Field and Virtual Testbeds for Cost-effective Sustainable Remediation Enhanced Attenuation and Long-Term Monitoring

White Paper

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Summary

This white paper presents an innovative approach for sustainable and cost-effective groundwater remediation and monitoring. Our approach integrates recent advances in various technologies: (1) enhanced attenuation-based remediation technology, (2) *in-situ* autonomous sensors, (3) big data analytics, (4) non-invasive remote mapping, and (5) parallel high-performance computing for flow and reactive transport modeling. Bringing these new technologies is expected to transform the long-term management and closure strategies of the Department of Energy (DOE) Environmental Management (EM) sites and to result in enormous cost saving (50 – 90%).

Enhanced attenuation (EA) uses engineered approaches to enhance natural geochemical processes to immobilize contaminants when monitored natural attenuation (MNA) is not sufficient to meet remedial goals. Compared to typical active remediation approaches, EA is cost-effective and sustainable remediation, often reducing cost by 50-90% and producing minimal waste. EA and MNA requires a paradigm shift in monitoring to ensure the long-term stability of sequestered contaminants. To achieve cost-effective monitoring, we have developed an innovative long-term monitoring strategy that focuses on measuring the key variables that control contaminant plume mobility and their spatial and temporal distribution (such as pH, redox potential, electrical conductivity, and groundwater level). We measure these variables using in situ autonomous sensors and use data analytics methods to determine the most important variables and the values that trigger additional monitoring in space and time. The strategy of emphasizing leading indicators of plume change reduces point measurement of contaminants, thereby reducing costs, but more importantly facilitates proactive rather than reactive response to the changes. Coupled with sparse groundwater sampling, the data analytics methods – data mining and machine learning – allow us to estimate contaminant concentrations, and also to detect which changes of the plume mobility are significant. In addition to point well-based sensors, advanced spatially integrating techniques – such as fiber optics sensing, geophysics, and UAV-based mapping technologies – will be evaluated for use in monitoring heterogeneity in plume characteristics.

The state-of-art parallel numerical flow and reactive transport simulator allows us to improve the physical and mechanistic understanding of the plume system, and to predict the long-term plume migration and distribution under a variety of scenarios. Such modeling will lead to improved estimates of the life-cycle cost of both remediation and monitoring, as well as optimized

strategies under long-term changes caused by the geochemical evolution of the plume as well as overarching changes caused by climate change. We demonstrate this integrated approach at the Savannah River Site (SRS) F-Area Seepage Basins, where groundwater is contaminated with various radionuclides. The historical monitoring and characterization datasets in combination with a 3D reactive transport model for SRS F-Area provides a unique opportunity for development of a field and virtual *testbed* for the EM technology development.

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1. Introduction

Nuclear weapon production during the Cold War has resulted in groundwater contamination at many locations in the United States. Low-level radioactive waste solutions were often disposed into unlined seepage basins with minimal or no engineered barriers between disposal point and groundwater. In addition, some of high-level radioactive waste tanks have been reported to have leaks, causing soil and groundwater contamination. The Department of Energy, Office of Environmental Management (DOE-EM) is responsible for the remediation of these sites, which is considered one of the most technically complex cleanup challenges in the world [NRC, 2000]. The overall cost is predicted to exceed $200 billion over the next few decades.

There have been a large variety of remediation techniques that have been implemented at the DOE-EM sites, such as soil excavation or pump-and-treat. Although many remediation efforts have been successful or are expected to reach regulatory clean-up limits in the next several decades, there are still dozens of sites that have contaminant concentrations above regulatory standards. The biggest challenge is to address the large volume of contaminated soil and groundwater with relatively low contamination levels that still exceed regulatory cleanup limits, cases where soil removal is not practical and pump-and-treat systems are not effective due to low contaminant concentrations. Monitored natural attenuation (MNA) has been considered as a preferred alternative for such sites when natural flow and geochemical processes are expected to reduce contaminant concentrations and public health is not compromised. However, several sites have reported that the contaminant concentrations have not decreased as rapidly as they were originally predicted [Zachara et al., 2013].

Enhanced attenuation (EA) has emerged as a promising strategy to achieve cost-effective and sustainable remediation. EA remedies use an amendment or an engineered feature to enhance contaminant attenuation bringing concentrations down to maximum concentration levels (MCLs) until natural processes take over. At the Savannah River Site F-Area Seepage Basins, for example, base injection was performed to increase pH and immobilize uranium, which resulted in groundwater concentrations lower than the regulatory standard with a remedial cost reduction of approximately $9 million per year compared to the existing pump-and-treat system [Denham and Eddy-Dilek, 2016]. Other successful examples of enhanced attenuation include adding oils to a groundwater contaminated with VOC’s to establish anaerobic zones that facilitate destruction of primary contaminants followed by aerobic zones in which daughter products degrade.

The potential impact and cost-saving from EA for metals and radionuclides is enormous yet it has not explored fully at other sites, in part because of the burden of proof required to demonstrate that stabilized contaminants will remain over time. Different from the conventional remediation approaches, contaminants are sequestered in subsurface rather than removed or degraded. Continued groundwater monitoring will be required for several decades to centuries to ensure the long-term stability and safety. The current practice of monitoring – obtaining and analyzing the contaminant concentrations in groundwater samples at numerous wells – becomes costly over a long time frame and accounts for a large fraction of the projected life-cycle cleanup costs at the DOE sites.

To reduce the long-term monitoring cost, SRNL and LBNL have been developing an innovative approach by taking advantage of recent advances in *in situ* automated sensor technologies [Eddy-Dilek et al., 2016; Wainwright et al., 2016]. The new approach emphasizes measurement of physical and chemical variables that control contaminant mobility in groundwater. The physical variables – such as groundwater tables and infiltration – reflect the forces responsible for groundwater movement. The chemical variables – such as pH, redox potential, electrical conductivity – control adsorption and precipitation of contaminants. When these leading indicators change within the groundwater system, it signals potential remobilization of stabilized contaminants. In contrast, traditional long-term monitoring relies on measuring the arrival of contaminants at sentry wells – a lagging indicator of contaminant mobilization. Measuring leading indicators allows proactive responses that are more effective and less costly than the reactive responses produced by relying on lagging indicators.

At the Savannah River site, for example, *in situ* measurable properties are found to be almost perfectly correlated to the contaminant concentrations [Wainwright et al., 2016]. Since these *in situ* variables are also leading indicators of the plume mobility, the *in situ* sensors can serve as an early warning system so that actions can be taken before the contamination migrates. Data analytics methods are the key in this monitoring strategy to estimate contaminant concentrations and to detect meaningful changes from the noise that is inherent in environmental data. Importantly, the leading indicators such as pH, redox potential, groundwater level, and electrical conductivity are typically easily and inexpensively measured by *in situ* automated sensors. In addition, data from *in situ* sensors can be streamed through wireless communications directly to desktop computers or other devices, eliminating the need for people to visit the sensor locations to download data.

In addition to such point-based continuous sensors, mapping contaminants and other plume-related parameters is important for successful implementation of EA and long-term monitoring. Geophysical techniques have made significant advances in the recent decades to map subsurface heterogeneity, contaminant distributions and plume dynamics in two- and three-dimensional domains [e.g., Rubin and Hubbard, 2005; Wainwright et al., 2014; Binley et al., 2015]. There have been significant advances in fiber optics technologies for measuring temperature, soil properties, soil moisture, gamma radiation, and several chemical properties [e.g. Gaebler, 1983; Potyrailo and Hieftje, 1998; Suarez et.al. 2011]. Unmanned aerial vehicle (UAV) and gamma-ray imaging technologies have transformed mapping of near-surface and surface radionuclide contaminations [Barnowski et al., 2015; Martin et al., 2015; Martin et al., 2016]. These techniques show great promise in efficiently identifying areas in the plume where significant changes are occurring.

Critical to assessing both EA and *in-situ* monitoring strategies over a long time frame is the predictive understanding of long-term plume mobility and dynamics. Modeling of flow and contaminant transport enables the prediction of contaminant plume evolution under various conditions for many years. Such predictions, for example, can provide the time frame of transition from EA to MNA and also provide the spatial-temporal distribution of plume for monitoring, necessary to secure regulatory agreements and assess the life-cycle cost at the site.

Over the last five years, the ASCEM project, funded by DOE-EM, has made a significant advancement to develop a flow reactive transport code that takes advantage of DOE's state-of-art high performance computing capabilities. The 3D reactive transport simulator can now include realistic geology, boundary conditions and artificial structure (such as engineered barrier systems) as well as complex geochemical reactions (such as the interactions between contaminant concentrations and controlling variables) [Wainwright et al., 2016].

In this paper, we first review recent technical advances, and identify research needs associated with EA and long-term monitoring. There are five key components, including (1) enhanced natural attenuation remedies, (2) *in-situ* remote monitoring approaches, (3) fiber optics technology, (3) autonomous geophysical monitoring, (4) UAV-based gamma ray imaging and mapping, (5) Big Data analytics for large spatial and temporal datasets, and (6) predictive simulation capabilities of contaminant transport. In addition to the existing scope with DOE-EM of EA and modeling capabilities, we highlight innovative sensing and characterization technologies, as well as data analytics. Those sensing and analytics capabilities have been rapidly expanding in other areas (such as computer science, artificial intelligence, climate science) which can be transferred to the DOE-EM applications rapidly.

We then propose to demonstrate the effective integration of these components at Savannah River Site (SRS) F-Area, where contaminant plumes and subsurface structure are well characterized and historical monitoring and geological datasets have been effectively curated. The ASCEM project has developed a 3D flow and reactive transport model with complex geochemical reactions and engineered systems. Having real data and established models, the SRS F-Area will be a *testbed* for technologies to achieve sustainable and cost-effective remediation and monitoring at the DOE-EM sites. Such an integrated technology can also be transferable to other types of groundwater contamination such as the DOE Office of Legacy Management sites, nuclear power plants, and other contaminated sites.

2. Key Technologies

2.1. Enhanced Attenuation Remedy

Attenuation-based remedies are those that rely on *in-situ* processes to retard the migration of contaminants to "mitigate contaminant risk to receptors" [Denham et al., 2016]. Monitored natural attenuation (MNA) relies solely on natural processes, whereas enhanced attenuation (EA) uses engineered processes to assist the natural attenuation. The attenuation processes for radionuclides can be both physical and chemical. Physical processes include dilution, dispersion, and the engineered processes of blocking or diverting the migration path or reducing the hydraulic driving force for migration. Chemical processes for radionuclide attenuation include the partitioning of contaminant from the aqueous to the solid phase by adsorption, absorption, or precipitation, as well as radioactive decay. The partitioning of contaminant to the solid phase can involve microbial reactions, particularly when redox transitions are required. With the exception of radioactive decay, all of these attenuation processes result in radionuclides being left in the ground rather than being extracted or degraded.

Proof of the sustained effectiveness of an EA remedy is made much easier by strategic design. In multi-contaminant plumes, it is important to prioritize contaminants according to the risk they pose, with the recognition that one remedy rarely treats all contaminants. The goal for the engineered portion of the remedy should be maximum risk reduction with minimal engineering. Natural attenuation or negotiated alternate concentration levels will often allow low risk contaminants to go untreated. Whenever possible, the remedy should be consistent with the geochemical evolution of the waste site. This insures that the treated immobilized contaminants are likely to remain relatively immobile for long time frames. Finally, use what nature provides geologically, hydrologically, geochemically, or microbiologically in the remedy design.

2.2. Point In Situ Sensors

Recently there have been rapid advances in *in situ* sensors to measure groundwater qualities such as soil moisture, pH, water table, electrical conductivity. These sensors are typically placed inside wells to monitor groundwater or surface water continuously. Many sensors are commercially available at relatively low cost with less than hundred dollars per sensor. There are also more sophisticated sensors under development such as optical sensors for nitrate and organic matter [Pellerin et al., 2012]. In addition, recent advances in wireless communication technology allow transmission of data through wireless communication or mobile phone networks, to store in cloud platforms, and to visualize real-time streaming data continuously. Now wireless sensor networks can deploy dozens or hundreds of sensors and to characterize spatially heterogeneous properties [e.g., Kerkez et al., 2012].

At DOE-EM sites, these sensors can be a powerful alternative to conventional groundwater sampling and laboratory analysis. This is particularly useful for the sites that are remote or the ones that have numerous wells. Although they may not measure the contaminant concentrations directly, such *in situ* variables (such as electrical conductivity) can be considered as proxies for contaminant concentrations of interest, or considered as controlling or master variables that dictate contaminant plume mobility and dynamics. Although *in situ* sensors will not eliminate groundwater sampling as regulations in RCRA and CERCLA specify a minimum number of wells, this strategy can be used to reduce the number of wells and the frequency of sampling. In addition, those controlling and master variables are often leading indicators of changes prior to plume movement. Our approach based on measurement of master variables and explicit monitoring of hydrologic boundary conditions combined with traditional metrics should lead to improved monitoring with early warning systems while simultaneously reducing the overall cost of monitoring.

2.3. Distributed Sensing with Fiber Optics Cables

Over the last 20 years, significant advances have been made in the utilization of optical fibers for measuring environmentally relevant parameters including temperature [Suarez et.al. 2011], strain, soil moisture, acoustic waves [Bao & Chen, 2012, Cox et.al. 2012], gamma radiation [Gaebler, 1983], and limited chemical parameters [e.g. Potyrailo and Hieftje, 1998]. Each pulse of light samples the state of the fiber at all locations, yielding property measurements along the entire length at a fine lateral resolution (~ 0.25 - 1m). Optical time or frequency domain reflectometry are used to measure localized differences in optical scattering (Rayleigh, Brillouin, or Ramen) along fibers caused by changes in the environment. Since the fibers themselves are passively interrogated by coherent laser pulses, they do not require power sources along their length. They also function effectively over a broad range of temperatures and can be packaged to withstand harsh conditions in either near-surface or borehole environments. To date, distributed temperature sensing and distributed acoustic sensing have found a wide variety of applications in monitoring infrastructure including leak detection in buried pipelines [Frings, 2011], the structural health of buildings [Lopez-Higuera et.al. 2011], and road conditions in tunnels [Krohn & Nicholls, 2009]. LBNL has recent experience in deploying distributed fiber optic systems in the context of deep borehole monitoring [Daley et.al. 2013] and integrated temperature/strain monitoring of shallow arctic test sites.

A host of parameters relevant to both MNA and EA are accessible using distributed fiber-optic sensing. While direct distributed measurement of contaminant analytes at MCL levels is currently not possible, a variety of secondary variables relevant to the efficacy of EA can be quantified including pH, soil moisture, saturated flow rate, temperature, and possibly gamma dosimetry. Soil moisture in particular is a key target since spatial variation in infiltration and flow regime can significantly impact plume evolution. Soil moisture can be measured directly by coupling a resistive heater to a fiber-optic distributed temperature sensing (DTS) system, a distributed version of a classical heat-pulse measurement (e.g. Weiss 2003). Alternatively, soil moisture can be measured passively by using DTS to quantify the impact of soil moisture on diurnal temperature cycles in the soil column. The resulting datasets could prove useful in constraining the hydrologic components in coupled reactive transport models. While measurement of pH and fiber-optic gamma dosimetry (using scattering variations, e.g. Gaebler et.al. 1983) are more challenging, either dataset could significantly improve knowledge of redox-sensitive contaminant state as a function of time. More generally, the advantage of distributed fiber-optic sensing, in contrast to autonomous point sensors, is the pervasive nature of the datasets combining large extent and fine space/time sampling.

2.4. Autonomous Geophysical Monitoring

Geophysical methods – including electrical resistivity, seismic, and radar – have been increasingly used to characterize subsurface in a non-invasive manner [e.g., Binley et al., 2015]. They can image subsurface contaminant plumes [e.g., Johnson et al., 2010, 2012; Dafflon et al., 2012], as well as map flow and biogeochemical properties that are important for predictive modeling and understanding [e.g., Johnson et al., 2010; Dafflon et al., 2011; Johnson et al., 2012; Wainwright et al., 2014]. High-performance computing capabilities have led to high-resolution imaging (sub-meter) in a large spatial extent (up to several kilometers).

Autonomous electrical resistivity and phase tomography (ERT) monitoring, in particular, has the potential to achieve rapid and automated detection and identification of contaminant plumes. This approach can bridge the gap in sparse wellbore locations, by providing high-resolution and spatially extensive information in a minimally invasive manner. While the feasibility of such subsurface imaging technique has been demonstrated by a number of different field experiments [e.g., Johnson et al., 2015], real-time monitoring was still challenging until recently. LBNL has been developing software and hardware capabilities for automated and autonomous acquisition and processing of large time-continuous data, so that actionable information can be generated cost effectively in near real time. Laboratory experiments have been successful in detecting complex electrical resistivity signatures associated with salt water movement, temperature variations and changes in chemical conditions (Figure 1).

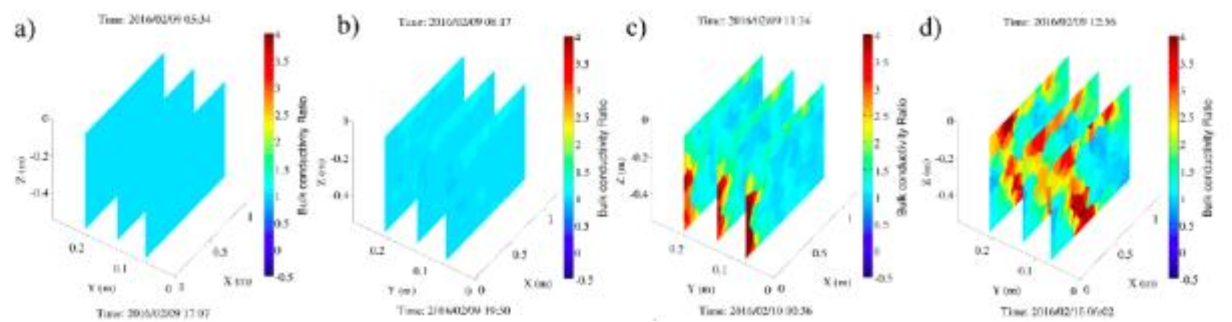

Figure 1. Increase in bulk electrical conductivity (inverted) during a contaminant (i.e., dissolved CO_2) injection experiment.

2.5. UAV-based Surface Contamination Mapping

Exposure points for groundwater contaminants are often in wetlands and along seeplines where groundwater contamination plumes crop out, ultimately leading to contamination of associated streams. Such surface contamination causes serious public health concerns and worker safety issues during monitoring or remediation activities. The ability to accurately map of spatial contamination would represent a fundamental change in the approach of monitoring low level plumes in these settings. In addition, the surface-subsurface interface is considered to be a critical interface for contaminant transport, since near-surface soil contains a large amount of organic carbon, which affects the mobility of key metals (e.g., Tc, U). At many sites these seepline areas are the primary location of contaminant attenuation, but the dynamic physical and chemical changes in these areas can result in periodic releases of contaminants. Hence, long-term monitoring of any site where a plume has reached a seepline must include periodic surveys of the area to detect changes in contaminant distribution.

Over the past several years, LBNL has developed unique capabilities in mapping and reconstructing complex environments and to fuse them with radiological information in three dimensions (3D) [Haefner et al., 2015; Barnowski et al., 2015]. This so-called Scene-Data Fusion (SDF) capability enables the effective and accurate detection and mapping of radiological and nuclear materials and its visualization in indoor and outdoor domains. Hand-portable and unmanned aerial system (UAS) based platforms have been deployed effectively in the contaminated area in the Fukushima Prefecture and in several locations in the U.S. (Figure 2).

SDF is based on the integration of contextual and visual sensor information with compact gamma-ray detection and imaging data. The contextual sensors provide the 3D map of the scene and the position and orientation (i.e. the so-called pose) of the gamma-ray detection or imaging instrument in this scene, enabling the reconstruction and fusion of gamma-ray activities with the scene in 3D. To-date, the LBNL-developed High-Efficiency Multimode Imager HEMI and the Location and Mapping Platform LAMP in combination with a range of contextual sensors have been deployed.

The goal of the new project is to demonstrate this new SDF capability in contaminated areas at the EM sites with the focus on aerial mapping of radiological materials utilizing small UAS systems.

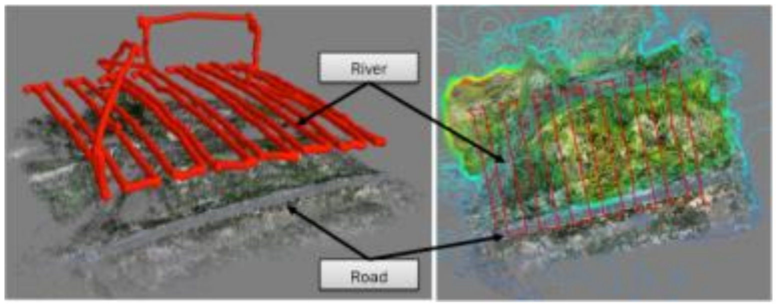

Figure 2 Reconstructed path of RMAX UAS with HEMI and a visual camera on board (left). Projection of path, 3D surface reconstruction, and fused gamma-ray intensity with 662 keV energy gate (right). The boundaries of the road and river with less contamination can be clearly distinguished and separated with a resolution of ~1 m, consistent with the achievable angular resolution and altitude of the flight.

2.6. Data Management and Analytics

Many of the EM sites have accumulated very large amounts of monitoring and characterization data over the last three decades. Many sites have numerous wells and different types of information gathered during environment characterization and remediation studies. Recently, DOE has invested significant efforts to develop a framework for curating those datasets and for making the datasets easily accessible and interpretable [Agarwal et al., 2015; http://phoenix.pnnl.gov/]. For example, the DOE-EM's ASCEM database provides a comprehensive and unified storage for various datasets (e.g., contaminant concentrations core-analysis data), as well as visualization capabilities such as geological and plume maps [Agarwal et al., 2015]. In addition, *in situ* sensors and new sensing technologies produce large datasets that require large-scale data analysis to be efficiently utilized in EM site monitoring.

To make efficient use of such a large amount of datasets, data science – including data mining and machine learning methods – have been one of the most rapidly expanding areas of science and technology in the past ten years [NRC, 2013]. It has been transforming various fields such as marketing, entertainment, and national intelligence. In contaminant science applications, the potential of data science has been largely unexplored. Historical datasets at the EM sites would provide a great opportunity to discover hidden correlations or processes that are key for understanding contaminant transport.

At the EM sites, data mining methods enable mining of historical datasets to find relationships among different variables, and testing of implemented monitoring and remediation approaches in a retrospective manner. In the specific application of long-term monitoring, data mining methods (e.g., time series analysis, cluster analysis, and principal component analysis) was performed on the long-term time series of contaminant concentrations and *in situ* variables to (a) determine temporal and spatial correlations between the controlling variables and contaminant concentrations, and (b) cluster wells into a set of clusters. This analysis includes evaluation of the effect of meteorological parameters (precipitation and evapotranspiration) and groundwater level fluctuations. Such analysis can lead to continuous estimation of contaminant concentrations based on in situ variables [Wainwright et al., 2016], and also to identifying a reduced set of field measured parameters, as part of the development of a cost-effective monitoring program. Data analytics are also the keys to differentiated changes resulting from seasonal, climatic or other fundamental processes, rather changes due to the noise that is inherent in environmental data.

2.6. Predictive Capabilities: ASCEM

Predicting the fate and transport of contaminants is crucial to develop cost-effective remediation and closure strategies. To tackle this challenge, DOE-EM initiated the development of the Advanced Simulation Capability for Environmental Management (ASCEM). ASCEM software is an open source, modular computing framework that incorporates new advances and tools for predicting contaminant fate and transport in natural and engineered systems. ASCEM includes a state-of-art numerical code (Amanzi) for simulating complex flow and reactive transport, and toolsets for data management, visualization, uncertainty quantification (UQ) and parameter estimation (PQ). Amanzi can take advantage of state-of-art high-performance computing systems, and include the Mimetic Finite Difference (MFD) framework and a new optimization capability to minimize the effect of mesh distortion [Beirao da Veiga et al., 2014].

To accommodate complex geochemical reactions, a biogeochemistry application program interface – Alquimia – has been developed to provide a unified connection between Amanzi and biogeochemical codes. Instead of building a new reaction library, Alquimia provides unified data structures and subroutine signatures so that existing mature geochemical codes perform these calculations. Alquimia has brought a new paradigm in modeling complex geochemistry, allowing any subsurface flow and transport simulator to access a wide range of functionality. Currently, Alquimia provides access to the geochemical codes PFlotran and CrunchFlow and can be used for the simulation of aqueous complexation reactions, radioactive decay, ion exchange, surface complexation and mineral dissolution-precipitation.

ASCEM has been transformative in modeling reactive transport over the past five years. Before ASCEM, the complex geochemical reactions, such as pH dependency of uranium sorption kinetics, were only considered in a simplified domain [Bea et al., 2013]. Now ASCEM can solve this system in 3D with more than one million grid cells (Figure 3). In addition, the robust flow solver allows us to include complex geological contrasts and artificial structures such as low-permeability barriers. It has come to the point where we can have a realistic domain and reactions in 3D, and implement different remedial options to be a virtual test bed [Wainwright et al., 2016].

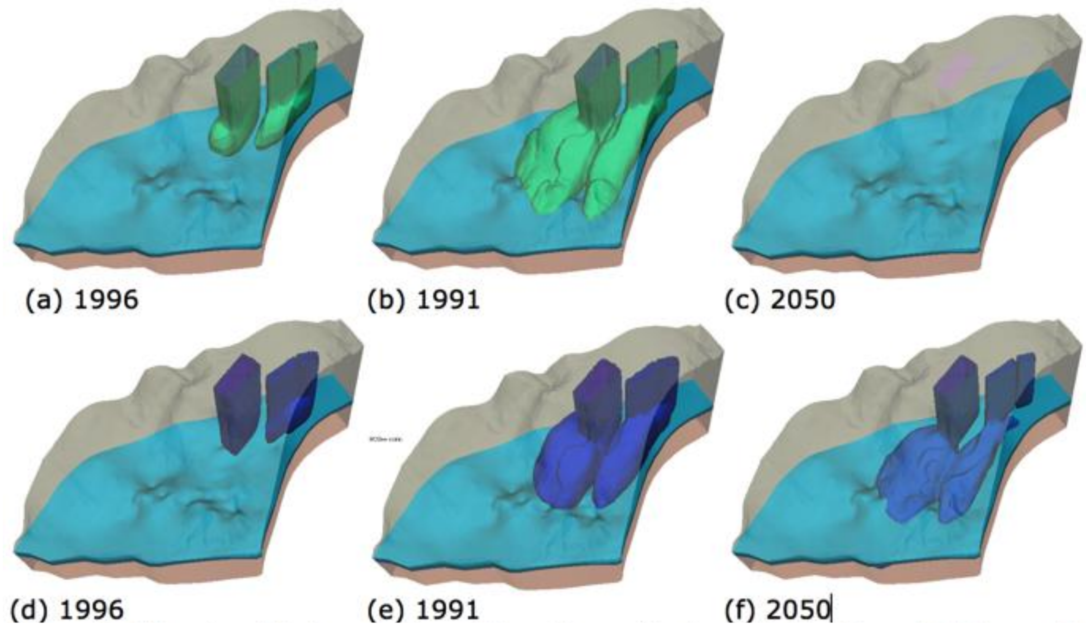

Figure 3. The simulated evolution of (a-c) low-pH plume (pH> 4) and (d-f) uranium plume (concentration>1x10^{-6}mol/L). The sky blue region is the low permeable TCCZ, which separates the upper and lower aquifers. Vertical exaggeration=15X.

3. Three-year Vision: Testbed Concept

Although each individual technology has made a remarkable progress over the past decade, all the technologies have to come together for achieving sustainable remediation and cost-effective monitoring. Demonstrating such an integrated strategy requires a testbed – a site that is well characterized and that has a reliable conceptual model.

The Savannah River Site (SRS) F-Area is a two square kilometer field site located down gradient of the F-Area separations facility. Liquid process waste was disposed into unlined seepage basins during the period between 1955 and 1988. The associated groundwater plume contains dissolved uranium, strontium, iodine, technetium, tritium, as well as other radionuclides and metals. Despite many years of active remediation including pump-and-treat, the groundwater still remains acidic, and the concentrations of U(VI) and other radionuclides are still significant.

The vast historical datasets at the SRS F-Area provide an opportunity to test various remediation and monitoring strategies. The mature conceptual site model includes detailed information on site hydrology, geologic features, and contaminant distribution. In addition, implementation of a phased remedial strategy that combines standard and innovative remedial approaches over several decades has resulted in the development of a rich database of supporting measurements. In parallel, a 3D flow and reactive transport model has been developed under ASCEM, which is critical to provide mechanistic and predictive understanding of the contaminant plume behavior, and also to evaluate the sensitivity and effectiveness of new monitoring approaches in the future.

Combination of these two components enables us to transform the F-Area to be a real/virtual *testbed* for DOE-EM applications.

3.1. Enhanced Attenuation

In addition to the existing focus on enhanced attenuation remedy for uranium, we include Tc-99, which is major risk driver for large complex plumes at Paducah and Hanford due to its relative mobility and long half-life. At F-area, Tc is present in the plume but is not considered a major risk as it naturally attenuates at the seepline interface (i.e., groundwater-surface water interface), where the groundwater seeps into surface and the plume has the first contact with organic carbon-rich soil. We have hypothesized that organic carbon will enhance the Tc-99 sorption, and reduce release of Tc-99 into surface waters. We are actively investigated the natural attenuation processes and plan to evaluate methods to enhance these natural processes. The plume geochemistry and geologic environment are analogous to the Paducah site, where Tc is migrating from the C-400 toward a similar seepline to the Ohio River.

We will evaluate chemical/physical reactants to immobilize, precipitate, transform, or fix soluble Tc-99 in a shallow, multi-contaminant groundwater plume and shallow surface water. A key issue is the episodic nature of outcropping of the seepline plume. The seepline is a dynamic environment. We need new monitoring approaches that will allow us to map the contamination reliably in a contaminated area. To achieve this goal, we propose to map the spatially heterogeneous distributions of Tc-99 and other radionuclides using UAV and/or three-dimensional gamma/Compton camera. Since the system understanding of geochemistry is critical for EA, various characterizations will be performed such as carbon composition in the organic-rich sediment. Characterization will be done periodically so that maps can be subtracted so that areas with significant changes can be quickly and cost effectively identified leading to an understanding of the dynamic processes.

In addition, 3D reactive transport modeling will be used to evaluate the impact of EA, and predict if and when it will be possible to transition from active to passive cleanup of contaminated groundwater using MNA. Such results will provide an estimate of life-cycle cost and cost saving of EA and other alternative options. The uncertainty quantification (UQ) capabilities are particularly useful to evaluate the impact of hydro-geochemical and other uncertainties on the life-cycle cost.

3.2. Long-term Monitoring

At the SRS F-Area, data mining of the historical datasets has showed excellent correlations between in situ measurable variables (e.g., pH, and nitrate) and contaminant concentrations, including tritium and uranium. Note that nitrate can be measured in situ at this site, since it dominates total dissolved solid, and hence determines electrical conductivity. Such strong correlations suggest the feasibility of inferring contaminant concentrations based on the *in situ* sensors, by describing the contaminant concentration as a function of the in situ measurable parameters. There is some uncertainty in the correlations, suggesting the importance of quantifying the uncertainty at each location, as well as the quantifying critical points for different variables at which contaminant concentrations begin to deviate from the correlation.

To achieve more accurate estimation of the contaminant concentrations, the future work focuses on implementing the automated estimation method based on the Kalman filtering approach. The Kalman filter was originally developed to control spacecraft or robots by tracking (i.e., estimating) their trajectory and movement based on indirect data or noisy signals. For groundwater monitoring, the Kalman filter enables us to continuously estimate contaminant concentrations based on *in situ* measured data. It also provides a systematic approach to update the correlation parameters real-time (which leads to more accurate estimation), as well as to quantify the uncertainty of the estimates, given noise and measurement errors. The approach will be demonstrated using the in situ sensors currently deployed at the F-Area. In addition, other machine learning approaches such as artificial neural network will be tested and compared.

The virtual testbed (i.e., A 3D flow and reactive transport model) has provided a mechanistic understanding of the correlations between *in situ* variables and contaminant concentrations. In addition, the virtual testbed has proved to be useful to project the correlations into the future, and to investigate the effectiveness of the proposed monitoring strategy. The uncertainty quantification has been used to investigate the impact of the uncertainties and variability in hydrological and geochemical parameters.

In the future, the virtual testbed will be used to explore the impact of climate change and other hydrological shifts on monitoring strategies, as well as to develop a better strategy for optimal spatial-temporal placements of the monitoring well locations. The UQ simulations will be used to estimate the life-cycle cost of monitoring and other key metrics (e.g., how many wells are enough, and how long we need monitoring).

4. Relevance to the Other Contaminated Sites

4.1. Nuclear Power Plants

Soil and groundwater contamination has been reported at several nuclear power plant sites [NRC, 2006]. Nuclear industry has initiated a voluntary monitoring program, which includes regular (annually or quarterly) groundwater sampling at wells [EPRI, 2008]. The main contaminant of interest is tritium, although other radionuclides such as strontium have been detected. Tritium is a weak beta emitter, which does not produce gamma ray. Quantifying tritium concentrations requires laboratory measurements, which could take several days and delay the response against leaks. Since most of the plumes are within the site boundaries, none of the plumes have caused significant health risk. However, the groundwater contamination has led to a large increase in decommissioning cost due to remediation (i.e., excavation) and soil waste disposal.

High-resolution hydraulic modeling at commercial nuclear sites could be used delineate flow pathways, and it may be possible to design barrier systems to slow and delay radionuclide transport especially where contaminant flow is highly stratified. For example, analysis of the barrier system at the F-area Seepage Basin site at SRS indicates that barriers have isolated highly contaminated flow paths from exiting the barrier system allowing time for decay of tritium and uranium in situ. These types of strategies may prove effective at other sites.

Monitoring needs at nuclear power plants are to (1) develop a rapid detection system of contaminant leaks, and (2) decrease routine monitoring cost. The new monitoring technologies described above can be directly applicable to tackle these challenges. Although current *in situ* sensors do not measure tritium directly, they can detect many of controlling variables or indicators of leaks from reactors (such as electrical conductivity, pH, soil moisture and temperature). *In situ* monitoring of tritium surrogates at wells can be done remotely and continuously in time, which can reduce the cost of groundwater sampling and analysis, as well as enable the rapid detection of leaks. In addition, spatially extensive monitoring techniques – fiber optics and geophysics – are particularly powerful for such detection due to their large spatial coverage.

4.2. DOE Legacy Management

One responsibility of the DOE Office of Legacy Management is management of former UMTRCA sites (Title I of Uranium Mill Tailing Radiation Control Act) where uranium ore to support the national defense was processed. At some of these former processing sites, the residual radioactive material is sequestered in place; at others, the residual radioactive material was moved from the processing sites to offsite disposal locations. The remedial strategies implemented to treat the residual contaminated groundwater use either active pump and treat or passive natural flushing of groundwater. At the larger complex sites, there is a need to ongoing need to monitor residual contamination. The new monitoring approaches discussed are directly applicable at these arid sites where groundwater flow is slow and surface vegetation is sparse allowing the effective use of spatial geophysical methods described above.

Looney et al (2013) evaluated the potential use of existing high resolution aerial gamma surveys overseen by Region 9 of the U.S. Environmental Protection Agency (EPA) for monitoring. Aerial radiation surveys were flown over 41 blocks in the Navajo Nation during October 1994 through October 1999, and covered areas of known or suspected uranium mining and milling operations. The surveys were conducted by the DOE Remote Sensing Laboratory to assist with locating and characterizing abandoned uranium mines (AUMs) and to quantify the potential radiation exposures associated with these and other historical uranium mining/milling activities. The surveys were flown using a helicopter-based acquisition platform. These local surveys were particularly useful because they were flown at low altitude (45 m above the terrain) with 100% coverage and provide high resolution information. The data were integrated at one second intervals and provide an average radiation level for each 90 m diameter footprint under the helicopter as shown.

Looney et al (2013) provided the following analysis for the Tuba City facility in Arizona. Despite the low levels of radioactivity, the pattern of gamma exposure levels revealed significant information about background conditions and about the potential transport pathways for milling related radionuclides. One of the most notable features shown in Figure 4 is the band of high natural gamma exposures along the entire surveyed reach of Moenkopi wash. This gamma is associated with the evaporative accumulation of minerals in a regional discharge area and the accumulation of heavy minerals due to physical sorting and concentration in a surface water environment that is subject to flash flooding. The background gamma exposure levels are the lowest in areas of surficial sand dunes and relatively higher in other areas. In the vicinity of the mill site, there are detectable gamma signatures toward the east, north and south. The eastern gamma signature was previously observed (Havens and Dean, 1967) and attributed to windblown dust from the mill site during the period when the tailings were not consolidated and covered. The north signature is associated with the "Highway 160 Project Site" – an area that was later characterized and cleaned-up (see NNEPA, 2011). The gamma exposure levels are notably low over the area of the tailings cell. Importantly, the data indicate that the LM cleanup actions – collection/consolidation and covering of contaminated tailings and soils – were relatively effective since the terrestrial gamma exposure levels measured in the post-1990 aerial gamma were significantly below the historical values measured by Havens and Dean (1967) and, as noted above, meet applicable guidelines to protect humans and the environment. The gamma signature to the south of the mill site is particularly interesting because it is indicative of groundwater transport and discharge (either at seeps or evapotranspiration boundaries) and/or indicative of erosion and overland transport of contaminated soils from the middle terrace to the lower terrace. The southern transport pathway is limited in scale and is consistent with the expected short flow distances for the uppermost groundwater flow lines or limited erosion. Note that there is no southern transport pathway observed for the windblown area (the surficial contaminants to the east) – possible evidence that supports the subsurface/groundwater pathway from the main mill site area. For the southern transport direction, however, the aerial gamma surveys alone do not allow the alternative pathways to be differentiated and additional lines of evidence (e.g., study of contaminant profiles above the water table) would be needed to further refine and quantify the conceptual model.

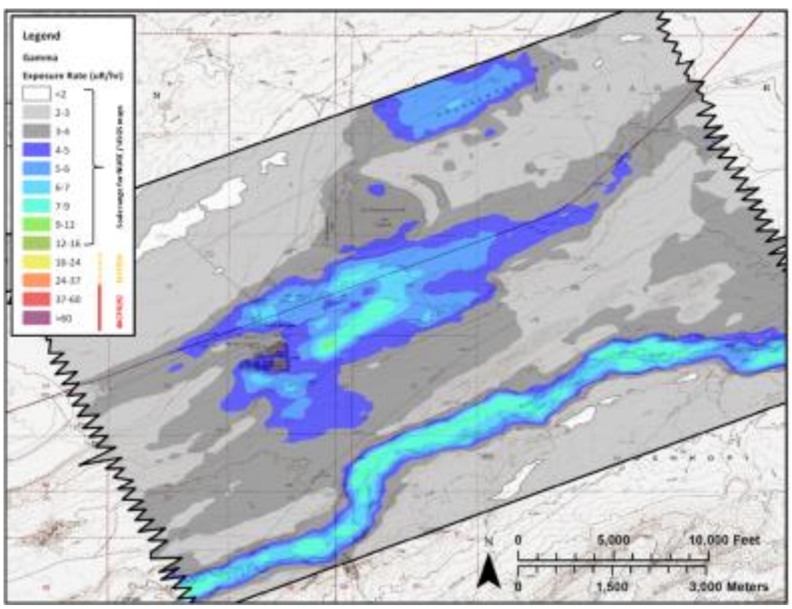

Figure 4. High resolution terrestrial gamma exposure map (uR/hr) in the vicinity of the Tuba City Mill Site from Looney et al, 2013. Tuba City UMTRA tailing pile shown lower center, large linear area to the right shows Moenkopi wash

In this example, the aerial gamma data provide a powerful and cost effective technique to test and explore conceptual models of radionuclide transport near the Tuba City Site. Additional geophysical surveys could be used to easily identify areas where significant changes in gamma deposition would require additional sampling. This example provides an example of how this type of information can be used to improve technical understanding of the site, facilitate clear communications with regulatory agencies and the Navajo and Hopi Nations, and inform future environmental management decisions at Tuba City and at other LM sites.

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