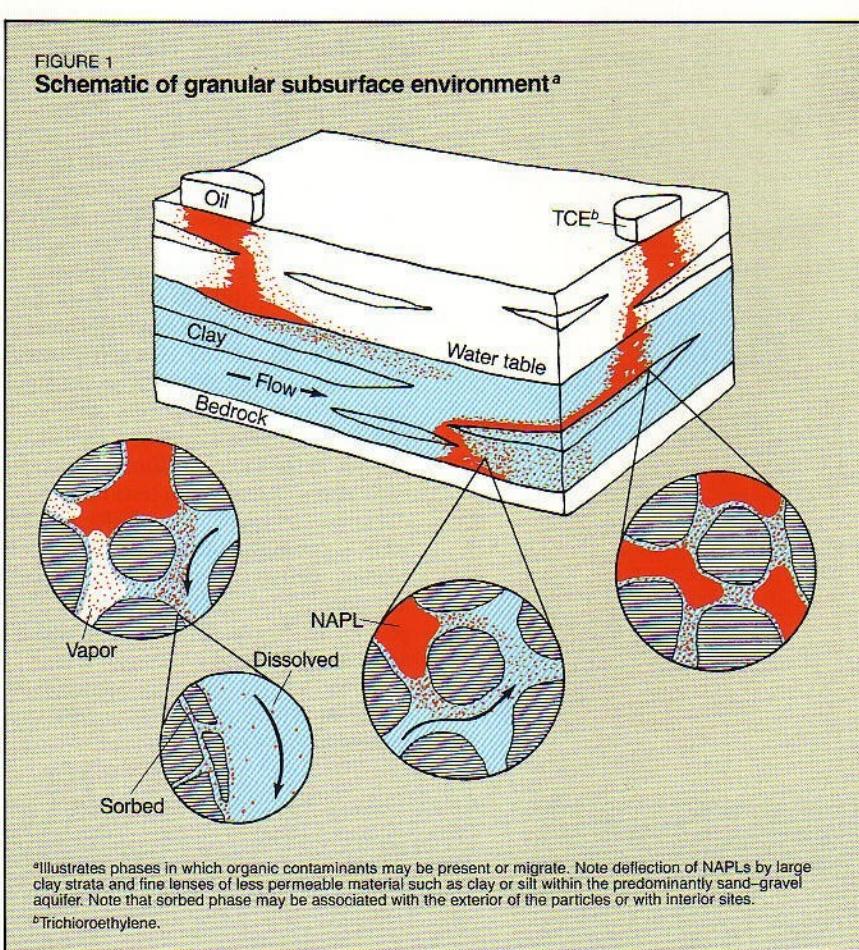
The table also illustrates that plumes often contain less mass in the dissolved form than would be present in a few drums of NAPL. This may be a minuscule fraction of the total NAPL mass that entered the subsurface at many sites where the total amount of organic chemicals used or disposed of would have been extremely large, often measured in hundreds of tanker truck loads.

These examples ignore the contaminant mass that would be sorbed to the aquifer media contacted by the plume, which may be on the same order as or significantly greater than the dissolved mass (8). Nevertheless, it is clear that the primary challenge in groundwater cleanup is to remove the organics masses that serve, in effect, as subsurface sources and cause the plumes to grow and persist, rather than simply to remove the mass of dissolved contaminants that defines the plume. This issue, so fundamental to the proper diagnosis and efficient solution of the problem, is explored further below.

Cleanup of sand and gravel aquifers

Ideally, a remediation project for contaminated sand and gravel aquifers would be designed based on a solid understanding of the mass and types of pollutants released, the current location of all the mass remaining in the subsurface, and the processes controlling the removal of the mass from the subsurface (or its destruction *in situ*). Unfortunately, none of these requirements is generally met in practical investigations, leading to a considerable amount of guesswork in developing a cleanup plan.

Figure 1 illustrates a few of the many known complexities of granular subsurface environments as well as the phases in which organic contaminants may be present and migrate through the porous media that comprise the unsaturated (above the water table) and saturated



(below the water table) zones. Upon release to the subsurface, the total mass of each pollutant will be distributed among the various phases by the movement of vapors and liquids and diffusion of the pollutants within them. NAPL may be present in pools at near saturation, having displaced most of the pore water, or in "residuals" at "residual saturation," on the order of 1–10% of the pore volume (6, 9) remaining from contact with a migrating NAPL slug. The mass of organic pollutant present in a given volume of NAPL-contaminated soil may be many orders of magnitude greater than the mass present if the soil volume were contaminated only with vapor, dissolved and sorbed phases. We thus refer to the pools and residuals as subsurface contaminant sources.

With regard to cleanup efforts, it is clearly advisable to first remove the NAPL sources from the subsurface if at all possible, because they may contain most or nearly all of the total mass of fugitive contaminants. For NAPLs such as benzene and other petroleum products, which tend to float on groundwater, there have been successes in pumping a significant fraction of the NAPL to the surface (10). Yet for others more dense than water (e.g., chlorinated solvents, creosote, and PCB-

rich oils), very little success has been achieved in even locating the subsurface NAPL sources, let alone removing them (11).

For NAPLs that cannot be removed directly or that remain in residual saturation, the pollutant mass they contain will generally have to be removed in a much more dilute form, such as by vaporization into the soil gas or by dissolution in groundwater. These removal methods require the extraction of considerable volumes of gas or water. Practical experience indicates that the cleanup process is lengthy and expensive, especially for contaminants in the saturated zone.

The problem of groundwater cleanup is exacerbated by the desire to return aquifers to drinking-water quality, which for many important organic contaminants requires concentrations less than 100 parts per billion (ppb) and in some cases less than 5 ppb. However, the concentrations in groundwater withdrawn by wells are controlled in part by transfer of contaminant mass to the flowing water from other phases acting as contaminant reservoirs: contaminant sorbed by the aquifer solids, contaminant present in immobile pockets of contaminated groundwater in less permeable but porous strata or lenses, vapor spreading from residual or

pooled NAPL in the vadose zone, or dissolution of residual or pooled NAPL in the saturated zone. As discussed below, such transfers of contaminant mass can cause the extracted groundwater to fail to meet drinking-water standards for prolonged periods of time, a problem compounded by the slow rate at which these transfers often occur.

Effects of contaminant desorption from solids

Dissolved organic contaminants generally move more slowly through granular aquifers than the groundwater itself because of sorptive interactions with the aquifer solids (12–16). Although there have been only a few field studies that have yielded quantitative understanding of the relative mobility of organic contaminants, a review of them indicates—as expected from laboratory studies and basic geologic knowledge—that field retardation varies among contaminants for a given site and among sites for a given contaminant (8).

The greater the retardation, the more time will be required to remove the

contaminants for a given pumping rate. Furthermore, the removal of dissolved and sorbed contaminants by pumping requires the extraction of more water than is contaminated at the onset of remediation. Figure 2a illustrates this in an idealized case for a contaminant with a retardation factor of two (i.e., in which the sorption/desorption interactions cause the contaminant to move at a constant fraction, 1/2, of the groundwater velocity). The retardation factors observed in sand–gravel aquifers for contaminants such as those listed in Table 1 vary from 1 to 33 (8); in other media or for other contaminants, the retardation may be even greater. Thus, unless injection wells are used to supply the clean water, which is rarely the case, the “pump-and-treat” approach may utilize a considerable volume of uncontaminated groundwater surrounding the site to flush the contaminants from the polluted area.

However, kinetic limitations to desorption can occur during groundwater extraction programs, as has been observed in field studies (17, 18) and implied by laboratory investigations (19–22). The practical effect of these kinetic

limitations is to slow the removal of the contaminants from the aquifer, thereby increasing both the time required to achieve cleanup and the total volume of water that must be extracted to flush the contaminated zone. Furthermore, if pumping is ceased before all of the contaminant is removed, the contaminant concentrations in the groundwater will rise as desorption continues (1). Whiffen and Bahr (17) observed such results.

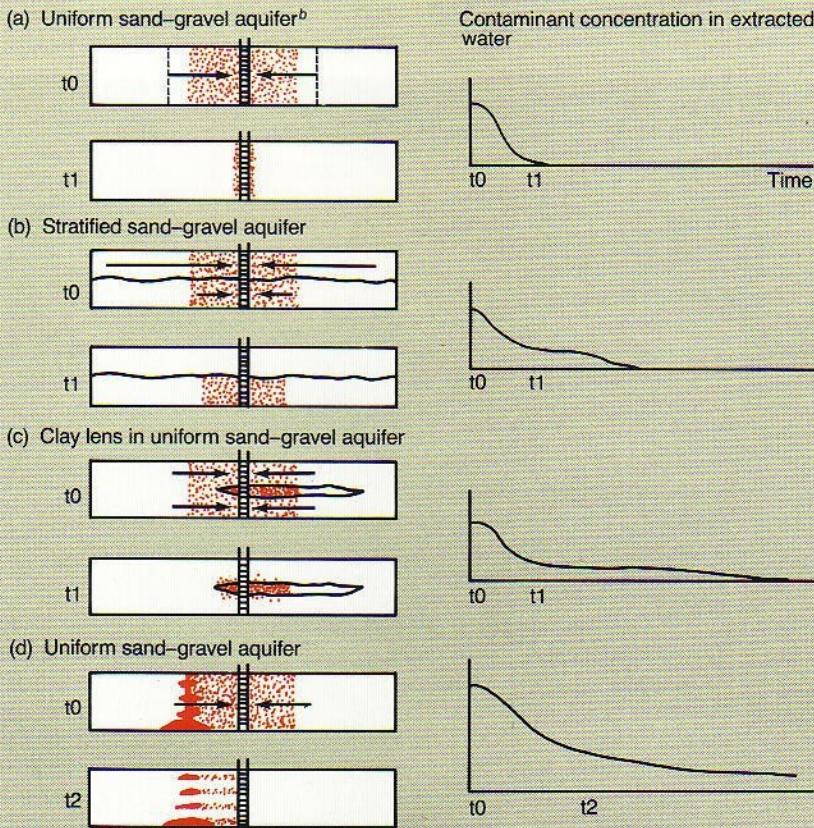
Effects of geologic complexity

Although the processes that affect organic contaminant transport are essentially the same in various sites, the optimal design of a remediation program is very site specific, mainly because of geologic complexity. Sand and gravel aquifers typically have silty or clayey strata above, within, or beneath them (Figure 1). These strata normally are less permeable than the aquifer by a factor of 1000 to 10 million. Groundwater flow and contaminant migration are distorted because of these strata. The positions of NAPL sources in the aquifer are commonly complex and unpredictable because exact locations and volumes of spills or leakages are unknown, and, as illustrated in Figure 1, the positions of subsurface NAPL sources are determined by deflections and pooling caused by stratification. Current research suggests that even slight heterogeneities can influence NAPL penetration into porous media, particularly in the saturated zone (23).

As plumes spread through aquifers, the dissolved contaminants move quickly through more permeable zones while they slowly invade the less permeable ones by flow or diffusion (24). Over the years and decades, this invasion can cause the plume to occupy large volumes of low permeability material. To obtain clean water from wells, it is generally necessary for the lower permeability parts of the aquifer system to be cleaned as well as the high permeability zones.

Figure 2b illustrates an idealized case in which the aquifer is composed of two relatively distinct and horizontally continuous sand–gravel strata, the upper stratum having a somewhat higher hydraulic conductivity than the lower. The contaminant is assumed present in dissolved and sorbed phases, uniformly spread in the volume indicated. If groundwater is extracted from a fully penetrating well or a well with most of its screen in the more permeable zone, as is common practice, the bulk of the water will be moving through the upper stratum. Thus, even in sand–gravel aquifers, some strata may be flushed of contaminants long before others. Figure 2b assumes that the retardation factor of the contaminant is two in both

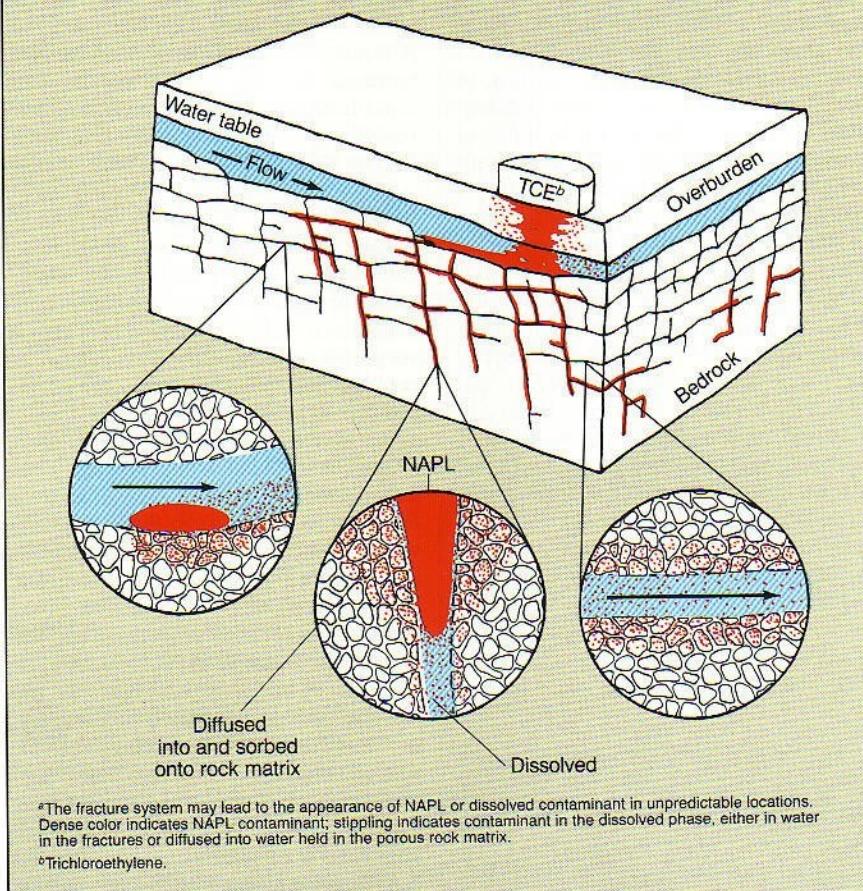
FIGURE 2
Hypothetical examples of contaminant removal from aquifers^a



^aDense color indicates NAPL contaminant, stippling indicates contaminant in dissolved and sorbed phases (assumed uniformly distributed initially), and arrows indicate relative velocity of groundwater flow. The groundwater is assumed to be extracted from the well at the same rate in the four cases.

^bDotted lines enclose total volume of water that would be pumped to remove contaminant with retardation factor of 2.

FIGURE 3
Schematic of subsurface environment composed of fractured rock under the overburden^a



^aThe fracture system may lead to the appearance of NAPL or dissolved contaminant in unpredictable locations. Dense color indicates NAPL contaminant; stippling indicates contaminant in the dissolved phase, either in water in the fractures or diffused into water held in the porous rock matrix.

^bTrichloroethylene.

strata. If the retardation is greater in the lower stratum and desorption is kinetically limited, then the flushing will be even more inefficient (25).

Figure 2c illustrates another situation: a very low permeability clayey stratum in the middle of an aquifer that has been contaminated for decades. Dissolved contaminants have permeated the clayey stratum during this period primarily by molecular diffusion. Although the permeability of a clayey stratum is low, the porosity is usually as large as or larger than that of the adjacent aquifer, thereby facilitating diffusion into the clay. Furthermore, the capacity of clayey strata to sorb contaminants may be much greater per unit volume than that of the aquifer. When the aquifer is flushed by clean water, the only significant process for release of the contaminant from the clay will be a reversal of the diffusion direction. The relatively slow rate of release of contaminants from the clay by diffusion and the potentially appreciable contaminant mass contained in dissolved and sorbed form in the clay causes a long-term bleed of contaminants into the aquifer during remediation (1). In many aquifers there are numerous thin beds of silt and clay that

compound this problem of delayed diffusive release. Such processes may maintain contaminant concentrations in the extracted water above typically low cleanup criteria for very long periods of time, greatly increasing the duration and cost of cleanup. In most investigations, however, the details of site geology are not well enough understood to allow prediction of the effect of delayed diffusive release.

Effects of fugitive NAPL

As described above, organic NAPLs are often suspected to be present in the subsurface, but reliable estimates of the volume of NAPL spilled or disposed of exist for very few groundwater contamination sites. Furthermore, even after exceptionally detailed site investigations are conducted, it is normally not possible to predict reliably where these NAPL pools are. Not knowing the size and location of NAPL pools and zones of residual NAPL makes it impossible to predict how long a pump-and-treat program must operate in order to clean the aquifer. Figure 1 shows how clayey lenses in the aquifer can cause the NAPL to be deflected laterally so that most of the NAPL mass exists as isolated pools away from the spill origin. Perched pools of NAPL can jeopardize site investigations because it is very easy to unknowingly drill through the pool and the bed it sits on, causing the pool to drain down the hole into a



deeper part of the aquifer or into a different aquifer. When pumping causes lowering of the water table from a position above perched NAPL pools or NAPL residual zones to below them, remobilization of the pools and residuals may occur, allowing them to drain deeper into the aquifer.

Figure 2d illustrates the effect that even relatively small quantities of NAPL might have on a typical cleanup program based on groundwater extraction. In the general case the extraction well is located at some distance from the NAPL that has penetrated the saturated zone. The groundwater extraction might quickly remove the bulk of the dissolved and sorbed contaminant that had migrated from its NAPL source(s) and thereby achieve an initial decrease in concentration in the extracted water. However, the NAPL present in pools or residual saturation may be dissolved slowly by the groundwater flowing around or through them, maintaining contaminant concentrations at significant levels for a long time, even in relatively uniform aquifers. When zones of lower permeability are contaminated with NAPL, the cleanup time would be expected to increase considerably. Feenstra and Cherry (11) present a more complete review of these and other issues regarding NAPL behavior and cleanup of NAPLs denser than water.

Cleanup of fractured rock aquifers

Generally, rock aquifers contain a myriad of cracks (fractures) of various lengths, widths, and apertures. Most rock aquifers in North America are permeable primarily because of the effective porosity provided by these fractures rather than that of the rock matrix, which is relatively impervious. The effective fracture porosity of fractured-rock aquifers is generally in the range of 0.001–0.1%, which is much smaller than the porosities of typical granular aquifers (20–40%). For example, a rock mass with one fracture per linear meter with fracture apertures of 500 µm would be very permeable but at saturation would have a very small storage volume of mobile groundwater (only about one-half liter per cubic meter of rock).

When NAPL enters such aquifers, it flows mainly through the interconnected fractures and settles out in dead-end segments of the fracture system (Figure 3). Relatively small volumes of NAPL can move deep and far into the rock because the retention capacity offered by the dead-end fractures and the immobile filaments and globules in the larger fractures is so small—much less than the percentage given above for water in saturated fractured rock. Al-

though the rock matrix typically has a relatively small intergranular porosity, it is commonly large enough to allow dissolved contaminants from the fractures to enter the matrix by diffusion and be stored there by adsorption, as shown in Figure 3.

The prognosis for cleanup of fractured rock aquifers, particularly those containing NAPL contaminants, is worse than for sand and gravel aquifers. Even if the location of the spill is known exactly, the location of the NAPL is typically difficult or impossible to determine from site investigations. This is because NAPL pathways

the plume toward its source(s), but for the shrinkage to persist it is necessary for the pumping to continue. Even if contaminants remain in some portions of the aquifer, it is often possible by such hydraulic influence to eliminate real, potential, or perceived risks to public health. However, the long-term cost of such pumping with treatment of the extracted water is often high whether measured in dollars spent for system operation and maintenance or in gallons of previously uncontaminated groundwater used to flush out the contaminants. The mass of NAPL at or below the water table is not known with sufficient detail at most sites to make reliable predictions of the time necessary for cleanup by pump-and-treat programs. In general, it is appropriate to view such approaches as remediation in perpetuity.

At quite a few sites, the dissolved mass of organic contaminants in relatively large plumes is quite small, less than that present in one drum of the pure chemical. This suggests that seemingly innocuous and often unnoticed leakages or spills of a few gallons per day or less, so common at many industrial and military sites, may pose a major threat to groundwater. Considering the extremely high cost per equivalent NAPL gallon of removing the contaminant from aquifers, it is clear that the economic advantage of preventing such small leaks or spills is immense.

A number of new technologies are under development for groundwater remediation, as explained in later articles in this series, which may accelerate contaminant removal from the subsurface (e.g., injection of steam, surfactants) or destroy the contaminant in situ (e.g., bioreclamation). For many if not most sites, it is important to recognize that all of these technologies will be severely hampered by geological complexities and the difficulty of locating the subsurface contaminant sources. Laboratory studies and small-scale field prototype trials are likely to yield overoptimistic expectations for the application and efficiency of these technologies.

Site characterization programs often seem to the public and regulators to be inefficient and excessively lengthy. Shortening site characterization efforts, a currently popular demand, may be appropriate for pump-and-treat programs intended to prevent contaminant migration, but cannot be expected to be satisfactory for permanent aquifer cleanup. At sites where permanent cleanup is the goal, detailed and accurate site characterization is a prerequisite for a reasonable probability for success (1). As reviewed in other papers in this series and elsewhere (1, 8), there is

"...cleanup of groundwater contamination by organic chemicals typically proceeds slowly using the common pump-and-treat approach"

through the fracture system are exceptionally complex and distribute the NAPL into many small and scattered amounts (Figure 3). When attempts are made to clean such fractured rock aquifers by pumping water, major improvements in water quality are exceedingly slow because little or no water flushes through dead-end fracture segments or through the porous but impervious rock matrix, both of which are likely to retain the bulk of the contaminated mass. Such has been the experience at an organic liquids disposal site in Ville Mercier, Quebec, where the effectiveness of pump-and-treat remediation of the large plume has been severely hampered by the penetration of the NAPLs into the fractured bedrock (26).

Summary and implications

We have explored, via simplified examples and illustrations, many of the reasons cleanup of groundwater contamination by organic chemicals typically proceeds slowly using the common pump-and-treat approach. At many sites of significance, a relatively large mass of contaminants has been leaked, spilled, or disposed into the subsurface, and in comparison the rate of contaminant mass removal by pumping wells is exceedingly slow. In such cases the pump-and-treat option is best thought of as a management tool to prevent, by hydraulic manipulation of the aquifer, continuation of contaminant migration.

This option often effectively shrinks

much potential for improvement in site characterization and remediation, through both the development of new tools and the continued training of everyone involved: site owners, consulting engineers and hydrogeologists, regulators, and the public.

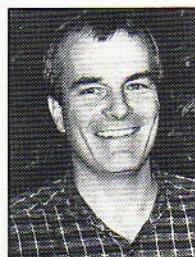
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