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MTBE treatment by air stripping, carbon adsorption and advanced oxidation

Sutherland and others (2004) examined five different MTBE-contaminated groundwater sites (A, B, C, D, and E) across Missouri and the techniques used to clean up MTBE. The most common MTBE treatment processes include

- air stripping
- granular activated carbon (GAC)
- adsorption
- advanced oxidation
- soil vapor extraction

The effectiveness and costs of some of these processes, which this article will examine, are determined primarily by

- water quality characteristics (e.g., organic carbon, alkalinity)
- process parameters

Site waters

Table 1 lists some groundwater characteristics from the five sites, A, B, C, D, and E (Sutherland and others, 2004).

	A	B	C	D	E
Total alkalinity (mg/l as $CaCO_3$)	214	152	432	390	106
pH	7.0	7.1	7.6	7.7	7.3
Total hardness (mg/l as $CaCO_3$)	267	238	408	259	140
Ca hardness (mg/l $CaCO_3$)	112	168	166	214	94
Mg hardness (mg/l as $CaCO_3$)	156	70	242	45	46
Fe(II) (mg/l Fe(II))	0.01	0.01	0.01	0.00	0.01
Total (mg/l Fe)	0.09	0.02	0.05	0.01	0.15
NH_3 (mg/l NH_3)	<0.01	<0.01	<0.01	<0.01	0.03
NO_2^- (mg/l NO_2^-)	<0.004	0.05	<0.004	<0.004	0.01
NO_3^- (mg/l NO_3^-)	0.40	2.50	0.30	0.30	0.40
Turbidity (NTU)	0.23	0.47	1.11	0.31	1.73
TDS (ppm)	250	273	424	298	169
COD (mg/l O_2)[a]	1	29	61	5	5
Influent MTBE (mg/l)	5.03-5.31[b] (spiked)	0.963-1.26	0.023-0.029	0.198-0.224	0.033-0.039
Influent TBA (mg/l)	<0.001	<0.001	<0.001	<0.001	<0.001
Influent BTEX (mg/l)	<0.0005	<0.0005	1.1-3.6	0.052-0.17	0.011-0.018
Influent benzene (mg/l)	<0.0005	<0.0005	0.49-1.09	<0.0005-0.012	<0.0005
Influent toluene (mg/l)	<0.0005	<0.0005	0.16-0.91	0.03-0.009	<0.0005
Influent ethylbenzene (mg/l)	<0.0005	<0.0005	0.009-0.15	<0.0005-0.04	0.0009-0.0017
Influent p-xylene (mg/l)	<0.0005	<0.005	0.48-1.46	0.021-0.11	0.01-0.016

[a]COD for site A waters was non-detect; COD was set to the MDL of 1 mg/l for calculational purposes.

[b]Influent MTBE was non-detect in unspiked sample.

Table 1. Characteristics of site waters (Sutherland and others, 2004).

Air stripping

Air stripping was performed by an insulated packed tower that was "0.30 m in diameter, contained 2.9 m of Jaeger 2.54-cm polypropylene TriPack and was operated in countercurrent mode... The tower was a forced draft system with air blown into the tower at up to 0.033 m^3/s with a 0.75 kW DR404 regenerative blower" (Sutherland and others, 2004).

Granular activated carbon

Researchers used a RSSCT (rapid small scale column test) to estimate the performance of GAC (granular activated carbon). "In the RSSCT setup, groundwater containing MTBE was pumped through a pulse dampener to two or three glass columns in series (30 x 1.1-cm I.D., inside diameter) using 0.32-cm stainless-steel tubing or fluorocarbon tubing. Samples were periodically collected for analysis from the effluent of each column. Calculational adjustment in the volume fed to the second (and third) columns was made to account for the

amount of water collected from preceding columns. The breakthrough curves for MTBE for each column in series overlay each other when plotted versus bed-volumes, which checked that premature exhaustion or the pre-loading effect were not significant. Carbon was ground for the RSSCT experiments using a dry blend technique detailed elsewhere. The columns contained 3.0 or 5.8 g of either pulverized Calgon F-400 or F-600 GAC sieved to provide a 80 x 140-mesh fraction (0.18 x 0.11 mm). The water temperature was held at 21.5 (±2) °C during the experiments. The columns were loaded at a flowrate of 7 ml/min (4.4 m/h)" (Sutherland and others, 2004).

UV/H_2O_2 system

"The UV/H_2O_2 system was continuous flow with treatment rates maintained between 3.8 and 7.6 l/min. The system used a 1-kW medium-pressure mercury vapor arc lamp controlled by an ARC power/control cabinet. The reactor was a 0.050-m-long by 0.015-m inside diameter, 316-liter passivated stainless-steel chamber with an annular volume of 8.7 l. Hydrogen peroxide, sodium hydroxide and MTBE were metered into the system as necessary with Masterflex L/S positive displacement (peristaltic) pumps" (Sutherland, 2004).

O_3/H_2O_2 system

"The O_3/H_2O_2 system was continuous plug-flow configuration with treatment rates maintained at 3.8 liter/min using centrifugal pumps. Oxygen for the ozone generator was produced with an oxygen generator. Ozone was generated in an Ozat-0 ozone generator and was introduced into the system with an in-line Venturi injector system. Researchers controlled ozone injection rate using a mass flow controller. Gas-phase ozone concentrations were monitored using a PCI HC 12 UV-absorbance-based ozone monitor. Hydrogen peroxide, sodium hydroxide, and MTBE were metered into the system with Masterflex L/S peristaltic pumps. Ozone gas mass transfer to solution, excess entrained gas removal, and offgas destruction were achieved using a compact injection, centrifugal degas system with thermal-catalytic ozone destruct process system" (Sutherland and others, 2004).

Analytical methods

Analytical methods performed on samples included the following:

- a gas chromatograph/mass spectrometer (GC/MS) system for measuring MTBE, benzene, toluene, ethylbenzene and o-, *m*- and *p*-xylene
- pH/temperature probe for pH and temperature
- a spectrophotometer for water quality analysis
- Standard Method 4500-CL G for free and total chlorine
- salicylate method for monochloramine and ammonia
- cadmium reduction method and diazotization chromotropic acid method to measure nitrate and nitrite
- phenanthroline method and TPTZ method for measuring ferrous and total iron
- dichromate reactor digestion method for COD, chemical oxygen demand

Results

Air stripping. Researchers found that the MTBE mass transfer coefficient for each flow rate was low due to a low Henry's constant and MTBE's high water solubility. They determined that the "process temperature would be expected to have a significant effect with lower removal efficiency resulting from lower process temperature. Furthermore, fouling of the packing can be a concern especially for groundwater that contains significant ferrous iron (which is oxidized to ferric hydroxide precipitate), high hardness (which leads to carbonate scaling as carbon dioxide is stripped) or nutrients (which may lead to biofouling)."

Data indicated

- the required packing height for a 75:1 G/L (gas/liquid) ratio was 1.5-3.0 times greater than for a 150:1 G/L ratio
- for equivalent MTBE removals, the 75:1 G/L ratio resulted in higher unit treatment costs ($1,000 per liter) by a factor from 1.03 to 2.1
- the higher G/L (150:1) resulted in both shorter towers and lower estimated treatment costs
- tower heights for 99.5 percent MTBE removals were estimated to be 3.2-4.4 times greater than for 80 percent removal

GAC. Researchers used either Calgon F-400 or Calgon F-600 GAC. "For all groundwaters, F-600 had significantly greater breakthrough and ultimate capacities for MTBE than F-400 by a factor of from 1.5 to 2.2 times" (Sutherland and others, 2004). Results also indicated:

- a strong correlation exists between influent MTBE concentration and capacity on both carbons
- low influent (equilibrium) concentrations should yield lower capacities than higher influent (equilibrium) concentrations
- RSSCT (rapid small scale column tests) indicated preferential retention of BTEX compounds over MTBE; BTEX displaces MTBE-reducing capacity of carbons for MTBE
- in all groundwaters, the COD (chemical oxygen demand) attributable to BTEX was less than 1.1 percent of the total COD
- GAC sorption capacities for MTBE are enhanced in situations where MTBE concentration is relatively high (e.g., > 1 mg/l) and competitive adsorption effects are low

- carbon costs were independent of assumed flow rate on a per volume basis
- associated carbon costs required to treat each groundwater ranged from $0.58-2.39/1,000 per liter for F-400 and $0.49-1.54/1,000 per liter for F-600
- treatment costs for the lesser expensive carbon (F-400) were greater than for F-600 in all waters except at site C due to F-400's lower capacity
- unit costs were significantly higher for the low-flow system than for the high-flow

UV/H_2O_2. "The UV dose varied generally from 2.2 to 4.4 kWh (kilowatt hour) per 1,000 liters by adjusting the influent flowrate to the process. The pH was controlled to 7 and 9 (±0.1) in these experiments to examine the influence of pH on hydroxyl radical scavenging and overall process efficiency... Under only the most efficient conditions in the study did the EE/O (electrical energy per order of magnitude removal) values for UV/H_2O_2 treatment of MTBE fall within typical ranges for other groundwater contaminants" (Sutherland and others, 2004). Thus, it is important to examine the factors that affect the process efficiency, which include

- absorbance efficiency of applied UV by H_2O_2
- fraction of generated hydroxyl radicals reacting with MTBE (the target) versus scavenging species
- the effect of pH on these processes (for instance, increasing the pH from 7 to 9 decreased the fraction of •OH reaction with MTBE by a factor of 2 to 3)
- the water's chemical makeup

Treatment costs ranged from $0.2 to $2.8 per 1,000 liters for four of the sites; site C was an exception, although researchers weren't sure of the cause of its low efficiency. Site C did have the highest COD, TDS and alkalinity of any groundwater tested, each of which can contribute to •OH scavenging. "For the more efficient waters, (i.e., lower EE/Os at sites A, D and E), annualized capital costs were generally 70 percent of total unit costs for a 38 l/min system, but only 20 percent for a 3,800 l/min system. Due to economies-of-scale with respect to capital costs, larger flow systems have lower unit treatment costs" (Sutherland and others, 2004).

O_3/H_2O_2. "Calculations for the O_3/H_2O_2 process showed that unit treatment costs (capital plus O&M) ranged from $0.4 to $2.9 per 1,000 liters for the study waters, excluding site C in which little or no MTBE was removed."

A problem with advanced oxidation processes is the formation of oxidation byproducts. Thus, "the required oxidant dosage for MTBE treatment may be significantly higher for total treatment (e.g., MTBE plus TBA) than if byproduct removal is not required... Results show that the required oxidation time and energy were approximately 100 percent greater for MTBE plus TBA removal than that for MTBE alone. The corresponding energy costs would thus be much greater. Other oxidation byproducts of MTBE will tend to be more readily biodegradable and less of a concern (e.g., formate, acetate, methyl acetate)" (Sutherland and others, 2004).

Conclusions

Researchers concluded the following:

- water quality and/or process conditions significantly affect removals and unit treatment costs
- the lowest unit treatment costs for high treatment flow rates (i.e., 3,800 l/min) for each site was with air stripping
- relatively tall packed towers were often necessary where a high removal efficiency was required
- the least costly treatment for MTBE is often high flow rate air stripping
- air stripping towers are often subject to fouling by iron floc, carbonate scaling and biofouling
- air stripping becomes less efficient at low temperatures
- other processes, such as GAC, become more efficient at low temperatures
- air stripping of VOCs may also require additional offgas treatment
- highest treatment cost (with the exception of site C) was with GAC
- GAC is subject to fouling by iron and carbonate and biological growth
- an advantage of GAC, however, is ease of use
- the O_3/H_2O_2 AOP had the lowest unit costs for four of five waters, but it was ineffective at site C
- UV/H_2O_2 provided the second lowest costs at low flowrates
- efficiency of UV/H_2O_2 relies on presence of radical scavengers, pH and background UV absorbance
- if treatment requires TBA or other oxidation byproduct removal, costs can increase (double or more)
- of the four techniques examined, air stripping is the most robust, i.e., least affected by variable water quality

Finally, Sutherland and others (2004) contend that their results demonstrate that "the selection of the best process for MTBE treatment is site specific and must include consideration of a variety of factors."

Reference

Sutherland, J., Adams, C. and J. Kekobad, "Treatment of MTBE by Air Stripping, Carbon Adsorption and Advanced Oxidation: Technical and Economic Comparison for Five Groundwaters," *Water Research*, Vol. 38, 2004; http://www.elsevier.com/locate/watres

UTTU thanks Dr. Adams, adams@umr.edu, for his help on this article.

Aboveground treatment of contaminated water

By Evan K. Nyer and James M. Bedessem

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After a decade of focusing on alternative technologies—air sparging, natural attenuation, in-situ bioremediation and in-situ oxidation—environmental professionals who found themselves on the cutting edge of innovative technologies now must face the challenges of groundwater extraction and aboveground treatment. Professionals who have entered the field in the last 10 years have not had to deal with these complexities and so may have to call on the scientists and engineers crossing over from water supply and sanitary engineering. However, even these designers do not have all the answers. Remember, pumping and treating contaminated water from marginal aquifers presents unique challenges compared to pumping clean water from prolific aquifers.

Why go back to pump-and-treat?

There are two main reasons why we will be designing more aboveground treatment systems. First, several chemicals being discovered at contaminated sites are making it necessary to re-evaluate and even implement pump-and-treat remedial solutions. Second, specific limitations in the geology of the aquifer, or the specifications for the final use of the groundwater, require the use of pump-and-treat technology.

Chemicals that are driving the return to pump-and-treat technology include

- 1,4 dioxane
- 1,2,3, TCP
- perchlorate
- MTBE
- NDMA
- arsenic
- hexavalent chromium
- endocrine disruptors
- radionuclides

These organic compounds have very similar physical and chemical properties. Primary among them are limited biodegradability or biodegradability under very specific, non-naturally occurring environmental conditions. They also do not react with the natural geology of the aquifer. In other words, dilution is the only natural attenuation method that decreases compound concentrations—not a good method when we are required to achieve low parts-per-billion or even parts-per-thousand levels of treatment. *(Reviewer's comment: Sorption may also work for these compounds.)* Other common characteristics that may result in selection of the pump-and-treat approach include

- high solubility
- high mobility (low retardation)
- moderate to high toxicity—carcinogens
- high public awareness or concern

The inorganic compounds also have similar chemical properties. These compounds will not easily be removed by natural interaction with the aquifer's geology and geochemistry. Some, like arsenic, may be naturally occurring in the aquifer at equilibrium levels above new drinking water standards.

Final use of the water can also have a large impact on the treatment method used. We have to be very careful when we alter the natural environment of a drinking water aquifer. The problem is that the chemicals listed previously either do not naturally attenuate or need a very specific environment in order to attenuate. Logically, we do not want to cause more harm to the drinking water aquifer than we solve by removing these chemicals. Also, it is difficult to apply enhanced in-situ remediation techniques to drinking water aquifers; these aquifers are usually deep and very permeable. All of the enhanced methods require that we add some type of environmental altering chemical. It is **very** difficult to deliver any chemical over a large area in a deep, fast-moving aquifer. In other words, it does not matter that you have great data from a laboratory or microcosm: the aquifer controls. The difficulty or cost of applying enhanced methods to deep, fast-moving aquifers may drive your design above ground, and include a pump-and-treat system.

There are several difficulties when trying to apply in-situ techniques:

- in-situ biodegradability can be difficult given the very specific conditions that many of these compounds require to be substantially degraded
- in-situ enhanced biodegradation is also not readily accepted as a treatment technology for water supply aquifers immediately upgradient of existing water supply wells that may be impacted and require treatment
- in-situ oxidation is able to address most of these emerging chemicals; however, it is proving to be best applied in source areas
- application of oxidation is limited because of cost; the cost is driven primarily by the fact that in-situ oxidation is chemical specific; non-target chemicals create an oxidant demand that must be overcome to treat the target contaminants

All of these reasons may combine to create a situation in which we will be forced to use wells and pumps to control contaminant movement. Once we use pumps to control aquifer flow, we are forced to provide treatment methods to remove these compounds above ground. But water treatment systems are unique and require

specific skills and experiences to design equipment that will operate over a long period of time. We will need those old design engineers.

Aboveground treatment challenges

There are two main areas of challenges for designing an aboveground treatment system. First is simply getting the water aboveground. Finding a hydrogeologist who knows how to design and develop a production well is becoming difficult. Second, there are several extra challenges when we design a treatment system to remove chemicals from groundwater. Frequently the natural material from the aquifer controls the design of the treatment system, not the chemicals that you originally set out to treat.

One of the most significant challenges is getting the water above ground. Contaminants often exist in groundwater systems that are marginal water producers or have other characteristics that make them undesirable potable water sources. Design, installation and development of wells for groundwater extraction are different from wells used for groundwater monitoring. Environmental professionals have standardized monitoring well installations, and these designs often minimize screen slot sizes and filter packs to minimize turbidity at the expense of water production. This may be acceptable for monitoring wells where only several liters of water need to be pumped during a sampling event. For water production, the well screen and filter pack must be matched to the aquifer materials to maximize well efficiency.

Once the proper well screen and filter pack have been selected and installed, the well must be properly developed to ensure effective water production. This is not a one- to two-hour effort typical of monitoring wells; proper well development can take several days or weeks, depending on the formation. Well installation method can also affect well efficiency. To minimize investigative derived waste (IDW), environmental professionals have been using techniques such as direct-push technology (DPT). To maximize drilling speed and lithologic screening, they are using rotosonic and other techniques. If these methods are used for extraction well installations, the boreholes can be smeared or the formations compacted due to vibration. These issues can have a detrimental effect on well efficiency and water production. Again, aggressive well development may be necessary to overcome these problems.

Presence of contaminants can also change the aquifer's geochemistry and produce water that is more difficult to treat. Biodegradable contaminants may create reducing conditions that cause high-dissolved metals contamination. Metals may precipitate on well screens, in pipelines, or on treatment system components. Pretreatment may need to be considered before primary treatment for contaminants of concern. Systems should be designed to be easily serviced and maintained.

Sample problems recently observed

We have observed several of these problems with systems that we recently installed. As part of a pilot test in which a pump-and-treat system was installed to capture impacted groundwater discharging to a river, rotosonic techniques were used to install a 120-foot-deep extraction well. Lithology data collected during installation were used to locate the screen interval, as well as size and match the well screen and filter pack to the formation. Following installation, the well was initially developed using surge-block and pumping techniques common to environmental consultants and drilling companies. These standard methods resulted in a well yield of less than 2 gallons per minute (gpm).

So, what do you do when you have an underproducing well? You consult an old water supply hydrogeologist with more than 35 years of experience. With a new development plan consisting of jetting combined with airlift pumping, which, by the way, requires equipment not normally carried on rigs by environmental drilling companies, several additional days of development improved the sustainable water production by almost 100 percent to between 2 and 3 gpm.

The preliminary hydraulic characteristics of the aquifer prior to installing this well indicated a well yield of 10 gpm. Again, the old water supply hydrogeologist helped implement a more aggressive development plan consisting of chemical treatment to break down clays that had smeared the borehole, followed by concurrent jetting and airlift pumping. Finally, after several more days of development (and several thousand dollars, too), the well was able to sustain a yield of more than 20 gpm.

We also recently observed some metals precipitation problems in an aboveground system designed to biologically treat a groundwater highly impacted with organics. The average biochemical oxygen demand (BOD) of the groundwater entering the treatment system was about 1,000 mg/l, and a two-stage, submerged, fixed-film bioreactor was being used to reduce that concentration to < 250 mg/l for discharge to a local publicly owned treatment works (POTW). With the high amount of organics in the groundwater system, reducing conditions were prevalent and contributed to high dissolved metals concentrations, particularly iron and magnesium. During treatment, we typically see these dissolved metals precipitate, and some attachment to the submerged media occurs along with the biogrowth; however, the force of aeration is usually sufficient to shear the solids as they build up, and then those solids carry over to the clarifier where they are removed. In this instance, however, the combination of significant metals precipitation combined with extremely rapid biological growth due to high loading rates overwhelmed the system sufficiently to plug the submerged media.

The solution to this aggressive metals precipitation problem was to convert the first stage of the system to a suspended growth reactor and use an intermediate clarifier ahead of the second-stage fixed-film reactor, thus allowing the intermediate clarifier to remove a majority of the precipitated metals prior to reaching the fixed-film unit. Solids from the intermediate clarifier were also returned to the first-stage suspended-growth reactor to control sludge age and the mixed-liquor suspended solids (MLSS) concentration in that reactor, creating a classic activated sludge treatment operation.

We originally selected the submerged fixed-film system because this type of reactor is less sensitive to influent concentrations and flow fluctuations than suspended-growth systems. Also, because of the high bacterial populations growing on the media, loading rates up to 180 lbs/1,000 ft^3 of reactor media (three times the loading rate typically used in suspended-growth systems) can be achieved. In this instance, however, the combination of metals precipitation and biological growth occurring under suspended-growth conditions has generated a sludge that can be recirculated from the intermediate clarifier to maintain a high MLSS concentration, > 5,000 mg/l. Operating under these high MLSS conditions has permitted the suspended-growth system to operate as desired, and the entire system was able to achieve > 90 percent BOD removal.

The second metal precipitation problem that occurred with this system pertains to the nutrient feed system. Nutrients in the form of urea-ammonium-phosphate mixture were injected into a 2-inch recirculation loop of a bioreactor; however, a high magnesium concentration in the water resulted in the formation of struvite ($MgNH_4PO_4$) when the nutrient was added. The resulting precipitate caused mechanical problems with the recirculation pumps that required excessive maintenance. The solution in this case was to relocate the nutrient injection into the 10-inch influent header pipe and eliminate the high concentration of ammonia and phosphate within the smaller-diameter recirculation loop.

Conclusions

These types of problems can occur on large- or small-scale pump-and-treat systems. The point of this article is that groundwater treatment systems are unique and will require a certain amount of experience to overcome the inherent problems associated with bringing groundwater from less-than-perfect aquifers to aboveground treatment systems.

One current project that we are working on is a biological treatment system that may be as large as 1,000 gpm and require treatment of 5,000 lbs/day of BOD. We are still gathering the final data that will set the size of the pumping system required to capture the entire plume. While the situation that required this aboveground treatment system is unique, in other parts of the country there are several large treatment systems being designed and installed for perchlorate, MTBE, 1,4 dioxane, and similar chemicals. Pump-and-treat has always been a viable design for controlling and remediating groundwater contamination. In-situ methods have generally proved to be more cost-effective and successful than pumping methods; however, new chemicals and aquifer conditions are preventing some successful in-situ applications.

Reference

Nyer, E.K. and J.M. Bedessem, "Aboveground Treatment Equipment: Back in Fashion," *Ground Water Monitoring & Remediation*, Vol. 23, No. 4, Fall 2003; http://www.ngwa.org.

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In-situ sequenced bioremediation

Devlin and others (2004) evaluated the performance of an in-situ groundwater treatment system that consisted of two sequenced technologies, the first anaerobic and the second aerobic. They created "an artificial plume of carbon tetrachloride (CT, 1-2 mg/l), tetrachloroethene (PCE, 1-2 mg/l) and toluene (TOL, 1-10 mg/l)" and treated the plume "inside an isolated section of aquifer subjected to a constant rate of groundwater flow." Next, they evaluated the system against "an identical adjacently located section of aquifer in which natural attenuation was permitted to occur."

Anaerobic bioremediation

Researchers previously tested "an approach to bioremediation that minimizes pumping while maximizing dispersive mixing at the Borden aquifer... Briefly, the method involves the injection of a nutrient or primary substrate solution from a nutrient injection wall (NIW) at regular intervals determined by the average linear groundwater velocity. The solution pulses are timed so that they overlap due to hydrodynamic dispersion at some predetermined distance (on the order of a few meters) from the injection wells. Where the overlap occurs, subsurface bacteria receive a continuous supply of nutrients/substrate, and a bioactive zone is sustained. Although the NIW reported by Devlin and Barker (in Devlin and others, 2004) was successful in creating and sustaining a modified anaerobic redox zone in which CT was transformed, it was unsuccessful at stimulating the reduction of TCE" (Devlin, 1994; Devlin and Muller, 1999 in Devlin and others, 2004).

Aerobic bioremediation

Bioremediation of aromatic hydrocarbons and lesser chlorinated ethenes can occur relatively rapidly in aerobic environments. Methods to accomplish this include

- biosparging
- release of an oxygen-releasing metal peroxide compound (ORC)
- oxygen introduction through diffusive emitters

Devlin and others (2004) tested a variation of biosparging in an area of the aquifer downgradient of the NIW. "Sparging was conducted only to emplace or renew residual oxygen bubbles in the porous medium, with minimal excess gas delivery. The residual oxygen bubbles dissolved into the flowing groundwater, providing a readily available electron acceptor" (Devlin and others, 2004).

In this study, a distinct geochemical boundary was formed between the anaerobic and aerobic treatment systems. Another goal of the researchers was to "evaluate any effects of the anaerobic-aerobic boundary on the performance of the system, with emphasis on the possible flow interferences that might result" (Devlin and others, 2004).

Site description

The Borden aquifer, at the Canadian Forces Base Borden, is

- pristine
- well-sorted
- 3 m deep
- underlain by a 6-m-thick aquitard, an effective barrier beneath the aquifer
- buffered by a significant carbonate component that tends to keep waters at a near-neutral pH

Experiment at the test site

"The water table varied between 0.3 and 1.5 m below ground surface (bgs), depending on time of year and location. A sheet pile wall bounded the test section on three sides, creating an alleyway measuring 2 m wide by 24 m long, and hereafter referred to as the gate. An identical alleyway was constructed immediately next to the current one, and served as a control; no treatment measures were attempted in that section of the aquifer. The gates were covered with a 30-m-long by 9.2-m-wide greenhouse structure consisting of steel tube framing and a flame retardant, vinyl-coated polyester cover to protect the equipment and instrumentation from the elements. The structure also facilitated sampling throughout the year and prevented direct local recharge due to rainfall or snowmelt, which might have influenced the hydraulics.

"The groundwater flow inside the treatment gate was maintained by continuous pumping of a fully screened 5.08 cm (2 in.) well, located at the closed end of the structure. The pumping rate was maintained at 130 ml/min. The uniformity of the flow was assessed by a bromide tracer experiment tracked with fences of multilevel monitoring wells. Average linear groundwater velocities were determined to be in the range of 4-15 cm/day, expressed as fence averages. A value of 14 cm/day was considered most representative of conditions in the gate and was used in all calculations" (Devlin and others, 2004).

Thus, the test section of the aquifer

- was hydraulically separated from the bulk of the aquifer by sheet piling
- had a gate that was enclosed on three sides with a pumping well at the closed end
- left open the fourth side to permit native groundwater to enter the test section

Researchers isolated the test section in this way to enable consistency of flow and allow contaminant transformations to be interpreted with minimal ambiguity. Using diffusive emitters at the end of the open gate, they introduced the three test compounds: tetrachloroethene (PCE), carbon tetrachloride (CT) and toluene (TOL).

"This design permitted a long-term source that could be discontinued without excavation at the conclusion of the experiment. The target concentrations for the contaminants were 1-10 mg/l for TOL, and 1-2 mg/l for PCE and CT. Previous work established that the retardation factors for these compounds in the Borden Aquifer were about 1.2, 2.7 and 1.2, respectively" (Devlin and others, 2004).

Other characteristics of the experiment included

- a NIW (nutrient injection wall) constructed about 5 m into the gate and designed to introduce benzoate and inorganic pulses to the aquifer downgradient from the source; this became an 8-meter-long anaerobic zone for reductive dechlorination
- on day 207 and for each injection afterward, supplementation of the solution with nutrients
- analysis for determining a pulsing period (28 days) using velocity and dispersion data
- a residual oxygen barrier (ROB; the aerobic treatment zone) installed about 16 m into the gate, downgradient of the NIW; it consisted of about six horizontal sparge wells (made of 5.1-cm outer diameter PVC) constructed at the aquifer's base and oriented perpendicular of flow direction; each sparge well contained four 0.62-cm-diameter polyethylene tubes terminating at different distances along the horizontal pipe; at the surface, tubes connected to a manifold were also connected to a cylinder of oxygen, and gas injections were performed at regular intervals
- test sparging, which was completed once the ROB was backfilled with gravel up to water table level
- a control (a second, three-sided sheet pile alleyway of identical dimensions and also instrumented with monitoring wells) located next to the gate where degradation resulting from bioremediation could be

compared to degradation due to natural attenuation processes

- sampling that used multilevel wells to give detailed vertical and lateral data on contaminant fate

For more detail on the sampling methods, analytical methods and data reduction, please see the original paper.

Conclusions

Based on analysis of data, researchers concluded the following:

- sequenced bioremediation is an "advantageous and technically viable method of treating mixed contaminant plumes" (for instance, cDCE (cis-dichloroethene) formed in the anaerobic zone and was degraded in the aerobic zone)
- benzoate and nutrient additions stimulated the degradation of PCE and TCE to form cDCE
- TOL degraded with a half-life of about 60 days by anaerobic natural attenuation; the anaerobic environment inhibited TOL degradation; however, TOL degradation rates in the aerobic treatment zone were triple those treated only by natural attenuation
- brief daily sparging for the emplacement of residual oxygen bubbles had no significant effect on the aqueous concentration of TOL or cDCE; however, the sparging did create and sustain an aerobic environment in which target compounds could be degraded
- for chlorinated methanes, the advantage of sequenced treatment over natural attenuation was minimal
- results suggest that for the transformations of both CT and CF, TOL served as electron donor
- "both TOL and PCE degraded more rapidly in the treatment gate than by natural attenuation; also in both cases, the degradation rates were apparently increasing when the experiment ended, so the maximum rates have yet to be determined"

Devlin and others (2004) add: "Finally, there was no evidence of hydraulic interferences due to pore clogging, despite a sharp transition from highly anaerobic conditions to aerobic conditions. Further work is needed to examine the longer performance of these coupled technologies and the transition zone between them."

Reference

Devlin, J.F., Katic, D. and J.F. Barker, "In-situ Sequenced Bioremediation of Mixed Contaminants in Groundwater," *Journal of Contaminant Hydrology*, Vol. 69, 2004; http://www.elsevier.com/locate/jconhyd

UTTU thanks Dr. Devlin, jfdevlin@ku.edu, for his help on this article.

The City of Los Angeles Former Gas Station Sites Program, part 1

This is an excerpt from the "Guide to Resolving Environmental and Legal Issues at Abandoned and Underutilized Gas Station Sites," originally published by the City of Los Angeles at http://www.lacity.org/ead/labf/Gas%20Station%20Guide.htm

When the City of Los Angeles started its gas station program, regulators expected to find a large number of former gas station sites that were vacant or abandoned by owners who could not afford to remove underground tanks or contamination. The reality was more complex. As it turned out, there were few vacant sites. There were a significant number of former gas station sites with old, obsolete USTs still in the ground and no longer being used, but the sites functioned as auto repair, tire sales, used car sales and storage businesses. Often these sites present a poor appearance due to obsolete signage, decrepit buildings in need of paint, growth of weeds, and inadequate screening of automotive repair activities.

When the city reviewed former gas station sites, it found that most were occupied; and as a first step, the city's program focused on redeveloping vacant sites. Therefore, this guide begins its focus there as well. Vacant sites are less complicated because they do not require the interruption or displacement of an ongoing business. Also, vacant sites produce a higher level of blight. Therefore, this guide will be most helpful for such sites, but it also contains information that can be helpful in addressing any former gas station site.

The problem

Until the mid-1980s, most USTs were made of bare steel, which is likely to corrode over time, allowing the contents to leak. Faulty installation or inadequate operation and maintenance procedures also can cause USTs to release their contents into the soil or groundwater.

Unused USTs remain in the ground because of the high cost to remove them—anywhere from $5,000 to $10,000 per tank. If the tank has leaked, remediation may cost anywhere from under $50,000 to more than $500,000, with most sites falling in the under-$200,000 range. Such sites often cannot be sold unless the USTs are removed. In neighborhoods with high property values, the cost to remediate a site can come out of the purchase price or rental income. However, in communities with lower real estate values, the cost to bring a site into compliance can exceed its value. Some owners have not used the tanks for many years and are not even aware of the regulations. Others use or lease the site, hoping no one will force the issue. In some cases, owners who cannot afford to remediate simply abandon the site.

Federal requirements

In 1984, Congress added Subtitle 1 to the Resource Conservation and Recovery Act (RCRA) that required the U.S. Environmental Protection Agency (U.S. EPA) to develop a comprehensive program for regulating USTs used for storing petroleum and certain other hazardous substances. In 1986 the Superfund Amendments and Reauthorization Act (SARA) created the Leaking Underground Storage Tank Trust Fund and required owners of USTs to show they had the financial resources to clean up future potential UST contamination.

Administration in California

In California, the U.S. EPA has delegated the implementation of the federal UST requirements to the California Water Resources Control Board, which has in turn delegated certain responsibilities to its regional boards and other local enforcement agencies. The following table gives a summary of each agency's area of jurisdiction.

Agency	Area of responsibility
Local agency (fire, health, etc.)	Tracks UST registration, assuring that notification and technical requirements (described above) are met, including removal of out-of-service USTs. If owners cannot or will not remove the tanks, local agencies may not have the funds to do so. How to address this problem is the subject of this guide.
Regional Water Quality Control Board	Steps in when groundwater contamination is discovered to assure it is remediated properly. Oversees groundwater remediation and other related remediation at sites.
California Department of Toxic Substances Control	Oversees remediation of soil contamination beyond the scope that can be handled by the local agency.
State Water Resources Control Board	Administers California programs to implement the LUST Trust Fund, cost recovery, and financial responsibility requirements described above.

Table 2. Areas of responsibility of California agencies.

The local agency may be a city or county agency—usually a fire department or environmental health department. If not, these agencies should be able to direct you to the proper agency in your area. As this guide describes, these agencies work together to implement these programs. In addressing problem UST sites, it is important to understand how the agencies work, identify appropriate individuals within these agencies and cultivate a good working relationship with them.

Redevelopment perspective

Redeveloping old gas station sites may be facilitated by cooperation between the agency tasked with enforcing UST regulations and a redevelopment agency. The redevelopment agency may be able to contribute resources that the regulatory agency lacks. Such cooperation works best when each agency comes to the problem with the aim to achieve both compliance and redevelopment. However, redevelopers must remember that a regulatory agency's first responsibility is to protect public health and the environment by enforcing the law, and the regulatory agency can help only to the extent that their goal is complementary to the goal of site development. Finding common ground between regulator and redeveloper is the key to success in addressing these sites.

Advice for UST owners

Many regulatory agencies were not able to identify all cases of non-compliance immediately after January 1999, when all unused USTs and UST delivery systems were required to be upgraded or removed. Increasingly as the years go by, these agencies are catching up on the backlog of non-compliant sites. Some agencies are issuing orders to owners to remove the tanks. In some jurisdictions, those owners with the least financial resources to take action are now being contacted because they are the only sites left. Prior to receiving an order to remove the tanks, many of these owners had no idea that removal was required. Many are at a loss as to how to fund removal.

To owners who find themselves the subject of such an order, we advise cooperation with the regulating agency whether the owners have the funds to remove the tank or not. The eligibility of a site to receive funds from the California State Cleanup Fund to pay for remediation (see Chapter 5, "Funding UST Removal and Remediation) may depend on whether the UST owner/operator was cooperative or knowingly avoided compliance. Some regulating agencies are working with owners to help them identify loans or other resources to comply. In some jurisdictions, non-cooperative owners are being referred to the district attorney or city attorney for legal action. Some jurisdictions are also applying for special state funding known as the Emergency Abandoned, Recalcitrant (EAR) Account of the California Petroleum UST Cleanup Fund, to remove USTs.

City of Los Angeles Gas Station Program

The City of Los Angeles Brownfields Program helps owners of sites that have or once had an underground storage tank to bring their site into compliance and redevelop their property. The program offers over-the-phone technical assistance on accessing the various programs discussed in this guide. There also may be financial assistance available to qualifying owners who lack the financial resources to remove USTs or otherwise bring their site into compliance. Owners of USTs with property located in the City of Los Angeles are urged to contact the city's Gas Station Program personnel for additional assistance.

Reference

"Guide to Resolving Environmental and Legal Issues at Abandoned and Underutilized Gas Station Sites," LA Brownfields Program, http://www.lacity.org/ead/labf/

UTTU thanks Maxine Leichter, mleichte@mailbox.lacity.org, for her help on this article.

Research notes

Characterizing vehicle emissions from the burning of biodiesel made from vegetable oil

Zou, L. and S. Atkinson, *Environmental Technology*, Vol. 24, 2003; http://www.environtechnol.co.uk/history.htm

Benzo[a]anthracene, chrysene, Benzo[b]fluoranthene, Benzo[k]fluoranthene, Benzo[a]pyrene, Dibenzo[a,h]anthracene and Indeno[1,2,3-c,d]pyrene are PAH carcinogens (or genotoxic PAHs) found in particulate matter that is produced from the burning of diesel and biodiesel fuels. Also found in the particulate matter are the co-carcinogens fluoranthene, pyrene and Benzo[g,h,I]perylene. Zou and Atkinson (2003) studied these two fuels and compared amounts and types of particulate matter produced. While burning biodiesel fuel seemed to produce a smaller amount of particulate matter and a "marked reduction of the less toxic PAHs such as naphthalene," researchers caution others that such conclusions are still preliminary.

Researchers found a smaller "impact of using biodiesel on concentrations of NOx (nitrogen oxides), HC (hydrocarbons) and CO_2 (carbon dioxide). Based on the real-time monitoring of gaseous pollutants emission, marginal reductions (about 10 percent) in emission were found for NOx, HC and CO_2 when burning 100 percent biodiesel, compared to burning 100 percent petroleum diesel."

Researchers believe that "The information obtained from this study can be used to gain a better understanding of the beneficial environmental impacts of using biodiesel. Although some research has reported a lower mutagenic potency of the particulate matters from biodiesel emission, further study on understanding the genotoxicity and other health effects of biodiesel emissions on human cells is needed to more fully explore the benefits of using biodiesel."

Enhancing attenuation of petroleum pollutants by modifying natural soil with surfactants

Zhu, K., Chen, H., Yang, R. and W. Zhou, *Practice Periodical of Hazardous, Toxic and Radioactive Waste Management*, April 2004; http://www.pubs.asce.org/

The loess plateau of Northwest China is a land of large oil fields where severe soil contamination by petroleum hydrocarbons is not uncommon. Remediators sought to create environmental conditions conducive to natural attenuation of these contaminants by using a cationic surfactant (hexadecyltrim-ethylammonium bromide (HDTMA-Br)) and an anionic surfactant (sodium dodethylbenzene sulfonate) to modify the soils.

According to Zhu and others (2004), "Among all natural attenuation mechanisms, sorption is a major process since the intrinsic mobility of petroleum hydrocarbons is often inversely related to their sorption on soil surfaces. Physically, sorption can occur if the leading forces from London-van der Waals act on the solid sorbents, or chemically by the chemical bonding forces. However, the distinction between physical and chemical adsorption is not always explicit. The realization of the potential for natural attenuation is, however, dependent on many factors, such as soil properties, moisture, pH values, temperature, residue concentration, organic contents and co-contaminants in soil environment in the vadose zone where the petroleum residues are present... Adsorption may also affect other cleaning reactions such as biodegradation, plant uptake and volatilization, etc., while soil minerals and coating organic matters are the principal sorbents in the soil matrix. It has been shown that sorption to natural soils has little impact on attenuation and fate of the organic contaminants when the distribution coefficient (K_d) value is low. Therefore, more research is needed to investigate techniques for enhancing adsorption capacity of natural soils."

Based on their laboratory work with the cationic and anionic chemicals, researchers concluded that these chemicals can enhance the sorption capacity of the loess soils.

Estimation of primary drainage three-phase relative permeability for organic liquid transport in the vadose zone

Oliveria L.I. and A.H. Demond, *Journal of Contaminant Hydrology*, Vol. 66, 2003; http://www.elsevier.com/locate/jconhyd

In this article, Oliveria and Demond assessed the ability of eight models "to predict the drainage permeability relative to oil in a three-phase system." They used models that "provided a closed equation for the relative permeability of oil when water is draining, whose implementation required a minimum number of assumptions, and whose parameters could be determined independently. In addition, consideration was given to models commonly used in the literature and recently proposed models that have yet to be evaluated" (Oliveria and Demond, 2003).

Previous research indicated that for water-wet three-phase systems:

- "the relative permeability to water is determined by the water saturation only and is not affected by whether the remaining void space is filled by air, an organic liquid or both
- "similarly, the relative permeability to gas is a function of its own saturation only
- "the relative permeability to oil arises in a more complex manner, being dependent on both water and gas saturations"

The goal of the two researchers was "to compare eight different predictive models for the relative permeability to an organic liquid using the more complete set of data produced by Oak" (1990, in Oliveria and Demond, 2003). Researchers found that the goodness of fit of the models did not increase with amount of data or computation, because "the data sets were not uniform in the direction of saturation change."

Integrated simulation-optimization approach for real-time dynamic modeling and process control of surfactant-enhanced remediation at petroleum-contaminated sites

Huang, Y.F., Li, J.B., Huang, G.H., Chakma., A. and X.S. Qin, *Practice Periodical of Hazardous, Toxic, and Radioactive Waste Management*, April 2003; http://www.pubs.asce.org

The authors of this article state that "the cleanup of contaminated groundwater has been less successful than originally anticipated," in addition to its cost and time. Most remediation processes are complex, "with a variety of temporal and spatial variations. Also, many process factors, such as pumping rate, oxygen addition rate, additives addition rate, and groundwater temperature, have impacts on remediation efficiency; determination of the optimal combination for these factors is difficult due to lack of insight about the subsurface processes. Thus, improvements in the site remediation practices are desired."

To address this problem, Huang and others (2003) suggest an enhancement of the process control of remediation systems. Other researchers, for instance, have developed approaches for "simulating surfactant-enhanced remediation processes, through bench-scale modeling and field investigation. Generally, to improve remediation efficiency and cost-effectiveness, a number of control factors, such as surfactant supply, nutrient injection and pumping/recharge rates, need to be able to dynamically match the desired levels for microorganisms to degrade the hydrocarbon effectively. Therefore, the designed system must be able to facilitate on-line adjustment and control of these factors according to varying site conditions. However, most of the existing surfactant-enhanced processes tend to be operated in a simplified way with a fixed set of values for process parameters, leading to relatively unsatisfied removal efficiencies. This is mainly due to difficulties in incorporating a complicated numerical simulation model that is needed for process forecasting within a real-time nonlinear optimization framework that is critical for supporting process control."

The authors here developed a 3-D multiphase and multicomponent model to simulate subsurface contaminant transport. Next they employed a dual-response surface technique to "support development of an optimization model to determine optimum process operation conditions." Finally, authors developed a decision support system to "guide decisions of remediation process control...under various site conditions" (Huang and others, 2003).

Reviewer's comment: Thousands of surfactants are commercially available. Because hydrogeologic conditions and mixtures of contaminants vary from site to site, each site must be evaluated. Furthermore, the specific surfactant added may need to be adjusted as chemicals and contaminant ratios may change at a site.

Information sources

U.S EPA publications and information

Reports available to view or download at http://clu-in.org/techpubs.htm include:

Abstracts of Remediation Case Studies, Vol. 8 (EPA 542-R-04-012)

Guidance for Obtaining Representative Laboratory Analytical Subsamples from Particulate Laboratory Samples (EPA 600-R-03-027)

From U.S. EPA http://clu-in.org/studio:

IRTC In-Situ Chemical Oxidation

ITRC Phytotechnologies Technical and Regulatory Guidance and Phytoremediation Decision Tree

What is Remediation Process Optimization and How Can it Help Me Identify Opportunities for Enhanced and More Efficient Site Remediation?

Other EPA publications and Web sites:

Center for Subsurface Modeling Support (CSMoS), which provides public domain groundwater and vadose zone modeling software and services, http://www.epa.gov/ada/csmos.html

Cost and Performance Information on Cleanup Technologies, http://www.frtr.gov

New and Improved FRTR Remediation Optimization, http://www.frtr.gov/optimization

Performance Monitoring of MNA Remedies for VOCs in Ground Water (EPA 600-R-04-027), http://www.epa.gov/ada/download/reports/600R04027/600R04027.pdf

Other publications:

Assessing Ground Water Vulnerability to Contamination: Providing Scientifically Defensible Information for Decision Makers (USGS Circular 1224), http://water.usgs.gov/pubs/circ/2002/circ1224/pdf/circ1224_ver1.01.pdf

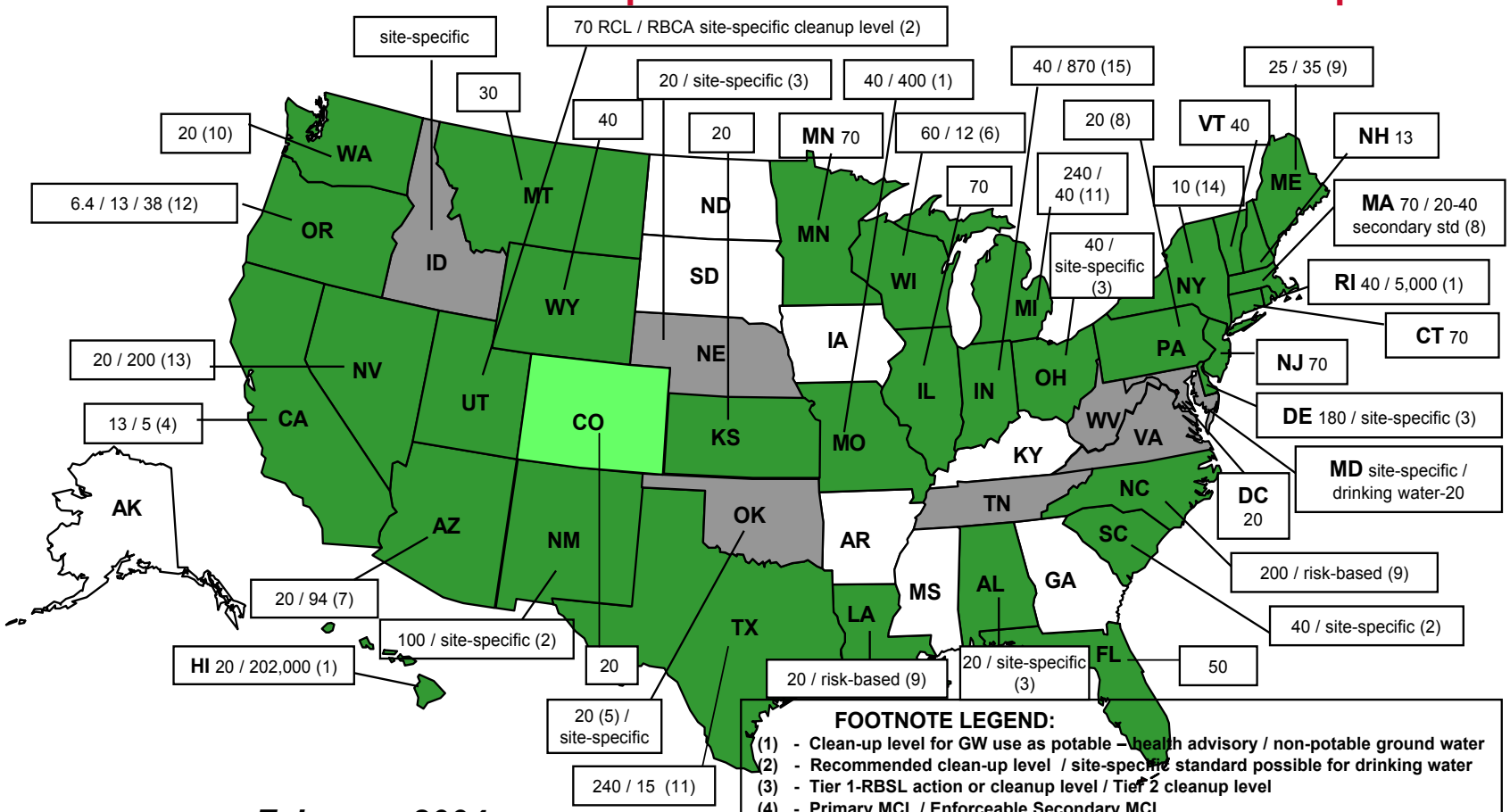
Innovative Remediation and Site Characterization Technologies Resource CD-ROM (EPA 542-C-04-002), http://www.epa.gov/ncepi

Nanoscience and Nanotechnologies: Opportunities and Uncertainties, http://www.nanotec.org.uk/finalReport.htm

Triad Resource Center, http://www.triadcentral.org

UTTU obtained many of these sites from TechDirect (http://clu-in.com/techdrct) and Ground Water Monitoring & Remediation (http://www.ngwa.org) and other publications. We thank the editors and writers for allowing us to reprint this material.

MTBE Groundwater Clean-up Levels for LUST Sites: Current & Proposed



FOOTNOTE LEGEND:

(1) - Clean-up level for GW use as potable – health advisory / non-potable ground water
(2) - Recommended clean-up level / site-specific standard possible for drinking water
(3) - Tier 1-RBSL action or cleanup level / Tier 2 cleanup level
(4) - Primary MCL / Enforceable Secondary MCL
(5) - Action level or level of concern / site specific cleanup levels
(6) - WDNR NR 140 Enforcement Standard / Preventative Action Limit goals
(7) - Receptor threatened or impacted / no receptor
(8) - Groundwater resource used as residential drinking water
(9) - Action level / cleanup level (i.e., ME 25 / 35)
(10) - Method A MTBE groundwater clean-up level at 20 ug/L
(11) - Health-based / aesthetic criteria
(12) - Tap water standards: residential / urban-residential / occupational uses
(13) - MTBE risk-based action levels (i.e., NV 20 / 200)
(14) - Enforceable groundwater guidance value & drinking water standard
(15) - Cleanup level: residential / industrial

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MTBE Cleanup Level (ug/L)

Site-Specific MTBE Clean-up Level (ug/L)

Projected 2004 MTBE Cleanup Level (ug/L)

Waiting for EPA MCL

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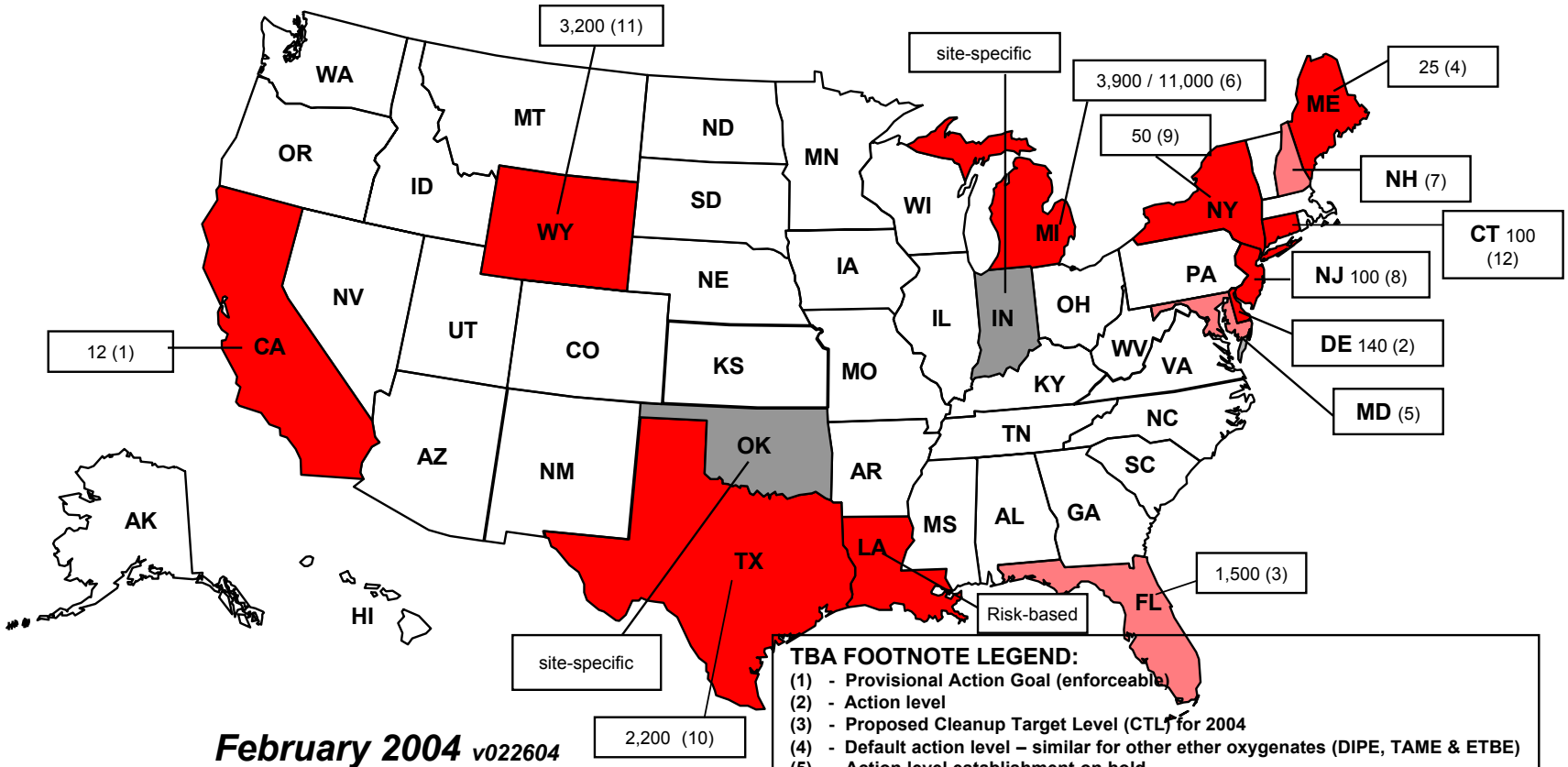
tert-Butyl Alcohol (TBA) Groundwater Clean-up Levels for LUST Sites: Current & Proposed



February 2004 *v022604*

- TBA Action / Cleanup Level (ug/L)
- Site-Specific TBA Clean-up Level (ug/L)
- Possible 2004 TBA Cleanup Level (ug/L)

TBA FOOTNOTE LEGEND:

(1) - Provisional Action Goal (enforceable)
(2) - Action level
(3) - Proposed Cleanup Target Level (CTL) for 2004
(4) - Default action level – similar for other ether oxygenates (DIPE, TAME & ETBE)
(5) - Action level establishment on hold
(6) - Residential drinking water / commercial-industrial land uses
(7) - Possible 2004 Rule Making Process for TBA, ETBE, DIPE & TAME
(8) - Interim Specific (IS) criteria for TBA cleanup value at 100 ppb
(9) - Enforceable Unspecified Organic Contaminant (UOC) regulated at 50 ppb - similar for other ether & alcohol oxygenates
(10) - Petroleum Storage Tank program (PST) typically does not request analysis, but if TBA is found, then development of cleanup levels may be required.
(11) - Action or cleanup goal. Proposed TBA change to 220 ppb.
(12) - Interim drinking water target level. Cumulative [MTBE + TBA] > 100 ppb may require drinking water supply replacement or groundwater cleanup.

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