Overview of U.S. Department of Energy Office of Legacy Management Applied Science and Technology Program - 11347

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ABSTRACT

The Applied Science and Technology (AS&T) program has a critical long-term surveillance and maintenance (LTSM) role for the U.S. Department of Energy (DOE) Office of Legacy Management (LM). DOE applies science and technology advances to ensure that the implementation of LTSM will be efficient and cost-effective. Objectives include (1) apply advances in science and technology to improve sustainability of remedies, (2) share technologies and lessons learned with others, (3) publish project results to provide a measure of credibility and to bring visibility to LM, and (4) collaborate and share project costs.

This paper highlights the following ongoing AS&T projects:

- System Operation and Analysis at Remote Sites
- Monticello Permeable Reactive Barrier and Treatment System
- Radon-222 as a Natural Tracer for Permeable Reactive Barriers
- Chelation Enhancement of Zero-Valent-Iron-Based Treatment Cells
- Natural Contamination in Mancos Shale
- Monticello Water Balance Cover Lysimetry
- Soil Water Fluxmeter Evaluation
- Cover Renovation Lysimeter Test Facility
- Remote Sensing Case Study at Monument Valley

INTRODUCTION

The U.S. Department of Energy (DOE) Office of Legacy Management (LM) has responsibility for the management of environmental impacts from 87 sites nationwide that resulted from legacy operations, particularly during the Cold War. Many of these sites have contaminated groundwater and 41 have waste disposal cells. LM established an Applied Science and Technology (AS&T) program to address application-based science for these sites and support technology advances to ensure that the implementation of long-term surveillance and maintenance (LTSM) will be efficient and cost-effective. In general, this means moving the "state of the science" in long-term stewardship strategies and methods into the "state of the practice" at LM sites. Site stewards also need better information and resources to work more effectively with regulators and stakeholders in exploring whether new or improved approaches may work better than baseline technologies. The overriding goal is to explore and apply innovative ways to reduce LTSM costs and risks to human health and the environment.

The LM AS&T strategy focuses on the following objectives:

- Ensure that sound engineering and scientific principles are used to conduct LTSM.
- Evaluate and improve the effectiveness of LTSM practices.
- Track and apply advances in science and technology to improve sustainability of remedies.
- Share technologies and lessons learned with stakeholders; regulators; and state, tribal, and local governments.
- Publish project results to provide a measure of credibility in the scientific and regulatory communities, to bring visibility to LM-applied science and technology initiatives, and to enable others to utilize the results.
- Collaborate and share project costs with other DOE offices, other agencies, academia, and industry.

This paper highlights some ongoing and proposed future AS&T projects.

System Operation and Analysis at Remote Sites (SOARS)

SOARS was implemented in 2006 to improve data collection at LM sites. This project established the feasibility of collecting data remotely in real time and transmitting them to LM servers. Many LM sites are in remote locations, and field visits are costly. Uses of SOARS include operation and monitoring of pump-and-evaporate groundwater remediation systems at Shiprock, New Mexico; zero-valent iron treatment cell operation and monitoring at Monticello, Utah; disposal cell cover performance evaluation at Monticello; phytoremediation at Monument Valley, Arizona; groundwater tracer testing using rhodamine dye injection at Mound, Ohio; and assessment of groundwater plume dynamics at Rifle, Colorado, Durango, Colorado, and Green River, Utah. Data are collected from 539 sensors on 102 dataloggers, located at 15 LM sites in 9 states (Fig. 1). Sensor measurements include flow rate, water levels, in-line pressure, specific conductance, temperature, pH, oxidation-reduction potential, oxygen levels, wind velocity, relative humidity, rainfall, solar energy, water content, unsaturated water flux, and chemical injection rates.

Data are collected on frequencies ranging from 1 minute to 24 hours. More than 250,000 data values are collected daily. A centralized post-processing program automatically captures the data and updates graphical displays. Alarms are sent to project personnel if specific triggers are exceeded (for example, if flow rates have ceased at a pump-and-treat remediation project or if data have not been collected). All graphs and raw data are provided to site personnel in real time on a Web-based server. The

immediate availability of data greatly facilitates interaction among personnel who are sometimes located in different parts of the country.

The data are able to capture changing site conditions such as flow direction or equipment failures. The post-processor is used to make real-time calculations such as changes in the slope of the water table. SOARS data and graphs are available immediately, and corrective actions can be expedited. SOARS greatly improves the ability to diagnose problems and make timely repairs and adjustments. Use of SOARS also reduces the need for on-site support, reducing the need for travel.



Fig. 1. Location of LM sites and SOARS stations.

SOARS also incorporates a surveillance system. Three still cameras and a Web-based real-time camera are included in the system. The photos are used to detect the presence of birds and other wildlife that might access contaminated ponds and to provide security at the site.

Monticello Permeable Reactive Barrier and Treatment System

A permeable reactive barrier (PRB) was constructed at LM's Monticello site in 1999 to contain the spread of uranium-contaminated groundwater in a shallow alluvial aquifer. The PRB consisted of 250 tons of granular cast zero-valent iron (ZVI) placed in a 30-m-long reactive zone within the path of the contaminated groundwater [1]. The reactive zone was flanked by 100 m of impermeable slurry wall to direct all of the groundwater in the alluvial aquifer through the reactive zone. The PRB effectiveness decreased over time because of precipitation of calcium carbonate and iron oxide

minerals in the pores of the reactive media. For example, more than 8 metric tons of calcium carbonate had precipitated in the reactive media within 2.7 years, as indicated by the chemical analysis of more than 250 core samples [2]. This mineralization in the ZVI led to a four-orders-of-magnitude reduction in hydraulic conductivity in 5 years of operation [3]. Although concentrations of contaminants remained low in water samples collected from wells completed in the ZVI, the flux of groundwater through the PRB was uncertain and likely had diminished considerably. Further analysis using results of tracer tests, regular monitoring of groundwater chemistry, and groundwater elevations indicated that contaminated groundwater, unable to pass freely through the reactive zone, had found pathways to bypass it without being treated.

Chemical reactions in the ZVI media worked remarkably well for uranium containment, as the system achieved a nearly complete removal of uranium from the groundwater within a few minutes. Other contaminants such as selenium and arsenic were also removed. The performance issues concerned the reactions that caused undesired mineral precipitation and flow diversion rather than contaminant treatment. Thus, it seemed reasonable to use the same reactive media (ZVI) but place it in an ex situ treatment cell so that the media could be exchanged as it became fouled. To test this concept at field scale, DOE built an experimental ex situ treatment system at the Monticello site in June 2005 (Fig. 2). Successful operation of the treatment cell led to the construction of a second treatment cell in April 2007. The treatment cells have been operating for 5 years with minimal intervention except for media change-out about once every 1.5 years. The cost to construct, operate, and monitor the treatment system is significantly less than for the PRB based on treatment of equivalent volumes of groundwater.

Radon-222 as a Natural Tracer for Permeable Reactive Barriers

During operation of the Monticello PRB (see previous section), it was recognized that permeability reduction from mineralization was a main cause of system failure. The mineralization led to modification of the groundwater flow paths. Delineation of the groundwater flow paths in such a setting is not straightforward. Because hydraulic conductivity distribution was changing at a rapid pace, conventional flow delineation using Darcy's Law principles was not useful in evaluating the flow field. Tracer testing using gaseous (argon and helium) and ionic (bromide and iodide) tracers injected upstream of the PRB resulted in mixed and uncertain results. Conventional tracer tests such as these are costly, requiring introduction of the tracer over a long time period and collection and analysis of numerous samples. Potential problems include maintaining the injected tracer at a known and constant concentration and collecting sufficient spatial and temporal samples to intercept the flow paths. Recovered masses of the tracers during the Monticello tests were much less than the masses injected, and thus, much of the tracer found flowpaths outside the monitored area despite a fairly dense array of monitoring wells.





Fig. 2. Groundwater treatment cells at Monticello, Utah.

As an alternative to conventional (injected) tracer methods, it was recognized that radon-222 that occurs naturally in all aquifers could be used as a tracer to determine the residence time in a ZVI-based PRB [4]. If much of the groundwater was flowing past the ZVI rather than through it, we would expect the groundwater within the ZVI to stagnate and have a long residence time. Radon-222 concentrations are relatively constant in most groundwater systems and are at high enough levels to be measured accurately by conventional liquid scintillation methods. Radon-222 in groundwater results from steady-state emanation from its parent radium-226 contained in aquifer minerals. Concentrations of radon-222 range from about 500 to several thousand picocuries per liter in most shallow alluvial groundwater. The background concentration of radon-222 in the Monticello alluvial groundwater is about 1000 pCi/L. Radon-222 has a half-life of 3.8 days, making it ideal for the study of residence times in PRBs within time periods up to about 400 hours (Fig. 3).



Fig. 3. Determination of residence time from radon-222 concentrations in samples collected from a ZVI-based PRB [4].

The design residence time in the ZVI was about 20 hours. Radon-222 concentrations in groundwater samples collected from the ZVI zone yielded much longer residence times, averaging 286 hours. Residence times calculated from the radon-222 concentrations correlate well with chemical signatures of residence time expected from modeling the progressive corrosion of ZVI [5].

Chelation Enhancement of Zero-Valent-Iron-Based Treatment Cells

The treatment cells at the Monticello site (see previous sections) have been reasonably cost-effective in removing contamination from groundwater using ZVI. However, the main cost is due to the need for media replacement. Significant cost savings are possible if the longevity of the ZVI can be extended. Over time, precipitation of calcium carbonate and iron oxide minerals causes media degradation similar to that which occurred in the PRB (see previous section). Results of bromide tracer testing of the Monticello treatment cells indicate that hydraulic conductivity decreases and chemical dispersion increases over time. This project investigated the idea that mineral precipitation in the reactive media could be reduced using a chelate that combines with iron and calcium to keep them in the aqueous phase.

EDTA and citrate are known to chelate calcium and iron and were investigated. Based on results of initial column testing, it was apparent that citrate was capable of maintaining both iron and calcium in solution. Column longevity, based on uranium uptake, increased by about 30 percent. However, tests conducted with different solution compositions

indicated that uranium breakthrough was earlier than without chelation, suggesting that uranium was also chelated. This study is still in its early stages, and efforts will be made to better understand the chemical conditions controlling iron, calcium, and uranium mass transfers between aqueous and solid phases.

Natural Contamination in Mancos Shale

Selenium, and to a lesser degree uranium and arsenic, is known to occur naturally in shallow aquifers, particularly in areas where groundwater contacts dark gray shale such as the Cretaceous Mancos Shale. Because these same constituents contaminate groundwater at uranium mill tailings disposal sites, establishing background standards for contaminant plumes can be difficult if the disposal cells are built on Mancos. The Mancos Shale is thought to be a good substrate for disposal cells because of its low permeability and the presence of salts and metals that render the natural water of limited use. Several uranium mill waste disposal cells are located on the Mancos Shale (e.g., Shiprock, New Mexico, and Grand Junction, Colorado), and one of the largest tailings piles (Moab, Utah) on the Colorado Plateau is currently being relocated to a site at Crescent Junction, Utah, which is underlain by a thick section of Mancos Shale.

This is a new project that is investigating the chemical composition of groundwater from the Mancos Shale. Sampling locations were identified over much of the Mancos depositional basin. Reconnaissance identified more than 30 Mancos seeps in Colorado, Utah, and New Mexico that were sampled in the fall of 2010. Uranium-234/238 activity ratios are included in the study to determine if they are useful in determining whether the source of uranium is mill-related or natural. Preliminary analyses indicate that activity ratios are close to 1 nearer uranium tailings sites, whereas farther away the ratios are closer to 2.

Water Balance Cover Lysimetry

LM is investigating alternatives to conventional cover designs for uranium mill tailings disposal cells. A cover constructed in 2000 near Monticello was a redundant design with a conventional low hydraulic conductivity composite cover overlain with an alternative cover designed to mimic the natural soil water balance as measured in nearby undisturbed native soils and vegetation. Conventional cover designs for uranium mill tailings include compacted soil layers (CSLs) to slow radon diffusion and to impede percolation into underlying contaminated materials. Several studies have shown that saturated hydraulic conductivity of CSLs in conventional covers for uranium mill tailings, and in similar covers for other applications, are higher than expected, often by several orders of magnitude.

An alternative design approach is to manipulate the cover ecosystem with the goal of enhancing soil water storage and evapotranspiration (ET). The long-term sustainability of this water balance cover design depends on interactions of the climate, soil hydrology, and plant ecology of the site. In many arid and semiarid ecosystems, relatively low precipitation, high potential ET, and thick unsaturated soils limit recharge. Water balance covers are designed to mimic this soil water conservation [6]. Capillary barriers composed of coarse-textured sand and gravel placed below the soil "sponge" can enhance soil water-storage capacity and limit unsaturated flow. The Monticello water balance cover [7] relies on the water-storage capacity of a 163-cm, fine-textured soil and rock layer overlying a 38-cm sand capillary barrier layer. Hydraulic performance can be evaluated as the probability that ET is sufficient to prevent water accumulation in the soil sponge from exceeding the storage capacity in any given year.

The Monticello design includes frost protection, deterrents for biointrusion, and attributes for plant establishment and growth. The soil depth is more than adequate to protect underlying components (CSL and geomembrane) from frost damage. The topsoil layer has physical and hydraulic properties similar to those of the rest of the water-storage layer but also contains the nutrients, propagules, and microorganisms (e.g., mycorrhizae) needed to establish a sustainable plant community. The soil thickness is the primary biointrusion deterrent. Water retention in the storage layer should limit deep root penetration; the layer also exceeds the depth of most burrowing vertebrates in the area. A 30-cm layer of cobble-sized rock above the capillary barrier is an added deterrent should deeper burrowers enter the area. Fine-textured soil fills the interstices of this rock layer and prevents it from behaving like a second capillary barrier.

Water balance monitoring (Fig. 4) within a 3.0-ha drainage lysimeter, embedded in the Monticello cover during construction, provides convincing evidence that the cover has performed well over a 10-year period, 2000–2010. The total cumulative percolation, 4.8 mm (approximately 0.5 mm per year), satisfied a regulatory goal of <3.0 mm per year. Most percolation can be attributed to the very wet winter and spring of 2004–2005, when soil water content exceeded the storage capacity of the cover. Diversity, percent cover, and leaf area of vegetation increased over the monitoring period, indicating that natural plant succession can increase ET and the long-term performance of the water balance cover.



Fig. 4. Soil water balance parameters (precipitation, soil water storage, surface runoff, evapotranspiration, and percolation) measured in a 3-ha lysimeter embedded in the Monticello, Utah, disposal cell cover. Evapotranspiration was calculated using both on-site precipitation measurements and data from a nearby National Weather Service station.

Soil Water Fluxmeter Evaluation

Monitoring the hydrologic performance of disposal cell covers has proven to be a challenge. Water content and water potential sensors are generally inadequate because they do not measure flux rates directly. Water-sensing data must be coupled with estimates of the soil's unsaturated hydraulic conductivity, giving rise to water flux estimates that may be off by an order of magnitude or more. Similarly, large uncertainties exist with water balance models used to predict drainage, particularly at low flux rates. The only direct and proven way to verify percolation flux rates is using lysimetry. However, large pan lysimeters cannot be easily installed post-construction in an operational final cover.

As an alternative, LM is investigating a new type of passive wick lysimeter, or water fluxmeter (WFM), which can be used to directly monitor percolation flux through existing disposal cell covers. The WFM, developed by Pacific Northwest National Laboratory, features a funnel to direct water from the soil into a passive wick for moisture tension control, a miniature tipping bucket for real-time flux measurements that can be calibrated from the surface, and a pipe or chimney extending above the funnel to minimize divergent flow [8]. Three WFMs were installed in the top slope of the Lakeview, Oregon, disposal cell during the fall of 2005. WFMs were placed in holes augered into the upper tailings material just below the radon barrier. The results of the pilot study show that WFMs can be installed in existing disposal cell covers to directly monitor percolation flux. Cumulative percolation remained exceptionally high during a 4-year monitoring period [9], possibly because the WFMs were strategically placed in downgradient locations where there may be a water-harvesting effect. The WFMs may have a relatively short operating life. One of the three WFMs employed in the test failed within 2 years of its installation.

Four WFMs were placed at depths of 3.0–3.5 m in phytoremediation test plots at the Monument Valley site to evaluate percolation below the root zone of native desert phreatophytes planted to remove nitrate and control the soil water balance [10]. The WFMs are capable of directly monitoring saturated and unsaturated water fluxes ranging from 0.02 mm per year to more than 1,000 mm per year. The four WFMs have recorded zero percolation since they were installed in March and July 2006, supporting the hypothesis that infiltration from the combination of ambient precipitation and irrigation can be stored in the fine-sand profile and not percolate and leach nitrate into the alluvial aquifer.

Cover Renovation Lysimetry

A lysimeter test facility was constructed in 2007 to evaluate a renovated cover design at an LM disposal cell constructed with a conventional, low-permeability cover design. Testing the renovation procedure on the existing cap was not practical. Thus, two identical large-scale lysimeters simulating the cover were constructed adjacent to the Grand Junction disposal cell [11]. One of these lysimeters will be renovated, whereas the other will be monitored as a control that simulates the existing cover. The renovation treatment will be applied to one test section after surface conditions indicate deposition in the riprap is occurring, and the monitoring data indicate that the radon barrier has become more permeable.

The lysimeters were modeled after the test sections used in the U.S. Environmental Protection Agency's Alternative Cover Assessment Program [12]. Each 10 m \times 20 m lysimeter includes instrumentation to monitor percolation from the base of the cover, a runoff collection system, a suite of instruments to monitor state variables within the cover profile, and a weather station to monitor meteorological conditions. This system permits quantification of all components of the water balance, namely precipitation, runoff, soil water storage, percolation, and ET. The latter is obtained by difference.

The lysimeters were constructed using the same earthen materials employed for the fullscale cover, which had been previously stockpiled in 1992 for future closure activities at the Grand Junction disposal cell. Therefore, all cover materials used in construction of the test facility are identical to those in the cover over the disposal cell. Heavy equipment similar to that used for the full-scale cover was employed for construction.

Remote Sensing Studies

LM is investigating the efficacy of using remote sensing technologies to improve performance monitoring of disposal cells and eventually reduce the costs of LTSM at LM sites. Remote sensing can provide nondestructive and spatially comprehensive reconnaissance and also reduce the frequency and enhance the effectiveness of on-site LTSM inspections.

Several important cover performance parameters can be remotely monitored. Multispectral and hyperspectral sensors can be used to map spatial patterns and temporal changes in vegetation growing on and surrounding disposal cells. Changes in vegetation may alter the performance of covers in different ways. For conventional covers, Long-Term Surveillance Plans often require control of plant encroachment and root intrusion, especially infestations of noxious weeds. In contrast, alternative covers often rely on vegetation to extract soil water and limit deep percolation and contaminant leaching. Vegetation on covers might be monitored as a surrogate for other performance parameters. Changes in vegetation patterns and health can occur in response to disturbances, such as erosion or animal burrowing. Changes in the growth and health of vegetation may also reflect changes in soil moisture patterns or the presence of heavy metals. Variation in water content is often manifested in vegetation biophysical parameters, such as leaf area index (LAI) and biomass, and can be detected in optical spectral reflectance characteristics. Vegetation spectral responses have also been successfully used for the detection of contaminant leaks at Superfund sites and on landfills. Remote sensing can therefore be used to survey the spatial and temporal variation of vegetation as a surrogate measure of other changes taking place on covers.

A combination of field measurements and remote sensing was used to measure transpiration by two native desert phreatophytes, black greasewood and fourwing saltbush, growing over a nitrate plume at Monument Valley [13]. LM is studying the use of black greasewood and fourwing saltbush to control hydraulic gradients and slow the spread of the plume. Heat balance sap-flow sensors were used to measure transpiration by the two phreatophytes, and results were scaled to larger landscape units and longer time scales using LAI, fractional vegetation cover, meteorological data, and the enhanced vegetation index from the Moderate Resolution Imaging Spectrometer (MODIS) sensors on the Terra satellite. Transpiration was high, depending on leaf area, and was controlled by vapor pressure deficit in the atmosphere. Black greasewood tended to have higher transpiration rates than fourwing saltbush and had a steeper response to vapor pressure deficit, but both exhibited midday depression of leaf conductance. Over most of the site, fractional vegetation cover and area-wide LAI were low due to heavy grazing by cattle and sheep. However, a portion of the plume that had been protected from grazing for 10 years had higher transpiration rates. Transpiration rates on a ground-area basis varied with LAI. Controlling grazing could, theoretically, slow or halt the movement of the contamination plume by allowing the shrub community to extract more water than is recharged in the aquifer.

Proposed Future Projects

Future projects may include (1) a catalog of natural analogs to support long-term cover performance evaluations, (2) an evaluation of bio-uptake of contaminants on covers, (3) an evaluation of the effects of cover soil development processes on radon attenuation, (4) the impacts of naturally occurring contaminants, (5) mechanistic studies of monitored natural attenuation processes, (6) an evaluation of groundwater monitoring tools, and (7) an assessment of groundwater remediation technologies.

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