

# Underground Tank Technology Update

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## Greenhouse gases, vehicle emissions and innovative vehicles

Greenhouse gases are emissions released by human activity into the atmosphere, insulating the earth and causing the earth's temperature to rise. The release of greenhouse gases has increased dramatically since the Industrial Revolution. According to Revkin (2000), "So far the insulating effect of these gases is the equivalent of turning on two one-watt Christmas tree light bulbs over each square yard of the earth's surface." In comparison, scientists calculated that the "extra wattage accounting for the difference between the last ice age and current equable conditions is 6 to 9 watts per square yard. If greenhouse gas concentrations continue to rise, the human-caused increase in energy bathing the surface could exceed four watts per square yard, a rise that begins to approach the change from the last ice age until now."

The greenhouse gases that contribute to the greenhouse effect of warming the earth include, in order of volume output to the atmosphere:

- CO<sub>2</sub>, carbon dioxide, 72.7%
- CH<sub>4</sub>, methane, 16.6%
- N<sub>2</sub>O, nitrous oxide, 7.6%
- CFCs, HFCs and PFCs, 2.7%
- SF<sub>6</sub>, sulfur hexafluoride, 0.4%
- black carbon soot, 10 million tons/yr

(Revkin, 2000)

Black carbon soot—microscopic soot particles released from inefficient diesel engines, unfiltered power plants or burning forests—is not a gas. However, soot does absorb heat and forms seeds for cloud droplets that darken clouds and reduce cloud tendency to reflect sunlight.

Some sources of greenhouse gases are:

CO<sub>2</sub> — heating and cooling, transportation, oil and gas production and industries such as petroleum refineries

CH<sub>4</sub> — natural gas and oil production, coal mining, waste decomposition in landfills, digestive processes of livestock, manure decomposition and rice cultivation

N<sub>2</sub>O — nitrogen fertilizers, nitrogen-fixing crops, livestock manure, waste management and runoff, emissions from fossil fuel use and chemical manufacturing

CFCs, HFCs, PFCs — aerosol cans, packaging materials, refrigerants, emissions from the manufacture of semiconductors and from aluminum smelting

SF<sub>6</sub> — electrical equipment insulation, magnesium production and medical treatments like asthma inhalers

**Black carbon soot** — incomplete combustion of fossil fuels and vegetation fires

Another way to analyze greenhouse gases is to compare the global warming potential, G.W.P., a measure of the relative radiative effects of various greenhouse gases. G.W.P. values, in terms of the ratio of global warming from one unit mass of a gas to that of one unit mass of carbon dioxide over 100 years, is as follows:

- carbon dioxide, 1
- methane, 21
- CFC-11, 3,800
- sulfur hexafluoride, 23,900

Scientists don't agree about which gases the world should focus on reducing: the more common and voluminous, or the more powerful? According to Revkin (2000), "The argument for attacking the less common greenhouse ingredients is mainly based on the idea that these substances contribute to harmful air pollution as well as climate change, and so an easier case can be made to policy makers and the public to clean them up." Reducing carbon dioxide output is difficult because carbon dioxide is a basic product of coal and oil burning and thus an intrinsic part of modern life. Soot and other greenhouse gases, scientists argue, "constitute an extraordinarily complex brew whose influence on climate and interactions with each other and with clouds and other atmospheric ingredients are still poorly understood." Scientists maintain that interactions between greenhouse gases and other compounds are almost impossibly complex.

For example, the extremely reactive hydroxyl radical, OH, created as ozone (O<sub>3</sub>), destroys methane molecules. Thus, one greenhouse gas "eats" the other. Future trends for ozone and methane suggest that the two would largely cancel each other out. "Ozone is continually formed and destroyed when sunlight hits air, particularly when that air is laden with nitrogen oxides, hydrocarbons or other emissions from burning fuels or forests" (Revkin, 2000). This poisonous ozone is also known as smog. Another ozone is formed high in the stratosphere, where it shields earth from radiation and traps heat.

### Vehicle emissions

The transportation sector generates an estimated one-fourth of the total U.S. greenhouse gas emissions (GHG). In addition, vehicle emissions contribute to major health problems, costing the health industry approximately \$20 to \$50 billion annually. Even small amounts of toxic substances emitted by vehicles can be a concern since millions of vehicles emit these small amounts (Maclean and Lave, 2000).

### Some background on the internal combustion engine (ICE)

The internal combustion engine (ICE), one source of GHG, powers more than 90 percent of the automobiles worldwide. Although non-ICE alternatives such as battery-powered vehicles and hybrids exist, and fuel-cell vehicles are being developed, these vehicles cannot yet compete economically with ICE vehicles.

Only 20 to 25 percent of the energy from gasoline actually propels a car, but at least one organization wants to increase fuel economy to 80 mpg—requiring engine efficiency to reach 40 percent thermal efficiency. “At present, efficiency generates more exhaust pollutants, especially nitrogen oxides (NO<sub>x</sub>). For example, a DI (direct injection) diesel vehicle has the highest efficiency of ICE options but produces relatively high NO<sub>x</sub> emissions and particulate matter (PM).”

Alternative fuels have the advantage of emitting fewer toxins to the air. Such fuels are used by only about 1 percent of the world’s motor vehicles. Furthermore, vehicles that use alternative fuels are not optimized for either fuel economy or efficient emission control.

### Alternative-fueled automobiles

Using alternative-fueled automobiles has three goals:

- to increase fuel economy by decreasing GHG/pollutant emissions
- to satisfy regulations
- to have consumers voluntarily purchase vehicles

Maclean and Lave (2000) have identified “two alternative engines not currently in large-scale use that have the potential for significant market entry in a 15-20-year time frame:”

- direct injection stratified charge gasoline engine, equivalent to the spark ignition DI (SIDI) gas engine
- direct injection diesel engine, or compression ignition DI (CIDI) diesel

Direct injection engines are more efficient than indirect ones. “However, operating at maximum efficiency (and therefore maximum fuel economy) does not result in low vehicle emissions. To achieve their maximum efficiencies, these engines must operate lean. In contrast, conventional catalyst technology is most efficient at stoichiometric conditions. Thus, emissions control results in lower efficiency (and resulting CO<sub>2</sub> cost). In addition, lean NO<sub>x</sub> catalyst technologies for emissions control with DI engines require low sulfur fuel, and more complex emissions technologies require energy, reducing the operational efficiency.”

### Greenhouse gases in vehicle emissions

Greenhouse gases that make up vehicle emissions include CO<sub>2</sub>, methane, nitrous oxide, and ozone. Methane and nitrous oxides produced from low-emission gasoline and diesel vehicles are small. Compressed natural gas (CNG) vehicles emit about 10 times the amount of methane as gasoline vehicles, but the quantity is small compared to CO<sub>2</sub>. Researchers in this study focused on CO<sub>2</sub> emissions.

### Vehicle emission conditions

Three general driving conditions lead to vehicle emissions:

- on-cycle driving, conditions covered in the Federal Test Procedure (FTP)
- off-cycle driving, conditions other than those in the FTP, e.g., high speed, high load; approximately 15 percent of vehicle driving time is in the off-cycle mode
- driving with malfunctioning emissions control systems

The largest source of emissions is from 5 to 10 percent of primarily older vehicles with malfunctioning control systems. Emissions from such vehicles are 10 to 1,000 times greater than the overall average emissions.

Researchers created a simplified method for calculating exhaust emissions based on California’s certification standards for baseline vehicles and came to these conclusions:

- hydrocarbon emission rates from off-cycle driving exceeded rates from the FTP (Federal Test Procedure) for less than half of the fleet/fuel combinations
- the carbon monoxide emission rate is two to five times higher during off-cycle driving
- NO<sub>x</sub> emissions are about 40 percent higher during the off-cycle than the FTP across all fuel/fleet combinations

Researchers also compared, for 18 vehicle types including gasoline indirect injection, diesel to methanol 85 and compressed natural gas (indirect and direct injection):

- vehicle operational energy in terms of maximum and minimum efficiency
- carbon dioxide emissions during vehicle lifetime

The diesel and direct injection vehicles showed the lowest operational energy; compressed natural gas vehicles have heavy fuel storage systems, which burden efficiency. The largest amount of CO<sub>2</sub> emitted was from a California Phase 2 reformulated gasoline spark-ignition indirect injection vehicle; the lowest was from a spark-ignition direct injection compressed natural gas vehicle.

Researchers also found that vehicles designed for extremely low emissions are more sensitive to sulfur than other vehicles and that “using high sulfur fuel will increase emissions immediately; the higher emissions might continue even after low sulfur gasoline is used due to poisoned (possibly permanently) catalysts. CNG vehicles are less dependent on specific fuel properties in meeting low-emissions standards than are gasoline or diesel. In addition, key issues for the future are the fraction of low-emission vehicles that become high emitters (and the resulting emissions). These issues will have significant impacts on air quality. Experts suggest that the increasing redundancy in emissions control systems and other improvements in engine and emissions control are likely to lead to improvements. However, in the absence of stringent inspection and maintenance programs and effective onboard diagnostics, these gains may be lost.”

Maclean and Lave (2000) state that “Lifetime GHG emissions resulting from the operation of any vehicle fueled by a fossil fuel are significant. Light-duty vehicles account for 18 percent of the U.S. total CO<sub>2</sub> emissions. This implies the importance of increasing efficiency, using smaller, more fuel-efficient vehicles and renewable energy sources. The GHG differences among the options have the potential to be significant. If the entire light-duty fleet instantaneously was replaced by vehicles whose operation resulted in an average 40 percent lower CO<sub>2</sub>, the GHG reduction would be close to the level required to satisfy the Kyoto requirement. In the near-term, efficiency improvement can lower resource use and GHG emissions; however, it is apparent that improving efficiency of vehicles

fueled by nonrenewable energy sources will not achieve the reductions likely to be required to satisfy Kyoto and will not lead to sustainability. Further reductions will be necessary through the use of renewable energy sources."

The authors concluded:

- vehicles fueled by CNG have the potential to be a significant benefit with respect to toxic reduction
- although the primary focus of the ULEV (ultra-low emission vehicle) standards is on lowering regulated exhaust emissions and not reductions in the toxic air pollutants, the emissions control systems, engine controls, and fuel formulations that will be required to meet the standards are likely to have a favorable impact on toxic emissions
- malfunctioning emissions control systems can offset this benefit (above)
- focusing on GHG and resource use, vehicles fueled with renewable energy sources such as alcohols and diesel from biomass result in no net CO<sub>2</sub> emission
- in the absence of renewables, diesel and SIDI (spark-ignited direct injection) vehicles are highly efficient
- with respect to air quality, CGN and low-emission vehicles are preferred
- CGN-fueled vehicles are favorable with respect to GHG, exhaust and evaporative emissions of regulated and toxic pollutants
- considerable CGN storage technology development is necessary for cars with ranges equivalent to those of gasoline-fueled vehicles
- a conventional reformulated gasoline-fueled ULEV, properly maintained and not driven excessively, is presently the best option in terms of environmental and human health

For more information, see the following articles:

- "Emissions from Two Outboard Engines Operating on Reformulated Gasoline Containing MTBE," Gabele, P.A. and S.M. Pyle, *Environmental Science & Technology*, Vol. 34, No. 3, 2000.
- "Effects of a Biodiesel, Biodiesel Blends, and a Synthetic Diesel on Emissions from Light Heavy-Duty Diesel Vehicles," Durbin, T.D., Collins, J.R., Norbeck, J.M. and M.R. Smith, *Environmental Science & Technology*, Vol. 34, No. 3, 2000.

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Maclean, H.L. and L. B. Lave, "Environmental Implications of Alternative-Fueled Automobiles: Air Quality and Greenhouse Gas Tradeoffs," *Environmental Science & Technology*, Vol. 34, No. 2, 2000; <http://www.pubs.acs.org>.

Revkin, A.C., "Fossil Fuels Will Be Hard to Give Up, and Some Scientists Say There May Be Another Way," *The New York Times Science Times*, Tuesday, October 3, 2000.



## Detailed geochemical study of hydrocarbon bioremediation

Researchers studied an active natural gas production site contaminated with gas condensate hydrocarbons, including BTEX, to determine if intrinsic bioremediation was occurring. To achieve this, they collected data and assessed multiple lines of evidence (*Gieg and others, 1999*).

### Groundwater monitoring

Five groundwater monitoring wells were installed in a contaminated area downgradient of the natural gas source while five wells were drilled in an upgradient background zone. Well locations were based on vadose zone measurements of O<sub>2</sub>, CO<sub>2</sub>, and volatile organic compounds (VOCs).

### Sediment/vadose sampling, chemical analyses and microbiological enumerations

Field workers took sediment cores annually (from 1993 to 1997), using hollow-stem augers with split-spoon samplers. Samples from the background areas and contaminated areas were analyzed for the following:

- BTEX
- sulfate
- nitrate
- Fe (II)
- Fe (III)
- pH
- moisture content
- porosity
- bulk density
- microbial populations and numbers

Researchers sampled the vadose zone for

- O<sub>2</sub>
- CO<sub>2</sub>
- volatile organic compounds

In addition, researchers searched for putative hydrocarbon metabolites from groundwater samples. For details on the chemical analyses, see the authors' text.

Researchers also created aquifer slurries, incubations in the lab to determine the "susceptibility of BTEX compounds to anaerobic biodegradation under sulfate-reducing and methanogenic conditions . . . Three viable and two autoclaved control incubations were prepared for each of the six BTEX components under each condition." Researchers monitored BTEX and methane concentrations using a GC-FID; sulfate concentrations were measured by ion chromatography.

## Groundwater chemistry

By multiplying BTEX migration velocity (groundwater flow rate/retardation factor for xylene, neglecting the effects of longitudinal dispersion) with time elapsed since release, researchers were able to estimate plume migration. They found the BTEX plume center was almost stationary for 4 years at about 18 to 20 m from the contaminant source. After major rainfall events increased water table elevation, the plume center did move as far as 39 m from the source but receded as water table elevation decreased. Because researchers found BTEX compounds only in shallow wells of the contaminated area, deep well data were not presented in this study.

In the areas of contamination:

- groundwater was black and had a strong hydrogen sulfide odor
- dissolved sulfide ranged from 1 to >10 mg/L (sulfide was not detected in background wells)
- alkalinity, as determined by CaCO<sub>3</sub>, averaged 500 mg/L, nearly double that of background wells
- dissolved O<sub>2</sub> was substantially depleted (<0.5 mg/L) compared to uncontaminated wells (1 to 4 mg/L)
- nitrate concentrations were lower in the contaminated region (undetectable to <5 mg/L) as compared to background (undetectable to 12 mg/L)
- dissolved methane ranged from 5 to 17 mg/L
- dissolved H<sub>2</sub> concentration
  - varied spatially and seasonally; lower concentrations indicated more oxidizing environments while higher values indicated reducing environments
  - 0.2-0.8 nM (nanomolar) correlated with iron reduction
  - 1-4 nM correlated with sulfate reduction
  - 5-25 nM correlated with methanogenesis
  - limits of detection were about 0.05 nM

From this data, researchers concluded that groundwater with H<sub>2</sub> concentrations near 1 or 2 nM (which occurred between March 1997 and 1998) were characterized by iron or sulfate reduction. Background wells that showed less than 0.05 nM H<sub>2</sub> indicated more oxidizing environments. Some wells did shift to a more reduced process (methanogenesis).

Researchers also concluded that while dissolved methane (5 to 17 mg/L) was always detected in the contaminated wells, it did not always correlate with H<sub>2</sub> concentrations. "This lack of correlation was also true for other geochemical parameters measured. Sulfate remained depleted as compared to the background zone even when H<sub>2</sub> measurements suggested that methanogenesis or iron reduction predominated, and Fe (II) was always detected in the contaminated wells, regardless of the predominant anaerobic process suggested by H<sub>2</sub> measurements" (*Gieg and others, 1999*).

Researchers also found several putative bacterial metabolites of gas condensate hydrocarbons in contaminated groundwater that were not detected in the uncontaminated groundwater or gas condensate. The metabolites included aromatic alcohols, aldehydes and acids such as 3-methylbenzylsuccinic

acid (dimethyl ester), methylbenzaldehydes, benzoic acid, toluic acid and tolylacetic acid isomers, dimethylbenzoic acids, hexadecanoic and octadecanoic acids. These were identified using GC retention times and spectral profiles or mass spectral match alone. Gieg and others (*1999*) state: "These metabolites were present in the groundwater at all sampling events, but there was no discernable pattern as to their location within the contaminated portions of the aquifer."

## Contaminated sediments

Field workers characterized the vadose zone soils as follows:

- BTEX concentrations were highest in the sediment samples removed from just below the water table in the contaminated area
- BTEX compounds were not detected below a depth of 3 m in the contaminated area
- BTEX concentrations dropped from >1,000 to <100 mg/kg in the contaminated area
- sediments were black and had lower sulfate concentrations (3–15 mg/kg); those in the background region had about 40 mg/kg sulfate
  - sediments in background wells had 220–550 mg/kg Fe (III) in a microbially reducible form
  - sediments from the contaminated area were depleted in bioavailable Fe (III); but Fe (II) concentrations ranged from 450 to 1,000 mg/kg
  - background sediments had no measurable Fe (II)
- O<sub>2</sub> concentrations were greater in background (20 to 20.5% v/v) than in contaminated zones (0.5% v/v)
- CO<sub>2</sub> concentration in the background zone was 0.05 to 1.2% v/v, and 8.5% v/v in the contaminated area

## Laboratory confirmation

Gieg and others (*1999*) reported that "microbial enumerations performed on background and contaminated sediments were consistent with the geochemical measurements of depleted sulfate and increased sulfide and methane in the contaminated area as compared to the background site." The numbers of types of heterotrophic microorganisms derived from contaminated and uncontaminated sites were the same; however, the most numerable cultivable species differed in type.

Laboratory studies—slurries amended with benzene and sulfate—confirmed that benzene oxidation was coupled to sulfate reduction, as were ethylbenzene and the xylenes. Toluene consumption was coupled to methane production as indicated by methanogenic incubations.

## Analyses

Gieg and others (*1999*) believe that the rainfall events that caused the plume to migrate temporarily were likely "due to solubilization of residual hydrocarbons in the vadose zone that were subsequently transported downgradient as the elevated water table dropped back to its original elevation. After sufficient time, the center of the BTEX plume receded to its original position, presumably due to attenuating

factors. Although BTEX concentrations in groundwater remained relatively steady, sediment-associated BTEX concentrations decreased over time in all locations sampled, indicating a reduction in the total mass of these hydrocarbons. On the basis of these results, however, one cannot conclude that biological processes alone contributed to this measured attenuation. Abiotic processes such as leaching, dilution, or volatilization could also explain the observed loss."

If microbiological processes were operative, researchers would expect to see "substantial changes in the geochemistry and microbiology of the contaminated area relative to the reference area." Gieg and others (1999) did find the following in contaminated zones relative to uncontaminated zones:

- decreased O<sub>2</sub> concentrations, increased CO<sub>2</sub> concentrations and the presence of VOCs in the vadose zone of the contaminated region
- depleted sulfate and dissolved O<sub>2</sub> in BTEX-contaminated water
- detection of sulfide, Fe (II) and methane in contaminated water
- absence of bioavailable Fe (III) and sulfate depleted (in the contaminant zone) relative to Fe (III) and sulfate-rich background sediments
- higher populations of sulfate-reducing bacteria and methanogens in the contaminated area

Researchers suggest these findings indicate that "both aerobic and anaerobic microbial populations were stimulated in response to hydrocarbon contamination. However, even in light of reduced BTEX concentrations in the sediments, these measurements did not firmly attest to the ongoing activity of subsurface microorganisms. . . ."

By measuring dissolved H<sub>2</sub> concentration in groundwater, researchers determined predominant electron-accepting processes. A dynamic molecule in anaerobic environments, H<sub>2</sub> can indicate iron-reducing, sulfate-reducing and methanogenic processes and thus demonstrate active anaerobic subsurface microbial populations. At the site, H<sub>2</sub> concentrations suggested that sulfate reduction was most active, although H<sub>2</sub> concentrations varied spatially and seasonally, sometimes markedly. Researchers observed that "mixed geochemical signatures associated with anaerobic processes had no discernable pattern: depleted sulfate and enhanced amounts of sulfide, Fe (II), and methane were measured in virtually every well within the contaminated plume at every sampling event despite the predominant electron-accepting process suggested by H<sub>2</sub> measurements." This lack of correlation "may be due to the measurement of a soluble geochemical indicator such as methane, which originated from an upgradient zone where it was originally produced." Or the lack of correlation "may be due to different anaerobic processes occurring simultaneously in heterogeneous microenvironments. Groundwater extraction from variable, closely spaced redox zones can result in the volumetric averaging of H<sub>2</sub> concentrations, resulting in misleading interpretations of the dominant electron-accepting process" (Gieg and others, 1999).

However, researchers maintained that dynamic processes were operative in the contaminated area not only because of shifting H<sub>2</sub> values (denoting sulfate reduction and methanogenesis), but also due to microbiological evidence of anaerobic microbial populations. Researchers did ascertain that active microbial populations were biodegrading BTEX hydrocarbons: "Laboratory incubations using sediments from the site showed that all six aromatic hydrocarbons were biodegraded under sulfate-reducing conditions and that toluene was degraded under methanogenic conditions" (Gieg and others, 1999). Benzene degraded also after a 115-day lag time.

Laboratory results, interpreted in light of finding putative BTEX metabolites in contaminated groundwater, pointed strongly to in-situ biodegradation of BTEX: this appearance of signature microbial metabolites affirmed biological rather than abiotic contaminant loss.

The putative metabolites identified (the alcohols, aldehydes and aromatic acids) could have resulted from aerobic or anaerobic processes. Researchers theorized that the shallow water table here allowed O<sub>2</sub> infiltration from rainwater, and this O<sub>2</sub> may have generated partially oxidized intermediates amenable to anaerobic degradation. In addition, researchers found benzylsuccinic and methylbenzylsuccinic acids, signature acids formed during anaerobic decay of toluene and the xylenes. They also detected other intermediates found in similar BTEX-contaminated aquifers that may serve as indicators of in-situ biodegradation.

## Conclusions

Gieg and others (1999) were able to document substantial changes in geochemical and microbial parameters in uncontaminated and contaminated portions of an aquifer. From their data, they concluded:

- BTEX concentrations decreased
- substantial geochemical changes were evident
- numbers of anaerobic microorganisms increased
- hydrogen concentrations indicated that anaerobic populations were active
- anaerobic populations had the potential to biodegrade all BTEX components
- putative microbial metabolites were detected in the contaminated portion of the aquifer

By invoking multiple lines of evidence, researchers were able to conclude that intrinsic bioremediation was occurring, even under anaerobic conditions.

## Reference

Gieg, L.M., Kolhatkar, R.V., McInerney, M.J., Tanner, R.S., Harris, S.H., Sublette, K.L. and J.M. Suflita, "Intrinsic Bioremediation of Petroleum Hydrocarbons in a Gas Condensate-Contaminated Aquifer," *Environmental Science and Technology*, Vol. 33, No. 15, 1999; <http://www.pubs.acs.org>.

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## Success of pump-and-treat in heterogeneous geologies

Large and small heterogeneities can significantly impact a pump-and-treat effort and consequently cleanup success. The optimal operation of a remedial scheme depends on site-specific factors when sites are lithologically, stratigraphically and structurally complex (Lee and others, 2000).

Remediators in some instances have ignored measuring and characterizing heterogeneities because obtaining adequate data can be difficult. Pump-and-treat and bioremediation operations, however, can be severely hampered by a lack of knowledge. As media become more heterogeneous, for instance, longer time periods are required for aquifer decontamination. "Recent numerical modeling studies of pump-and-treat remediation have illustrated the complicated capture zones that result from aquifer heterogeneity." Numerical simulations have shown that geologic heterogeneity influences the efficiency of oxygen-demanding (aerobic) bioremediation, thus comprehensive characterization of aquifer heterogeneity is important to the success of in-situ bioremediation.

### Modeling using TBM

Lee and others (2000) used the Turning Bands Method (TBM) to generate spatially correlated two-dimensional permeability fields. TBM simulations of random permeability use a spectral method—a technique easily "integrated with various one-dimensional covariance functions such as the Gaussian, exponential, Bessel and Tellis methods. A weighted sum of the corresponding values of the line processes is assigned to each point of the 2D problem domain. The TBM model can be easily generalized to produce anisotropic fields."

Researchers fed this data—scenarios of permeability heterogeneities—into the BIOPLUME® II model which simulates plume transport and degradation and "predicts the transport of dissolved hydrocarbon, oxygen and microbes by combining various physical, chemical and biological processes including advection, hydrodynamic disruption, pumping/injection and first-order decay." The model can be used to simulate:

- pump-and-treat remediation by groundwater extraction
- in-situ bioremediation operations by oxygen-charged water injection

*Editor's note:* Modeling NAPL degradation and transport requires variable-density and multiple phase codes, which Lee and others (2000) did not consider in this study.

### Geologic complexity

It is common knowledge that stratification and interbedding can produce significant variation in permeability. Low-permeability areas can trap contaminants for long periods of time. "Studies have shown that in-situ remediation is usually limited to aquifers with hydraulic conductivity of at least  $10^{-4}$  (~ 0.1 darcy or  $3.2 \times 10^{-6}$  ft/s), which represents the low end of the

range of permeability measurements in silty sand ( $10^{-5}$  to  $10^{-1}$  cm/s). For this reason, if local geology is poorly understood, the location and spreading of a plume are expected to be unpredictable" (Lee and others, 2000).

Researchers performed a series of simulations and showed that a two-layer system with clay results in less than a 60 percent removal of contaminants over 10 years; in contrast, researchers projected that complete cleanup of a homogeneous aquifer would take 7 years. Furthermore, if in-situ bioremediation is to succeed, injected materials such as electron acceptors and nutrients must have access to the contaminants. Modeling using TBM and BIOPLUME® showed that plume degradation varied with applied conductivity field—uniform vs. random. Researchers modeled plumes using a log conductivity variance of 0.1 and a variance of 1. In this scenario, oxygen and water were injected into a recharge well while contaminated water was withdrawn from a pumping well. The resulting flow velocities produced highly irregular plume evolution in heterogeneous aquifers because of flow velocity variations over short distances.

According to Lee and others (2000), "After about 3 years, the plume has split into two lobes, with the larger one being retained in low-permeability zones. The larger plume has been displaced to the left by almost 25 ft after 2 years of simulation. The smaller plume has migrated ahead of the larger one because it has moved through a more permeable zone and has experienced a fast mass loss. The result demonstrates the significant effect of heterogeneity structure on plume migration and position."

### Conclusions

"The presence of continuous, low-permeability strata could have the most adverse impact upon the success of the pumping treatment. Our results also show that the stochastic prediction is very sensitive to the degree of heterogeneity. The irregular plume transport and biodegradation patterns predicted by the transport and the biodegradation patterns predicted by the stochastic model differ markedly from the somewhat regular pattern shown in a 'homogeneous' aquifer. The rate of mass loss and the position and migration pathways of the plume are highly influenced by the heterogeneity feature of the host geologic materials. The preferential flow paths in highly heterogeneous media result in rapid bioremediation at the early stage, but tailing of contaminants continues for a long period of time. These results argue that detailed site characterization should be well carried out to allow the reliable prediction of the effectiveness of field remediations . . . [Our] generic model also provides a check on whether a specific site is suitable for a proposed remediation operation. Our modeling results indicate that pump-and-treat and bioremediation will not be successful in highly heterogeneous media" (Lee and others, 2000).

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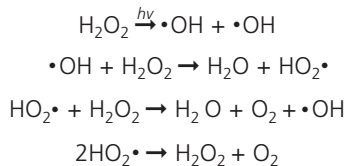
Lee, M.K., Sanders, J.A. and L.W. Wolf, "Effects of Geologic Heterogeneities on Pump-and-Treat and In-Situ Bioremediation: a Stochastic Analysis," *Environmental Engineering Science*, Vol. 17, No. 3, 2000; <http://www.libertpub.com>.



## MTBE treatment by UV/H<sub>2</sub>O<sub>2</sub>

A promising, advanced oxidation technology for treating MTBE-contaminated water uses UV light with an oxidizer such as H<sub>2</sub>O<sub>2</sub>, or ozone (O<sub>3</sub>), to generate hydroxyl radicals (•OH). This process is used worldwide for cleaning ground-water and drinking water supplies. This process is also superior to ozone-based processes because it does not form bromate ion, a suspected carcinogen (*Cater and others, 2000*).

In the UV/H<sub>2</sub>O<sub>2</sub> process, hydrogen peroxide is photolyzed (ultraviolet light cleaves the oxygen-to-oxygen bond), which generates hydroxyl radicals (•OH) that oxidize and mineralize most organic pollutants. "The hydroxyl radicals attack almost non-selectively any organic compound with very high reaction rate constants. The radical can be scavenged by an organic compound to oxidize the organic, recombine with other hydroxyl species to reform hydrogen peroxide or initiate a radical chain degradation of hydrogen peroxide in the series of reactions shown below (*Chang and Young, 2000*):



"Photolysis of hydrogen peroxide may be affected by suspended particles and other absorbing species . . . the presence of other radical-reactive species in a water sample such as carbonate, bicarbonate, humic substances or phosphate ions will affect degradation rate of organic contaminants by consuming hydroxyl radicals. Inorganic compounds present in water may also precipitate following UV exposure, coating lamp tubes and affecting the amount of UV light that is available for water treatment" (*Chang and Young, 2000*).

### Experimental conditions and kinetic model

By studying concentrations of the interacting components in the UV/H<sub>2</sub>O<sub>2</sub> process, *Cater and others (2000)* explored MTBE treatment efficiency. Researchers used Toronto municipal drinking water and MTBE obtained from a commercial establishment. The contaminated water was treated in a standard Rayox 1 kW batch reactor. Light source was a 1 kW medium-pressure Hg UV lamp. (For more description, see the text, pages 559 to 560.) Researchers used a gas chromatograph to measure MTBE concentration and devised a kinetic model based on initial destruction rates that estimated the pollutant decay. This kinetic model is described in the text.

### MTBE destruction

MTBE destruction is influenced by initial concentrations of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) and MTBE. MTBE decay, even with various initial concentrations, follows first-order reaction

kinetics with a pseudo-first-order rate constant. Researchers, therefore, analyzed the data in terms of the electrical energy per order (E<sub>EO</sub>) figure-of-merit. E<sub>EO</sub> is defined by the Photochemistry Commission of the International Union of Pure and Applied Chemistry as the number of kilowatt hours of electrical energy required to reduce the concentration of a pollutant by 1 order of magnitude (90%) in 1 m<sup>3</sup> of contaminated water. The E<sub>EO</sub> (kWh/m<sup>3</sup>/order) can be calculated from the following equations for a batch type reactor:

$$E_{EO} = P \times t \times 1,000 / V \times 60 \times \log (C_i/C_f)$$

$$E_{EO} = 38.4 \times P / V \times k_1'$$

P is power input (kW) from the wall to drive UV lamp(s)

t is irradiation time (min)

V is the volume (L) of the water in the reactor

C<sub>i</sub> and C<sub>f</sub> are initial and final pollutant concentrations

k<sub>1</sub>' is the pseudo-first-order rate constant (min<sup>-1</sup>) for the decay of pollutant concentration

38.4 is the conversion factor 1,000 x ln (10)/60

As k<sub>1</sub>' increases (E<sub>EO</sub> decreases), MTBE concentration decreases. "At higher concentrations of MTBE, higher concentrations of intermediates are formed, which are also highly reactive toward hydroxyl radicals. Thus, MTBE and its intermediates compete more effectively for OH radicals, reducing MTBE treatment efficiency. Increasing the MTBE concentration from about 100 ppb to 100,000 ppb increases the electrical energy per order by a factor of about 40" (*Cater and others, 2000*).

### Effect of BTX

Photooxidation of 10 mg/L MTBE is not affected by BTX concentrations of less than 2 mg/L. "At lower MTBE concentrations, lower levels of BTX will affect the E<sub>EO</sub> for MTBE at corresponding ratios of BTX to MTBE." MTBE E<sub>EO</sub> increases with increasing BTX concentration because of the following:

- BTX components are highly reactive toward hydroxyl radicals and compete with MTBE for their capture
- BTX are relatively strong absorbers in the UV region
- as treatment progresses, phenolic intermediates are generated, which are stronger UV light absorbers than their parent compounds
- phenolic intermediates are also highly reactive toward hydroxyl radicals, which further slows MTBE treatment

The kinetic model confirms the assertions above. Researchers can equate E<sub>EO</sub> to treatment costs. For MTBE of 12 µg/L, the E<sub>EO</sub> is 1.4 kWh/m<sup>3</sup>/order. If the treatment objective is 10 µg/L, a log reduction of 3.08 results and the total electrical energy required per m<sup>3</sup> is 4.3 kWh. If the cost of electricity is \$0.06 per kWh, the contribution to the treatment cost from electrical energy will be \$0.26 per m<sup>3</sup> or \$0.98 per 1,000 U.S. gallons. Hydrogen peroxide and lamp replacement are additional costs.

### Analytical methods and degradation pathways

Stefan and others (2000) describe the analytical methods used to define MTBE's degradation pathways by UV/H<sub>2</sub>O<sub>2</sub> as:

- gas chromatography
- ion chromatography
- UV-V spectrophotometry
- gas chromatography/mass spectrometry sample derivatization
- TOC analysis

Degradation reaction intermediates that these researchers identified during experiments included

- TBF, tert-butyl formate
- TBA, tert-butyl alcohol
- methyl acetate

Other intermediates detected and quantified were carbonyl compounds and organic acids. For detailed information and degradation pathways, see the text.

### Another UV/H<sub>2</sub>O<sub>2</sub> experiment

Chang and Young (2000) conducted UV/peroxide treatments of MTBE in a cylindrical, stainless steel flow-through reactor. Researchers added to the reactor 10 mg/L of MTBE and a 31.5 percent aqueous solution of hydrogen peroxide. They warmed the ultraviolet lamp 1 hour before each experiment. In some experiments, they added 500 mg/L of benzene "to permit estimation of the hydroxyl radical concentration from the rate of benzene disappearance and established rate constants." At specified intervals, researchers removed water samples and analyzed them using purge-and-trap GC-MS to monitor MTBE and benzene degradation.

Researchers found nearly complete MTBE removal (>99.9%) after 75 minutes in the reactor. The major by-product was tert-butyl formate, TBF. "No other purgable by-products in the mass scan range (m/z between 45 and 99) were detected in these analyses. Other by-products such as formaldehyde or acetone were probably formed but went undetected because they were not purgable or were too small and/or too volatile to appear in the analytical results."

"Both benzene and MTBE disappearance followed pseudo-first-order behavior in the initial period (10-75 min.) of each experiment. Pseudo-first-order rate constants for MTBE and benzene were obtained from this initial period data by linear regression on the natural logarithm of species concentration versus time" (Chang and Young, 2000).

Their study also showed that significant quantities of TBF are formed and persist beyond the time when MTBE is gone. "After 60 minutes in our experimental system the mass ratio of TBF:MTBE remaining was 35:1, 21:1 and 3.6:1; H<sub>2</sub>O<sub>2</sub>:MTBE ratios were 15:1, 7:1 and 4:1." Thus, TBF, with unknown toxicity and uncertain impact on human health, could be a large concern with this technology.

### References

Cater, S.P., Stefan, M.I., Bolton, J.R. and A. Safarzadeh-Amiri, "UV/H<sub>2</sub>O<sub>2</sub> Treatment of Methyl tert-Butyl Ether in Contaminated Waters," *Environmental Science & Technology*, Vol. 34, No. 4, 2000, <http://www.pubs.acs.org>.

Chang, P.B.L. and T.M. Young, "Kinetics of Methyl tert-Butyl Ether Degradation and By-Product Formation During UV/Hydrogen Peroxide Water Treatment," *Water Resources*, Vol. 34, No. 8, pp. 2233-40, 2000; <http://www.elsevier.com/locate/waters>.

Stefan, M.I., Mack, J. and J.R. Bolton, "Degradation Pathways During the Treatment of Methyl tert-Butyl Ether by the UV/H<sub>2</sub>O<sub>2</sub> Process," *Environmental Science & Technology*, Vol. 34, No. 4, 2000, <http://www.pubs.acs.org>.



## Oxygen-release compounds

Waite and others (1999) investigated the use of oxygen-release compound from solid peroxides. Oxygen-release compounds increase the oxygen content of contaminated areas, enhancing biological activity and thus promoting natural attenuation. The specific compound used will depend on soil chemistry, concentration of target organics, type of target organics and cleanup levels. Parameters of interest are release rate of oxygen at different effective partial pressures and ratio of oxygen released to amount of oxygen applied.

Researchers studied the solid oxidants below with respect to dissolution rate and ease of movement through other media:

- Na<sub>2</sub>CO<sub>3</sub>•1.5H<sub>2</sub>O<sub>2</sub> encapsulated sodium percarbonate
- free sodium percarbonate crystals
- CaO<sub>2</sub>, calcium peroxide
- MgO<sub>2</sub>, magnesium peroxide

### Oxygen movement

Oxygen movement in the subsurface is influenced by:

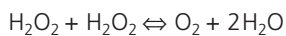
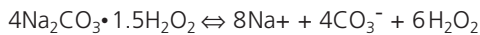
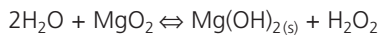
- soil heterogeneity
- moisture content, which can hinder O<sub>2</sub> movement
- pore size—a function of sediment age and history
- tortuosity, caused by small pore sizes, which increases O<sub>2</sub> path distance

Soil morphology directly influences O<sub>2</sub> diffusion through the soil and soil redox potential, and the biological degradation that will occur at interfacial areas. In the interstitial pores, microbes are protected from toxic compounds. "Interstitial pore space attachment also makes predation more difficult.

[Adding] a solid oxidant may provide both sources of O<sub>2</sub> and, in the case of encapsulated solid oxidants, locations for biological attachments. [Introducing] solid oxidants into the void spaces within the soil matrix may result in localized partial pressures above 21.3 kPa, resulting in DO concentrations well above what is possible for water in equilibrium with air (8.9 mg/L). The conditions may then alter the behavior of the solid oxidant, as well as microbe viability" (Waite and others, 1999).

"In developing an O<sub>2</sub>-rich zone, the soil's chemical oxidative demand must be met before the O<sub>2</sub> levels in the pore water are elevated. Only after the initial soil and water demands have been met can the amendment(s) be added (or released) at a rate that maximizes biological metabolism while limiting production of excess O<sub>2</sub>. The O<sub>2</sub> release rate of the solid oxidant has to be sufficient to overcome these potential O<sub>2</sub> sinks for successful application. This still does not ensure that the target organics are preferentially metabolized, but it does set up conditions more conducive to their microbial use."

"Solid oxidants can exhibit slow dissolution and fall into a reaction-limited domain. Conversely, these compounds can release oxygen from their surfaces rapidly, exhibiting transport limitations." Researchers predicted that the encapsulated Na<sub>2</sub>CO<sub>3</sub>•1.5H<sub>2</sub>O<sub>2</sub>'s release of O<sub>2</sub> was by diffusion-limited transport while the other studied oxidants were controlled by chemical reaction kinetics of dissolution. The kinetics of dissolution have both chemical and thermodynamic limitations. Reactions are as follows:



Some of the reaction products produced—Mg(OH)<sub>2</sub> and Ca(OH)<sub>2</sub>—have solubility values lower than the ions added. Such precipitates may coat reactant particles and block pores in both the soil and reactant particles, limiting transport of reacting ions and particles.

Sodium percarbonate would release O<sub>2</sub> by diffusion-limited transport whereas chemical kinetic reactions would control dissolution rate of other oxidants. Release rates of MgO<sub>2</sub> and CaO<sub>2</sub> could be limited because of self-encapsulation.

### Experiments and results

Researchers performed experiments using batch reactors to determine the rate of oxygen release. "Based on the saturation function, the empirical kinetic expressions accurately predicted the release of oxygen into water for three out of four solid oxidants. Computer-aided data acquisition enabled repeatable and consistent modeling (over five orders of magnitude of time) of O<sub>2</sub> release from solid oxidants. This model can be used to estimate times needed to reach DO concentrations that initiate aerobic respiration if the soil oxygen demand is known. In addition, if O<sub>2</sub> is the rate-limiting component, this model can be used as the first step in estimating the rate of bioremediation."

Researchers determined that the unencapsulated Na<sub>2</sub>CO<sub>3</sub>•1.5H<sub>2</sub>O<sub>2</sub> had the most rapid release rate, followed by CaO<sub>2</sub>, and encapsulated Na<sub>2</sub>CO<sub>3</sub>•1.5H<sub>2</sub>O<sub>2</sub>. MgO<sub>2</sub> had the slowest O<sub>2</sub> release by several orders of magnitude.

However, the large size of both forms of Na<sub>2</sub>CO<sub>3</sub>•1.5H<sub>2</sub>O<sub>2</sub> slows transport of bulk particles. CaO<sub>2</sub> and MgO<sub>2</sub> both have fractions small enough to permit migration where soil particles, and thus pore spaces, are larger than the particles of solid oxidant. In some cases, lack of movement of oxidant particles may be desirable in establishing stationary oxidative zones.

Adding oxidants to water also changes the water's pH, usually in the range of 10 to 12. "Shifts to high pH conditions generally have a negative effect on indigenous bacteria, but soils can have a buffering capacity to counteract or neutralize the pH shifts."

### Other conclusions

Release rates that are too rapid for biological uptake rates will prevent the utilization of all O<sub>2</sub>. Oxygen release rates below optimum may result in reduced aerobic metabolism or failure to maintain aerobic respiration. Of the oxidants tested, MgO<sub>2</sub> has the widest application based on

- O<sub>2</sub> release rate, which was the longest
- pH shift, which was lowest
- O<sub>2</sub> release per mass, which was highest

Na<sub>2</sub>CO<sub>3</sub>•1.5H<sub>2</sub>O<sub>2</sub> and CaO<sub>2</sub> showed the most rapid release, with half-lives of less than 0.25 h—desirable if instantaneous elevated oxygen levels are the goal.

Waite and others (1999) suggest that mixtures of these compounds may be formulated to develop other "time release" compounds. "Examining the release of dissolved oxygen in the presence of sediments would help identify the response of solid oxidants to both chemical and organic oxygen demands in saturated sediments."

### References

Waite, A.J., Bonner, J.S. and R. Autenrieth, "Kinetics and Stoichiometry of Oxygen Release from Solid Peroxides," *Environmental Engineering Science*, Vol. 16, No. 3, 1999; <http://www.liebertpub.com/EES/>.

See also

- Oxygen-release articles on the UTTU website, <http://epd.engr.wisc.edu/uttu/>
- "Contaminant Adsorption and Oxidation via Fenton Reaction," by Huling, S., Arnold, R.G., Sierka, R.A., Jones, P.K. and D.D. Fine, *Journal of Environmental Engineering*, July 2000; <http://www.pubs.asce.org>.



## Information sources

### U.S. EPA publications and information

The following publications can be viewed or downloaded from <http://clu-in.org/techpub.htm>

- *An Overview of the Phytoremediation of Lead and Mercury*
- *Subsurface Remediation: Improving Long-Term Monitoring and Remedial Systems Performance, Conference Proceedings* (EPA 542-B-00-002)
- *The Use of Plants for the Removal of Toxic Metals from Contaminated Soil*

Other EPA/government documents:

*Abiotic In-Situ Technologies for Groundwater Remediation, Conference Proceedings* (EPA 625-R-99-102); for hard copies call 800-490-9198 or 513-489-8190 or fax 513-489-8695.

*Automatic Tank Gauging Systems for Release Detection: Reference Manual for Underground Storage Tank Inspectors* (EPA 510-B-00-009), [http://www.epa.gov/swerust1/pubs/atg\\_0900.pdf](http://www.epa.gov/swerust1/pubs/atg_0900.pdf).

*Compact High Resolution Spectrometer* (DOE/EM-0548), <http://ost.em.doe.gov/ifd/itsrs/itsr1564.pdf>.

*Innovative Technology Summary Report: Spectral Gamma Probe* (DOE EM-0542); <http://ost.em.doe.gov/ifd/scfa/itsrs/itsr2364/itsr2364.pdf>.

*Institutional Controls: A Site Manager's Guide to Identifying, Evaluating and Selecting Institutional Controls at Superfund*

and RCRA Corrective Action Cleanups (EPA 540-F-00-005); view or download at <http://www.epa.gov/superfund/resources/institut/guide.pdf>.

*New Environmental Technology Verification (ETV) Reports* includes reports on All Systems Inc., Boreal Laser Inc., Opsis Inc., and Unisearch Associates; see <http://www.epa.gov/etv>.

*No-Purge Groundwater Sampling: An Approach for Long-Term Monitoring* (API Bulletin No. 12), <http://www.api.org/ehs/sgreshbul.htm>.

*Operating and Maintaining Underground Storage Tank Systems: Practical Help and Checklists* (EPA 510-B-00-008), <http://www.epa.gov/swerust1/pubs/ommanual.pdf>.

*Results-Based Corrective Action, Draft Guidance Document for Public Comment*, <http://www.epa.gov/correctiveaction>.

*Statistical Estimation and Visualization of Groundwater Contamination Data* (EPA 600-R-00-034, [http://www.epa.gov/ada/download/reports/epa\\_600\\_r00\\_034.pdf](http://www.epa.gov/ada/download/reports/epa_600_r00_034.pdf) or call Kay Cooper at 580-436-8651 or fax 580-436-8503.

*Tech Trends* (EPA 542-N-00-007) focuses on innovative technologies and information sources for site characterization involving contaminated soil and sediments; call 800-490-9198 or 513-489-8190 or fax 513-489-8695.

The following EPA documents can be ordered from the National Service Center for Environmental Publications (NSCEP; <http://www.epa.gov/ncepihom/ordering.htm>) and are available in a searchable database at <http://www.frtr.gov>:

- Bioremediation* (EPA 542-R-95-002)
- Bioremediation/Vitrification* (EPA 542-R-97-008)
- Ex-Situ Soil Treatment* (EPA 542-R-98-011)
- Groundwater Pump-and-Treat* (EPA 542-R-95-003)
- Groundwater Pump-and-Treat* (non-chlorinated solvents; EPA 542-R-98-014)

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### Other publications

*Evaluation of H.E.L.P. Mate 2000 for the Identification and Quantification of Petroleum Hydrocarbon Products* (TR-00-20). This report evaluates a spectrophotometric device for determining extent and type of TPH contamination in soils and water. View or download at [http://www.crrel.usace.army.mil/techpub/CRREL\\_Reports/reports/TR00-20.pdf](http://www.crrel.usace.army.mil/techpub/CRREL_Reports/reports/TR00-20.pdf).

*Guide Specification: Operation, Maintenance and Process Monitoring for Soil Vapor Extraction (SVE) Systems (CEGS-01830)*. This gives requirements that will ensure proper operation of SVE equipment. View or download at <http://www.hnd.usace.army.mil/techinfo/cegs/pdf/01830.pdf>.

*Subsurface Barrier Verification with the SEAttrace Monitoring System (Tech ID 308)*. This is a description of a barrier monitoring system that used tracer injection, automated multipoint sampling and real-time global barriers. View or download at <http://ost.em.doe.gov.itsr.itsr.308.pdf>.

The following publications are available from D. Glass Associates, Inc. (617-726-5474; DglassAssc@aol.com):

- *U.S. Remediation Markets*
- *U.S. Bioremediation Companies*
- *U.S. and European Bioremediation: Microbial Product Companies*

### Websites

For discussion of no-purge sampling, see <http://communities.msn.com/NoPurgeGroundwaterSampling/homepage>.

*Monitored Natural Attenuation*, <http://www.GW-MNA.com>, presently under construction, will contain documents, papers and software related to monitored natural attenuation.

*State Environmental Data Exchange Strategy Network (SEDEnet)*, <http://www.sedenet.org/EquIS/eqsindex.htm>.

### Miscellaneous

The Environmental Technology Verification (ETV) Site Characterization and Monitoring Center is testing field-portable technologies for MTBE measurement in groundwater. For information on this project contact Wayne Einfeld at Sandia National Laboratories (505-845-8314; weinfeld@sandia.gov) or Eric Koglin (702-798-2432; koglin.eric@epamail.epa.gov).

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*UTTU obtained many of these sites and other information from the Groundwater Mailing List (<http://groundwater.com>), the Bioremediation Discussion Group (<http://biogroup.gzea.com>) and TechDirect (<http://clu-in.com/techdrct.htm>).*

*UTTU thanks the moderators/editors from these groups—Ken Bannister of Groundwater, Richard Schaffner of Biogroup and Jeff Heimerman from U.S. EPA's TechDirect.*

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