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U.S. EPA OUST has two excellent websites:

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Contains state home pages for UST, LUST and Fund agencies

<http://www.epa.gov/swerust1/mtbe/index.htm>

Contains advisories, publications, and links on MTBE



Evaluating natural attenuation of MTBE

Although under certain conditions MTBE can be removed by destructive processes such as biodegradation, non-destructive processes—dispersion, in particular—may be the most important MTBE-reduction mechanism in the subsurface (*Anthony and others, 1999*). This article describes a methodology for assessing the natural attenuation of MTBE.

Review of MTBE properties and characteristics

The physical properties and characteristics of MTBE include

- minimal retardation; MTBE has an affinity for the aqueous phase and moves approximately at groundwater velocity
- low Henry's law constant, indicating a limited transfer of MTBE to the atmosphere by groundwater volatilization
- penchant to preferentially partition out of NAPL (non-aqueous phase liquid) into the aqueous phase; thus MTBE concentrations will vary depending on flux of water through NAPL
- unreactive over a wide range of conditions; in general, MTBE, an ether, is difficult to degrade
- ability to react with hydroxyl radicals in the aqueous phase, yielding tert-butyl formate (TBF) which is readily hydrolyzed to tert-butyl alcohol (TBA)
- ability to undergo microbially mediated hydrolysis and oxidation reactions; the only expected pathway for MTBE degradation (in surface water, soil and groundwater) is by production of TBA, which is degradable, but at very low rates
- recalcitrant in most situations, which means that no degradation was observed under the particular conditions and within the time frames examined; MTBE is resistant to microbial degradation because of its stable, unreactive ether bond
- degradation under certain conditions by
 - denitrification
 - iron reduction
 - sulfate reduction
 - possibly methanogenesis

MTBE has a possible field degradation rate of 0.000 to 0.008/day; however, the limited database of field investigations does not yet allow broad generalizations to be made regarding the particular environmental conditions that favor MTBE degradation (*Anthony, 1999*).

In addition, although TBA is the principal degradation product of MTBE, its appearance does not necessarily indicate degradation; TBA is also used as a fuel additive.

Thus many investigators regard dispersion as the most significant attenuation mechanism for MTBE.

MTBE properties will impact on the site investigation. For instance, because MTBE preferentially partitions out of NAPL into the aqueous phase, MTBE concentrations will vary depending on the flux of water through NAPL.

Evaluating natural attenuation of MTBE

The lines of evidence that support MTBE natural attenuation, which are similar to those of hydrocarbon attenuation, include

- documented loss of contaminants at the field scale
- evaluation of contaminant and geochemical analytical data
- direct microbial evidence

According to Anthony and others (1999), "The apparent recalcitrance of MTBE to biodegradation, together with its tendency to remain in the aqueous phase and to move virtually unretarded with advective groundwater flow, suggests that in many cases dispersion may be the most important attenuation process acting to reduce MTBE concentration in the subsurface. Therefore, evaluation of the distances and times required for dispersion to reduce MTBE concentrations to acceptable levels must be a primary objective of natural attenuation investigations that address MTBE in groundwater."

Quantification of contaminant migration and attenuation rates, and implementation of MNA (monitored natural attenuation) will require the following:

- reviewing existing site information
- developing a preliminary conceptual model and assessment of potential for natural attenuation
- refining the conceptual model based on site characterization data, using pre-modeling calculations, and documenting natural attenuation indicators
- simulating natural attenuation using analytical or numerical solute fate and transport models that allow incorporation of a biodegradation term
- conducting an exposure assessment
- preparing a long-term monitoring plan
- presenting findings to regulatory agencies and obtaining approval for the MNA option

References

Anthony, J.W., Henry, B.M., Wiedemeier, T.H., Gordon, E.K. and J.B. Bidgood, "Methodology to Evaluate Natural Attenuation of Methyl Tertiary-Butyl Ether," 1999, *In-Situ Bioremediation of Petroleum Hydrocarbon and Other Organic Compounds*, Battelle Press; 800-451-3543; <http://www.battelle.org/bookstore>.

Anthony, J.W., "MTBE half-life," e-mail to the Biogroup, February 23, 1999; see archives at <http://www.biogroup.gzea.com>.

UTTU thanks John Anthony, Parsons Engineering Science, Inc., Denver, Colorado, for his help on this article.



Quantifying effective vapor-phase diffusion coefficients

To refine the results of risk-based corrective action sites where vapor migration pathways exist, researchers are beginning to quantify the role of vapor migration. Vapor migration is of concern with respect to enclosed spaces, atmospheric venting and estimates of contaminant source longevity.

Johnson and others (1998) developed a field procedure—a transient concentration analysis—to estimate effective vapor-phase porous media diffusion coefficients. The protocol involves injecting into the soil a conservative gas tracer, waiting, then withdrawing and measuring. This article describes the protocol, results from a field study, and the relevance of the vapor-phase porous media diffusion coefficient (D_v^{eff}) for estimating risk.

Effective vapor-phase porous media diffusion coefficient

Diffusion is the process by which different substances mix as a result of the random motions of their component atoms, molecules and ions. The effective vapor-phase media diffusion coefficient has not always been defined consistently in the literature, thus values given in the literature will not always be comparable. Johnson and others (1998) define the effective vapor-phase porous media diffusion coefficient, $D_{i,v}^{\text{eff}}$ (m^2/s), in accordance with its use in the modified Fick's first law, which is

$$F_i = -D_{i,v}^{\text{eff}} \nabla C_{i,v} - D_{i,w}^{\text{eff}} \nabla C_{i,w}$$

F_i = flux of chemical i ($\text{mg}/\text{m}^2\text{-total soil area}/\text{s}$)

$D_{i,v}^{\text{eff}}$ = effective vapor-phase porous media diffusion coefficient (m^2/s)

$C_{i,v}$ = vapor phase concentration of chemical i (mg/m^3)

$D_{i,w}^{\text{eff}}$ = effective moisture-phase porous media diffusion coefficient (m^2/s)

$C_{i,w}$ = moisture-phase concentration of chemical i ($\text{mg}/\text{m}^3 \cdot \text{H}_2\text{O}$)

∇ = the del vector operator

According to Johnson and others (1998), "Under natural conditions, the migration of volatiles and semivolatiles within the vadose zone is often assumed to be dominated by vapor-phase diffusion. In some cases, such as the transport of aerobically degradable petroleum compounds, reaction may also play an important role. Thus if one wants to quantify vapor migration rates for these situations, then knowledge of site-specific effective vapor-phase porous media diffusion coefficients is essential. This knowledge is also critical if one wants to characterize or quantify the kinetics of aerobic biodegradation using in-situ measurements."

Models that incorporate the diffusion process require input of an effective porous media diffusion coefficient. This quantity "is a function of chemical-specific molecular diffusion coefficients in air and water as well as porous media and chemical properties. In some situations, such as those involving mixtures with high vapor concentrations, the diffusion coefficient may also vary with composition." This parameter is "a combination of effective porous media diffusion coefficients in the vapor and soil moisture phases when equilibrium between phases is assumed. . . For estimation purposes, empirical correlations are used to determine the individual effective vapor-phase and soil-moisture-phase porous media diffusion coefficients as functions of the moisture content and total porosity of the porous medium" (Johnson and others, 1998).

"Results from transport models can be very sensitive to small changes in the effective diffusion coefficient. This is true when trying to predict coupled diffusion and reaction in the vadose zone or when trying to back out kinetic biodegradation parameters from field oxygen and contaminant soil gas data" (Johnson and others, 1998.) If direct measurements are not possible, modelers use empirically derived effective diffusion coefficients. Because the uncertainty of the values is sometimes not acceptable, it is preferable to measure diffusion coefficients.

Johnson and others (1998) emphasize that empirical correlations of the coefficients have been evaluated using laboratory-scale soil column experiments. Duplication of laboratory values by field testing, however, has not yet been established.

Determination of effective vapor-phase diffusion coefficient

Johnson and others give the mathematical basis, 12 formulas, for computing effective diffusion coefficient. If the effective diffusion coefficient is known for a tracer (which is used in the field procedure), the effective diffusion coefficient can be calculated for the chemical of interest by correcting for differences in molecular diffusion coefficients in air. Other quantities that must be measured (usually in core samples) include total porosity, moisture content and soil bulk density. Given the difficulties in collecting and analyzing intact soil cores, errors in these quantities are likely.

Transient concentration analysis approaches require a measurement for air-filled porosity. "The unique feature of the transient volume-averaged concentration approach proposed here is that in many settings it is not necessary to independently measure air-filled porosity to determine with reasonable precision the effective diffusion coefficient." Researchers reason that by using average vadose zone vapor-filled porosity values for a particular soil type at most sites, then "this approximation error could easily be reduced to within 10 percent" (Johnson and others, 1998).

Considering all sources of error, researchers concluded "that in many settings independent measurement of the vapor-filled porosity is not necessary since little error (< 20 percent) will be introduced by assuming a reasonable value as described above. Furthermore, the unique feature of the transient volume-averaged concentration approach is that the functional dependence on air-filled porosity is reduced relative to the transient point concentration measurement-based approaches. . . One other advantage of the approach described here is that one can measure diffusion properties over different length scales by changing the injection and extraction volumes" (Johnson and others, 1998).

Limitations

Limitations to this approach include the following:

- low permeability or very wet soils complicate the injection or extraction of soil gas
- because radial symmetry is assumed, the approach's applicability is questionable in regions where significant changes in soil properties occur over relatively small distances, such as near the capillary fringe or ground surface

Protocol

The researchers' protocol characterized the following effective diffusivity coefficient:

- D_v^{eff} within a spherical volume of about a 9 cm diameter
- vapor-filled porosity of about $0.30 \text{ cm}^3\text{-vapor/cm}^3$, which corresponds to about a 1-L soil gas volume, although procedure modification could accommodate larger or smaller volumes

The protocol uses SF_6 as the tracer, but other gases such as helium could be used. Researchers used a gas chromatograph equipped with an electron capture detector (GC-ECD) to quantify SF_6 concentrations in the ppt to ppm volume range.

The protocol is as follows:

- 1) Prepare a gas mixture containing approximately 10 ppmv SF_6 , or other conservative tracer gas at a measurable concentration
- 2) Inject 5 ml of 1 ppmv SF_6 into a 1-L Tedlar bag containing 1 L of SF_6 -free air
- 3) Measure the resulting concentration in the 1-L Tedlar bag (should be approximately 5 ppbv); record and denote this value as C_{max}
- 4) Inject 5 ml of 1 ppmv SF_6 into the desired location in the vadose zone through small-diameter tubing ($\frac{1}{8}$ " stainless steel tubing is preferred); follow this injection with sufficient SF_6 -free air to ensure that the 5 ml of SF_6 has just been flushed from the tubing into the vadose zone (the total injected volume should be minimized; approximately 1 ml/ft for 304 SS $\frac{1}{8}$ " tubing with 2.1 mm i.d.)

- 5) Immediately withdraw enough soil gas to fill a 1-L Tedlar bag; analyze and record the SF_6 concentration in the 1-L Tedlar bag
- 6) Inject 5-10 L of clean air into the vadose zone at this sampling point before conducting a longer test
- 7) Repeat steps 4, 5, and 6 except wait for periods of 15, 60 and 120 minutes before withdrawing the soil gas sample for analysis
- 8) Reduce the data by dividing the measured concentrations by C_{max}

(Johnson and others, 1998)

Field testing

Researchers conducted tests at the U.S. Naval Base at Port Hueneme, California. Vadose zone soils there are fairly uniform sandy silts and clayey silts. Depth to groundwater at time of test was approximately 3 m. Workers constructed a multilevel monitoring network using 5-cm-diameter PVC pipe and 0.32-cm-diameter stainless steel tubing installed with direct push equipment. They collected vapor samples at depths of 0.6, 1.2, and 1.8 m below ground surface. Researchers did not measure actual soil moisture contents; however, capacitance probe logging nearby suggested that the soil moisture content increased with depth.

Researchers collected data in accordance with the procedure described above. They measured the tracer gas concentration (SF_6) with a Lagus AutoTrac instrument equipped with an electron-capture detector. Samples were taken at the three depths and for time periods of 15, 60 and 120 minutes. Researchers reported that data were most consistent with sampling intervals of 60 and 120 minutes, but overall results indicated a decrease in D_v^{eff} with depth. Additional studies are needed to

- refine the protocol
- define typical ranges of variability
- define scenarios where this approach is and is not appropriate

In addition, if field workers take measurements of moisture content, total porosity and air-filled porosity, then this approach can be used to assess the validity of commonly used empirical correlations.

Reference

Johnson, P.C., Bruce, C., Johnson, R. I. and M.W. Kemblowski, "In Situ Measurement of Effective Vapor-Phase Porous Media Diffusion Coefficients," *Environmental Science and Technology*, Vol. 32, No. 21, 1998; 703-552-0685; <http://www.pubs.acs.org>.



Air sparging patterns

In-situ air sparging (ISAS) produces two types of gas-phase flow patterns in groundwater. Why one flow pattern is preferred over the other is the topic of this article.

ISAS works by injecting a gas phase, typically air, into the saturated zone, which either volatilizes contaminants or promotes bioremediation. The process relies on mass transfer from one phase to another. This mass transfer "is dependent on the interface between the phases, which is dependent on the type of flow pattern" (*Brooks and others, 1999*).

There are two types of gas phase flow patterns:

- bubble flow, which involves discrete non-continuous air bubbles moving through the porous media
- channel flow, in which air displaces liquid along continuous paths of least resistance

Brooks and others (*1999*) give references for experimental testing for each flow pattern.

Channel flow, in this context, describes a continuous air phase. The literature describes several forms, the two extremes being

- a macro-scale dendritic distribution of air channels of limited cross-sectional extent, originating from the point of air injection
- an air distribution in which the air saturation varies continuously over the entire domain influenced by the air injection points

Factors that will influence channel flow patterns include

- viscosity differences
- capillarity
- air flow rates
- micro- and macro-scale heterogeneities

Brooks and others (*1999*) use the term "channel flow" " to imply a continuous air distribution, with no inferences intended for the nature of the air distribution in the continuum . . . recent ISAS articles imply that channel flow will prevail in sparging operations. An inherent assumption in this inference is related to the size of the porous media. . . It is noted that a transition from air channels to bubble flow is observed as the porous media size increases. It is evident from [several studies] that a change in the observed flow patterns occurs around 1 to 2 mm for air-water systems. . . Using the Uniform Soil Classification System, the transition zone falls in the medium to coarse sand classification. Therefore, as a gross predictive tool, ISAS in a porous media characterized by medium sand or smaller will have air channels, while the flow pattern in porous media composed of coarse sand or larger will have bubble flow. Since the majority of remedial sites are composed of media smaller than coarse sand, air channels should prevail."

Brooks and others (*1999*) note, however, that "in general, multiphase flow patterns in porous media are a complex phenomenon which depend on porous media properties and ISAS-operational characteristics. It should be understood that for the wide range of subsurface conditions and operational characteristics that may occur, both bubble flow and air channels are possible, and with changing conditions, one flow pattern may evolve to the other, or to an intermediate two-phase flow pattern." The importance of predicting flow pattern is essential to understanding remedial potential and developing predictive models. "Flow patterns must be known before flow calculations, such as the application of Darcy's law, can be attempted."

Flow pattern change as related to grain size

Pore space in the subsurface may be thought of as larger pore bodies interconnected by smaller pore throats. "Consider a single bubble within a pore body. Assume the bubble is on the same order as the pore body. The bubble will tend to rise due to the associated buoyancy force, and the capillary pressure associated with the pore throat above the bubble will oppose the upward motion. If the buoyancy force is sufficient to overcome the capillary pressure of the pore throat, then the bubble will rise. If the buoyancy force is not sufficient to overcome the capillary pressure, then the bubble will be trapped in the pore space. Additional pressure will have to be supplied to the air phase in order for it to advance through the pore throat. This additional pressure would be supplied through an air connection to the air source (i.e., a channel from the sparging well). It is proposed that this simplistic scenario defines the division between bubble flow and channel flow" (*Brooks and others, 1999*).

When buoyancy forces are large enough to overcome capillary pressures, bubble flow occurs. When capillary pressures prevent bubble advancement, channel flow occurs. Not included in this situation is the effect of viscous forces, which are very important in multiphase flow. "Viscous forces are a function of the velocity of the fluid phase . . . Since the onset of motion is the focus, the fluid velocities are negligible and consequently the viscous forces are negligible" (*Brooks and others, 1999*).

Capillary and buoyancy equations

The pressure difference across a gas-liquid interface is the Laplace equation of capillarity:

$$P_c = \sigma (1/r_1 + 1/r_2)$$

$$\sigma = \text{air-water surface tension [M-T}^{-2}\text{]}$$

$$r_1 \text{ and } r_2 = \text{principal radii of curvature [L]}$$

$$P_c = \text{capillary pressure [M-T}^{-2} - \text{L}^{-1}\text{]}$$

This equation shows that capillary pressure is inversely proportional to the radii of curvature.

Buoyancy force, F_b , for a spherical gas bubble is defined as

$$F_b = \left(\frac{4}{3} \pi r_b^3\right) g \Delta \rho$$

r_b = radius of bubble [L]

g = gravitational acceleration [L-T⁻²]

$\Delta \rho$ = density difference between the object and the surrounding fluid [M-L⁻³]

At this point, researchers use a dimensionless number called the Bond number (N_B) to compare buoyancy and capillary forces. The Bond number is defined as

$$N_B = \frac{\Delta \rho g r^2}{\sigma}$$

r = particle radius [L]

The above equation assumes

- a zero contact angle between the wetting and solid phases
- r is a measure of the pore space

If N_B is less than one, capillary forces dominate; if N_B is greater than one, buoyancy forces dominate. In this scenario capillary and buoyancy forces exist in a single spatial dimension. "The pore throat dimension will dictate the largest capillary pressure and the pore body dimension will dictate the largest buoyancy force. A single value of r cannot simultaneously characterize both the pore size associated with the maximum capillary pressure and the pore size associated with the maximum buoyancy force" (Brooks and others, 1999). Therefore the authors propose another Bond number (modified Bond number N_B^*) to compare buoyancy forces in the pore body to capillary forces:

$$N_B^* = \left(\frac{\Delta \rho g}{\sigma}\right) \left(\frac{r_b^3}{r_c}\right)$$

Where

N_B^* = modified Bond number

r_c = radius associated with capillary force (pore throat radius) [L]

"The ratio of the pore throat size to pore body size, referred to as the aspect ratio, α , can be used to eliminate r_c from the above equation. The pore body radius, r_b will be assumed equal to r (the grain radius) and r_c is assumed equal to r times the aspect ratio, α . Therefore, the modified Bond Number may be defined as

$$N_B^* = \frac{N_B}{\alpha} = \frac{\Delta \rho g r^2}{\sigma \alpha}$$

Brooks and others report from a literature search that "the radius associated with capillary forces should be between 0.05 and 0.5 times the radius associated with buoyancy forces."

"Figure 1 shows the modified Bond number for aspect ratios of 0.1 and 0.05. Values of α used to create the plot were 72 dyne/cm for air-water-glass and 60 dyne/cm for water-air-soil . . . For comparison, the Bond number has also been plotted on this figure. Note that the modified Bond number is approximately unity around 1 to 2 mm for the aspect ratios

used, which corresponds to the transition zone on Figure 2. This result fits the presumption that change in flow patterns occurs when capillary forces and buoyancy forces reverse their dominance" (Brooks and others, 1999).

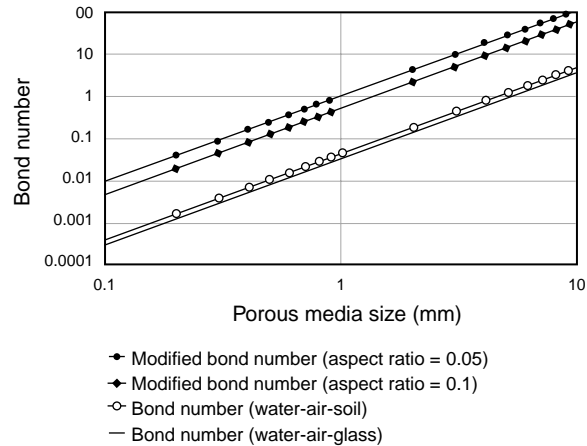


Figure 1. Modified Bond number for aspect ratios of 0.1 and 0.05 (from Brooks and others, 1999).

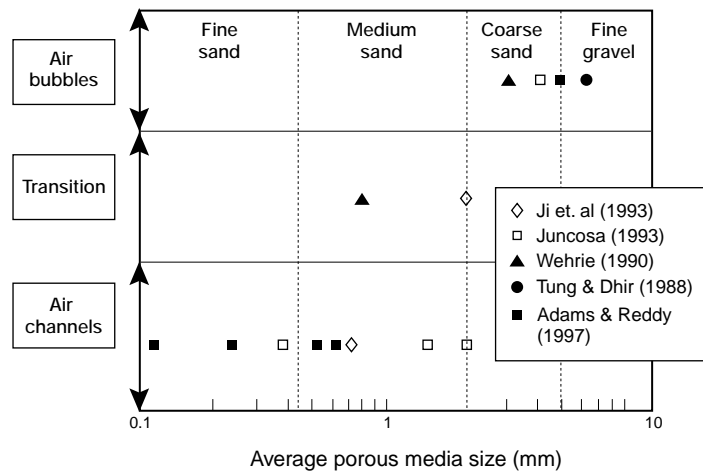


Figure 2. Air bubble, transition and air channel zones as related to grain size (from Brooks and others, 1999).

Experimental work

Researchers devised laboratory experiments to observe flow patterns as determined by porous media size and air flow rate. Major apparatus consisted of the following:

- an air-sparging column of borosilicate, 30 cm long with an outside diameter of 6.3 cm
- a pre-sparging column to water-saturate the air
- a compressor, to supply air
- a cylindrical diffusing stone, to inject air into the sparging column

Researchers used six glass bead sizes:

- 0.43 to 0.50 mm
- 0.71 to 0.80 mm
- 1 mm
- 1.5 mm
- 2 mm
- 3 mm

The calculated porosities for all beads (once in the glass cylinder) ranged from 33 to 36 percent. "These porosities fall between a loose cubic packing with a calculated porosity of 47.6 percent and a tight rhombohedral packing with a calculated porosity of 26.0 percent." During testing, researchers used flow rates of 20 to 3,000 ml/min. Brooks and others (1999) give thorough descriptions of cleaning, bead packing, and sparging procedures.

Conclusions

Because visual pattern identification is subjective, researchers identified flow patterns that fell into the following categories:

- channel flow, describing the pattern when areas within the columns suddenly desaturated and remained significantly unsaturated for the duration of the applied pressure
- bubble flow and slug flow, describing the patterns characterized by moving bubbles or moving slugs, respectively; the distinction between bubbles/slugs and channels was not always clear; in general, if an area was not significantly resaturated after initial desaturation, then the pattern was labeled channel flow
- mixed flow when either bubbles or slugs, and channels were combined

Researchers also found that wall effects prevent "tight packing of porous media adjacent to the wall and produce larger pore spaces. The result is that bubble and slug flow patterns may be produced at the wall, whereas channel flow prevails in the interior of the column. However, for the larger bead sizes, the size of the observed slugs and bubbles, and the ease with which they moved through the media, suggest that slug and bubble flow occurred throughout the sparging column. . . . As predicted, air channels correspond to values of the modified Bond numbers less than unity, and the pattern changes to slug and bubble flow as the modified Bond number increases above unity" (Brooks and others, 1999).

In addition, researchers contend that because Darcian theory is used in ISAS modeling, and Darcian theory is based on the concept of a representative elementary volume (REV), the results of their air-sparging column experiments merit further discussion. According to Brooks and others (1999), "This concept basically assumes that microscopic porous media properties can be described in terms of macroscopic values over some small volume. One such value is the air saturation. Observations during the column experiments lead to the conclusion that a REV smaller than the volume of the column would be necessary to describe the air saturation. At high flow

rates for the glass beads less than 1 mm, for example, the air saturation appeared to become sufficiently distributed over the column to apply an REV concept. However, at low flow rates, a large portion of the column remained saturated with water; therefore, a single air saturation value could not be used to describe the entire column. Care should therefore be used in application of these models to describe all ISAS situations."

Researchers concluded the following:

- the air phase distribution and dynamics during ISAS are complex due to the intricate nature of aquifer material, as are the forces controlling multiphase flow in porous media
- multiple air flow patterns may exist depending on aquifer characteristics
- channel flow will occur in porous media less than 1 to 2 mm, approximately the size of a medium sand according to the Unified Soil Classification System
- this observation, based on porous media composed of glass beads, raises questions of applicability in natural porous media; any porous media property that affects the pore space or aspect ratio will affect the flow pattern transition size
- decreasing pore space size will promote air channels
- grain-size distribution, sand grain angularity and packing configuration will affect the pore space dimensions
- grain-size distributions will most likely have the largest effect on pore space; a broad grain size range will complicate the relationship between the pore space and grain size; smaller grains will fill spaces between larger grains; therefore pore space size will be similar to the size of the smaller grains; in the smaller grain size, distribution should be used to calculate the modified Bond number
- a fundamental understanding of which pattern may occur is important to flow and mass transfer processes because of their dependence on the air phase pattern and distribution

Additional publications

Other publications of interest include:

Rabideau, A.J. and J.M. Blayden, "Analytical Model for Contaminant Mass Removal by Air Sparging," *Ground Water Monitoring and Remediation*, Fall 1998; 800-332-2104; <http://www.ngwa.org>.

Special feature on advances in air sparging, *UTTU*, Vol. 8, No. 4, 1994.

Special feature on air sparging, *UTTU*, Vol. 6, No.3, 1992.

Reference

Brooks, M.C., Wise, W.R. and M.D. Annable, "Fundamental Changes in In-Situ Air Sparging Flow Patterns," *Ground Water Monitoring and Remediation*, Spring 1999; 800-332-2104; <http://www.ngwa.org>.



Ethanol as a substitute for MTBE

The use of MTBE is being phased out in some states and may eventually be phased out in the entire United States. One suggested substitute for MTBE is ethanol (*Parkinson, 1999; Ellis and Kuhn, 1999*). Ethanol use has problems, which include the following:

- insufficient capacity; California alone would require 75,000 bbl/d; however, total U.S. capacity is 120,000-125,000 bbl/d
- higher Reid vapor pressure, 18 psi vs. 8 to 8.5 for MTBE
- volume percent limitations; use of ethanol at the 2 weight percent oxygen level adds 5.5 volume percent vs. MTBE's 11 volume percent to gasoline
- high water solubility, which prevents transportation by pipeline
- increased cost; ethanol is estimated, at current gasoline prices, to add 6 to 7 cents/gallon to the cost of gasoline production
- increased emissions of acetaldehyde
- higher mass and reactivity of evaporative emissions vs. MTBE-blended gasoline
- its co-solvency effect, which is described below

Brazilian study of ethanol-amended gasoline

In Brazil, which typically produces gasoline that is 78 percent standard gasoline and 22 percent ethanol, the co-solvency effect of ethanol on other gasoline components is an issue of concern (*Corseuil, 1999*).

A primary determining factor on the distribution, fate and transport of chemical compounds in the environment is the compound's solubility. Knowing the aqueous solubility of the pure compound and its fraction in gasoline, researchers can calculate the constituent concentration in the aqueous phase (Raoult's law). Previous studies have shown that the aqueous solubility of hydrophobic organic compounds (HOCs) increases log-linearly with increased fraction of completely miscible compounds (CMOS, or co-solvents). This can be expressed as

$$\log S_m = \log S_w + \beta f_c$$

S_m = HOC solubility in the mixture of binary solvents

S_w = HOC solubility in pure water

β = a measure of co-solvency energy

f_c = volume fraction of the co-solvent in the mixture of binary solvents

The co-solvency energy is determined by

$$b = a \log(K_{ow}) + b$$

K_{ow} = the octanol/water partition coefficient of HOC

a, b = regression parameters applied to a group of hydrophobic organic compounds

Corseuil (*1999*) created a series of experiments using batch reactors consisting of 60-ml glass vials with Teflon septum and aluminum seals. He calculated co-solvency energy by using pure benzene, toluene, o-xylene and ethanol in an ethanol-distilled water system. Following the tests with pure compounds, Corseuil used ethanol-free gasoline and Brazilian commercial gasoline to evaluate his co-solvency log-linear model. "The experiments with BTX were prepared in triplicate with a water-BTX ratio of 10:1 (experiments were performed using one hydrocarbon at a time) with 1.0, 2.2, 4.0, 16.0 and 22.0 percent ethanol. The experiments with ethanol-free gasoline and with the Brazilian commercial gasoline were prepared with gasoline-distilled water ratios of 1:1, 1:2, 1:5, 1:10 and 1:20. Regarding the Brazilian commercial gasoline, these ratios represented ethanol fractions of 15.8, 8.47, 3.62, 1.81 and 0.93 percent respectively."

Results and conclusions

Corseuil (*1999*) used data from these experiments to predict a co-solvency energy. "The values obtained for benzene (0.65), toluene (1.27) and o-xylene (1.66) indicate that the co-solvency energy increases according to the greater BTX hydrophobicity (lacking affinity for water). Thus, the more hydrophobic the contaminant, the greater the ethanol effect on solubility enhancement of these compounds."

Experiments with pure gasoline (i.e., no ethanol) indicated a relatively constant BTX solubility; experiments with Brazilian commercial gasoline (ethanol-amended) indicated enhancement of BTX aqueous concentration along with the enhancement of the co-solvent fraction. For instance, ethanol-free aqueous solubility for pure BTX components were

- 1,757 mg/L for benzene
- 567 mg/L for toluene
- 202 mg/L for o-xylene

With 10 percent ethanol in the aqueous phase, solubility of BTX compounds increased as follows:

- 20 percent for benzene
- 40 percent for toluene
- 50 percent for o-xylene

Corseuil's data and model (co-solvency energy and log-linear plots) can be used to predict solubility enhancement. For instance, "For ethanol fractions in groundwater of 1 and 10 percent, the model predicts that the total BTX mass in the aqueous phase would increase 3 and 33 percent, respectively . . . this effect is larger for xylenes, which are the least soluble compounds among BTX" (*Corseuil, 1999*). The largest unknowns that influence the model are the molar fractions of benzene, toluene and xylene in the gasoline.

References

Corseuil, H.X., "Co-solvency Effect in Aquifers Contaminated with Ethanol-Amended Gasoline," in *Natural Attenuation of Chlorinated Solvents, Petroleum Hydrocarbons and Other Organic Compounds*, 1999, Battelle Press, Columbus, Ohio; 614-424-6393; <http://www.battelle.org/bookstore>.

Ellis, P. and J. Kuhn, "The National Research Council (NRC) Report: 'Ozone-Forming Potential of Reformulated Gasoline,'" June 1999, *ASTSWMO MTBE and Fuel Oxygenates Workgroup Newsletter*, Vol. 2, No. 2; 202-624-5828; <http://www.astswmo.org/Publications/summaries.htm#MTBE>.

Parkinson, G., "All Sides Pumped Up for MTBE Ban," *Chemical Engineering*, June 1999; 212-512-2000; <http://www.che.com>.



Research notes

Evidence for MTBE in Heating Oil

Robbins, G.A., Henebry, B.J., Schmitt, B.M., Bartolomeo, F.B., Green, A. and P. Zack, *Ground Water Monitoring and Remediation*, Spring 1999, 800-332-2104; <http://www.ngwa.org>.

A study of heating oil releases in Connecticut showed evidence of MTBE releases, although heating oil does not generally contain MTBE. Maximum concentrations ranged from 1 to 4,100 µg/L. Data indicated that 51% of the releases were associated with aboveground storage tanks (mostly associated with leaks in fuel lines buried beneath basement floors), whereas 32% of the releases involved USTs, with the remaining 17% unknown.

"Equilibrium MTBE water concentrations could vary over the range of calculated MTBE heating oil concentrations . . . levels of MTBE contamination in heating oil in the range of 0.2 to 0.8 mg/L could result in groundwater contamination levels that exceed the EPA drinking water advisory levels. MTBE contamination in heating oil at levels greater than 1 to 2 mg/L could result in groundwater concentrations exceeding the CTDEP (Connecticut Department of Environmental Protection) groundwater protection."

No evidence of MTBE in heating oil samples was found—either because method limits of detection were higher than MTBE concentrations (which were 14 mg/L), or samples were not analyzed for MTBE—except for one sample where MTBE groundwater concentration was 4,100 µg/L.

The source of MTBE in heating oil has not been identified, but authors believe the contamination results from "the mixing of reformulated gasoline containing MTBE with heating oil during the bulk distribution of the fuel to intermediate storage facilities or during delivery to end users." They recommend the following:

- at sites where heating oil has contaminated groundwater, both groundwater and heating oil should be analyzed for MTBE; MTBE analyzers should strive to achieve low limits of detection
- possible causes of heating oil contamination with MTBE should be investigated and preventive measures taken
- consideration should also be given to MTBE contamination of diesel and other fuels

Caution Against Interpreting Gasoline Release Dates Based on BTEX Ratios in Ground Water

Alvarez, P.J.J., Heathcote, R.C. and S. Powers, *Ground Water Monitoring and Remediation*, Fall 1999, 800-332-2104; <http://www.ngwa.org>.

Authors of this article state that "Forensic chemical analysis and examination of spatial concentration trends of groundwater contaminants are bona fide components of liability assessment, but caution should be exercised to avoid stretching forensic inferences beyond scientific constraints."

Some investigators have ascertained that the age of a release "can be established in any situation by comparing aqueous concentration ratios such as benzene to xylenes (B/X) or benzene-plus-toluene to ethylbenzene-plus-xylenes ([B+T]/[E + X]). This provocative idea is based on the fact that benzene and toluene are more soluble than ethylbenzene and xylenes, and thus preferentially solubilize from the gasoline . . . However, inferring the age of a release based on BTEX ratios alone is a valid approach only in rare, well-constrained situations."

According to the authors, establishing empirical relationships between BTEX ratios and age of release requires knowing:

- the gasoline composition, which varies with octane rating, season, geographic area and period of manufacture
- volume of spill
- numerous physical, chemical and biological processes affecting the fate of the BTEX species (dispersion, sorption, volatilization and biodegradation)

In addition the gasoline weathering is affected by these variables:

- soil texture and soil composition
- microbial diversity; different sites and different zones within a given site are inhabited by different types and concentrations of microbial populations; furthermore, some phenotypes will prefer one substrate (for instance, benzene or toluene) over another, and therefore BTEX ratios will increase or decrease
- electron acceptor availability
- release size
- groundwater chemistry and hydrodynamic characteristics

Authors conclude that "The aquifer properties that control the rate and extent of different weathering mechanisms are site-specific. Therefore, one should not use the relative concentrations of individual BTEX compounds in groundwater to determine reliably the age of a petroleum product release. Such attempts bear a burden of proof that is often beyond the limits of scientific constraints and require far more data than can be obtained given common technical and economic constraints."

Natural Attenuation of BTEX and MTBE Under Complex Hydrogeological Conditions

Tyner, L., Grown, K., Daron, D., Perina, T., Daftary, D. and B. Sibbett, in *Natural Attenuation of Chlorinated and Recalcitrant Compounds*, 1999, Battelle Press, <http://www.battelle.org/bookstore>.

California researchers took BTEX and MTBE samples at a gasoline-impacted, shallow, perched groundwater site at Vandenberg Air Force Base. They used MODFLOW to model groundwater flow and MT3D to model contaminant fate and transport. Water uptake by eucalyptus trees and groundwater recharge from a nearby car wash complicated the modeling efforts.

Modeling results indicate risk to human health and environment was negligible; however, drinking water sources were not present 3 miles upgradient or downgradient of the site, and groundwater production wells were screened 200 to 400 ft deep. Researchers plan to study in more detail the effect of the eucalyptus trees on MTBE and BTEX fate. This will involve sampling of groundwater, tree roots, tree cores, xylem sap and transpired gases.

Biodegradation During Contaminant Transport in Porous Media. 2. The Influence of Physicochemical Factors

Brusseau, M.L., Hu, M.Q., Wang, J. M and R. M. Maier, *Environmental Science and Technology*, Vol. 33, No. 1, 1999; 202-872-4600; <http://www.pubs.acs.org>.

Researchers devised laboratory experiments to evaluate the impact of coupled physicochemical factors on biodegradation of dissolved contaminants during transport in porous media. Results from these experiments on residence time, substrate concentration, and electron-acceptor concentration should help remediators design better in-situ bioremediation systems.

"In systems wherein biodegradation is coupled with transport, the magnitude and rate of biodegradation are influenced not only by microbial properties but also by physicochemical properties." Researchers used petroleum-contaminated aquifer material (96% sand) and benzoate as a model compound. Benzoate was chosen as representative of aromatic hydrocarbons such as alkylbenzenes. Benzoate is nonvolatile (it is not expected to undergo much hydrolysis, thus mass loss by abiotic processes is unlikely), and unlikely to be sorbed by the aquifer material. Thus it should be available for biodegradation. The experiments used various influent benzoate concentrations and pore-water velocities. Results indicated that

- benzoate showed no retardation
- biodegradation was limited by insufficient oxygen
- type of transport behavior was dependent on both physicochemical and microbial properties
- contaminant transport was influenced by biomass growth, which occurs only under certain conditions
- residence time and oxygen availability constrained the magnitude and rate of benzoate biodegradation

Study of the 25-Year-Old Nipisi Oil Spill: Persistence of Oil Residues and Comparisons Between Surface and Subsurface Sediments

Wang, Z., Fingas, M., Blenkinsopp, S., Serby, G., Landriault, M., Sigouin, L. and P. Lambert, *Environmental Science and Technology*, Vol. 32, No. 15, 1998; 202-872-4600; <http://www.pubs.acs.org>.

Research on the degradation of an early 1970s oil spill in northern Alberta—one of the largest land spills—investigated

- the effectiveness of cleanup techniques
- long-term persistence of oil residues
- changes in oil character after 25 years of exposure to weathering and biodegradation
- relationship of residual oil degradation rate to depth at which samples were collected

Contaminated sediments fell into the following groups:

- subsurface samples, 0 to 4 cm, highly contaminated and highly weathered, showing degradation rates of 15-43%
- subsurface samples, 10 to 40 cm, highly contaminated and lightly to moderately weathered/degraded

- subsurface samples, 80 to 100 cm, mixtures of oil and vegetation hydrocarbons, lightly contaminated

Researchers found that "extent of contamination and degree of degradation correlated strongly with sample depth, and oil trends were similar across the site, even though different treatments had been used after the spill at the site." The article also describes in detail compositional changes of TPH and n-alkanes, and compositional changes of biomarker compounds.

Sorption of Volatile Organic Chemicals in Plant Surfaces

Welke, B., Ettlinger, K. and M. Riederer, *Environmental Science and Technology*, Vol. 32, No. 8, 1998; 202-872-4600; <http://www.pubs.acs.org>.

Researchers studied the partition coefficients (air/water, cuticular polymer matrix/water) of 50 VOCs including benzene, toluene, ethylbenzene and o-xylene. From the results of partition experiments, researchers concluded "The input of VOCs into the food chains of terrestrial ecosystems covered by vegetation may be estimated from analytical data on ambient concentrations and leaf surface/air partitioning parameters, determined experimentally or estimated according to equations [presented in the text]." For these experiments, researchers assumed:

- equilibrium partitioning
- estimates as only a first approximation of the real situation

"The important factors leading to deviations from equilibrium in air-to-cuticle partitioning are slow equilibration kinetics for highly lipophilic compounds as well as metabolism and translocation within the plant. The experimental results and predictive tools presented in this work may, nevertheless, be useful in assessing the tendency of various anthropogenic and biogenic organic volatiles to partition into the large lipophilic compartments making up the plant/atmosphere interface."

Population Heterogeneity Affects Transport of Bacteria Through Sand Columns at Low Flow Rates

Simoni, S.F., Harms, H., Bosma, T.N.P. and A.J.B. Zehnder, *Environmental Science and Technology*, Vol. 32, No. 14, 1998; 202-872-4600; <http://www.pubs.acs.org>.

Using flow-through sand columns, researchers found that differences in LPS (lipopolysaccharide) coating of bacteria were responsible for fractionation of the bacteria as the bacteria traveled through the sand. "We percolated dilute suspensions of *Pseudomonas* sp. Strain B13 through columns filled with purified sand at flow rates common in natural aquifers. The results are discussed in terms of a simple model accounting for two fractions with different adhesive properties. Characterization of the cell surface allowed us to develop a mechanistic interpretation based on energy iteration curves derived from the DLVO theory of colloid stability. . . . Our results indicate that the high travel distances of microorganisms might be due not only to the complex structure of aquifer material but also to heterogeneity in the adhesion properties within the bacterial populations."

"Apart from hydrodynamic heterogeneity in aquifers, intra-population variability may result in surprisingly high microbial travel distances in the subsurface. Additionally, extrapolations are uncertain since a change in flow parameters might lead to a different deposition regime. . . . the colloid filtration theory as well as the DLVO theory of colloid stability provide valuable frameworks for assessing transport of bacteria in aquifers, but interpretations should be subject to careful verification."

Genetically Altered Algae Remove Heavy Metals from Water, Sediment

K. Canning, *Pollution Engineering*, July 1999; 303-470-4280; cahnners.subs@denver.cahnners.com.

This article describes the work of Dr. Richard Sayre, researcher at Ohio State University, who genetically altered the *Chlamydomonas reinhardtii* algae to enable it to take up heavy metals. "The cells are incubated with the foreign DNA into the algal cells and shaken very hard in the presence of some glass beads. . . the glass beads break small openings in the cells that allow the DNA to get in. Then the DNA will integrate into the nuclear DNA and be stably maintained in the nuclear DNA."

The introduced protein, metallothionein, is found in most living organisms. It increases the algae's metal binding capacity almost twofold. The transformed organisms are more resistant to toxic concentrations of heavy metals. Removal time is site-specific and depends on the metal mobility and the matrix to which metals are attached. In a stirred solution, removal takes minutes, but Sayre states that by acidifying the environment, metals would become more soluble and easily recoverable. Sayre has also found that the algae will sequester a number of organic pollutants. Plans are to place the altered algae inside wells. For more information, see <http://www.biosci.ohio-state.edu/~rsayre/BIOREM.HTM>.



Information sources

Recent publications

A Mathematical Primer on Groundwater Flow, 1999, John Hermance, Prentice Hall, <http://www.prenhall.com>.

Characteristics of Dissolved Hydrocarbon Plumes. An API publication based on studies of more than 600 groundwater-contaminated sites. See the Groundwater Services, Inc. web page, <http://www.gsi-net.com/pubs/Docindex.htm> or the American Petroleum Institute page, <http://www.api.org/ehs/sgresbul.htm>.

Interim Final Report: Development of Health-Based Alternative to the Total Petroleum Hydrocarbon (TPH) Parameter, Massachusetts Department of Environmental Protection website, http://state.ma.us/dep/bwsc/vph_eph.htm.

EPA publications and information

Most EPA publications can be viewed or downloaded at <http://clu-in.org.techpubs.htm>, or ordered as hard copies by calling 800-490-9198 or 513-489-8190, or faxing 513-489-8695:

- *Bibliography for Innovative Site Clean-up Technologies*, August 1999 Update (EPA 542-B-99-004). References for more than 300 documents, databases and websites related to field analytical and remediation technologies.
- *Brownfields Technical Guides*, designed to facilitate assessment and cleanup of brownfield sites. A cost-estimating resource document with overviews of site characterization, a matrix listing of contaminants and media, site characterization tools and technologies for sample collecting.
- *Field Applications of In-Situ Remediation Technologies: Permeable Reactive Barriers* (EPA-542-R-99-002). A status report on the use of PRBs.
- *Groundwater Cleanup: Overview of Operating Experience at 28 Sites* (EPA 542-R-99-006). Contains 28 case studies.
- *Ground Water Currents* (EPA 542-N-99-004), a quarterly update. Descriptions and performance data for developments in innovative groundwater treatment.
- *Innovations in Site Characterization: Interim Guide to Preparing Case Studies* (EPA 542-B-98-009). Guidelines on collecting and documenting cost and performance data.
- *Phytoremediation Resource Guide* (EPA 542-B-99-003). Field studies, demonstrations, research articles and Internet sources.
- *RealVideo Presentation: In-Situ Alcohol Flushing for Remediation of NAPL Source Zones*. A 28-minute video on the goals and results of tests using in-situ alcohol flushing for extracting NAPLs. RealPlayer G2E, required to view the video, is available from RealNetworks at <http://www.real.com/products/player/downloadrealplayer.htm>.

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Expedited Site Assessment Tools for Underground Storage Tanks: A Guide for Regulators (EPA 510-B-97-001). Information on geophysical methods, soil-gas surveys, direct push technologies and on-site analysis methods. For hard copies (\$26.00), call 202-512-1800 or view at <http://www.epa.gov/swerust1/pubs/index.htm#sam>.

EPA's Water Quality Report Page. Information on usage and pollution of lakes, rivers, wetlands and estuaries in each state; view at <http://www.epa.gov/305b>.

Free-product Recovery of Petroleum Hydrocarbon Liquids. Basic physical/chemical parameters, recovery principles and remediation technologies. Download at <http://www.api.org/ehs/fpr/4682rh.htm> or call American Petroleum Institute, 202-682-8375.

Innovative Technology Summary Report: Direct Sampling Ion Trap Mass Spectrometry (DSITMS; DOE/EM-0421). Cost, performance and other key characteristics of direct sampling ion trap mass spectrometry. View or download at <http://ost.em.doe.gov/ifd/cmst/itsrs/dsitms2/dsitms2.pdf>.

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Nitrate-Based Bioremediation Homepage, developed by EPA's Robert S. Kerr Environmental Research Center. Information on nitrate use as an alternate electron acceptor in fuel-contaminated aquifers; a list of websites with summaries of laboratory and field work. See <http://www.epa.ada/bioremed.html>.

Oil Spill Program Update, produced by the EPA Office of Emergency and Remedial Response. Proposed rules to amend the Federal Response plan, DOT breakout tank regulations and Y2K information. View/download at <http://www.epa.gov/oilspill/docs/epaupd8.pdf>; hard copies, or e-mail oliveria.beatriz@epa.gov.

Reliability-based Uncertainty Analysis of Groundwater Contaminant Transport and Remediation (EPA 600-R-99-028). A probabilistic modeling tool to account for first- and second-order reliability methods. Download at <ftp://ftp.epa.gov/pub/ada/reports/reliability.pdf>. or contact Kay Cooper for copies, cooper.kay@epa.gov, or call 580-436-8651.

Using Field Methods: Experiences and Lessons: Defensibility of Field Data, 1994. View at <http://clu-in.org/techpubs.htm>.

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