BACK-DIFFUSION AND FUTURE ENERGY COST IMPLICATIONS FOR DNAPL REMEDIATION STRATEGIES

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ABSTRACT

A hypothetical DNAPL site is developed to facilitate a qualitative evaluation of the influence that back-diffusion and sustainability issues can have on remedial decision-making at DNAPL sites. The site source zone consisted of 12 individual pools with pool thicknesses ranging from 0.03 to 0.36 metres (m). A Tier 1 evaluation of five DNAPL source alternatives and two plume containment alternatives was conducted. Enhanced pump-and-treat was shown to have a significant mass removal benefit that is often ignored during remedial assessments. Thermal treatment appears to be the best alternative when long-term plume management (for decades or longer) is not required. When back-diffusion causes long-term groundwater exceedances of clean-up criterion, then enhanced in-situ bioremediation, enhanced pump-and-treat, Monitored Natural Attenuation, and thermal treatment have similar costs under a business-as-usual scenario. If future inflation rates are expected to increase due to rising energy or carbon dioxide offset costs, then a more detailed Tier 3 lifecycle analysis is recommended to consider the use of alternative energy sources and quantitative sustainability metrics before making a remedial decision. This study demonstrates that decision-making for DNAPL site management under today's economic and changing climate conditions is strongly influenced by the relative priority assigned to various decision-making metrics, and can be significantly influenced when back-diffusion causes long-term groundwater exceedances in the downgradient aqueous plume.

1. INTRODUCTION

Over the past five to ten years, a great deal of attention has been focused on the viability of DNAPL source zone treatment. Treatment approaches can include physical or hydraulic containment, or enhanced mass removal using chemical, biological, hydraulic, and/or thermal technologies. Less attention has been focused on decision-making considerations for integrated source zone and plume management remedial alternatives.

Recent findings (e.g. Chapman and Parker, 2005; Parker et al., 2008) indicate that even with complete isolation of a DNAPL source zone, contributions arising from back-diffusion from low-permeability sediments can cause long-term groundwater exceedances of clean-up criteria in downgradient aqueous plumes. This raises the question of how aggressively source zone treatment should be conducted when back-diffusion may create a longer-lasting problem than mass discharge from the remediated or contained source zone. Challenges involved with the remedial decision-making process for DNAPL sites is compounded by uncertainty in future inflation rates, costs of carbon dioxide emissions, and energy utilization rates. There is also uncertainty in the potential impact that remediation may have on the environment.

The purpose of this study is to qualitatively evaluate how back-diffusion and sustainability issues can influence remedial decision-making for DNAPL sites. The relevance and types of trends arising from back-diffusion are demonstrated. A hypothetical site scenario is used to compare remedial costs, mass removal efficiency, remediation timeframe, and sustainability issues for various source treatment and plume management alternatives.

2. BACK-DIFFUSION IMPLICATIONS FOR PLUME MANAGEMENT

Chapman and Parker (2005) and Parker et al. (2008) describe sites in Connecticut and Florida, respectively, where DNAPL source zones were physically isolated and long-term tailing in downgradient aqueous plume concentrations was observed. At the Connecticut site, the groundwater TCE concentration at a distance of 330 metres downgradient from the isolated source declined approximately 90% to 95% at three monitoring wells six years after the DNAPL source zone had been isolated. At the Florida site, total volatile organic compound (VOC) mass discharge had declined 90% to 99% in the five year period following source isolation. At both sites, the groundwater concentrations were expected to decline faster than had been observed because of the relatively fast travel time between the source zone and the downgradient monitoring locations. Chapman and Parker (2005) and Parker et al. (2008) demonstrate through high-resolution sampling and modelling that the persistent concentrations in the aqueous plumes at each site were caused by back-diffusion of TCE from silt and clay layers that had been exposed to high aqueous concentrations for a period of decades prior to source isolation. Parker et al. (2008) show with modeling that even clay layers with a thickness of only 0.2 metres can provide relatively high storage of organic pollutants, and clay layers as thick as 2 metres can cause groundwater problems that persist for many decades.

Figure 1 presents a schematic of how concentrations decrease over time at different distances downgradient from the source zone once it has been contained or remediated. The fastest decline in concentration occurs near the source zone, and changes to concentration occur more slowly with increasing distance from the source zone. Figure 1 also demonstrates that groundwater concentrations increase with distance from the source during back-diffusion, which is analogous to a line source of mass flux at the top of a low-permeability layer parallel to groundwater flow.

3. SITE SCENARIO

A hypothetical site scenario was developed to evaluate the potential influence of back-diffusion and sustainability considerations on remedial decision-making for DNAPL sites. The saturated thickness of the mildly heterogeneous sand aquifer is approximately 15 metres (m), and the aquifer overlies a thick, continuous clay aquitard. The average aquifer hydraulic conductivity is 0.01 cm/s, the horizontal hydraulic gradient is 0.01, porosity is 0.35, and the average linear groundwater velocity is approximately 0.3 m/day. A river representing a regional discharge boundary is 300 m downgradient of the site source zone, and the property boundary is located 50 m downgradient of the source zone.

The site is conceptualized as having had TCE DNAPL releases over time, the last release occurring approximately 20 years ago. Over the two decades since the last release, the vertical ganglia have been depleted due to natural dissolution, and only the horizontal DNAPL pools remain. The source zone for the hypothetical site included 12 distinct pools based on the example presented in Anderson et al., 1992. Initial pool lengths, widths and height are variable and are described in Carey and McBean, 2009. Pool heights ranged from 0.03 to 0.36 m, and initial pool surface areas ranged from 4 to 18 m^2. The total mass of TCE DNAPL in the pool phase in the source zone was approximately 1,200 kilograms (kg) at the start of this scenario 20 years ago, with individual pools having an initial DNAPL mass ranging from 31 to 271 kg. The overall length, width, and depth of the source zone are 4, 13.5, and 15 m, respectively, resulting in an approximate DNAPL source zone volume of 810 m^3.

The source zone dimensions are similar to the New Hampshire site PCE DNAPL source zone described by Guilbeault et al., 2005. The New Hampshire DNAPL source zone has a length, width, and depth of approximately 5, 17, and 14 m, respectively, resulting in a DNAPL source zone volume of approximately 1,200 m^3. A transect of vertical monitoring locations located 3 m downgradient of the New Hampshire site source zone indicated that there were 15 maxima of PCE concentrations, and a cross-section illustrates that individual DNAPL layer widths perpendicular to groundwater flow are on the order of several metres in the source zone, which is consistent with the hypothetical source zone pool dimensions.

A three-dimensional groundwater flow and chemical transport model was developed to simulate the extent of the aqueous plume downgradient from the hypothesized source zone. The model utilized uniform horizontal spacing of 5 m and a vertical spacing of 0.1 m. The model incorporated a longitudinal dispersivity of 3 m, a transverse horizontal dispersivity of 0.3 m, and a transverse vertical dispersivity of 0.003 m. Biodegradation and retardation of TCE were assumed to be negligible in the aerobic, sandy aquifer. Figure 2 shows the simulated horizontal extent of the aqueous plume, which has a width of approximately 90 m at the river boundary.

Figure 3 presents the simulated concentration of TCE directly above the aquitard, as well as the average concentration of TCE in the 1.5-metre portion of aquifer above the aquitard, which represents the concentration that would be measured using a monitoring well with a 1.5-metre screen length. Figure 3 demonstrates that the concentration of TCE just above the aquitard is relatively high in the downgradient plume, which confirms that there has been a persistently high gradient driving TCE diffusion into the clay aquitard prior to source remediation.

4. REMEDIAL ALTERNATIVES EVALUATION

Sale et al. (2008) present a 14-compartment model which can be used as a graphical tool to evaluate which portions of the subsurface represent long-term sources to groundwater and soil vapour contamination. The site scenario used for this study is assumed to have negligible vapour issues. Based on the 14-compartment model, a simplified analysis of the potential for back-diffusion from different regions of the subsurface is presented in Table 1. This demonstrates that the clay aquitard in the source zone, on-site and off-site regions of the aqueous plume will contribute to long-term groundwater exceedances of the clean-up criterion.

A simple Tier 1 analysis of remedial costs was conducted to compare the relative cost-effectiveness of different alternatives for source treatment and plume management. For this study, only containment of the plume just upgradient of the river was considered. A future study will be conducted to provide a more detailed quantitative assessment of lifecycle costs and sustainability metrics for each of the alternatives considered here.

4.1 Source and Plume Alternatives

Ten remedial alternatives were evaluated as part of this Tier 1 assessment, including five source remediation technologies: pump-and-treat (P&T), enhanced in-situ bioremediation (EISB), permeable reactive barrier (PRB), thermal treatment using electrical resistance heating (ERH), and Monitored Natural Attenuation (MNA). P&T was initially evaluated as a containment option utilizing a total pumping rate (7 gallons per minute, or gpm) equal to the hydraulic rate required to contain a 15-metre wide source zone, using one pumping well situated approximately 10 m downgradient of the source zone. A screening analysis indicated that this was not as effective an option as an "enhanced" P&T alternative that utilized a pumping rate equal to four times the rate required to maintain capture (i.e. 28 gpm) to enhance mass dissolution from the DNAPL source zone. The enhanced P&T alternative using the higher pumping rate was utilized in the remainder of this Tier 1 remedial analysis.

Each source treatment alternative was coupled with one of two plume containment technologies: P&T or PRB. The P&T plume containment alternative used three groundwater extraction wells situated approximately 20 m upgradient from the river. A three-dimensional groundwater flow model was used to confirm the pumping rates required for containment of the source zone and the aqueous plume. Model simulations indicate that a pumping rate of 60 gpm is required to contain the plume when the enhanced P&T source treatment alternative is not used. When the enhanced P&T alternative is coupled with the plume P&T alternative, then the total extraction rate required to contain the plume decreases to 50 gpm. The groundwater flow model was also used to determine that the horizontal hydraulic gradient was 3 times higher in the source zone for the enhanced P&T source treatment alternative. This enhanced gradient was utilized in the POOL model to evaluate remediation timeframe and mass discharge versus time for the enhanced P&T alternative (see next section).

Source treatment alternatives that cause a reduction in interfacial tension and thus potential DNAPL mobilization, such as surfactant enhanced aquifer restoration (SEAR) or EISB using solubility-enhancing electron donor concentrations, were not considered in this analysis because of their potential to cause expansion of the pool-dominated DNAPL source zone. These remedies may be suitable for other sites.

4.4 Remediation Timeframe Estimation

To determine the total remedial cost for each alternative, it was necessary to estimate the approximate remediation timeframe for each of the source treatment and plume containment alternatives. The remediation timeframe for thermal source treatment was assumed to be one year based on empirical data available at sites with similar geologic characteristics (e.g. Battelle, 2007). The POOL model (Carey and McBean, 2009) was utilized as a screening tool to estimate the remediation timeframe for source treatment alternatives including enhanced P&T, EISB, and PRB and MNA which were assumed to have the same remediation timeframe as the natural dissolution alternative evaluated using the POOL model. POOL includes input parameters for enhancements to the horizontal hydraulic gradient for a specific time period (e.g. enhanced P&T), as well as an enhanced mass dissolution rate (e.g. EISB). The EISB alternative was assumed to cause an enhanced mass dissolution rate equal to 3 times the natural dissolution rate, or an increase in 200% in the mass dissolution rate, as per ITRC (2008).

Figure 4 presents the simulated mass discharge versus time for natural dissolution (i.e. MNA or PRB source alternatives), typical P&T at low pumping rate, and the enhanced P&T alternative. As discussed in Carey and McBean (2009), the mass discharge versus time curve follows an exponential decline trend because of the large variability in pool heights assumed for this scenario.

Table 2 presents the estimated remediation timeframes for the source treatment alternatives, as well as the half-life calculated based on POOL-simulated mass discharge versus time for the enhanced P&T, PRB, and MNA alternatives. For comparison, the remediation timeframe and mass discharge half-life for the typical P&T alternative using a lower pumping rate is also presented in Table 2. The enhanced P&T alternative is shown to have a remediation timeframe of 26 years, compared to the remediation timeframes for typical P&T (lower pumping rates), EISB, and MNA or PRB of 51, 21, and 62 years, respectively. This illustrates that enhanced P&T can have a significant mass reduction benefit which is commonly not considered when evaluating remedial alternatives, because P&T is typically evaluated solely as a containment alternative. The extent of benefit that can be realized from an enhanced P&T alternative will depend on the magnitude of the increase in horizontal hydraulic gradient in the source zone, the degree of heterogeneity in hydraulic conductivity, and the potential for mass transfer limitations that may occur when the groundwater velocity is very fast.

It was assumed that other than thermal treatment, each of the other source treatment alternatives were limited to a 90% reduction in mass discharge from the source zone which was assumed to be sufficient. It is recognized that a reduction of 90% in mass discharge downgradient of a pool-dominated source zone may not be sufficient at some sites.

4.3 Capital and O&M Cost Calculations

Table 3 presents the estimated capital and annual operating and maintenance (O&M) costs, including the references used to provide a basis for derivation of the cost estimates. For the EISB source alternative it was assumed that a long-lived electron donor such as an emulsified oil solution (EOS) was injected every two years. The potential effect of enhanced biodegradation downgradient of the source zone was not considered in this analysis. As shown in Table 3, the PRB source treatment and plume containment alternatives assume a periodic change-out of the reactive wall materials, with a faster rate of change-out for the source treatment PRB alternative.

4.4 Net Present Value Cost Analysis

Two timeframes were considered for plume containment alternatives in this analysis:

a) back-diffusion is not significant and only 5 years of plume containment is needed after source remediation is achieved; or

b) back-diffusion is significant and requires long-term plume management for many decades after the start of remediation.

The sensitivity of the net present value (NPV) costs to future inflation rates was evaluated by using two discount rates: a) 2.8% (USOMB, 2008) for a business-as-usual scenario; and b) 0.8% to consider a higher-inflation rate scenario. (Note - the discount rates are adjusted for inflation as described in USOMB, 2008.)

Scenario A - Negligible Back-Diffusion Influence

Scenario A represents the situation where back-diffusion is assumed to have negligible contributions to groundwater concentrations in the plume downgradient of the source zone, relative to the clean-up criterion. Figures 5.a and 5.b compare the total NPV for each of the combined source-plume management alternatives based on discount rates of 2.8% (business-as-usual) and 0.8% (higher inflation), respectively.

Comparison of Figures 5.a and 5.b demonstrates that the thermal alternative for source treatment, combined with either P&T or PRB for short-term plume containment, is the most cost-effective alternative. Thermal remediation often results in an order of magnitude or higher source mass depletion relative to the other source treatment technologies, which is another reason why thermal treatment is the most effective alternative for this scenario. Thermal treatment is an energy-intensive process, although the short remediation timeframe and relatively low energy prices under current economic conditions indicate that this is still an attractive alternative relative to EISB assuming that sustainability is a lower priority than cost-effectiveness and mass removal efficiency. Even if sustainability is a high priority in the decision-making process, the significant cost difference between the EISB and thermal source treatment alternatives suggests that thermal treatment is the best solution.

If an alternative energy source was used for the P&T alternative for source treatment and plume containment, then this may be a reasonable alternative candidate to thermal under the business-as-usual scenario (although this needs to be confirmed with a Tier 3 lifecycle analysis). However, given the uncertainty in estimating future inflation rates (e.g. due to rising energy costs or the need to purchase expensive carbon offset credits at some point in the future), thermal appears to be the least risky of the source treatment alternatives (Figure 5.b).

Given the short-term nature of the plume management requirements with negligible back-diffusion influence, either a PRB or pump-and-treat could be used for containment of the downgradient plume if there is relatively low risk of increases to future inflation rates (Figure 5.a). If it is expected that energy and carbon offset costs will increase significantly in future (Figure 5.b), then using a PRB appears to be a better alternative than pump-and-treat for plume containment because it is a more sustainable alternative (i.e. PRB has less intensive energy and labour costs). A Tier 3 lifecycle cost and sustainability metrics analysis is needed to confirm that a PRB is a better alternative than pump-and-treat when there is a high risk of increasing inflation or energy costs.

Scenario B - Significant Back-Diffusion Influence

For the scenario where back-diffusion causes sustained long-term exceedances of the cleanup criterion in the aqueous plume downgradient from the source zone, the remedial decision is not as clear. For example, Figure 5.c compares the total NPV costs for each of the combined source-plume management alternatives for a discount rate of 2.8% (after adjusting for inflation) for this back-diffusion scenario.

MNA (i.e. natural dissolution of the DNAPL source zone) is the most cost-effective source treatment alternative because there is no incremental benefit (with respect to cost) for early clean-up of the source zone. This is because back-diffusion under this scenario requires plume management for a longer period than the time required for natural dissolution to exhaust the DNAPL source. For the MNA source alternative, the NPV cost for the pump-and-treat or PRB plume containment alternatives are similar assuming the business-as-usual inflation rate.

There is a relatively small cost difference between MNA and the enhanced P&T alternatives for source treatment when P&T is used for plume containment. This is due to the small incremental cost of using enhanced P&T for source treatment when the same P&T technology is also being used for containment of the downgradient plume. When considering the efficiency in mass removal and remediation timeframe, the best alternatives may be EISB or thermal treatment combined with a PRB for plume containment because of the relatively small difference in cost for these and the former alternatives discussed above. If sustainability is a high priority in the decision-making process, then perhaps EISB would be the best alternative for source treatment because it may require less intensive energy requirements than thermal treatment (although this needs to be confirmed with a Tier 3 lifecycle analysis). If a fast remediation timeframe is a high priority for remedy selection, then thermal is by far the most effective source treatment alternative.

When the risk of higher inflation is considered by using a 0.8% discount rate (Figure 5.d), then the most effective solution more clearly involves the use of a PRB for plume containment, and either EISB or thermal for source treatment. These two alternatives are similar in price to the MNA alternative but result in faster remediation timeframes. The final decision would depend on the relative priority assigned to sustainability, mass removal efficiency, and remediation timeframe considerations.

5. SUMMARY

To study the influence of back-diffusion and sustainability considerations in DNAPL site remediation, a hypothetical site scenario was developed. The TCE source zone consisted of 12 DNAPL pools in a relatively small soil volume (approximately 810 m^3), with a total initial DNAPL mass of 1,200 kg and a range of 31 kg to 271 kg DNAPL in each individual pool. Groundwater flow was assumed to be relatively fast (0.3 m/day) in the mildly heterogeneous sand aquifer overlying a thick, continuous clay aquitard. Five remedial alternatives were considered for source treatment involving mass removal or containment approaches: MNA, enhanced P&T, EISB, PRB, and thermal. Two remedial alternatives were considered for plume containment: P&T and a PRB.

For the scenario where long-term management of the downgradient plume was not a consideration, the best alternative for the pool-dominated DNAPL source zone was thermal treatment. For this source treatment alternative, there appears to be little difference in benefit for P&T or a PRB for short-term plume containment although this needs to be confirmed with a Tier 3 lifecycle assessment.

If long-term plume containment is required because of sustained back-diffusion contributions from the underlying aquitard, then it becomes less clear which remedial alternative is the most effective. Under the scenario where there is a low risk of increasing inflation rates, then MNA, EISB, enhanced P&T, and thermal treatment are generally similar in total NPV costs using either P&T or a PRB for plume containment. Thermal treatment has a substantially faster remediation timeframe, and EISB or MNA may be more sustainable then a thermal alternative. When considering the potential for higher inflation due to rising energy and carbon credit costs, then a PRB may be the most efficient choice for plume containment. The final decision will depend on the relative priority assigned to cost, remediation timeframe, sustainability metrics, and the degree to which alternative energy sources can improve the sustainability and reduce long-term costs of P&T alternatives.

These findings are subject to uncertainty due to the simplified Tier 1 remedial alternative analysis. A Tier 3 lifecycle cost and sustainability metrics analysis is recommended prior to making any conclusions regarding the relative merits of each alternative. This study demonstrates that decision-making for DNAPL site management under today's economic and changing climate conditions is strongly influenced by the relative priority assigned to various decision-making metrics, and can be significantly influenced when back-diffusion causes long-term groundwater exceedances in the downgradient aqueous plume.

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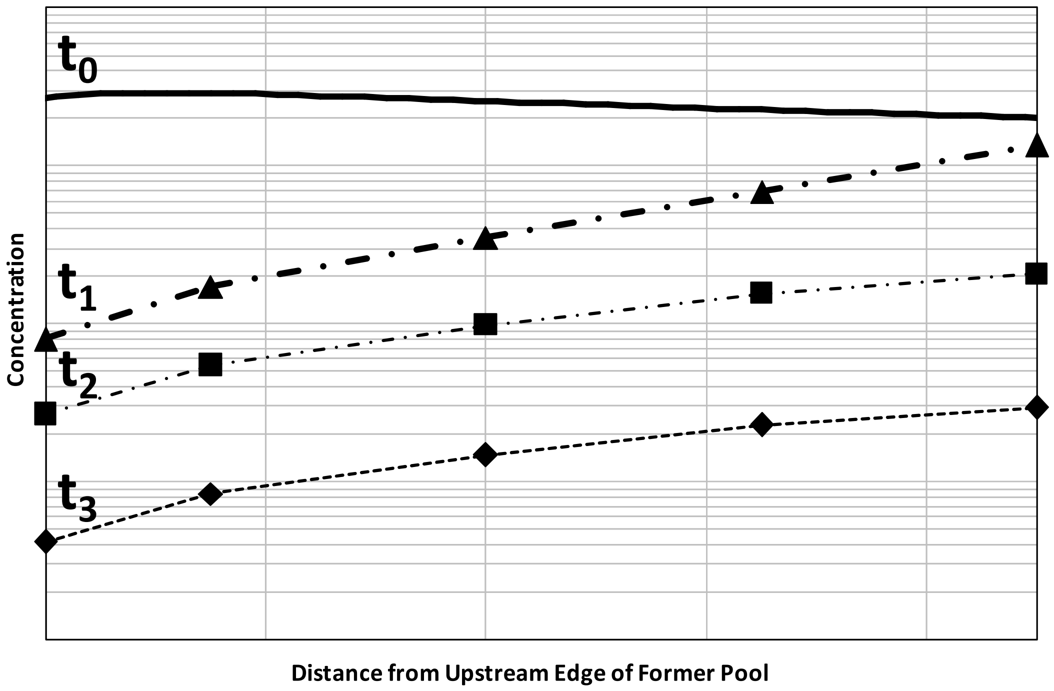


Note: t = time after pool mass is exhausted.

Figure 1 – Potential Concentration versus Distance Trends Due to Back-Diffusion Downgradient of a DNAPL Source

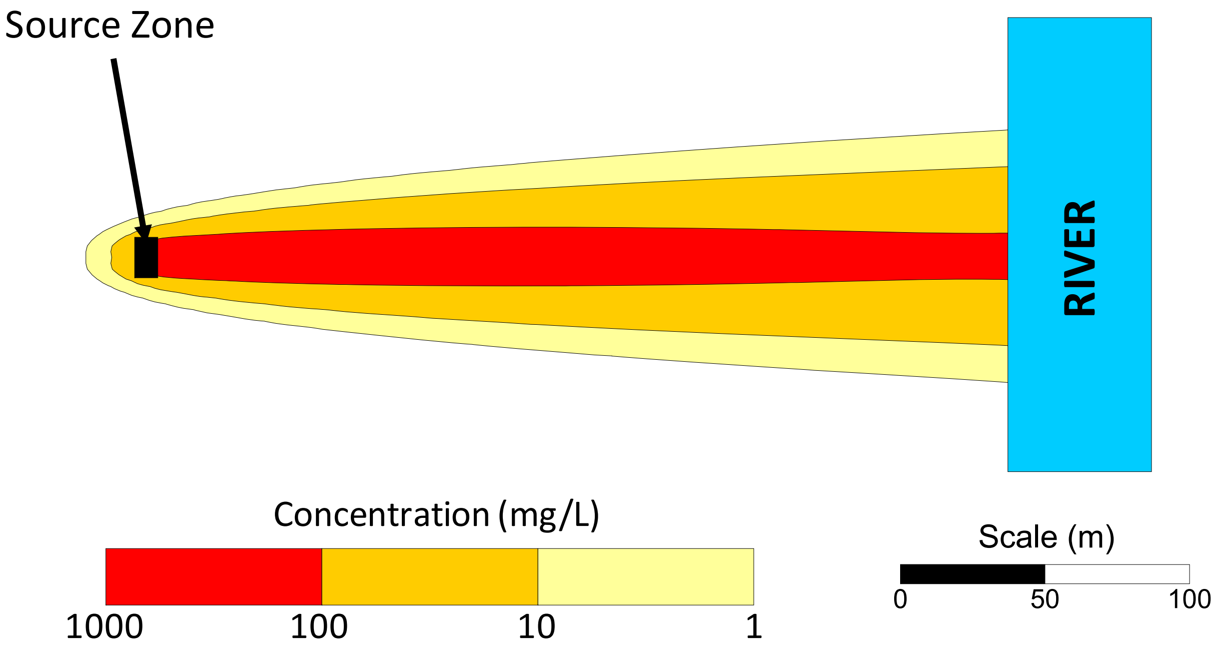


Figure 2 –Aqueous Plume Downgradient of Source Zone

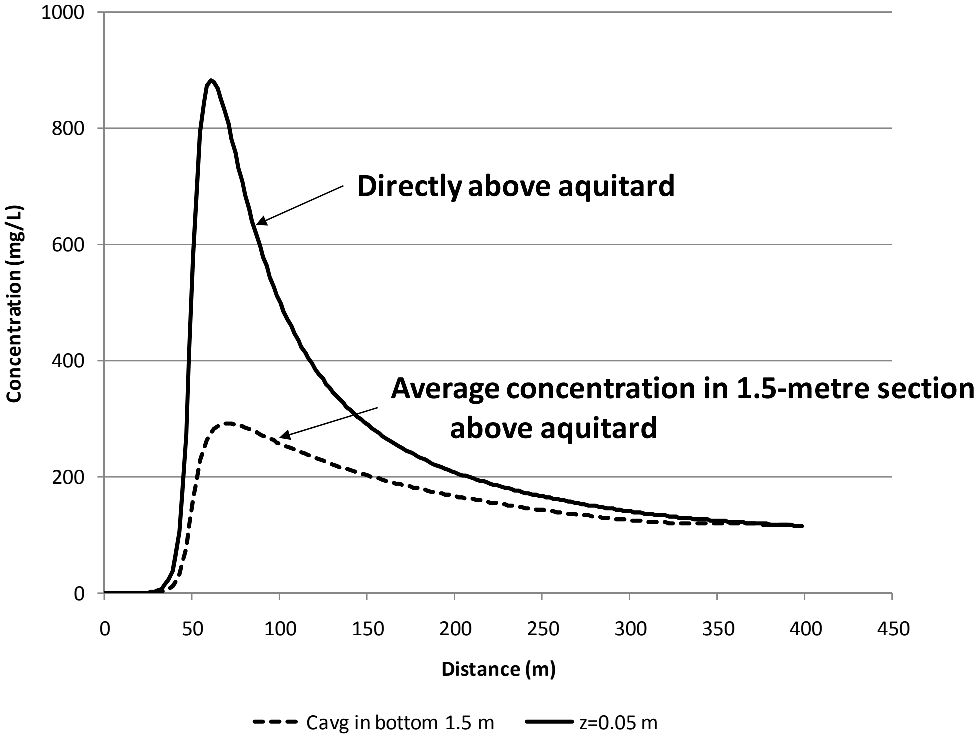


Figure 3 – Concentration versus Distance Downgradient of Source Zone Prior to Remediation

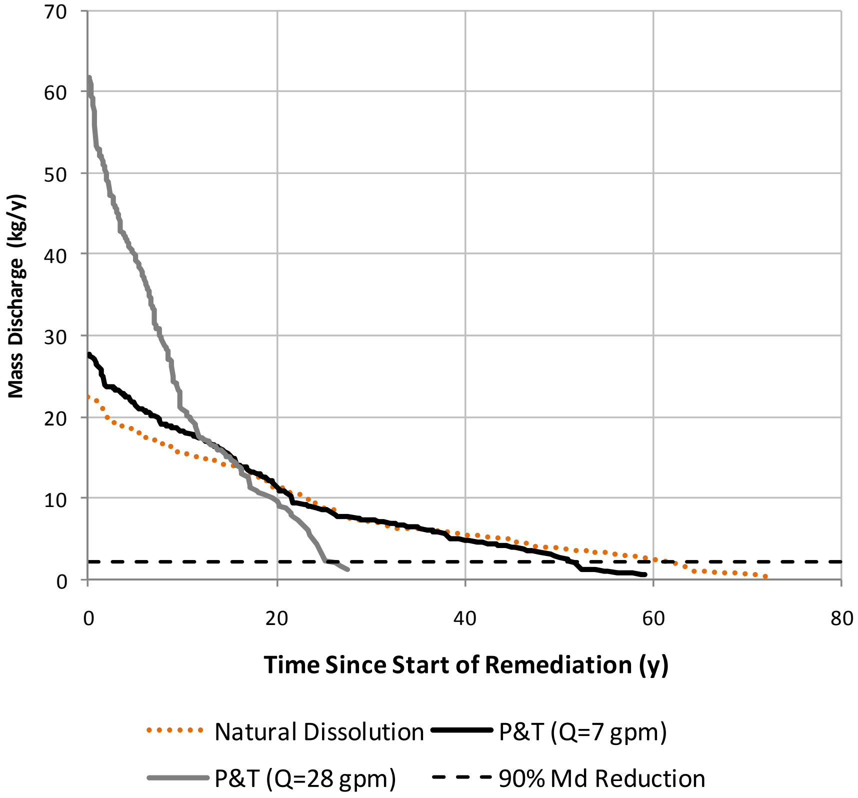
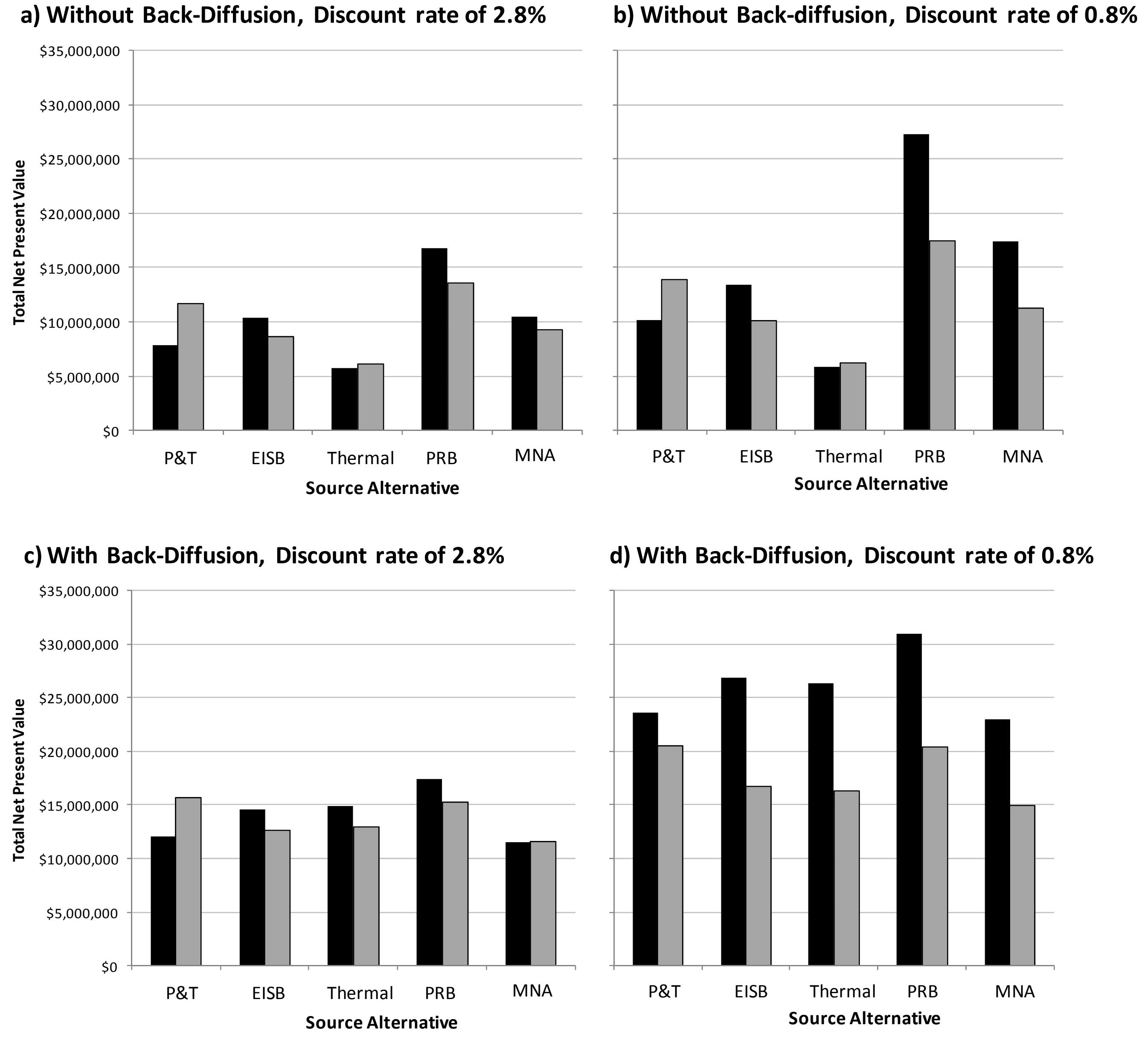


Figure 4 – Simulated Mass Discharge versus Time


a) Without Back-Diffusion, Discount rate of 2.8%

b) Without Back-diffusion, Discount rate of 0.8%

c) With Back-Diffusion, Discount rate of 2.8%

d) With Back-Diffusion, Discount rate of 0.8%

Notes:
P&T is pump-and-treat
EISB is enhanced in-situ bioremediation
PRB is permeable reactive barrier wall
MNA is Monitored Natural Attenuation

Figure 5 – Net Present Value of Remedial Alternatives

Table 1 – Back-Diffusion Exceedance Regions

Low-Permeability Region	**Back-diffusion Contributing to Groundwater Exceedance?**
Source Zone	YES
Plume (On-Site)	YES
Plume (Off-Site)	YES

Table 2 – Source Alternative Simulated Performance

Source Alternative	**Remediation Timeframe (y)**	**Mass Discharge Reduction Half-life (y)**	**R^2 (1)**
Thermal (ERH)	1	n/a	n/a
EISB	21	n/a	n/a
P&T (Q=28 gpm)	26	7	0.96
P&T (Q=7 gpm)	51	14	0.94
PRB or MNA	62	17	0.95

Notes:

(1) R^2 is calculated based on an exponential regression of the mass discharge versus time curve

Table 3 – Summary of Remedial Alternative Costs

Alternative	**Capital**	**O&M**	**Periodic/ Alternative O&M**	**References Used to Support Cost Estimates**
P&T (source only, Q=28 gpm)	$600,000	$300,000		ESTCP (2008), EPA (2001), AFCEE (2009)
P&T (plume only, Q=50 gpm)	$650,000	$325,000		ESTCP (2008), EPA (2001), AFCEE (2009)
P&T (plume + source, Q=60 gpm)	$700,000	$350,000		ESTCP (2008), EPA (2001), AFCEE (2009)
EISB (source)	$250,000	$110,000	$200,000	ESTCP (2008), SRT (2009)
In-situ thermal (ERH) for source	$3,250,000	$100,000		Battelle (2007); average cost per cubic yard for Cape Canaveral & NAS Alameda sites
PRB (source)	$912,500	$90,000	$273,750	EPA (2001), capital is avg of 16 sites, O&M is assumed to be 20,000 less than EISB O&M because less analysis and fewer analytes; replace every 8 years
PRB (plume)	$2,281,250	$95,000	$684,375	2.5x higher capital cost than source PRB; replace every 15 years if source undergoing treatment

Notes:

25% contingency on capital costs

30% contingency on O&M costs