

# Underground Tank Technology Update

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Comments and suggestions are welcome and may be directed to John T. Quigley, Project Director, 432 N. Lake St., Madison, WI 53706. Tel 608/265-2083.

If you have a problem locating a reference cited in *UTTU*, please contact Pat Dutt: [pmduddt@msn.com](mailto:pmduddt@msn.com) or 607/272-3212

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*Darrell Petska*  
*Debbie Benell*  
*Susan Kummer/Artifax*

Project Director  
Geologist/Writer  
Copy Editor  
Program Assistant  
Graphics

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## Natural attenuation assessment by the NRC

Since 1993, more than a dozen national protocols on natural attenuation have been published by U.S. EPA and other agencies, including state agencies. Using these reports and other information, The National Research Council (NRC) recently published a consensus document on the current knowledge of natural attenuation with respect to groundwater contaminants. The NRC consists of volunteer experts from government environmental laboratories, environmental consulting firms, industry and environmental groups.

As to when natural attenuation is applicable, Bruce Rittmann, NRC chairman, states: "On the one hand, natural attenuation is a valid concept and may be a remedy for a range of groundwater contaminants . . . under the right circumstances. On the other hand, natural attenuation should be selected as a remedy only when the following conditions are met:

- mechanisms responsible for destroying or immobilizing the contaminant are scientifically recognized
- natural attenuation has been documented to be working now at the site, and is sustainable"

"Natural attenuation's 'protective technology' is knowledge. This means that natural attenuation is a valid remedy only when we have confidence in the cause-and-effect relationship between loss of contaminant and the mechanism responsible for the loss" (*Rittmann in MacDonald, 2000*).

Natural attenuation use has grown in response to high costs and technical limitations of cleanups and therefore may be an appropriate strategy for managing groundwater cleanup. According to one scientist at the Natural Resources Defense Council, however, natural attenuation is a "low-cost, ineffective approach that delays the inevitable need to clean up a site." Industry representatives, on the other hand, assert that increasing scientific evidence warrants the greater use of natural attenuation. The NRC takes the position that "Natural attenuation is an established remedy for only a few types of contaminants . . . and . . . should be accepted as a formal remedy for contamination only when the processes are documented to be working and are sustainable" (*MacDonald, 2000*).

### High, moderate and low ranking of sites

The NRC rated several chemical classes as to their likelihood that natural attenuation would succeed as a remedial strategy. The chemical classes included hydrocarbons, oxygenated hydrocarbons, halogenated aliphatics, halogenated aromatics, nitroaromatics, metals, nonmetals, oxyanions and radionuclides. The ratings provide "a qualitative indication of the probability that any given site will have the right conditions for natural attenuation of the particular contaminant." The rating terms are:

- high, that is "scientific knowledge and field evidence are sufficient to expect that natural attenuation will protect human health and the environment at more than 75 percent of contaminated sites"
- moderate, that is, "natural attenuation can be expected to be protective at about half of the sites"
- low, that is, "natural attenuation is expected to be protective at less than 25 percent of contaminated sites . . . although a low rating can also result from a poor level of scientific understanding"

The NRC also rated chemical classes in terms of levels of understanding:

- high indicates that a good scientific understanding of the process involved exists, and field evidence confirms attenuation processes can protect human health and the environment
- moderate indicates that studies confirm that the dominant attenuation process occurs, but the process is not well understood scientifically
- low indicates that scientific understanding is inadequate to judge if and when the dominant process will occur and whether it will be protective

BTEX, for instance, earns a high level of current understanding, and the likelihood of natural attenuation success given its current level of understanding is high. Current understanding of MTBE and PAHs (polycyclic aromatic hydrocarbons) is moderate, while likelihood of natural attenuation success given current level of understanding is low. (*Reviewer's note: Understanding would be low because we do not know if MTBE biodegrades naturally.*)

### Definition of natural attenuation

In 1999, the U.S. EPA released its formal policy on natural attenuation, which defined it as "reliance on natural attenuation processes . . . to achieve site-specific remediation objectives within a timeframe that is reasonable compared to that offered by other methods." EPA's relevant processes include biodegradation, dispersion, dilution, sorption, volatilization, radioactive decay, chemical or biological stabilization, transformation or destruction of contaminants. (See also *UTTU*, Vol. 13, No. 6, 1999, "EPA's monitored natural attenuation directive 9200.4-17P".) The NRC's definition of natural attenuation is narrower. For instance, environmental advocates at the NRC disagreed with including processes of dilution and dispersion, noting that "The multiprocess definition of natural attenuation fuels [the] controversy because it includes mechanisms such as dilution and dispersion that are, in the view of many environmental advocates, unacceptable ways to manage contamination."

The NRC report stresses that "it is important to keep in mind that natural attenuation processes are always site-specific. They depend on the hydrogeology and biogeochemistry of the site in question." Furthermore, a decrease of contaminant concentration in monitoring wells is not sufficient evidence for documenting natural attenuation. "Documenting that

contaminant concentration has become very low or undetectable in groundwater samples is an important piece of evidence that natural attenuation is working . . . [but] is not sufficient to show that natural attenuation is protecting human health and the environment . . . Contaminants can

- bypass monitoring wells
- decrease in concentration near the monitoring well but not in other locations
- transform to a harmful byproduct

For these reasons, environmental regulators and others should not rely on simple rules of thumb—such as maximum contaminant concentration data or trends in these data over a relatively short time—to evaluate natural attenuation's success (*MacDonald, 2000*).

To adequately evaluate natural attenuation at contaminated sites, NRC recommends the following:

- provide a conceptual model of the site
- look for footprints—changes in water chemistry left by attenuation reactions—of expected natural attenuation processes
- monitor the site

The type of data required will vary depending on contaminants present, groundwater chemistry, site geologic characteristics and cleanup goals.

### National protocols

Concerning the national protocols reviewed, the NRC maintains the following:

- important gaps in the body of protocols need to be filled, which may entail EPA leading “an effort to develop national-consensus guidelines for protocols on natural attenuation”
- protocols give insufficient direction with respect to
  - when and how to involve the public
  - when and how to implement contingency plans should natural attenuation fail
  - when contaminant sources need to be removed
  - how to conduct long-term monitoring, and what level of training users need to implement the protocol
- the “scoring systems,” which rate the likelihood that natural attenuation will occur based on site data, should be replaced with the three-part evaluation: conceptual model, footprints and monitoring

### Reference

MacDonald, J.A., “Evaluating Natural Attenuation for Groundwater Cleanup,” *Environmental Science and Technology*, August 1, 2000; <http://www.pubs.acs.org>.



## Benzene removal using air sparging

Adams and Reddy (*2000*) designed a laboratory study to test removal of dissolved-phase and free-phase benzene pools from groundwater. Their goals were twofold:

- to assess how air injection rate affects the mass transfer and transport of dissolved-phase and NAPL (non-aqueous-phase-liquid) pools
- to determine the effect of groundwater flow on the removal of dissolved- and NAPL-phase pools during air sparging

### Experiment description

Researchers created a two-dimensional aquifer simulation apparatus with a Plexiglas™ tank measuring 111 cm in length, 72 cm in height and 10 cm in width. “The tank’s interior consists of three compartments: a soil chamber measuring 91 cm in length is centered within the tank and flanked by two groundwater reservoirs measuring 10 cm in length. The water reservoirs on either side of the soil chamber are connected to constant-head clean water source reservoirs. The source reservoirs are adjustable in height, allowing for a specific head to be maintained in each reservoir. Additionally, the source reservoirs may be adjusted to create a hydraulic gradient across the tank, thereby inducing groundwater flow. The tank reservoirs are separated from the soil chamber by a geotextile-lined heavy-gauge perforated stainless steel screen, allowing water to freely enter or exit the soil chamber but preventing soil particles from entering the water reservoirs. On the face of the soil chamber, 20 sampling ports are arranged in four rows of five, each protruding into the soil chamber. The ports allow for pore water sampling from the soil profile throughout the course of a test” (*Adams and Reddy, 2000*).

Researchers performed five tests using a coarse, homogeneous soil (coarse sand) with a hydraulic conductivity of  $4.64 \times 10^{-2}$  cm/s. Groundwater flow conditions were both static and non-static. Hydraulic gradient was 0.011, and all tests had similar initial dissolved- and NAPL-phase benzene conditions.

### Conclusions

Adams and Reddy concluded the following from their tests:

- air sparging can readily remove NAPL- and dissolved-phase pools
- injected air promotes NAPL removal by enhancing the mass transfer/transport mechanisms of dispersion, dissolution and diffusion
- injected air traveled within a parabolic zone of influence (in channel mode) when subjected to both static and groundwater flow conditions, indicating that groundwater flow (for the groundwater velocities tested) did not affect the injected air zone of influence

- an increase in air injection rate led to faster contaminant removal; however, at higher air injection rates, a threshold of removal was reached above which further increases in injection rate are a waste of effort
- air injected into the soil profile reduced the hydraulic conductivity within the zone of influence
- reduced hydraulic conductivity produced lower groundwater flow rates, permitting interception of the migrating NAPL plume
- trapped NAPL can act as a source for further groundwater contamination, thus sudden increases in contaminant level should be expected
- trapped NAPL can be treated by increasing air flow
- if air is injected at a rate too high, soil fracturing, excessive advective-dispersive transport and possible off-site contaminant migration could occur
- low air flow produces low air saturation, hindering groundwater flow through the zone of influence and possibly resulting in substantial off-site migration
- groundwater flow has little influence on the size and shape of the zone of influence of a sparge well, yet air injection will slow groundwater flow through the zone of influence and ultimately reduce or eliminate advective-dispersive contaminant mass transport

### Reference

Adams, J.A. and K.R. Reddy, "Removal of Dissolved- and Free-Phase Benzene Pools from Ground Water Using In Situ Air Sparging," *Journal of Environmental Engineering*, August 2000, Vol. 126, No. 8; <http://www.pubs.asce.org>.



## Vertical profiles of hydraulic conductivity

Researchers recently developed a field procedure that uses a Geoprobe™ direct-push device to obtain vertical profiles of the subsurface. The Geoprobe™, which offers a quick way to take in-situ conductivity measurements, has a small diameter and only minimally disturbs the site. With this field procedure and mathematical formulas for computing hydraulic conductivity, several in-situ hydraulic conductivity measurements could be estimated for a site. Researchers were particularly interested in vertical hydraulic conductivity measurements. Vertical profiles would shed light on the convective groundwater path and contaminant movement along this path.

"Locating the contaminant plume pathway is a very important step in site characterization, requiring detailed profiles of conductivity in the aquifer due to the heterogeneity encountered in the subsurface environment" (Cho and others, 2000).

### Field site and field procedure

Researchers performed the test at a former fuel farm site in North Carolina that had stored aviation gasoline for more than 50 years. Results of the study can facilitate plume management. The water table here was 195 cm below ground surface while depth to the aquifer's base was 715 cm. Testing took place 15 m downgradient from the fuel tank.

Researchers constructed hydraulic conductivity devices using aircraft structural steel pipe attached to an open-slotted section of pipe at whose base was a driving tip. The length of the open-slotted section was 45 cm, and the pipe's outside diameter was 2.54 cm. Field workers drove pipes below the water table, using the Geoprobe™ direct-push drive. Several water well volumes were purged to remove the fines.

"Groundwater sampling was conducted and the tubing was raised so that the tip of the tubing was located 30 cm below the initial water table. Pumping continued until the flow reached the steady state and the flow rate was measured with a graduated cylinder and a stopwatch. In order to measure the recharge rate from surrounding soil under a given hydraulic drawdown, the entire volume of recharge water had to be pumped and collected into the graduate cylinder during the measurement period. In order to achieve this, the pumping was set to exceed the recharge rate. With this excessive pumping, water and air bubbles should be seen flowing together in the tubing. After the triplicate measurements of hydraulic conductivity, additional pipes were connected at the top and pushed down farther to the desired depth, then the whole procedure was repeated. Hydraulic conductivity measurement and water sampling continued until the screened section reached into the clay zone at the aquifer's base" (Cho and others, 2000).

### Hydraulic conductivity estimation

Cho and others (2000) then estimated hydraulic conductivity by deriving the recharge rate, at steady state, through an open-slotted pipe. Conductivity is linearly proportional to the recharge rate over the hydraulic drawdown applied in the pipes. The equations (12) used to compute hydraulic conductivity are given in the text. In addition, field workers performed a slug test in a nearby monitoring well to confirm results.

### Comparison of slug test and Geoprobe™ testing

The monitoring well and Geoprobe™ test were separated by one meter at the site. The monitoring well had a 10-cm ID and an open-slotted section from 150 cm to 600 cm below ground surface. The slug test performed in this well gave a hydraulic conductivity of 0.0108 cm/s.

Vertical profiles of hydraulic conductivity from the Geoprobe™ test indicated that the highest conductivities—0.022 cm/s—were at 600 cm below ground surface. This conductivity was two orders of magnitude higher than conductivity measured at a shallower depth. Average hydraulic conductivity between 195 cm and 600 cm was 0.0098 cm/s, which was close to the slug test value of 0.0108 cm/s.

## Conclusions

Cho and others (2000) developed a simple field procedure to obtain vertical profiles of hydraulic conductivity and also derived appropriate mathematical formulas to estimate it. "These vertical profiles of hydraulic conductivity provided a good insight concerning contaminant fate and transport. For instance, using the average hydraulic conductivity from slug tests in long-screened wells to estimate groundwater flow velocity may result in erroneous estimation of contaminant travel times and retention times for degradation."

## Reference

Cho, J.S., Wilson, J.T. and F.P. Beck, "Measuring Vertical Profiles of Hydraulic Conductivity with In Situ Direct-Push Methods," *Journal of Environmental Engineering*, Vol. 126, No. 8, 2000; <http://www.pubs.asce.org>.



## Phytoremediation basics

This article describes the biology of phytoremediation, a low-tech, low-cost, non-intrusive and aesthetically pleasing technique. Plants that can immobilize, biodegrade and evapotranspire contaminants can restrict the spreading of a contaminant plume. Phytoremediation is most applicable at sites that do not have heavy contamination (which can be toxic to vegetation) and where aquifers are shallow or moderately deep (Narayanan and others, 1999; Al-Yousfi and others, 2000).

### Plant species selection

Ideally the plants chosen will draw up large quantities of water from the subsurface and evapotranspire the water and volatilize some of the contaminant. Evapotranspiration rate will be a function of vegetation cover, temperature, humidity and sunlight. By drawing up large quantities of water, the plant acts as a pump-and-treat system, thereby creating a cone of influence in the subsurface. In addition, transpiring vegetation will help prevent infiltrating water from reaching the plume and expanding it. If infiltration exceeds evapotranspiration, however, a cone of influence will not be formed. Ideally, plant species chosen for phytoremediation will have the following characteristics:

- the ability to draw up large quantities of water
- rapid growth over the entire season and persistence for several seasons
- easy establishment
- a depth and distribution of root zone amenable to the problem
- the ability to withstand dry and wet conditions
- the proper degradative enzymes in plant or root zone
- the ability to bioaccumulate contaminants (Narayanan and others, 1999; Al-Yousfi and others, 2000)

Species commonly used include mulberry trees, poplars and hybrid poplars. Trees in arid and semiarid regions have been shown to lower aquifer levels by as much as five feet during the growing season. A willow tree was shown to "consume" more than 5,000 gallons of water in one summer day. Hybrid poplars, which can grow more than 3 m/yr, can act like a 100-ft straw, drawing up contamination from soil and groundwater. "Tree pumping rate is directly correlated with evapotranspiration, which varies with total leaf surface area, whether for an individual tree canopy or an entire plot. . . . A dense arrangement will have less leaf-surface area per tree, but the combined leaf-surface area of a dense plantation will be greater than the combined surface area of a thin and sparsely distributed planting" (Al-Yousfi and others, 2000).

### Site conditions

Excess rainfall and surface water ideally will be directed off-site. The surface should be prepared to prevent ponding and encourage runoff, which may be accomplished with the placement of a tile system.

### Fate of organics

To understand the fate of organics in vegetation, researchers investigated plant root zones with respect to adsorption and biodegradation, then plant uptake and evapotranspiration.

In the plant root zone, organics may

- adsorb onto soil particles and plant roots
- biodegrade in the soil

**Adsorption.** According to Narayanan and others (1999), "vegetation creates a favorable natural ecosystem in the rhizosphere (root zone of the plants) for indigenous soil microbiota. These native soil microorganisms exist in symbiotic relationships with plant roots in the soil. The sphere of influence of a single plant root is usually less than 1 cm, but a root system explores the upper meter of soil fairly extensively" and its occupied area usually exceeds that of the leaf cover. Root depth depends on soil moisture, although roots normally extend only as far as necessary to find water. Alfalfa roots have been found at depths greater than 10 m.

"Roots of vegetation act as additional sorption sites for pollutants. Well-established plants have an extensive, sometimes dense root system, and roots may grow radially to a distance of more the 50 cm from the center root. Plant roots and the associated rhizosphere may effectively trap components of the water phase because net water movement is usually into the plants" (Narayanan and others, 1999).

A contaminant's ability to sorb onto soil particles and plant roots is a function of contaminant hydrophobicity, which is determined by  $\log K_{oc}$  and  $\log K_{ow}$  values, the octanol-carbon and octanol-water partition coefficient, respectively. Sorption sites are quickly saturated, however, and when sorption equilibrium conditions are approached, there is no further reduction in dissolved concentration due to sorption.

**Biodegradation.** The indigenous microbial community together with plant enzymes may degrade some organic contaminants in the rhizosphere. Root exudates provide

carbon and energy, which maintain an active microbial population. The exudates together with plant enzymes affect degradation extent. To further enhance aerobic biodegradation, remediators must lower the water table.

Beyond the root zone, plants may take up organics and then evapotranspire them to the air.

### Contaminant uptake by plants

The tendency of a contaminant to volatilize, translocate through plants, or be microbially biodegraded depends on contaminant properties, plant type, soil properties and environmental conditions. Compounds of intermediate polarity are most readily taken up by plants.

Researchers perform most plant uptake studies in closed hydroponic cultures; however, extrapolating results to actual open soil conditions may be justifiable. The possibility that a contaminant will be absorbed by a plant is reflected in its octanol-water partition coefficient,  $\log K_{ow}$ . Compounds with low  $\log K_{ow}$  ( $\leq 1$ ) are relatively mobile in both the plant's xylem and phloem, whereas those with high values are considered to be xylem mobile and phloem immobile. Compounds with high  $\log K_{ow}$  values ( $\geq 4$ ) are usually not substantially translocated into the plants. Soil type, clay content, organic matter content and rooting structure relative to the contaminant location also influence compound uptake.

### Evapotranspiration

Evapotranspiration helps plants to draw up water and contaminants into the vadose zone and can potentially lower the groundwater table. The plant-induced drying of soil causes increased oxygen transfer and gas-phase diffusion in the soil.

### Atmospheric release

Chemical properties that influence release rate to the atmosphere include

- specific gravity
- solubility
- Henry's law constant
- vapor pressure
- gas phase diffusivity
- $\log K_{oc}$

Environmental factors include

- surface area
- soil porosity
- temperature

Narayanan and others (1999) conclude that the "rates of contaminant release to the air are very small relative to dispersive mixing in the atmosphere." Dissolution rates may be a factor in intermedia transfer if the contaminant is non-aqueous. "If the route for dispersal is through water, only the concentration measured in water needs to be considered in estimating rates. A non-aqueous source (NAPL) will obviously influence the total amount that could be released over time. If a NAPL is directly exposed to air deep in the soil, its evaporative flux may be far greater than if not exposed to air . . .

If a NAPL is covered by a thin layer of water, solubility in water will become another limiting factor. At steady state, by definition, the flux from the surface for volatile compounds dissolved in groundwater will be proportional to the rate of water transfer stimulated by plants."

Narayanan and others (1999) describe in detail several release scenarios. For instance, consider MTBE, which, because of its high solubility and volatility, is readily transported through the soil and plant transpiration stream. Using a scenario presented in the text, and starting with a concentration of 100 mg/L MTBE, "researchers estimated that maximum contaminant concentration at a short distance outside the site is below the long-term level of concern on an annualized basis; within the site, MTBE releases would commonly exceed the long-term action level."

Furthermore, in some states, such as California, the median outdoor air concentration of MTBE is 2 ppbv or  $7\mu\text{g}/\text{m}^3$ ; however, concentrations at gasoline service stations are usually much larger. In contrast, MTBE releases to the atmosphere from a phytoremediation site would be small. MTBE's half-life in the atmosphere has been estimated to be about 3 days at 25°C.

According to Narayanan and others (1999), "Releasing the same amount of material over a somewhat longer or shorter period of time would give a similar overall risk factor when calculated on a carcinogenicity basis, because the carcinogenicity calculation really is using a fixed exposure amount distributed over a specified time of 70 years." Calculations can show that knowing contaminant level and volume of contaminated water, one may estimate conservative, safe emission rates relatively easily by assuming there is no biodegradation or transformation. Another variable in estimating allowable emissions is the source-to-receptor distance: with larger distances more contaminant dispersal will occur. Contaminant levels also decline exponentially in the atmosphere; concentration levels at 0.7 km (about  $\frac{3}{8}$  mile) and 3.5 km (about 2 miles) are estimated to differ by an order of magnitude. Researchers use dispersion modeling to estimate these values.

### Dispersion modeling

The most stable atmospheric conditions will occur at night, when evapotranspiration rates are low; during the day, wind and atmospheric movement cause natural convection of air and other gases. Following is an example of dispersion modeling from Narayanan and others (1999). With a flux rate of 0.35 g/s for a 30,000 m<sup>2</sup> area (obtained from evapotranspiration rates), if the wind velocity is 1 m/s (about 2 mph), 300 seconds will be required for 1 m<sup>3</sup> of incoming air to pass over the longer dimension (300 m) of the contaminant source. In this time, the entire air mass over the site will receive 105 g of contaminant. Assuming a reasonable mixing depth of 10 m, the contaminant will be dispersed in a volume of 300 x 100 x 10 or 300,000 m<sup>3</sup>. At steady state, each incoming m<sup>3</sup> of air will experience the same exposure to contaminant, about 0.35 mg/m<sup>3</sup> at the downwind edge of the area. This is the same value obtained for a distance of 0.7 km from

the source in the EPA guidance document using the estimate of 1 mg/m<sup>3</sup> for each g/s emission.

The 0.35 mg/m<sup>3</sup> value is about 8-fold lower than a 2.5 mg/m<sup>3</sup> value obtained using other wind velocity and mixing depth estimates. "On a breezier day with a velocity of 3 m/s (a little over 6 mph), the residence time of air over the site decreases by a factor of three, while with an unsteady breeze, the mixing depth might be as much as 30 m, together leading to a nearly 10-fold lower steady state concentration immediately downwind of the source" (Narayanan and others, 1999).

"For most of the Midwest over much of the year, the average wind velocity on a warm day exceeds 3 m/s. In only a few locations in the U.S. at certain times of the year will the velocity be less than 1 m/s for many hours at a time. By applying the combined transpiration and dispersion calculations we can estimate the likelihood of intermedia transfer of contaminants exceeding allowable limits outside a site. The fundamental physics of water vaporization and saturation in air sets about a 50,000-fold dilution factor on-site, provided all contaminant moves with water" (Narayanan and others, 1999).

### Conclusions

Narayanan and others (1999) conclude that "Design strategies should incorporate an understanding of surface and groundwater flow, contaminant properties and plant evapotranspiration processes. Volatile contaminants are released from the saturated zone as plants move water up into their roots. The evapotranspiration of water into the atmosphere provides a controlled release process for volatile contaminants that are not readily biodegraded by soil microorganisms. The unsaturated zone, with plant roots, microorganisms supported by root exudates and humic matter, provides an excellent environment for contaminant sorption and biodegradation."

### Petrochemical site in Texas

Al-Yousfi and others (2000) investigated a sandy, silty aquifer site in Texas that is contaminated with bis ether and 1,2-dichloroethane. The groundwater recovery system installed had 10 extraction wells pumping at 7 gpm. Researchers reported that "Twenty-plus years of natural growth of vegetation led to a diverse community of trees, bushes, and grasses, including hybrid poplar and mulberry species. Naturally occurring substances (sugars and amino acids) released by plants were also discovered in the vicinity of root systems, which could have served as a cometabolite for bacterial degradation of PAHs. In a single gram of rhizospheric soil at one location, 10,000 different microbial species were found."

Because the plume could not be treated or prevented from migrating off-site without a complementary groundwater recovery system, remediators created a "phytofence," approximately 40 poplar and mulberry trees in two rows along and within the plume. Planting and maintenance instructions for this phytodefence are given in the text. Researchers expect the mature trees to extract 100 to 200 L/day/tree of groundwater. In addition, "Long-term routine monitoring of the system will commence soon by measuring groundwater elevation and depicting the anticipated reversal trend in flow direction.

Additional enhancement and future tree planting will be decided upon and implemented if deemed necessary" (Al-Yousfi and others, 2000).

### Costs

Cost of the phytodefence was approximately \$20,000, which included trees, installation, initial irrigation and fertilization. This cost was significantly less than upgrading the existing pump-and-treat system. Researchers caution that costs vary, based on treatment strategy; for instance, harvesting plants that accumulate contaminants can drive up the cost in comparison to treatments that do not require harvesting.

"Because phytoremediation systems are primarily used as hydraulic barriers powered by solar energy, the most closely related engineering technology is a pump-and-treat system." Remediators estimated that a pump-and-treat system of three to five wells would cost more than \$100,000, including well installation, pumps and associated piping. "Our own experience indicates an approximate cost of \$10/1,000 gallon-groundwater" (Al-Yousfi and others, 2000). Maintenance costs, less than \$2,500/yr, can be attributed to pruning, mulching, and tree replacement.

### References

Narayanan, M., Erickson, L.E. and L.C. Davis, "Simple Plant-Based Design Strategies for Volatile Organic Pollutants," *Environmental Progress*, Vol. 18, No. 4, 1999; <http://www.aiche.org/publications>.

Al-Yousfi, A.B., Chapin, R.J., King, T.A. and S.I. Shah, "Phytoremediation: The Natural Pump-and-Treat Hydraulic Barrier System," *Practice Periodical of Hazardous, Toxic, and Radioactive Waste Management*, Vol. 4, No. 2, 2000; <http://www.pubs.asce.org/journals/97hz.html>.

See also the phytoremediation site, <http://www.engg.ksu.edu/HSRC/phytoform/>.



## Plant-based remediation of MTBE and other compounds in gasoline

By L.E. Erickson, Q. Zhang and L.C. Davis

This article, based on two manuscripts and a doctoral dissertation, summarizes some current studies on plant-based remediation technologies for MTBE, and to a lesser extent, for BTEX compounds.

The U.S. EPA has tentatively classified MTBE as a possible human carcinogen and issued a draft lifetime health advisory of 0.02 to 0.2 mg/liter, a dosage that is not expected to cause

carcinogenic effects over a lifetime of exposure. The EPA also has a drinking water advisory for MTBE of 0.02 to 0.04 mg/liter for taste and odor. Surveys have reported finding MTBE in 5 to 10 percent of potable groundwater supplies.

Because of its limited soil adsorption and its high water solubility, MTBE moves faster than BTEX compounds in an aqueous phase plume. Pump-and-treat is one of the most common methods to achieve plume control, although phytoremediation of MTBE is gaining attention. Table 1 compares MTBE, benzene and toluene properties.

### Plume control

Vegetation alone may be sufficient to provide plume control, while in other applications pump-and-treat may be required. Consider that a pressure gradient causes a subsurface flow of groundwater in the saturated zone, while at the earth's surface, precipitation, evapotranspiration and surface water flow control the water budget. To achieve plume control, researchers need to establish a cone of influence such that the removal of water by the vegetation through evapotranspiration, together with the mechanical pumping, exceeds the normal inputs associated with groundwater flow and infiltration. Therefore, precipitation is directed to run away from the site while efforts are made to discourage infiltrating surface water. Plants can pump up to about 2 m of groundwater per year per surface area ( $2\text{m}^3/\text{m}^2$ ) (Davis and others, 1998).

Alfalfa and poplar trees have been identified as potential plants for sites where high water use is required.

The design of a vegetated system to achieve plume control is similar to the design of a traditional pump-and-treat system:

- a mass balance is made to estimate pumping requirements to achieve plume control
- the area of planted vegetation required to remove groundwater at the calculated rate is determined
- a safety factor to compensate for uncertainty with respect to rainfall and seasonal variation is included

While plume control has been obtained in some phytoremediation applications, there is very little quantitative field data available to determine the magnitude of the safety factor.

### Fate of MTBE and BTEX compounds in vegetated soil

Zhang (1999) and Zhang and others (1999) investigated the fate of MTBE in vegetated chambers equipped with six channels (see Table 2). Some chambers were sparged and some were given additional bacteria. When vegetation is present, the water and MTBE flow to the soil surface is enhanced. Figure 1 shows the cumulative loss of MTBE over time from each of six channels. Table 2 describes the experimental conditions for each of the six channels. Table 3 provides mass balance information. The MTBE entering a vegetated channel may:

- be in the groundwater that flows out of the channel
- escape through gas phase diffusion into the atmosphere through the surface of the soil bed
- move up into the plant with the water that is transpired by the plant
- become transformed in the soil bed by aerobic microorganisms

	Planted with 10 alfalfa plants	Air sparged at 0.24 L per channel/day	Additional bacteria (2 L of suspension added 6/20/98)
Channel 1	Yes	Yes	#33 (Rhodococcus)
Channel 2	Yes	No	#33 (Rhodococcus)
Channel 3	Yes	No	No
Channel 4	No	No	No
Channel 5	Yes	No	#41 (Arthrobacter)
Channel 6	Yes	Yes	#41 (Arthrobacter)

For all channels, dimensions are 110 cm long, 65 cm deep, and 10 cm wide; soil is alluvial silty sand soil (with <10% silt) collected near a landfill in Riley County, Kansas; the system was illuminated with 12 cool white fluorescent lights (40W) at a height of 50 cm above the soil; light intensity was approximately  $160 \mu\text{E}/\text{m}^2/\text{s}$ ; channels were fed with MTBE water solution of 0.84 mM at 1 L/day from June 30 to September 20, 1998 (83-day duration); distilled water was fed from September 21 to December 25, 1998—a 96-day duration.

Table 2. Experimental conditions for the 6-channel chamber.

	Benzene <sup>a</sup>	Toluene <sup>b</sup>	MTBE <sup>c, d</sup>
Water solubility of pure phase	1,787 mg/L @25°C	550 mg/L @26°C	51,260 mg/L @25°C
Vapor pressure, mmHg	85	30	245 @25°C
Henry's law constant (dimensionless)	0.225	0.276	0.0216 @25°C
logK <sub>oc</sub> (octanol-carbon partition coefficient)	2.00	2.18	1.05
logK <sub>ow</sub> (octanol-water partition coefficient)	2.13	2.69	1.24
Molecular weight	78	92	88.2
Specific gravity	0.879	0.867	0.740 @25°C

<sup>a</sup>Source: Burken and Schnoor, 1998. <sup>b</sup>Source: Narayanan et al., 1998. <sup>c</sup>Source: Squillace et al., 1997 and <sup>d</sup>Source: Zhang, 1999.

Table 1. Physical properties of benzene, toluene and MTBE.

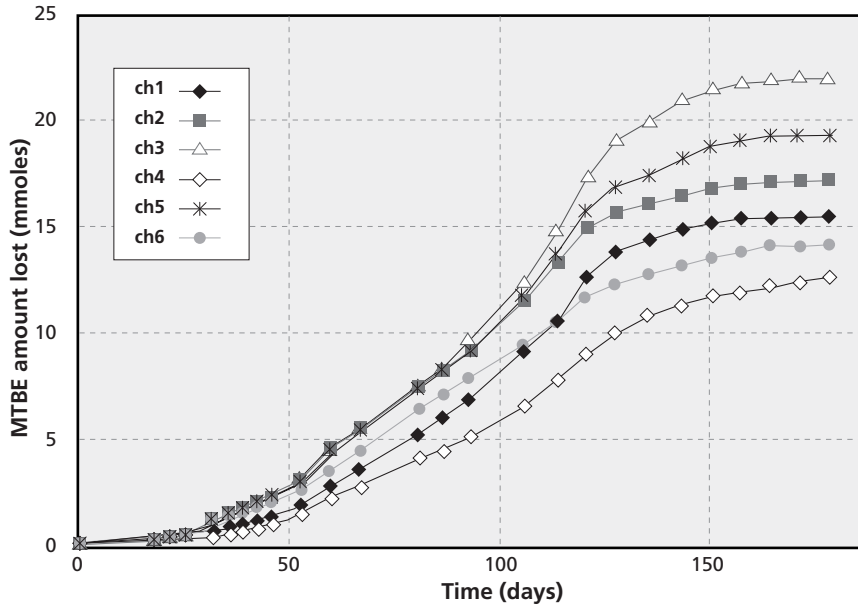


Figure 1. Cumulative loss of MTBE through soil surface to atmosphere for each channel.

In Figure 1, the largest loss to the atmosphere is associated with channel 3, which is vegetated but not inoculated with MTBE-degrading organisms. The smallest loss to the atmosphere is for channel 4, which is not vegetated. Microbial degradation appears to contribute to MTBE disappearance.

In contrast to MTBE, benzene and toluene are more likely to be biodegraded in the plant's aerobic root zone. In earlier studies with toluene, no detectable quantities of toluene were present in the atmosphere. Models based on aerobic biodegradation in the root zone fit the experimental data for toluene (Narayanan and others, 1998).

**Time and cost parameters**

Phytoremediation will often require less time than that required for in-situ bioremediation or in-situ natural attenuation; however, in some cases there may not be a very significant difference. The costs associated with phytoremediation are modest, on the order of \$10 to \$20/ton of soil.

**Fate of MTBE in the atmosphere**

MTBE released to the atmosphere through normal phytoremediation processes is rapidly dissipated. The published information on MTBE degradation and half life in the atmosphere indicates that MTBE readily degrades chemically when hydroxyl radicals (OH) are present. Squillace and others (1997) report that its half life in the atmosphere can be as short as 3 days at 25° C.

Researchers use a conservative mass balance approach to estimate MTBE concentration in the atmosphere above an MTBE release site. If the MTBE flows upward into the unsaturated zone together with the water as vegetation draws up the water, air volume required to hold the water vapor becomes a concern. For example, with 88 mg/liter of MTBE in groundwater, water and MTBE move into the atmosphere together at 25° C where the vapor pressure of water is 23.8 mm Hg

or 0.0313 mole fraction at saturation in air at one atmosphere pressure. A liter of water contains 55.6 gram moles, which requires 43,434 liters of air to dissolve the water under saturation conditions at 25° C and atmospheric pressure. The 88 mg/liter of MTBE is 1 mM in water, but only 0.001/1776 = 0.56 ppm by volume in air. Thus, MTBE concentration in air is diluted because water vapor dissipation partially controls evapotranspiration rate. Because MTBE concentrations in groundwater are often much smaller than 88 mg/liter, and because the air is usually not saturated with water vapor and the ambient air contains some water vapor, MTBE concentrations in the atmosphere due to phytoremediation are very small.

**Concluding remarks**

Phytoremediation can be used alone or with other technologies to remediate fuel spills. Mechanical pumping has been used at several sites where phytoremediation is used. In some cases, recovered water is used for irrigation of vegetation.

Channel number	1	2	3	4	5	6
Added (mmoles)	68.1	69.1	68.5	69.1	68.8	71.7
from groundwater (mmoles) <sup>a</sup>	42.4	34.5	37.6	63.0	44.7	41.2
from gas (mmoles) <sup>b</sup>	15.3	17.0	21.8	12.0	19.1	14.0
Total recovered (mmoles)	57.7	51.5	59.4	75.0	63.8	55.2
Groundwater recovery (fraction)	0.62	0.50	0.55	0.91	0.65	0.57
Soil gas recovery (fraction)	0.22	0.25	0.32	0.17	0.28	0.20
Estimated average plant uptake of MTBE (fraction)	0.015	0.029	0.024	0.0	0.032	0.035
Total recovery	0.87	0.78	0.89	1.08	0.96	0.81

<sup>a</sup> Strain #33 (*Rhodococcus*) was added    <sup>b</sup> Strain #41 (*Arthrobacter*) was added

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Table 3. Approximate mass balance data for MTBE for each of the six channels.

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## Research notes

### *An Array of Possibilities for Evaluating Environmental Health*

Technology Solutions, *Environmental Science and Technology*, Vol. 34, No. 13, 2000; <http://www.pubs.acs.org>.

The new genomics technology may help toxicologists and epidemiologists gain insight into the impact of pollutants on people, animals and ecosystems. At present, scientists extrapolate from "the high doses used in the lab to the low levels of pollution to which people are actually exposed. Evaluating data from chronic exposures and dealing with the groups of everyday chemicals to which people are routinely subjected are also major challenges for environmental health scientists. The new technology holds promise for these complicated endeavors, and it may one day provide a means of proving that exposure to one particular chemical is the cause of disease."

The genomic technology evolved from efforts to characterize the human DNA sequence. Companies in this field have produced DNA data sets, known as microarrays, on silicon chips or microscope slides. Currently, the pharmaceutical industry uses the microarrays for discovering new drugs and assessing potential toxicity of novel compounds. Applying microarray technology to environmental health assumes that "exposure to environmental agents like pollutants can affect humans and other organisms at the cellular level, causing mutations in an organism's DNA that may result in cancer or immune dysfunction . . . To compare how exposure to a pollutant may change gene expression, scientists harvest mRNA from tissue or cell samples that have been exposed to the pollutant as well as mRNA from an untreated control population. By generating the DNA associated with that mRNA and applying it to the microarray, researchers can compare the gene expression caused by the two samples by looking for DNA matches."

In late 2000 an epidemiological study in Europe used "homemade" arrays to study cancers in Central Eastern Europe. The scientists plan to use the arrays to evaluate the gene expression changes associated with human exposure to

- asbestos
- polycyclic aromatic hydrocarbons
- heavy metals
- select organic chemicals and solvents

Some predict that when issues concerning the use of microarrays are resolved, this new technology may be used in regulatory decisions.

### *Airborne Particle Emissions from 2- and 4-Stroke Outboard Marine Engines: Polycyclic Aromatic Hydrocarbon and Bioassay Analyses*

Kado, N.Y., Okamoto, R.A., Karim, J. and P.A. Kuzmicky, *Environmental Science and Technology*, Vol. 34, No. 13, 2000; <http://www.pubs.acs.org>.

Particle emissions (PM) from 2-stroke and 4-stroke outboard marine engines are a concern in California where approximately 141 tons of reactive hydrocarbons are emitted to the atmosphere per day. Although 2-stroke engines have been investigated for regulated pollutants, carbon monoxide and oxides of nitrogen, particulates emitted have been largely ignored. "Recently the U.S. EPA studied a 9.9 hp 2-stroke and 9.9 hp 4-stroke engine for air and water pollutant emissions when the exhaust was channeled into the water. The 2-stroke engine had higher levels of airborne volatile and particle emissions. Marine engines typically exhaust most of their emissions into the water through the propeller shaft, where a turbulent vortex is created. Since PM from vehicular combustion is hydrophobic, it has the potential to be resuspended into the air from the water. The emissions of PM into the air could be substantial, considering many 2-stroke engines historically emit high levels of PM" (*Kado and others, 2000*).

Researchers collected and analyzed PM emissions from three outboard engine types, all rated at 90 hp. The study took

place in a specially designed marine facility, did not analyze waterborne particles. Researchers identified particle sources that could contribute to the total particulate sources:

- direct engine emissions
- re-entrainment of PM from water aerosols generated by the engine propeller
- ambient air PM from dilution air
- particles derived from the enclosure walls and tunnel

Researchers found higher levels of emitted PAH in the tests, which may be due to differences in the characteristics of combustion, such as temperature, spray location or spray pattern of the fuel and oil, timing of fuel delivery, and high levels of unburned fuel and oil in the 2-stroke engines. Two-stroke engines also emitted the highest levels of semivolatle PAHs. "From these studies, the 2-stroke marine engines tested, including a direct fuel-injected model, had significantly higher emissions for PH, PAHs and mutagenic activity than the 4-stroke engine design . . . results indicate that pollutants such as PM and PM-associated toxic compounds are released into the ambient air from outboard marine engine activity in water and that engine design dramatically affects these pollutant concentrations and total emissions" (Kado and others, 2000).

**Stochastic Analysis of Oxygen- and Nitrate-Based Biodegradation of Hydrocarbons in Aquifers**

Kaluarachchi, J.J., Cvetkovic, V. and S. Berglund, *Journal of Contaminant Hydrology*, Vol. 41, pgs. 335-365, 2000; <http://www.elsevier.com/locate/jconhyd>.

Researchers modeled field-scale aerobic biodegradation using a Lagrangian stochastic framework and Monod-kinetics. Using various values for hydraulic conductivity, researchers

simulated subsurface heterogeneity and observed reactions between the contaminant, oxygen, other electron acceptors and microbes. Other studies have reported that oxygen is the most effective electron acceptor, followed by nitrate, and that reaction kinetics related to degradation are complex. In addition, studies have showed "that subsurface heterogeneity plays an important role in determining the fate of different species during biodegradation, and that the impact of heterogeneity, even on the smallest scale, cannot be ignored . . . aerobic degradation may be responsible for only 15 to 20 percent of the overall degradation in the long-term decay of a contaminated plume, with other electron acceptors such as NO<sub>3</sub><sup>-</sup>, Mn<sup>4+</sup>, Fe<sup>3+</sup> and SO<sub>4</sub><sup>2-</sup> being responsible for the majority of degradation."

Researchers had difficulty "in attempting to extrapolate results of controlled laboratory experiments to complex field-scale conditions. In addition, modeling work showed that the representation of reaction kinetics can be performed by many approaches ranging from simple instantaneous kinetics to flexible nonlinear Monod expressions, but that the associated analysis with Monod expressions can be complex and computationally intensive." (Reviewer's comment: *This is not necessarily true: "instantaneous" is not a valid approach. Monod computations are trivial if operation splitting is used.*)

Researchers concluded the following:

- remediation time is highly dependent on sorption, electron acceptor availability and heterogeneity
- when subsurface heterogeneity is present, risk assessment and remediation designs need to account for higher contaminant variability
- increased heterogeneity reduces the consumption of the electron acceptor and thereby biodegradation affectivity

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- the presence of nitrate and the consideration of nitrate as a second electron acceptor produced significantly different results under intrinsic conditions for different scales of heterogeneity and sorption
- as a second electron acceptor, nitrate was able to lower peak contaminant concentration and effectively increase remediation

Researchers suggest they could better analyze the potential field-scale biodegradation scenarios with this stochastic methodology.

### ***Comparison of Hydraulic Conductivity Values Obtained from Aquifer Pumping Tests and Conservative Tracer Tests***

Niemann, W.L. and C.W. Rovey II, *Ground Water Monitoring and Remediation*, Summer 2000; <http://www.ngwa.org>.

Researchers in Iowa used various methods to measure hydraulic conductivity (K) in a shallow sandy aquifer in a glacial outwash setting. Testing consisted of pumping tests (six each), and two types of tracer tests: natural gradient (two each) and forced gradient (two each).

Niemann and Rovey (2000) state that "The presented results indicate that K values obtained from aquifer pumping tests are approximately 10 to 20 times greater than those determined from conservative tracer tests at the site. If the Camp Dodge site is representative of other sandy aquifers, groundwater solute velocities may be overestimated if pumping test values are used as the applicable measure of K." Tracer test values are closer to values determined from 55 slug tests, and also closer to values estimated from grain-size distribution analyses.

Niemann and Rovey report that "K values calculated from the natural-gradient tracer tests depend on the value of effective

porosity measured during the radial-convergent tracer tests. Factors such as wellbore mixing/flushing in a finite-diameter well and non-uniform flow can affect breakthrough curves of such tests. Chiefly, these factors may alter the general shape of breakthrough curves by increasing spreading and introducing marked asymmetry between the rising and falling limbs. The next effect is a shift in the time of arrival for the tracer's center of mass past the time of peak concentration. The main concern arising from these effects is a bias in dispersivity calculations."

"The difference in magnitude of K may be related to the scale of the different tests: large-scale tests generally give larger mean values of K than do small-scale tests, except in extremely homogeneous media. Pumping tests affect head distribution over a large volume of aquifer, whereas tracer tests sample a small volume at any given time."

Another idea put forth by the authors is that "transient solute migration is less affected by high-conductivity heterogeneities than are pumping tests. Dispersion and diffusion may prevent a solute from traveling exclusively within thin high-conductivity pathways that account for a large proportion of water flux during pumping tests . . . the center of mass of a conservative tracer moves more slowly than would be predicted from the average K. In these cases the solute has to cross both high and low-conductivity heterogeneities and cannot migrate exclusively within the higher K zones. Thus, the solute velocity reflects an average weighted more toward lower-conductivity zones."

Authors conclude that the differences in values of K reflect varying degrees of influence of different pore systems on the two types of tests; K values based on water flux in the aquifer studied are distinctly larger than those based on transient solute flux.

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