ASSESSING AEROBIC NATURAL ATTENUATION OF TRICHLOROETHENE AT FOUR DOE SITES

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ABSTRACT

A 3-year Department of Energy Environmental Science Management Program (EMSP) project is currently investigating natural attenuation of trichloroethene (TCE) in aerobic groundwater. Determining whether TCE degradation occurs at meaningful rates under aerobic conditions via biological cometabolism has important implications for the assessment of natural attenuation. This presentation summarizes the results of a screening process to identify TCE plumes at DOE facilities that are suitable for assessing the rate of TCE attenuation under aerobic conditions. In order to estimate aerobic degradation rates, plumes had to meet the following criteria: TCE must be present in aerobic groundwater, a conservative co-contaminant must be present and have approximately the same source as TCE, and the groundwater velocity must be known. A total of 127 TCE plumes were considered across 24 DOE sites. The four sites retained for the assessment were: 1) Brookhaven National Laboratory, OU III; 2) Paducah Gaseous Diffusion Plant, Northwest Plume; 3) Rocky Flats Environmental Technology Site, Industrialized Area – Southwest Plume and 903 Pad South Plume; and 4) Savannah River Site, A/M Area Plume. The analysis indicates that TCE is being preferentially attenuated at environmentally significant rates under aerobic conditions with degradation half-lives ranging from 0.85 to 12 years in 8 of the 9 plumes evaluated.

INTRODUCTION

This paper summarizes and documents activities performed under one project task during the first 18 months of a 3-year project that is investigating natural attenuation of trichloroethene (TCE) in aerobic groundwater. TCE degradation under aerobic conditions has already been demonstrated in the Snake River Plain Aquifer at the Idaho National Engineering and Environmental Laboratory (INEEL), and this has led to the utilization of monitored natural attenuation (MNA) in place of pump-and-treat (Wymore et al. 2004). The purpose of this project was to identify TCE plumes at DOE facilities that are suitable for assessing the rate of TCE cometabolism under aerobic conditions and performing the assessment. This involved compiling and screening characterization information that describes TCE plumes throughout the DOE complex and evaluating spatial trends in contaminant concentration data to determine if TCE was being preferentially attenuated.

This paper includes a background summary of results of aerobic TCE attenuation at Test Area North (TAN) within the DOE’s INEEL, a description of the criteria and procedure for identifying and screening candidate DOE sites, and a description of the four DOE sites selected for further study, and the results of the analysis for selected sites.

Background

There are numerous plumes of TCE-contaminated groundwater at sites owned by DOE. Various approaches are employed to manage or remediate this contaminated groundwater. At the TAN site at the INEEL, a plume of TCE-contaminated groundwater extends approximately 2-miles downgradient from a well that was formerly used for injecting liquid wastes into the Eastern Snake River Plain Aquifer. Characterization and pilot studies performed under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) indicated that TCE was attenuating in the aerobic groundwater system, apparently by a microbial process. This process is believed to be cometabolism, in which enzymes produced by microorganisms to perform metabolic processes fortuitously also oxidize TCE to chloride, water, and carbon dioxide. Further studies at TAN indicated that the microorganisms that are probably responsible for cometabolizing TCE are methane-oxidizing and phenol-oxidizing populations.

The investigations at TAN that demonstrated that TCE attenuation was occurring at an environmentally significant rate (now estimated to have a half-life of about 13 years) and identified a credible mechanism led to selection of MNA as the CERCLA-defined remedy for most of the areal extent of the TCE plume. Selection of MNA, along with biostimulation in the source area, in lieu of the previously identified remedy, pump-and-treat (P&T), resulted in a life-cycle cost savings to DOE of approximately $23 million. To the best of our knowledge, the aerobic TCE plume at TAN is the first site in which MNA was selected as the remedy for a large TCE plume in an aerobic aquifer.

TCE also degrades under anaerobic conditions via a well-documented process known as anaerobic reductive dechlorination. This process commonly occurs in anaerobic groundwater but does not occur in aerobic conditions. Cometabolism is the only known microbial process by which TCE degrades biologically under aerobic conditions.

Determining that TCE degrades at an environmentally significant rate in aerobic groundwater and selection of MNA as a groundwater remedy are contrary to the widely-held belief that natural attenuation of TCE occurs only under anaerobic conditions, as reflected in *Technical Protocol for Evaluating Natural Attenuation of Chlorinated Solvents in Ground Water* (Wiedemeier et al., 1998). If it can be shown that TCE attenuation under aerobic conditions occurs at sites in addition to TAN, then it may lead to MNA being considered as a remedial alternative at other sites. Selection of MNA as a remedy could result in considerable cost savings.

If the cost savings projected for the TAN plume can be extrapolated to other DOE sites, then it is reasonable to expect that cost savings in excess of $100M could be achieved if MNA were selected as the remedy for other TCE plumes in aerobic aquifers. A major impediment to the widespread adoption of MNA is that the degree to which TCE cometabolism occurs in aerobic aquifers, and the rate at which it occurs, are poorly understood. It is not known if TCE cometabolism like that observed at TAN is a common occurrence, nor is the rate of TCE attenuation by cometabolism known for sites other than TAN.

A critical step in demonstrating that TCE was attenuating at TAN via a process in addition to dispersion, sorption, and volatilization, was demonstrating that TCE was preferentially removed or degraded relative to conservative solutes or tracers (i.e., tracers that are not sorbed, volatilized, or degraded). This was shown at TAN by comparing changes in TCE concentrations downgradient of the source to changes in concentrations of conservative tracers in the same plume. All solutes in groundwater are affected by dispersion, and thus, conservative tracer data can be used to account for the effects of dispersion on TCE concentrations along a flow path. At TAN, tritium and tetrachloroethene (PCE) are co-contaminants in the TCE plume. Tritium is a conservative solute in the aquifer. PCE may be slightly retarded by sorption but it is not biodegraded under aerobic conditions. The deep water table (>200 feet below ground surface) and thick contaminated zone (extending >200 feet below the water table) cause volatilization to be a very weak process in this plume. The analysis involved plotting the natural logarithm (Ln) of the ratio of TCE: tritium or TCE:PCE as a function of distance downgradient of the contaminant source. If there were no process that preferentially removed TCE, the Ln(ratio) values would be constant with distance, while a declining Ln(ratio) would indicate that TCE was being preferentially removed. The slope of a line through points at different distances downgradient of the source is related to both groundwater velocity and the degradation coefficient (assuming that TCE decays by a first-order process). This approach is described in "An Evaluation of Aerobic Trichloroethene Attenuation Using First-Order Rate Estimation" (Sorenson et al., 2000), and is referred to as the 'tracer-corrected method.'

At each of these sites, a co-contaminant derived from the same source area as TCE was used as a nonbiodegrading tracer to determine the extent to which concentration decreases can be attributed to abiotic processes such as dispersion and dilution. Any concentration decreases not accounted for by these processes must be explained by some other natural attenuation mechanism. Thus, "half-lives" presented herein are due to degradation processes in addition to attenuation that occurs due to hydrologic mechanisms. This "tracer-corrected method" has previously been used at the DOE's Idaho National Engineering and Environmental Laboratory in conjunction with other techniques to document the occurrence of intrinsic aerobic cometabolism. Application of this method to other DOE sites is the first step to determining whether this might be a significant natural attenuation mechanism on a broader scale.

For the purposes of this paper, the following initial requirements must be met in order to use the tracer-corrected method to calculate the rate of TCE cometabolism:

- TCE must be present in aerobic groundwater,
- A conservative co-contaminant must be present and have the same source and source function as TCE, and
- The groundwater velocity must be known.

IDENTIFICATION AND SCREENING OF CANDIDATE SITES

In order to evaluate the feasibility of utilizing MNA at other DOE sites , this project screened data from TCE plumes throughout the DOE complex to identify those with characteristics that are suitable for assessing the rate of cometabolism. The objective was to identify several TCE plumes that have characteristics needed for using the tracer-corrected method for determining the rate of TCE attenuation. After identifying suitable plumes, the degradation rate potentially attributable to cometabolism was determined. For an individual plume, determining the attenuation rate is the first step of evaluating the feasibility of using MNA as a component of a remedy for contaminated groundwater. In the bigger picture of all TCE plumes at DOE sites, determining whether or not TCE attenuation at environmentally significant rates under aerobic conditions is a common occurrence will indicate the likelihood that MNA can be used as a remedy component at numerous sites.

After the initial criteria were met, the screening criteria utilized to identify plumes for further study were:

- TCE concentrations >100 μg/L. Lower concentrations are more prone to analytical error and are unlikely to persist for substantial distances downgradient.
- TCE plume extent >1,000 ft.
- Aerobic conditions (defined here as dissolved oxygen concentration >2 mg/L)
- Several monitoring wells in the plume downgradient of the contaminant source, ideally located along or close to the same flow path.
- Conservative co-contaminant derived from the same source. The co-contaminants considered include tritium, PCE, technetium-99 (Tc-99), chloride, nitrate, and sulfate. These are all conservative under aerobic conditions, but only tritium would be conservative under anaerobic conditions.

The screening approach consisted of a data compilation and analysis process. This process included two main activities. First, candidate plumes were identified and geochemical characterization data were collected. Second, the characterization data were screened against the criteria listed above. Of these two activities, identifying candidate plumes and compiling relevant information was the more difficult and time consuming.

The four main avenues utilized to locate appropriate DOE sites and their associated data for this project were: data base queries, Internet searches, personal interviews, and report reviews. The *U.S. Department of Energy Ground Water Database*[a] was searched for DOE sites that contained TCE-contaminated groundwater plumes. The DOE sites that met the initial criteria were further researched to determine if they met the remaining criteria stated above. The DOE database led to Internet searches of DOE sites meeting the criteria. Many Internet searches were conducted using the “Microsoft Network” search engine. The name of the DOE site was used to conduct a search and find documents and data relating to the set screening criteria. Many of the DOE websites had links to their respective document libraries. One could then search the library for a particular document with its document number, a keyword(s) (e.g., TCE, groundwater, and aerobic), a specific title, etc. Conducting a keyword search within these DOE websites was the method used when specific titles or document numbers were unknown. These searches led to websites that related to the research being conducted for this project. These searches led to some appropriate documents and reports, but more importantly it led to contacts at the various DOE sites. Personal interviews allowed for individual contact of DOE personnel, and individuals were requested to identify plumes that might meet the screening criteria and to provide contact information for persons who would be able to provide site characterization data. Personal interviews were the most effective means of obtaining documents and data that were of concern to the project, and for identifying additional individuals who either were knowledgeable about groundwater contamination at specific DOE facilities or could provide data relevant to this evaluation. Various documents, reports, CDs, and data spreadsheets were obtained through personal contact via mail and email. Once characterization reports were obtained, they were examined to determine if the data needed were available and if plume maps and well location maps had been generated.

The plume identification and screening process led to a total of 127 TCE plumes at 24 DOE sites. Of these sites, four DOE sites were retained for assessment of aerobic TCE degradation rates: 1) Brookhaven National Laboratory, OU III; 2) Paducah Gaseous Diffusion Plant, Northwest Plume; 3) Rocky Flats Environmental Technology Site, Industrialized Area – Southwest Plume and 903 Pad South Plume; and 4) Savannah River Site, A/M Area Plume. It is important to reiterate that these four sites were selected not because they were the only ones that might have aerobic cometabolism of TCE, but because they had the best data available to evaluate whether this process might be important using the tracer-corrected method.

DESCRIPTION AND EVALUATION OF TCE ATTENUATION AT SELECTED SITES

The sites evaluated in this study are described in this section. They are presented in alphabetical order, as follows:

1. Brookhaven National Laboratory (BNL), Operable Unit (OU) III,

2. Paducah Gaseous Diffusion Plant, Northwest Plume,

3. Rocky Flats Environmental Technology Site (RFETS), Industrialized Area – Southwest Plume and 903 Pad South Plume, and

4. Savannah River Site (SRS), A/M Area.

Brookhaven National Laboratory, OU III

The BNL is located on Long Island near Upton, New York (BNL, 2003). The uppermost aquifer at the site is the Upper Glacial Aquifer, which is comprised of unconsolidated material. Groundwater at the BNL flows primarily to the south and southeast. There are numerous plumes of volatile organic compound (VOC)-contaminated groundwater beneath BNL, but we focused on OU III because our initial evaluation of the BNL site showed TCE is being preferentially attenuated. OU III is south of the southern site boundary and extends downgradient of a pump-and-treat system that is also located south of the site boundary.

TCE is a constituent of part, but not all, of the BNL OU III plume. Even when TCE is present, typically other VOCs are present at higher concentrations. Carbon tetrachloride (CT), which does not degrade under aerobic conditions, is a widespread contaminant at BNL and was used as the conservative tracer in this evaluation. Figure 1, the plot of Ln(TCE/CT), shows a trend of decreasing value with distance, suggesting that TCE is being preferentially attenuated relative to CT. This trend suggests, but does not prove, that preferential attenuation of TCE is occurring because the original ratio of TCE to other VOCs when the contaminants entered the groundwater is unknown. Hence, the possibility exists that the trend is an artifact of the ratio of VOCs released to groundwater changing over time instead of the result of ongoing TCE attenuation.

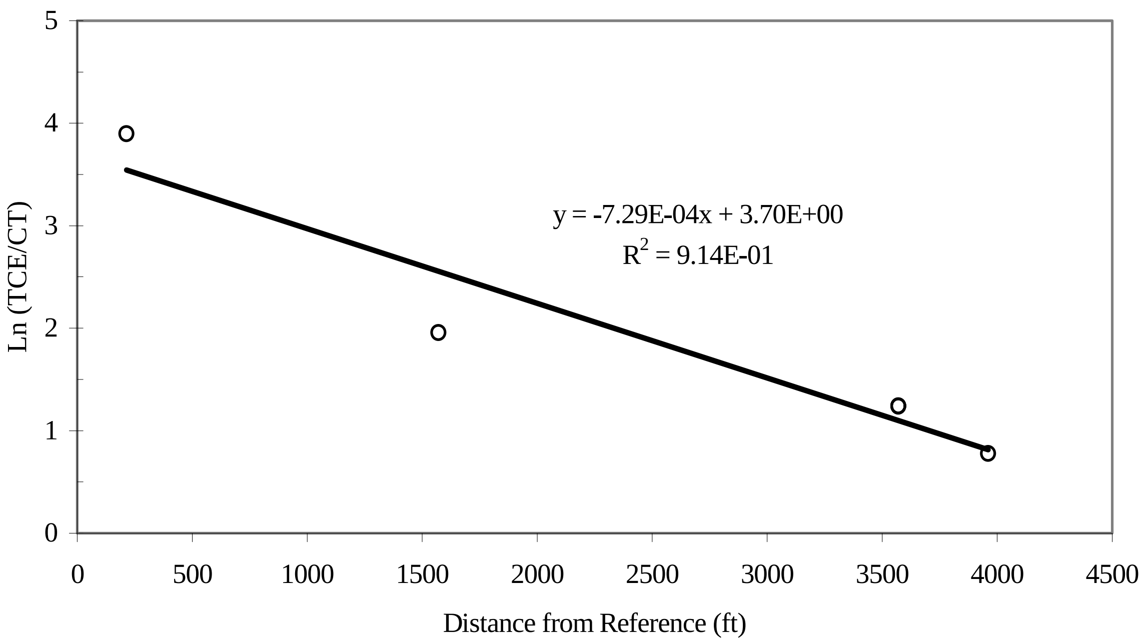


Fig. 1. Ln(TCE/CT) of the OU III plume at BNL.

The slope of the linear regression line through the Ln(TCE/CT) values for OU III (Figure 1) is the ratio of the first order decay coefficient and groundwater velocity. Multiplying the slope of the regression line by the average groundwater velocity, 100 to 500 ft/year assumed at BNL, yields the first order decay coefficient for TCE attenuation (Sorenson et al. 2000). The half-life of TCE attenuation is then determined by dividing Ln(2) by the decay coefficient. In OU III, TCE appears to be attenuating with a half-life of about 2 to 10 years, depending on the value assumed for groundwater velocity (500 to 100 ft/year, respectively). This rate appears to be faster than the degradation rate observed at the TAN site.

Paducah Gaseous Diffusion Plant, Northwest Plume

The Northwest Plume at the Paducah Gaseous Diffusion Plant, near Paducah, Kentucky, is in the Regional Gravel Aquifer (RGA) under aerobic conditions. The plume emanates from the industrialized portion of the site, where TCE is present in the subsurface as a dense, non-aqueous phase liquid, and extends about 3 miles northward far beyond the DOE property boundary. Paducah site contractor hydrogeologists have divided the RGA into Upper, Middle, and Lower horizons based on the depth where wells are completed. The lower portion of the Northwest Plume was chosen because it showed a more pronounced decreasing trend when compared to the middle and upper portions of the plume, in part because wells are located farther downgradient in the Lower portion of the plume.

Technetium-99 (Tc-99) was used as the conservative tracer for the lower portion of the Northwest Plume at Paducah. Preferential attenuation of TCE relative to Tc-99 was inferred from the decreasing trend with distance from the plot of Ln(TCE/Tc-99) (Figure 2). As noted above, the slope of the linear regression line through the Ln(TCE/Tc-99) values for the lower portion of the Northwest Plume (Figure 2) is the ratio of the first order decay coefficient and groundwater velocity. Multiplying the slope of the regression line by the average groundwater velocity, 1.3 ft/day at Paducah, (Clausen et al., 1997), yields the first order decay coefficient for TCE attenuation. The half-life of TCE attenuation is then determined as before, and results in a value of 7 years for the lower portion of the Northwest Plume. Again, this is a slightly higher degradation rate than that observed for the TAN site.

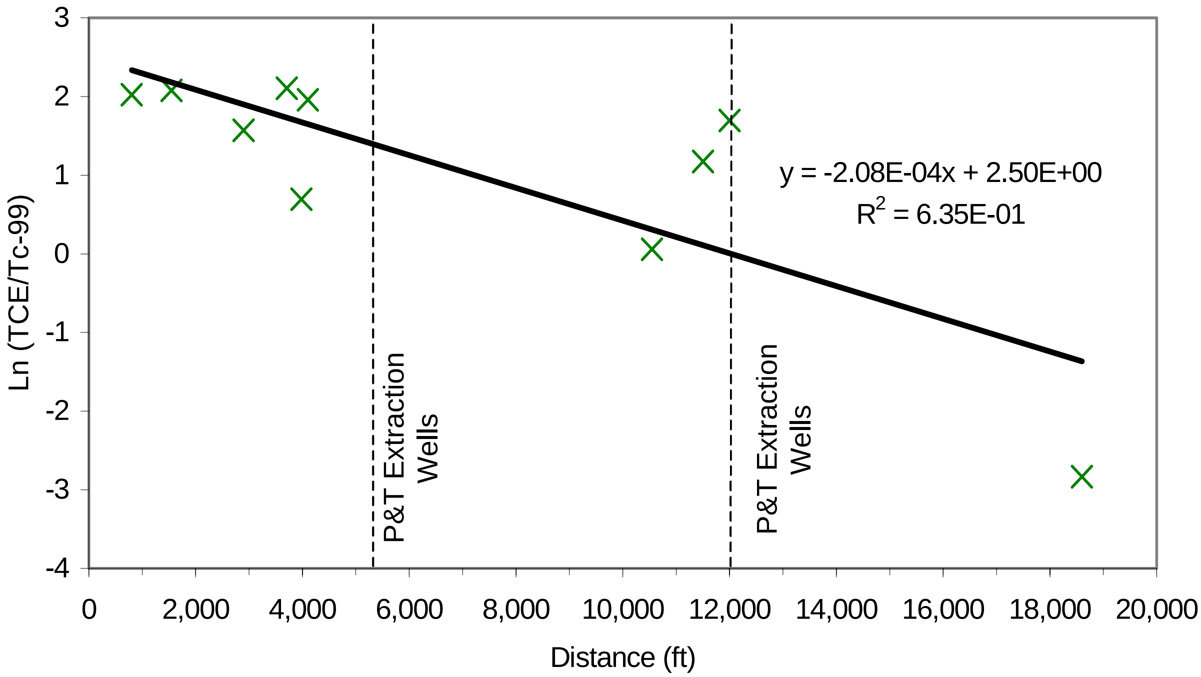


Fig. 2. Ln(TCE/Tc-99) in the lower portion of the Northwest Plume.

Rocky Flats Environmental Technology Site (RFETS), Industrialized Area – Southwest Plume and 903 Pad South Plume

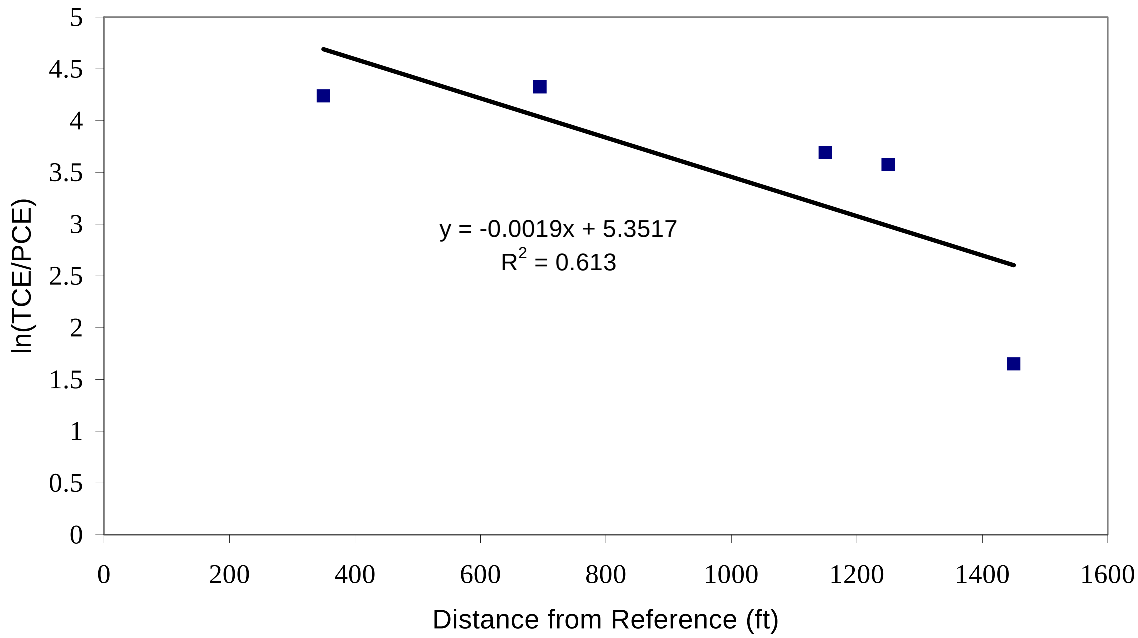
The RFETS is located near Denver, Colorado, and about 2 miles east of the Rocky Mountain Front Range. Information used in this evaluation was taken primarily from *Rocky Flats Environmental Technology Site Integrated Monitoring Plan FY 2002* (RFETS, 2002). The site is located on a pediment that slopes gently to the east. The industrialized portion of the site is on relatively flat area. Incised surface water drainages adjacent to the north, south, and east sides of the industrialized area affect local groundwater flow directions in the vicinity of the RFETS.

The evaluation of TCE attenuation focused on the upper aquifer, locally termed the Upper Hydrostratigraphic Unit (UHSU), which consists of the unconfined saturated zone including both unconsolidated and consolidated groundwater-bearing strata. Groundwater flow directions in the UHSU are influenced by local topography, with groundwater generally flowing from topographically high areas to topographically low areas. The incised channels adjacent to the industrialized area cause groundwater to flow in dramatically different directions in various parts of the site and therefore, plumes derived from individual contaminant sources extend in different directions in various parts of the site.

In the evaluation of TCE attenuation in aerobic groundwater at the site, five areas were identified in which a more-or-less discrete plume could be identified, or at least several wells were located along a flow-path downgradient of a contaminant source. In the two plumes retained for assessment, the 903 Pad South Plume and the Industrial Area Southwest Plume, the available data indicate the TCE may be attenuating relative to PCE, which is the conservative tracer in this environment.

The 903 South Plume emanates from the 903 Pad/Ryan’s Pit area and extends south toward a nearby incised drainage channel. TCE and PCE concentrations decline with distance along the flow-path (Figure 3). The ratio of TCE to PCE declines slightly throughout most of the plume and then sharply at the toe of the plume. Using the groundwater velocity estimated for the site of 155 ft/year as before yields a TCE degradation half-life of 2.4 years, quite short relative to TAN.

The Industrial Area Southwest Plume emanates from a source in the southwestern portion of the industrialized area and migrates generally southward along a curvilinear path toward an incised drainage. Although there is considerable scatter in the TCE and PCE concentration data, there is a trend of declining Ln(TCE/PCE) with distance suggesting attenuation of TCE (Figure 3). Calculating the TCE degradation half-life as before, with slope of the regression line through the Ln(TCE/PCE) values and a groundwater velocity of 170 ft/year, yields a TCE degradation half-life of 0.85 years.



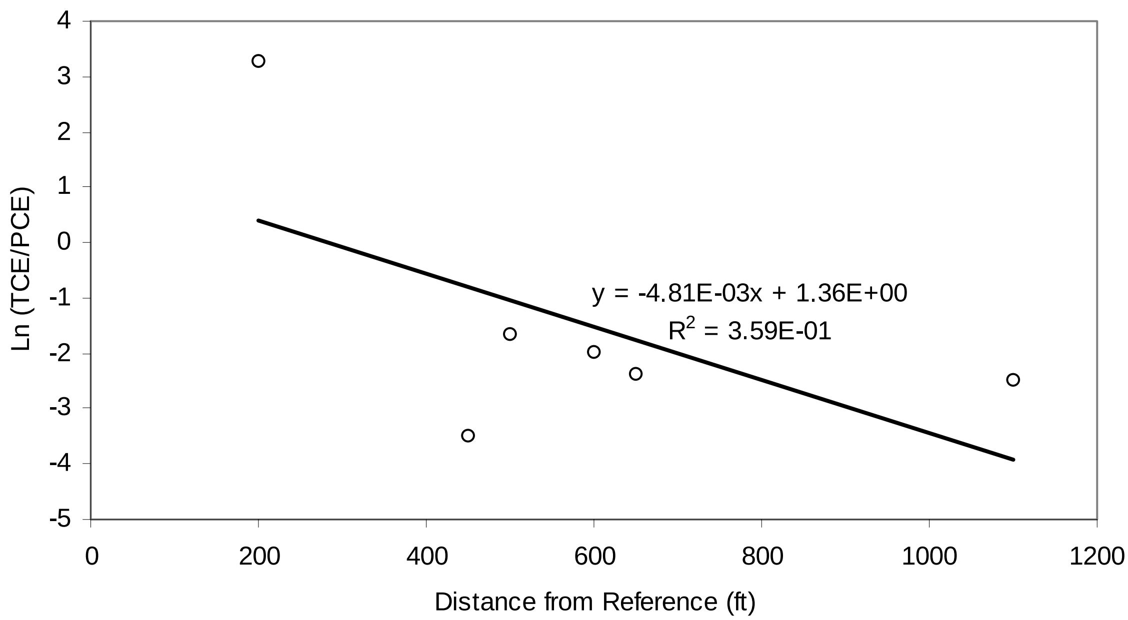


Fig. 3. Ln(TCE/PCE) for Rocky Flats Environmental Technology Site (903 Pad South Plume and the Industrialized Area Southwest Plume, respectively).

Savannah River Site, A/M Area

The SRS is located near Aiken, South Carolina. The portion of the site examined is the A/M Area, which is located in the northern portion of the site. The SRS lies within the Southeastern Coastal Plain. The Southeastern Coastal Plain is characterized by inter-bedded sands, silts, and clays that are typical of a shallow marine depositional environment. In the vicinity of the A/M Area, five permeable horizons contain TCE-contaminated groundwater. From shallowest to deepest, these include:

- M-Area Aquifer,
- Upper Lost Lake Aquifer,
- Lower Lost Lake Aquifer,
- Middle Sand of the Crouch Branch Confining Unit, and
- Crouch Branch Aquifer.

Each of the permeable horizons was evaluated, and four of the five aquifers show indications of TCE attenuation based on declining trends in the plots of Ln(TCE/PCE) (Figure 4). The Middle Sand of the Crouch Branch Confining Unit and the Crouch Branch Aquifer show trends with the largest decline, both with R^2 values of 0.7. The TCE degradation half-life is about 4 years in both units. The M-Area Aquifer and the Lower Lost Lake Aquifer have declining trends with smaller R^2 values, 0.27 and 0.38, respectively. TCE degradation half-lives of 4 and 12 years, respectively, were calculated for these aquifers. The trend in the Upper Lost Lake Aquifer has such a flat slope and low R^2 value, 1.29 x $^{10-4}$ and 0.03, that no degradation was apparent. These results are summarized in Table I.

Table I. Summary of TCE attenuation evaluation for the SRS A/M area.

Aquifer Zone	Regression Line Slope (ft^{-1})	R^2	Groundwater Velocity (ft/year)	Degradation Rate Coefficient ($year^{-1}$)	Degradation Half-Life (years)
M-Area Aquifer	-9.29E-4	0.27	192	1.78E-1	3.9
Upper Lost Lake Aquifer	-1.29E-4	0.03	241	NA	NA
Lower Lost Lake Aquifer	-2.53E-4	0.38	225	5.69E-2	12
Middle Sand of the Crouch Branch Confining Unit	-7.29E-4	0.73	235	1.71E-1	4.0
Crouch Branch Aquifer	-6.87E-4	0.67	262	1.80E-1	3.8

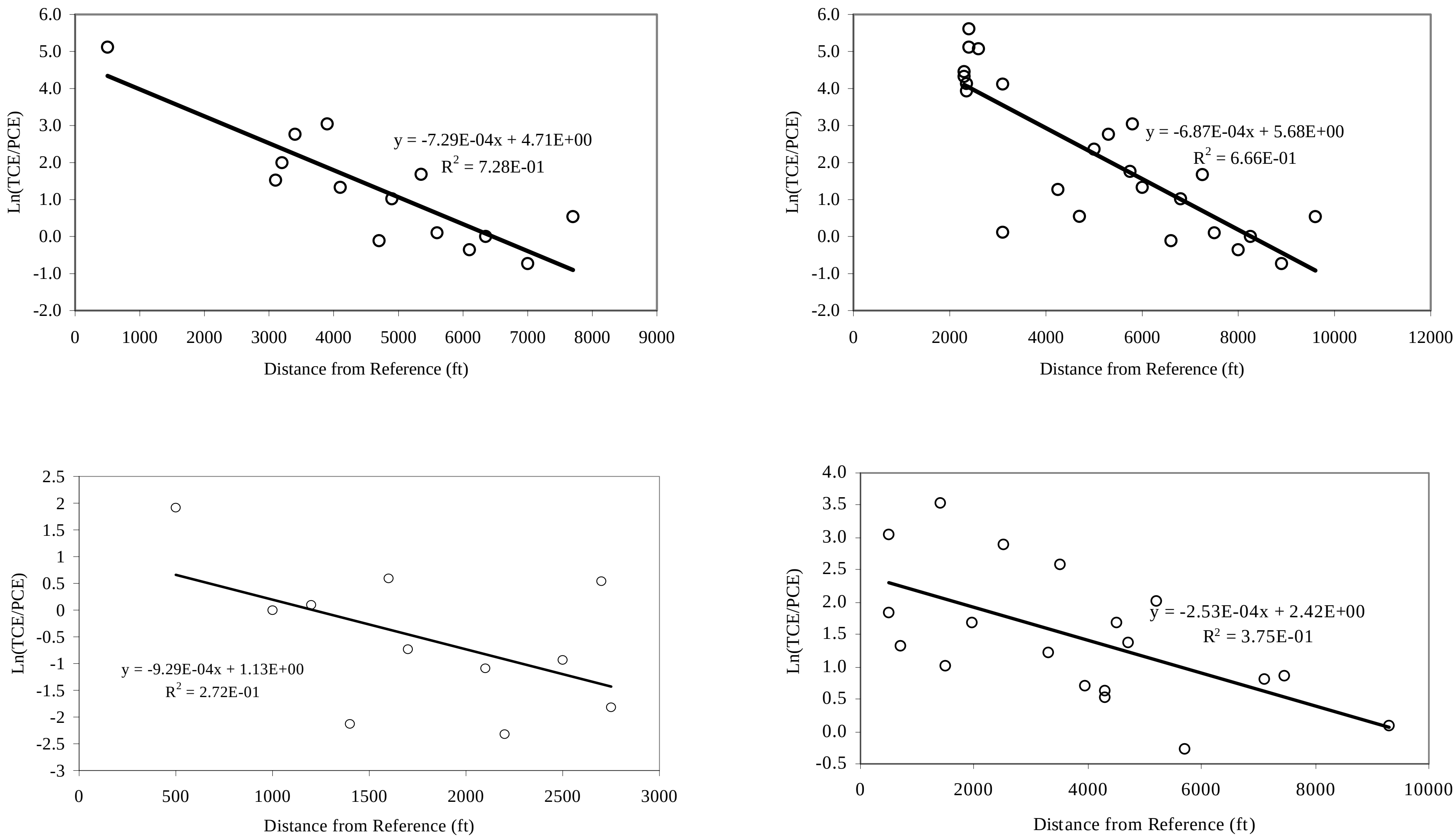


Fig. 4. Ln(TCE/PCE) of the SRS A/M Area (Middle Sand of the Crouch Branch Confining Unit, Crouch Branch Aquifer, M-Area Aquifer, Lower Lost Lake Aquifer, respectively).

CONCLUSION

At every one of the four sites evaluated, significant evidence of aerobic TCE attenuation relative to co-contaminants was observed. There were also plumes in which TCE attenuation could not be demonstrated, typically due to lack of a suitable conservative tracer or a well network configuration that was not suitable for this evaluation. The TCE degradation half-lives ranged from 0.85 to 12 years in the four DOE sites evaluated. The occurrence of aerobic TCE attenuation at such a high percentage of sites that had sufficient data for the analysis suggests that TCE attenuation under aerobic conditions may commonly occur at many sites.

While MNA might not be a feasible remedy at some of the sites evaluated due to the presence of other contaminants, these contaminants have made it possible to determine that TCE degradation under aerobic conditions might be more common than previously thought. If aerobic TCE degradation can be confirmed at these sites, this attenuation mechanism might be important much more often than previously thought, and might make MNA feasible where it was previously not considered. A further step in the evaluation of whether aerobic cometabolism of TCE in groundwater is responsible for the degradation observed is to apply novel analytical techniques that provide definitive evidence that TCE cometabolism is occurring by assaying the microbial enzymes responsible for TCE cometabolism. These techniques have now been demonstrated at the TAN site (Wymore et al. 2004, Wymore et al. 2005a, and Wymore et al. 2005b), and are ready for application at other sites.

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DISCLAIMER

Any opinions, findings, and conclusions or recommendations expressed in this material are those of the author(s) and do not necessarily reflect the views of the Department of Energy.

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FOOTNOTES

[a] This database, which is being added to the DOE Central Internet Database, can be obtained from Blaine Rowley (DOE; telephone [301] 903-2777).