Integrated Systems-Based Approach to Monitoring Environmental Remediation – 13211

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ABSTRACT

The US Department of Energy (DOE) is responsible for risk reduction and cleanup of its nuclear weapons complex. Remediation strategies for some of the existing contamination use techniques that mitigate risk, but leave contaminants in place. Monitoring to verify remedy performance and long-term mitigation of risk is a key element for implementing these strategies and can be a large portion of the total cost of remedy implementation. Especially in these situations, there is a need for innovative monitoring approaches that move away from the cost and labor intensive point-source monitoring. A systems-based approach to monitoring design focuses monitoring on controlling features and processes to enable effective interpretation of remedy performance.

INTRODUCTION

At many sites, remediation goals may include protection of an identified downgradient receptor. Multiple factors influence contaminant movement and the potential to impact the receptor. Evaluation of the contaminant and receptor setting (i.e., exposure pathway) can be organized as a system to facilitate understanding of contaminant transport and key controlling features and processes. These features and processes are then potentially useful in devising effective monitoring strategies [1]. Use of a system-based evaluation is especially important for inorganic and radionuclide contaminants in the vadose zone because of the constraints to monitoring approaches imposed by unsaturated conditions.

Inorganic and radionuclide contaminants in the deep vadose zone are at depths below the limit of direct exposure pathways, but may need to be remediated to protect groundwater [2]. The groundwater contaminant concentrations that result from vadose zone contamination are a function of the rate of contaminant movement through the vadose zone. For remediation, the magnitude of contaminant discharge from the vadose zone to the groundwater must be maintained low enough to achieve groundwater protection goals.

Vadose zone contamination is a concern at some sites due to the potential for long-term impact to groundwater. In this context, vadose zone contamination is a source, or potential source, to groundwater plumes. A system-based monitoring approach for the vadose zone focuses on developing a conceptual site model highlighting key features that control contaminant flux to groundwater. These features are derived with consideration of the unsaturated flow and contaminant transport processes in the vadose zone and the nature of the waste discharge. Diagnostic properties and/or parameters related to both short- and long-term contaminant flux to groundwater can be identified and targeted for monitoring. For instance, features that promote lateral moisture and contaminant spreading in the vadose zone, controlling factors for long-term water migration such as recharge rate, and indicators of geochemical interactions, especially in the context of the disposed waste chemistry, are potentially important elements of the conceptual model and for consideration in the associated monitoring design.

The resolution of monitoring data needed to correspond to a functionally useful indicator of flux to groundwater can be estimated using quantitative analyses and the associated unsaturated flow properties relevant to the targeted site and vadose zone features. This monitoring design approach follows the process of developing a quantitative conceptual model suitable for supporting projections of future flux to groundwater because it is likely that, in many cases, remediation decisions for the vadose zone will need to be made all or in part based on projected impacts to groundwater and monitoring will then be applied to verify that remedy goals are being met over a long period of time.

CONCEPTUAL APPROACH

Appling a natural attenuation assessment is a useful framework for developing a conceptual model for a vadose zone system. As an example, consider a vadose zone contaminated by a previous discharge of inorganic contaminants in an aqueous solution (Figure 1). There are significant natural attenuation processes inherent in vadose zone contaminant transport. Attenuation processes include both biogeochemical processes that serve to retain contaminants within porous media and physical processes that mitigate the rate of water flux. In particular, the physical processes controlling fluid flow in the vadose zone are quite different, and generally have a more significant attenuation impact on contaminant transport relative to those within the groundwater system. For example, lateral spreading of introduced fluids due to capillary forces can have a significant effect in decreasing the rate of downward movement toward the groundwater. Thus, while contaminants in the vadose zone act as a source to groundwater, there is also natural attenuation occurring in the vadose zone. Overall assessment of risk to receptors and remedy design should include consideration of these natural attenuation processes. Typically for Monitored Natural Attenuation (MNA) assessments in groundwater, the source and associated flux of contaminants to the downgradient plume are considered in comparison to the natural attenuation capacity within the plume to evaluate the expected plume dynamics over time, e.g., will the plume grow, be stable, or shrink [3, 4, 5, 6, 7]. When contaminants are in the vadose zone, natural attenuation within the source zone and its impact on the source flux to the groundwater plume need to be included in the assessment and in designing appropriate monitoring (Figure 2). Even though other remedial actions may be applied, the attenuation processes in the vadose zone are key targets for characterization and monitoring.



Figure 1. Conceptual depiction of inorganic contaminants in the vadose zone (adapted from Dresel et al. [2]).



Figure 2. Overall system components for MNA evaluation.

It's also useful to examine the overall contaminant transport processes to identify appropriate targets for monitoring. The rate of contaminant movement through the vadose zone (the mass flux and/or discharge) is a primary parameter used to assess potential impacts on groundwater.

Direct measurement of contaminant flux in the vadose zone is difficult and rarely feasible. However, this overall objective can be appropriately addressed using a lines-of-evidence approach, where the data collected are used to quantify the key factors related to contaminant migration and enable interpretation with respect to contaminant flux to the groundwater. While each site is unique, a general conceptual model can be used to describe key characterization and monitoring targets (Figure 3).



Figure 3. Generic description of the vadose zone as a transport path from the surface to groundwater for inorganic non-volatile contaminants (adapted from Truex and Carroll [8]).

A primary goal of monitoring for inorganic (non-volatile) contaminants is to provide information useful to understanding and quantifying the contaminant distribution in the vadose zone and transport to groundwater. Monitoring data can be, and should be, used as part of an overall strategy for predicting future states. Predictive (numerical) analyses are also part of that strategy, as are the iterative nature of monitoring and modeling. The results of predictive analyses and the refinement of these analyses with monitoring data can be important to the interpretation of monitoring data in terms of verifying appropriate remedy performance. For most sites, waste disposal (time "a" on Figure 3) has already occurred, but important information may be available to describe site conditions during the time of active disposal (i.e., the top curve on Figure 3). If these data are available, they can be related to predictions of, or monitoring approaches for, describing the contaminant flux to the groundwater (i.e., bottom curves). Key components of vadose-zone monitoring based on the generic transport framework (Figure 3) are discussed

below and include the categories of 1) water flux, 2) contaminant flux and its relationship with water flux, 3) remediation processes, and 4) volumetric moisture and contaminant measurements.

Water Flux. The contaminant flux to groundwater is directly related to the water flux through the vadose zone. Figure 3 shows the generic progression of water flux over time. One target for monitoring (and predictive analysis) is determining the shape of the water output (from the vadose zone to the groundwater) flux curve over time, and whether the site is currently at generic time "b," "c," or "d." At generic time "b," the water discharged with the waste disposal has not yet reached groundwater. At generic time "c," water flux at the water table is increased compared to pre-disposal conditions because the discharged water has reached the water table. At generic time "d," the peak of discharged water has passed into the groundwater and the water flux is at, or diminishing to a rate consistent with, long-term surface recharge conditions. Knowledge of whether the site is currently at generic time "b," "c," or "d" should be a goal of characterization, predictive analysis, or monitoring the water phase movement for those sites already impacted by anthropogenic activities. This type of knowledge may be useful in supporting streamlined monitoring approaches, especially for long-term monitoring if the dynamics of the contaminant flux are expected to minor. The actual timescale and magnitude of the water flux changes are controlled by the discharge conditions, vadose zone properties and their distribution (especially for controlling features such as interfaces between porous media zones with different properties), and the net surface recharge conditions. Thus, monitoring approaches that target these elements and focus on identifying changes over time are potentially critical components of a vadose zone monitoring program. The hydraulic gradient drives fluid flux, and measurements of fluid pressure or matrix potential at various locations over time can be used to evaluate changes in flux vertically and laterally.

Relationship Between Contaminant and Water Fluxes. Contaminant flux and its relationship with water flux (and therefore, also the relationship between the contaminant and water distribution) are controlled by contaminant interactions with sediment surfaces (e.g., adsorption or precipitation/dissolution processes) or by micro-scale transport phenomena such as intra-particle diffusion. Thus, lines-of-evidence vadose-zone monitoring may include monitoring for contaminant distribution (or surrogates such as ionic strength), factors that control the interactions (e.g., pore water chemistry), and/or indications of changes in interactions (e.g., sediment surface properties, ionic strength). The conceptual site model must indicate the type of contaminant interaction expected either under natural attenuation or remedy conditions. Thus, the conceptual site model would identify which contaminant curve shown on Figure 3 the contaminant flux to groundwater would be expected to resemble and potentially quantify expected fluxes for each contaminant (e.g., through geochemical, geological, and predictive analyses). The

focus of vadose-zone monitoring would then be to verify the expected fluxes and/or the conditions forming the basis of the expected flux.

Volumetric Moisture and Contaminant Measurements. Volumetric measurements or measures of the vertical distribution of moisture and contaminant distribution and changes in these distributions can be interpreted in terms of current or future contaminant fluxes to groundwater. This type of approach provides information about the mass distribution of water and/or contaminants in the vadose zone. In some cases, monitoring the water flux can be simplified, especially for long-term monitoring. For instance, if water flux conditions for the site are at or near the long-term water flux (i.e., approximately equal to surface recharge rate), monitoring for water flux to groundwater reduces to monitoring of net surface recharge.

Remediation Processes. Some remedies impart specific changes to the vadose zone or contaminants that can be directly or indirectly (e.g., indicators or surrogates) monitored over time and related to contaminant flux to groundwater. Approaches may include monitoring of remedy impacts to vadose zone properties (e.g., permeability, sediment surface properties, pore water chemistry), moisture conditions/distribution, contaminant properties (e.g., redox state, precipitates), or the remedy amendments (e.g., addition or transformation of organic carbon, concentration of oxidant or reductant, etc.). All remedies target one or more processes that decrease water and/or contaminant flux, which may be monitored to evaluate remediation progress, as noted above.

The vadose-zone monitoring components discussed above have the potential to provide data that can be interpreted in a lines-of-evidence approach. These components have been discussed in the context of an overall objective to understand and quantify the contaminant flux to groundwater and associated vadose zone remedy performance. For each site, this goal can be applied at different levels of rigor appropriate to the overall remediation approach and the relative importance of the vadose zone contamination. For instance, some sites may require only a threshold indication of contaminant flux from the vadose zone to the groundwater. At other sites, where the vadose zone is more closely tied to a groundwater remedy, more detailed monitoring and quantification of contaminant flux may be needed. Thus, the monitoring design and program will also be related to decisions that need to be supported with the monitoring data.

EXAMPLE QUANTITATIVE ASSESSMENT

A numerical model simulation was conducted to track water and non-sorbing solute (contaminant) in a model domain with a 10-m thick homogeneous vadose zone, hydraulic properties of a medium-grain sand, and a 1 m³ solute injection ($C/C_o = 1$) over 100 days in a 1 m² source area. The recharge rate was set to 4 mm/year, representing an arid site. In addition, a second simulation was conducted with the same configuration except that the solute injection

volume was 10 m³ over 1000 days. Simulation results in terms of the water and solute flux to groundwater are shown in Figure 4.



Figure 4. Simulated water and solute flux from the vadose zone to groundwater. Even non-sorbing contaminants migrate slower than the added water pulse and, as discharge volume decreases, more of the contaminant discharge to groundwater occurs under recharge-driven conditions.

Even though the vadose zone properties are homogeneous, lateral spreading is significant for the low discharge scenario such that contaminant reaches groundwater under recharge rate driven conditions. Therefore, monitoring and understanding recharge is a key factor in estimating risk to groundwater. As the volume of the discharge increases relative to the size of the vadose zone, a larger portion of the impact to groundwater occurs coincident with the pulse of high water flux and recharge is less important. Note, however, that some portion of the contaminant flux to groundwater still occurs during recharge driven conditions. Thus, between these two extreme examples, there may be conditions where impact to groundwater is a mixture of recharge control and input water control. This simple example demonstrates that by quantifying the expected transport behavior for the waste scenario, the importance of recharge rate on groundwater impact can be assessed and interpreted in terms of designing a monitoring strategy.

Similarly, the impact of other features such as low-permeability zones within the vadose zone can be assessed to determine their importance for monitoring relative to quantifying contaminant flux to groundwater. For instance, other simulations (data not shown) have indicated that the impact of low-permeability layers on migration of water and contaminants through the vadose zone is a function of the magnitude of moisture content change induced by the waste discharge. For relatively low moisture content changes, low-permeability layers have little impact on vertical migration. For relatively higher moisture content changes, low-permeability layers cause lateral spreading and/or perching conditions and the rate of contaminant migration to the groundwater can be significantly controlled by these layers until moisture conditions revert to recharge-driven conditions. Thus, specific monitoring of low-permeability layers is primarily diagnostic under high waste-discharge conditions, not low waste-discharge conditions.

CONCLUSIONS

Inorganic contaminants in the vadose zone are a potential source for long-term impact to groundwater. A system-based evaluation can be used to focus monitoring design on controlling features and processes to enable effective interpretation of remedy performance. A natural attenuation assessment is a useful framework for applying a system-based approach, especially in the vadose zone where there are significant natural attenuation processes that can occur. The resolution of monitoring data needed to correspond to a functionally useful indicator of flux to groundwater can be estimated using quantitative analyses and the associated unsaturated flow properties relevant to the targeted site and vadose zone features. In this way, a conceptual model and related quantitative analyses can help design an effective monitoring approach. This type of design approach can enable consideration of monitoring alternatives to traditional cost and labor intensive point-source monitoring.

REFERENCES

- Bunn AL, DM Wellman, RA Deeb, EL Hawley, MJ Truex, M Peterson, MD Freshley, EM Pierce, J McCord, MH Young, TJ Gilmore, R Miller, AL Miracle, D Kaback, C Eddy-Dilek, J Rossabi, MH Lee, RP Bush, P Beam, GM Chamberlain, J Marble, L Whitehurst, KD Gerdes, and Y Collazo. 2012. Scientific Opportunities for Monitoring at Environmental Remediation Sites (SOMERS): Integrated Systems-Based Approaches to Monitoring. PNNL-21379, Pacific Northwest National Laboratory, Richland, WA.
- Dresel, P.E., D.M. Wellman, K.J. Cantrell, and M.J. Truex. 2011. Review: Technical and Policy Challenges in Deep Vadose Zone Remediation of Metals and Radionuclides. *Environ. Sci. Technol.* 45(10):4207-4216.
- EPA. 2007a. Monitored Natural Attenuation of Inorganic Contaminants in Ground Water

 Volume 1, Technical Basis for Assessment. EPA/600/R-07/139, U.S. Environmental Protection Agency, Washington, D.C.
- EPA. 2007b. Monitored Natural Attenuation of Inorganic Contaminants in Ground Water

 Volume 2, Assessment for Non-Radionuclides Including Arsenic, Cadmium, Chromium, Copper, Lead, Nickel, Nitrate, Perchlorate, and Selenium. EPA/600/R-07/140, U.S. Environmental Protection Agency, Washington, D.C.
- EPA. 2010. Monitored Natural Attenuation of Inorganic Contaminants in Ground Water -Volume 3, Assessment for Radionuclides Including Tritium, Radon, Strontium, Technetium, Uranium, Iodine, Radium, Thorium, Cesium, and Plutonium-Americium. EPA/600/R-10/093, U.S. Environmental Protection Agency, Washington, D.C.
- ITRC. 2010. A Decision Framework for Applying Monitored Natural Attenuation Processes to Metals and Radionuclides in Groundwater. Prepared by The Interstate Technology & Regulatory Council Attenuation Processes for Metals and Radionuclides Team.

- Truex, M., P. Brady, C. Newell, M. Rysz, M. Denham, and K. Vangelas. 2011. The Scenarios Approach to Attenuation Based Remedies for Inorganic and Radionuclide Contaminants. SRNL-STI-2011-00459, Savannah River National Laboratory, Aiken, SC. Available at www.osti.gov, OSTI ID 1023615, doi: 10.2172/1023615.
- Truex, M.J. and K.C. Carroll. 2012. Remedy Evaluation Framework for Inorganic, Non-Volatile Contaminants in the Deep Vadose Zone. PNNL-21815, Pacific Northwest National Laboratory, Richland, WA.

ACKNOWLEDGEMENTS

A portion of the funds for this effort was provided by the U.S. Department of Energy Office of Environmental Management and the Richland Operations Office. Additional funding was provided through the Laboratory Directed Research and Development Program at Pacific Northwest National Laboratory, which is operated by Battelle for the United States Department of Energy under contract DE-AC06-76RLO1830.