CL:AIRE

SAB 6
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sabre bulletin

CL:AIRE SABRE bulletins describe specific, practical aspects of research from the LINK Bioremediation Project SABRE, which aimed to develop and demonstrate the effectiveness of *in situ* enhanced anaerobic bioremediation for the treatment of chlorinated solvent DNAPL source areas. This bulletin describes performance monitoring and assessment approaches undertaken as part of the SABRE project.

Source Area DNAPL Bioremediation: Performance Monitoring and Assessment

1. BACKGROUND

This bulletin is the final instalment of the SABRE (Source Area in situ BioREmediation) bulletin series. It describes performance monitoring and assessment (PMA) approaches undertaken as part of the SABRE project. SABRE was a collaborative R&D project undertaken by an international, multidisciplinary team between 2004 and 2008. The project comprised laboratory and field-pilot scale development of an accelerated anaerobic bioremediation process for dense non-aqueous phase liquid (DNAPL) chlorinated solvent contamination in groundwater.

With regard to PMA, the project team was guided by **Strategies for monitoring the performance of DNAPL source zone remedies** (ITRC, 2004) which defines performance assessment as, "*the task of evaluating the efficiency and effectiveness of a remedial action in meeting the remediation and operational objectives established for the project.*"

Subsequent to the SABRE project's completion, the Environment Agency published **Verification of remediation of land contamination** (Environment Agency, 2010), which introduced a lines of evidence approach to verification in the UK. Verification of remediation had already been defined by the Model Procedures for the Management of Land Contamination (Defra & Environment Agency, 2004) as, "*the process of demonstrating that the risks have been reduced to meet remediation criteria and objectives based on a quantitative assessment of remediation performance.*"

For the purposes of this document, the aims and objectives of PMA and verification are considered to be closely aligned, and their meaning interchangeable.

2. INTRODUCTION

Although defined above, put simply, PMA can be considered the process of gathering data to answer questions about the behavior of a remediation system. These can be generic questions, which can be i) applied to any site - Have remedial objectives been met? Are all risks being managed to acceptable levels? or ii) site-specific - Are the desired biodegradation reactions occurring? Has DNAPL dissolution been enhanced? Was bioaugmentation successful?

Pertinent PMA questions will be influenced by remedial objectives, the remediation technology and perhaps by site and contaminant properties. Qualified answers to these questions (yes, but…) may not be sufficient to satisfy site-owners, regulators or other stakeholders. In those cases, robust answers may need rigorously derived estimates of uncertainty.

Accounting for analytical accuracy, sample bias, and other measurement error is fairly straightforward. More challenging is quantifying the uncertainty in conceptual models, including error introduced as a function of spatial non-aqueous phase liquid (NAPL) distributions and hydraulic conductivity variability and possible temporal variability of flow conditions. It could be said that any attempt to reduce uncertainty must increase the level of confidence that can be attached to the assessment of a particular remediation's performance. However, the level of confidence in remediation performance will invariably be increased by collecting more data. As data gathering is expensive, effective PMA involves a balance of strategic data gathering and robust data processing with fiscal pragmatism to answer very specific questions with acceptable confidence. That, as mentioned above, makes it important that remediation criteria (and perhaps confidence intervals) are agreed upon even before a remediation system and its attendant monitoring network and sampling schedule can be designed and implemented.

PMA methods can be broken down into process indicators (qualitative), temporal concentration histories (semi-quantitative), and mass-based (quantitative). No single monitoring approach provides the data necessary to answer even the limited scope of questions above which is why PMA strategies should incorporate a number of monitoring approaches, devices and datasets that contribute lines of evidence.

The status of most remediation processes and temporal concentration histories can be effectively monitored using suitably placed standard monitoring wells. It is possible to apply mass-based metrics (contaminant mass discharge, mass turnover, etc) using typical monitoring well data, but the high degree of inherent uncertainty should be taken into account. Somewhat more advanced techniques (integral pumping, mass flux meters, multilevel sampler (MLS) transects) have been proposed to allow more accurate estimates of mass discharge (Bockelmann et al. 2003; Annable et al. 2005; Guilbeault et al. 2005). In the context of DNAPL source zones, flux meters or MLS transects offer certain advantages over integral pumping. The most useful is the preservation of spatial complexity, which permits monitoring of key processes that may be spatially discrete in complex heterogeneous aquifers. Further advantages are offered by transects of certain MLS designs: the ability to measure wellhead parameters and inorganic species, conduct hydraulic tests, monitor microbial factors, and alter sampling frequency in response to new data. Spatially intense data that can be provided by mass flux transects allows the application of advanced, but readily available geostatistical data processing methods that estimate mass discharge and provide rigorous estimates of uncertainty. These aspects of PMA are addressed herein using the extensive SABRE datasets.

3. SABRE INSTRUMENTATION AND MONITORING

As described in companion CL:AIRE bulletins (RB6, RB11, SAB1-5), the SABRE project involved bioaugmentation and biostimulation of a chlorinated solvent DNAPL source zone isolated in a U-shaped plastic sheet pile cell closed at the downgradient end (Figure 1). This configuration allowed for a degree of hydraulic control, which was anticipated to simplify performance assessment. A 3-4 m thick River Terrace Gravel aquifer bounded above by 2.5 m of alluvium and below by fractured and weathered Mercia Mudstone played host to the DNAPL source and associated dissolved plume. The principal objective of the project was to stimulate biodegradation near the DNAPL/water interface, thereby enhancing mass transfer from the DNAPL to the

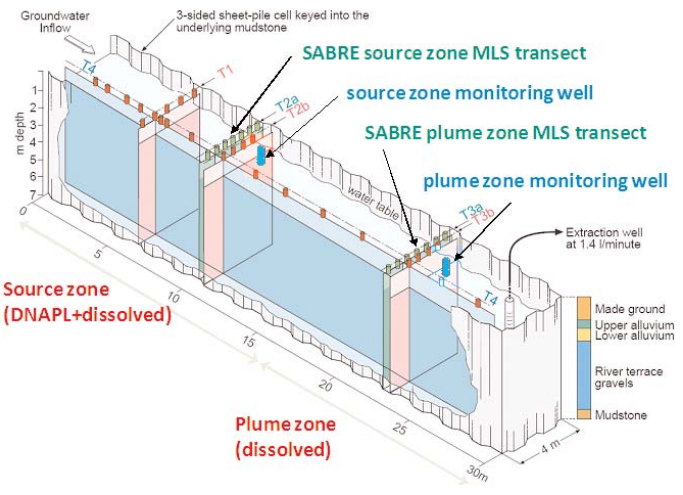

Figure 1: Schematic of the SABRE experimental test cell, showing location of conventional monitoring wells and MLS transects. Figure courtesy of the StreamTube project.

aqueous phase. The expectation was enhanced DNAPL dissolution and shorter DNAPL source zone life. The specific goal of the PMA therefore was to demonstrate that this objective was met.

Other PMA goals were to compare performance indicators derived using standard long screened monitoring wells and discrete point MLS transects, and to evaluate the utility of various remediation process markers. Whilst it is unquestionably more costly to install, sample and analyse the greater number of samples generated by MLS transects, our hypotheses were that such data gives important insight and understanding of processes relevant to the durability of the remediation. Also, transect data capture spatial detail that results in more accurate mass discharge estimates as well as quantifiable uncertainty.

Within the experimental test cell, two SABRE MLS transects were installed across the cell. One, referred to as the "source transect" was located roughly 10 m from the upgradient end of the cell. Characterisation work suggested this location was at or near the downgradient edge of the DNAPL zone. The second, or "plume" transect was installed 13 m further down the cell, in a zone believed to be free of NAPL. Both transects contained 7 MLS devices, spaced roughly 0.5 m apart. Each MLS featured nine 15 cm long screens, at depths based on the stratigraphy observed in core collected during installation. Vertical spacing of multilevel points ranged from 0.3 to 0.9 m, but was on average ~0.5 m. The purge/sample volumes taken mean that roughly 50% of the groundwater flowing across each transect is represented in the data (assuming a 5:1 anisotropy ratio). Standard 5 cm-diameter monitoring wells were installed 0.5 m downgradient of each transect. Both had 3 m screens, which meant that these wells only sampled 60% of the depth covered by the MLS devices.

During the field component of the project, on the order of 1000 water samples were collected, resulting in some 36,000 individual biogeochemical data (including wellhead parameters). In addition, some 25 cores were collected and sub-sampled, generating another 1000 chemical data. It is highly unlikely under normal circumstances that collecting such a comprehensive dataset will be feasible. The SABRE project afforded the opportunity to exceed such commercial practicalities in the pursuit of improved monitoring and assessment approaches, and better understanding of the impact of coarse sampling on confidence and uncertainty estimates.

4. PMA FROM LONG SCREENED MONITORING WELL DATA

For practical and economic reasons, the vast majority of monitoring strategies deployed at sites undergoing remediation involve conventional monitoring wells. As such, a component of the SABRE project was to explore what process and performance understanding could be teased from data collected from such devices. In the context of bioremediation in DNAPL source zones, evidence of reductive dechlorination is an obvious process indicator.

Figure 2 shows the concentration histories of trichloroethene (TCE), dichloroethene (cis-DCE), vinyl chloride (VC) and ethene observed in the source zone monitoring well during the experiment. A few useful qualitative observations can be made from these data. Low levels of cis-DCE and VC in the period before Day 0 (the start of biostimulation and bioaugmentation) is evidence of natural reductive dechlorination, albeit at a slow rate. This suggests that organisms capable of the desired reactions are present in the aquifer. These data do not, however, indicate whether slow TCE dechlorination is due to a lack of labile carbon to drive the reaction, some toxic or inhibitory effect, or a deficiency in the indigenous microbial consortium.

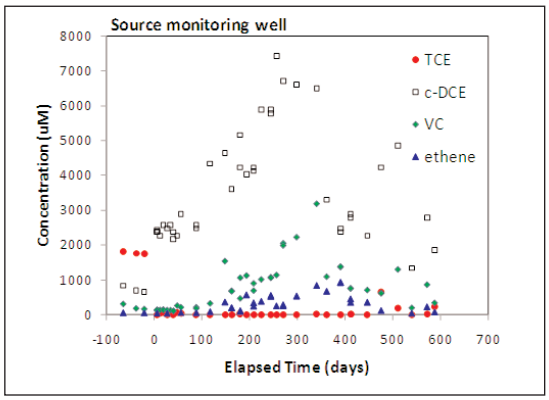

Figure 2: Temporal chlorinated ethenes and ethene concentration histories observed in the source monitoring well. Day 0 separates the pre-remediation from the post-remediation period.

After Day 0, the marked increase in cis-DCE, VC and ethene suggests that the rate of reductive dechlorination has been enhanced as a result of the remediation. This observation is important since the pre-remediation TCE concentrations prevailing in the source area can inhibit dechlorinating bacteria.

Also, it appears that cis-DCE and VC production exceeded their respective degradation rates for a period of time (the rising limb of concentrations), but then began to fall. Interpreting these data in a process context is not difficult, but extracting meaningful degradation rates is less straightforward because the reactions generate daughter products that also degrade.

Consideration of the temporal trend of total chlorinated ethenes (Figure 3) can also yield insight into TCE solubility enhancement. Since it is rare that DNAPLs contain anything other than trace levels of cis-DCE and VC, their presence in solution must be due to dechlorination of a parent compound. Since no PCE was found at the SABRE site, it is presumed that TCE is the parent. So, 1 mole of daughter represents 1 mole of TCE, and therefore an increase in total chlorinated ethenes (CEs) in solution must indicate an increase in TCE effective solubility. Figure 3 suggests that there was no appreciable increase in TCE solubility until after 100 days after the start of remediation. By Day 200, total CE concentrations reached ~6000 μM, which could only have been generated if TCE was dissolving to concentrations on the order of 790 mg/L. This is a two-fold increase over that observed during the pre-remediation (or natural attenuation) period. By Day 300, the total CEs observed suggest TCE solubility reached ~1400 mg/L. Pure phase TCE solubility is often reported as 1100 mg/L at 25C. However, various factors may result in effective field solubilities ranging from 1000 to 1300 mg/L. Nevertheless, it is clear that TCE dissolution was significantly enhanced during the early stages of remediation. It follows that a DNAPL dissolving twice as fast will deplete twice as quickly - a clear indication that, at least in a semi-quantitative sense, the remediation was effective.

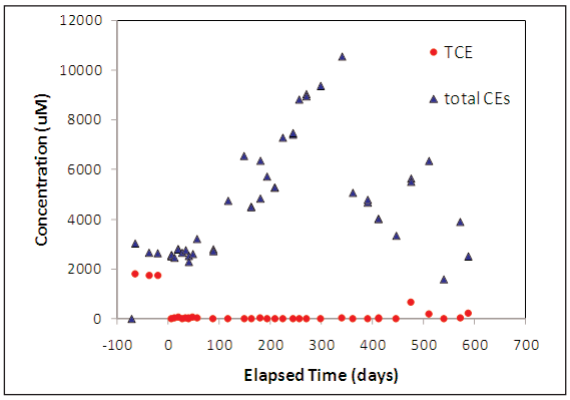

Figure 3: Temporal history of TCE and total chlorinated ethenes (TCE +cis-DCE + VC + ethene) in the source monitoring well.

Dechlorination also produces dissolved chloride, which is conservative (non-sorbing and non-degrading). Therefore, an increase in dissolved chloride concentrations over time indicates on-going dechlorination. However, noisy background chloride signals may make it difficult to quantify the process. In theory, complete dechlorination of TCE dissolving at ~365 mg/L (as during the pre-remediation period) should result in chloride concentrations on the order of 190 mg/L (Figure 4). If chloride concentrations increase in time, that is an indication of increased TCE dissolution and biodegradation. The dissolved chloride concentration of ~620 mg/L observed at Day 300 corresponds to a TCE concentration of roughly 950 mg/L. Note that the enhanced TCE solubility at Day 300 found using the total CE values (see above) was ~1400 mg/L - the difference is found in the residual chlorine-bearing daughter products still present at that time. In some cases, daughter product degradation may occur rapidly, leaving only chloride as a process signal. In those cases, the temporal chloride trend may prove valuable.

The longevity of the injected electron donor can be evaluated via temporal total organic carbon (TOC) monitoring. Figure 5 shows TOC concentration over time in both source and plume zone monitoring wells electron-donor (e-donor) being added in both zones on Day 0). The nature of TOC analysis is such that volatile compounds are often lost. So, it can be assumed that TOC is a rough approximation of the level of available e-donor (which was in this case non-volatile). It is apparent in these data that by Day 300 the carbon source began to deplete. In order to maintain dechlorination, it is obvious that reinjection would have been warranted around that time. Once depletion rates are established, it is conceivable that e-donor dosing could even be automated. However, given the frequency it will likely be more practical to conduct those additions manually.

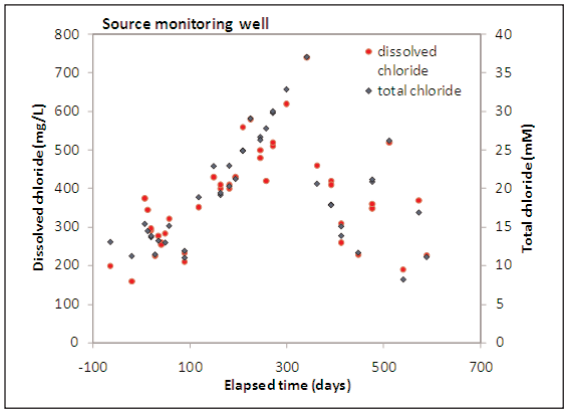

Figure 4: Total chloride and dissolved chloride observed in the source monitoring well during the experiment.

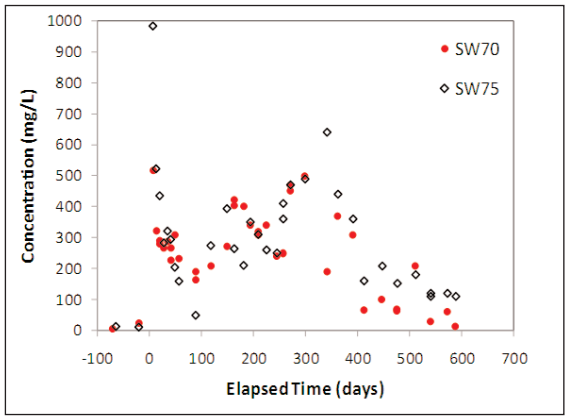

Figure 5: Total organic carbon (TOC) concentration histories in the source (red dots) and plume (open diamonds) monitoring wells. TOC is assumed to represent the amount of electron donor present.

5. PMA FROM TRANSECT MONITORING

It is not uncommon for monitoring devices installed as part of early site characterisation work to be used for remediation performance assessment. However, it is rare that the location of these wells provides data that leads to robust PMA with acceptable levels of confidence, because they would have been installed for a different purpose. Hence, PMA calls for a different positioning of monitoring devices. In DNAPL source zones, the discrete nature of non-aqueous phase distribution is such that very high concentration gradients can be expected. That spatial complexity is largely a function of lithologic heterogeneity, which also has an effect on dissolved phase reactive transport processes. Monitoring processes in a qualitative manner may be possible (as outlined above), but quantifying remediation using such networks carries considerable uncertainty.

An alternative strategy is to install devices (conventional wells or MLS devices) in a line transverse to the mean groundwater flow direction. This arrangement seeks to incorporate some level of the plume spatial variability, under the assumption that both NAPL and dissolved distributions will be highly variable. Capturing the status and variation of key processes (which may vary according to efficiencies in remedial amendment delivery, water velocity, etc) helps to reduce, but not remove, the uncertainty in monitoring mentioned above.

To illustrate how poorly monitoring well data captures spatial plume variability, Figure 6 compares the spatial distribution of the chlorinated species monitored across the plume MLS transect at two different times during the experiment, to the concentrations found in the monitoring wells on those two occasions. The well data suggest that they must have preferentially sampled higher permeability horizons adjacent to the screen. Further, those data do not in all cases faithfully represent the areal average concentration in that transect plane, nor give any indication that CE concentrations vary over 4 orders of magnitude.

In contrast, the spatially interpolated concentrations derived using the MLS data provides some useful insight into parent and daughter contaminant distributions that can help to refine engineering design. For example, the difference in TCE distribution across the plume transect in the baseline (pre-remediation) and 150 days after the start of remediation suggest that the source of the dissolved TCE on the left side of the cell may have been successfully removed, but the source on the right side persists. This may be due to differences in the rate of dechlorination reactions arising from

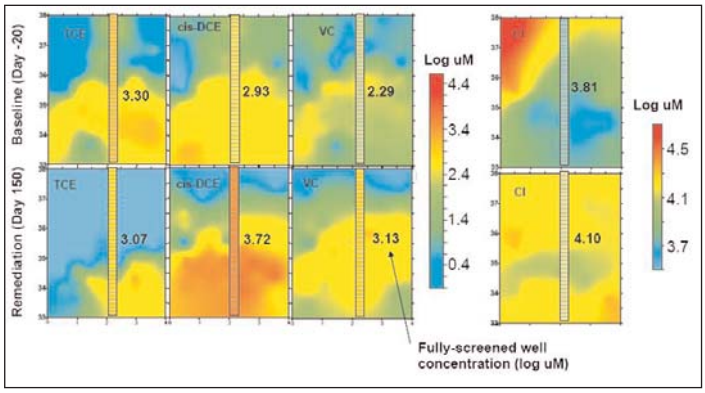

Figure 6: Comparison of plume area concentrations observed in monitoring wells and the spatial distribution of concentrations indicated by the MLS transects. Monitoring screen position is shown relative to the vertical and lateral MLS coverage. Top row is pre-remediation data; bottom row is 150 days after the start of remediation.

non-uniform distribution of e-donor and/or microbes, which would have been influenced by heterogeneity.

The spatial variability provides some insight into the nature of hydraulic conductivity heterogeneity. To get a better picture, modified falling head tests were performed in each MLS point (the devices being designed to allow these tests). The main value of the resulting data is that they provide point groundwater velocity estimates to allow more accurate contaminant mass discharge calculations.

Mass discharge (mass/time) is the product of mass flux (mass/time/area) and cross-sectional flow area. Mass flux is found by the product of concentration and Darcy velocity. In principle, data from any monitoring device could be used to estimate mass discharge, as long as the sampled area of flow and the Darcy velocity are known. Obviously, when the monitoring device samples spatial variability, estimates of the input components (concentration, area and Darcy velocity) are known. When monitoring data is collected at spatially and temporally distributed points there is variability and uncertainty in the data. The variability can be more accurately constrained, and the uncertainty reduced, by increasing the sample density, sampling frequency, or resolution. Thus, MLS installed in a transect at the same lateral density as a transect of monitoring wells will yield a more accurate estimate of mass discharge with lower uncertainty. An assessor ought to consider what level of uncertainty is acceptable in their decision making process (during remedy design) - having regard to its likely influence on a resulting management decision, cost, and the acceptability of a resulting decision - and collect data accordingly.

To allow mass discharge estimation using spatially discrete transect data, those data must first be interpolated. For the SABRE project, conditional simulation and Best Linear Unbiased Estimation techniques were used to spatially interpolate concentrations and Darcy velocity, and calculate mass discharge. The nature of the method is such that estimates of uncertainty are an inherent product of the process. Obvious trends were evident in the source transect data (not shown) that provided insight not evident in the monitoring well data. TCE discharge decreased after the start of remediation, but notably was not reduced to below detection as suggested by the monitoring well concentration data. There must therefore be mass flux paths in the test cell not sampled by that well. The degree of dechlorination along those flux paths may very well be different than that elsewhere in the cell since residence times will vary. By Day 580, the mass discharge of all three chlorinated species had returned to that observed during the pre-remediation phase. As the monitoring well data (Figure 5) suggest that the e-donor (as indicted by TOC values) appears to have started to exhaust itself starting about Day 300, it can be assumed that enhanced dechlorination activity cannot be sustained in the absence of the e-donor.

TCE mass discharge across the plume transect decreased to a very low level and remained low throughout the entire remediation period. Recall that an increase in TCE concentrations was observed in the plume monitoring well beyond Day 100, in some cases reaching concentrations similar to that during the pre-remediation period. That the mass discharge did not increase suggests that those TCE increases migrated in flux paths not sampled by the plume transect or that those concentrations were moving in a very slow flux path (not having arrived at the plume transect before Day 580). This ambiguity suggests that the most robust monitoring strategy may be a combination of MLS transects and longer screened monitoring wells. It was shown that total CE mass discharge across the source transect was approximately the same as that during the pre-remediation period, but markedly reduced across the plume transect.

The point mass discharge estimates can be temporally integrated to get an idea about the total amount of mass (and therefore dissolved DNAPL) discharging over a given period. Using the total CE mass discharge values, approximately 714 kg TCE equivalent (total CEs) crossed the source transect during the experiment. This translates to approximately 500 L of DNAPL. Approximately 570 kg of TCE equivalent crossed the plume transect. Given that the transects are presumed to capture more of the mass discharge spatial variability, it follows that these estimates should provide for more representative average estimates of first order decay rates than those found using data from monitoring points that may not lie along the same flow path. To make such estimates requires that an average flow velocity assumption must be made, but it is assumed that the mass crossing each transect is following dominant volumetric flux paths.

Changes in the DNAPL mass/volume during remediation provide an alternative performance metric, but without an independent estimate of starting DNAPL mass, estimates of percent DNAPL mass or volume removed/remaining cannot be made with any certainty. There were considerable efforts to quantify DNAPL mass in the characterisation phase of the SABRE project, but we recognise that such estimates will rarely be possible. Instead, a conservative approach could be adopted. If it is assumed that the cross sectional area fraction of the source fence in the pre-remediation period where TCE concentrations (data not shown) are high indicates the distribution of DNAPL, that DNAPL zone extends from the upgradient end of the cell to the source transect, and that DNAPL is at 2% residual saturation, then perhaps 840 L were present at the start of the experiment. Thus, on the order of 60% of the DNAPL may have been removed. Of course, the validity of such approximations is influenced by the assumptions made and should be supported by a rigorous uncertainty assessment.

Another powerful application of transect data is found when spatially interpolating the mathematical product of concentration and Darcy velocity (i.e. mass flux). Figure 7 shows the distribution of total CEs in the source fence at some point in time (left), the Darcy velocity across the source fence (centre) and the distribution of total CE flux across the fence at the same time (right). This shows that in this case, chlorinated species are distributed everywhere, but the mass flux map suggests that CE mass is significantly more mobile in some zones than others.

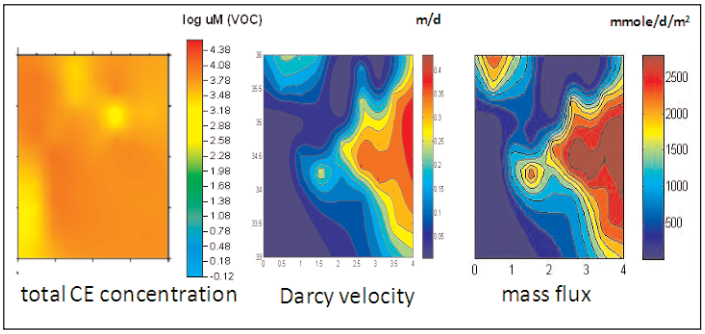

Figure 7: Total chlorinated ethene concentration distribution (left), Darcy velocity (middle) and total CE mass flux distribution (right) across the source fence during the remediation phase. Note that the concentration plot gives any indication of where mass is migrating. Flux maps allow monitoring of processes in space that are otherwise masked in the concentration plots.

6. CONCLUSIONS AND LESSONS LEARNED

Treatment PMA is an aspect of remedial system design, implementation and management that requires careful consideration in the early stages. Key decisions need to be made concerning the metric or metrics against which the effectiveness of a remediation will be measured, in order to develop multiple lines of evidence - qualitative, semi-qualitative and quantitative. These decisions have a bearing on the type and location of monitoring devices, frequency of sampling, analytical suite, and data processing methods.

In full-scale application, the likely remedial objectives for application of the SABRE technology include substantial depletion of source mass through enhanced dissolution and degradative losses, with a corresponding increase in mass discharge from the source. Therefore, mass flux was a key PMA metric for this work. At the SABRE site, mass discharge was compared to concentration based metrics that employ conventional, fully screened, wells. MLS transects were installed to capture the complex, heterogeneous, contaminant flow paths in the alluvial aquifer, and generate the data necessary to make mass discharge estimates of key dissolved species. An added benefit of transect monitoring was that 2-D mass flux maps, generated by multiplying concentrations by Darcy flux, revealed information not evident in spatially interpolated 2-D concentration plots; i.e. the location of high and low mass flux paths.

Ultimately, the design of performance monitoring systems is dependent on a number of factors, including the site conceptual model, the remedial technology, critical exposure pathways and points and regulatory drivers. At the time of publication of this Bulletin, mass discharge (or flux) is only accepted as a PMA metric in a limited number of jurisdictions. While mass discharge has received increasing attention from both regulators and academics, concentration-based standards (i.e., exposure point concentrations within the classical risk assessment paradigm) are still the focus of most regulatory programmes. Whether contaminant concentration in groundwater or mass flux is the best metric depends on potential exposure points, for instance:

- For vapour intrusion, contaminant concentration in groundwater is typically more important. However, if the assessment involved direct sampling of the vapour phase, then mass flux would be the more appropriate metric;
- For groundwater discharging to sediments and creating a sediment exposure point, concentration may be the better metric because the concentration in the discharging groundwater controls sediment concentrations;
- For groundwater discharging to a well-mixed water body, flux may be the better metric; and
- For a groundwater plume within the capture zone of a supply well, again, flux may be the better metric.

For remedial design, the best metric would depend on the remedial approach and the remedial objectives. Monitoring strategies and devices should be selected on a site specific basis, and the remedy design team should adopt a set of metrics that provide the best assessment of remedial action objectives. Adopting an adaptive approach to PMA design will ensure that overall remediation performance assessment is more robust and the selection of specific PMA tools ultimately meet the requirements of the end-users.

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