Encapsulation of flue-gas desulfurization wastewater using coal combustion by-product mixtures: Field lysimeter investigations

Jason J. Raucci [1] (jraucci@dbstephens.com), Jeffrey Samson [1] (jsamson@dbstephens.com), Kirk M. Ellison [2] (kellison@epri.com), Benjamin J. Gallagher [2] (bgallagher@epri.com), Joleen S. Hines [3] (jhines@dbstephens.com), James A. Kelsey [1] (jkelsey@dbstephens.com)

[1] Daniel B. Stephens & Associates, Inc., 6020 Academy Rd NE, Suite 100,
Albuquerque, NM 87111

[2] Electric Power Research Institute, 3420 Hillview Ave, Palo Alto, CA 94304

[3] Daniel B. Stephens & Associates, Inc. Soil Testing & Research Laboratory,
4400 Alameda Blvd NE, Suite C, Albuquerque, NM 87113

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Abstract

Wastewater management at coal-fired power plants is becoming a more prominent issue as additional emissions reduction equipment is used and regulations become more stringent. Wastewater derived from the flue-gas desulfurization process (FGD WW) is typically high in halides and has few cost-effective options for treatment and disposal. New regulatory limitations on FGD WW discharge may require water treatment facilities at many electrical generating units. The need to investigate alternative approaches for disposal of FGD WW has therefore acquired new urgency.

Solid-phase wastewater sequestration has been proposed as a potential breakthrough approach to FGD WW management. Sequestration is achieved by wastewater encapsulation within a cementitious, thixotropic paste consisting of concentrated wastewater, coal combustion by-products (fly ash), and binders that sets into a monolithic solid after emplacement within a landfill cell. To assist in analysis of ongoing encapsulation studies, a series of bench- and demonstration-scale experiments are being conducted to determine hydraulic transport properties of brine-ash encapsulated monoliths. At three sites in the southeastern United States, field lysimeters have been constructed using both paste encapsulation and conventional low-moisture compaction emplacement. Changes in moisture content and physical properties of the monolithic and compacted materials are evaluated through the use of embedded sensors, and are correlated with climate data generated by an on-site weather station. Runoff and

generated leachate are monitored using automated gauges, and are captured for volume verification and chemical analysis; these data also allow a water budget determination for the experimental cells. This paper summarizes findings from the first year of field lysimeter studies.

Introduction

The need for improved management of wastewater and coal combustion by-product (CCB) waste streams, especially for wet flue-gas desulfurization (FGD) systems, is being driven by changes in plant operations and environmental regulations. In a typical wet FGD system, an aqueous slurry containing lime or limestone passes in contact with flue gas to remove SO_2. Along with SO_2, other constituents of potential concern (COPCs), as well as halide salts, are collected in the FGD slurry.

Effluent limitation guidelines (ELGs) under consideration by the U.S. Environmental Protection Agency (EPA) would place stringent limits on the discharge of COPCs to receiving water bodies from FGD wastewater (FGD WW), and would likely increase treatment requirements for FGD waste streams. Wastewater from FGD processes may be managed through treat and discharge approaches or by eliminating discharge of the wastewater all together. Discharge elimination typically requires volume reduction, which can be achieved by a variety of methods (Renew et al., 2016; Ellison et al., 2017); however, the residual concentrated wastewater or brines contain significant concentrations of COPCs, as well as high concentrations of dissolved salts. The residual liquid waste must therefore also be managed and disposed of effectively.

The electric power industry has funded significant research in recent years working toward the prospect of achieving effective and holistic discharge elimination, including management of residual brines. One component of this effort has been investigating the potential for sequestration of leachable contaminants present in brine wastewaters and other CCB through solidification and stabilization (S/S) within a cementitious paste, a process known as wastewater encapsulation. Research conducted by the Electric Power Research institute (EPRI) and directly by electric utilities, at both the laboratory and field scale, suggests that such an approach could realize significant benefits over the practice of using the FGD WW to moisture-condition coal fly ash prior to compaction in lined landfill cells (Ellison, 2015; Sniderman, 2017). Co-disposal of FGD WW with CCB through S/S has the potential to be an effective discharge elimination option for FGD WW. This approach has the advantages of more effectively preventing the reintroduction of salts and COPCs from wastewaters and CCBs back into the environment than current methods, while co-disposing two abundant waste products generated at coal-fired power plants.

Previous Studies

The U.S. EPA regards encapsulation through S/S as an established treatment method for numerous waste categories (Ellison et al., 2016). A growing body of research has examined the potential of this approach for sequestering and immobilizing contaminants associated with the FGD waste stream. For example, Mooketsi et al. (2007) and Muntingh et al. (2009) established in principle that solidified brine/CCB pastes may sequester both salts and metals in a relatively impermeable matrix. Mahlaba et al. (2011) demonstrated that brine chemistry exerts significant control on the physical properties of solidified fly ash pastes. Recent laboratory studies have evaluated the potential for metals and salts in brine solutions to be effectively sequestered in fly-ash-based cementitious pastes for various compositions (e.g., Oza et al., 2015; Ellison et al., 2016; Renew et al., 2016 and 2017). Ellison et al. (2017) provided a broad overview of encapsulation and sequestration approaches to brine disposal, and recommended a robust program of laboratory and field studies to further explore and quantify the potential of this approach. Field lysimeter studies were suggested to assess the long-term performance of simulated disposal cells composed of fly ash encapsulated brine solutions and other materials.

Purpose of Investigation

In cooperation with EPRI and electric utility host sites, Daniel B. Stephens & Associates, Inc. (DBS&A) is currently conducting a series of field studies using lysimeters composed of various wastewater and fly-ash-based materials to assess and compare the physical and hydraulic properties of paste-encapsulated mix designs to conventional moisture-conditioned and compacted materials. Lysimeters consist of simulated waste cells containing the material being investigated and a basal drainage layer, emplaced in an aboveground vault. Movement of water through these simulated waste cells can be detected by embedded instrumentation and quantified as leachate or runoff, or change in storage within the waste cell. The principal objectives of these preliminary field studies are:

- Gain understanding of the quantity and quality of runoff and leachate water
- Assess the durability and usefulness of commercially available sensors to detect water movement through the highly saline and solidified cementitious materials
- Assess the hydraulic properties of these materials under long-term exposure to environmental conditions.

Experimental Design

Several studies, including bench-scale studies, column studies, and sensor evaluation studies, have been completed by the DBS&A Soil Testing & Research Laboratory in Albuquerque, New Mexico to provide insight into the physical and hydraulic properties of various wastewater and CCB material mixtures.

Objectives

The principal objectives of the current field lysimeter studies are summarized in the following subsections.

Assess Sensor Effectiveness

A principal goal of these initial field studies is to assess sensor responses and long-term performance in highly saline, alkaline, and cementitious materials. Each of the field lysimeters described in this paper is equipped with sensor arrays installed in vertical profiles within a lysimeter material matrix. Each sensor was evaluated in the laboratory prior to being chosen for further evaluation under field conditions. The specific layout of the sensor array at each installation is described later in this paper but, in general, each is equipped with the following sensors:

- Campbell Scientific 229 sensors to collect matric potential data
- Decagon 5TE or METER Teros 12 sensors to collect electrical conductivity and volumetric water content
- East-30 TR-1 sensors to collect thermal conductivity data

Each of the sensors also records temperature data.

Movement of water through the fly ash matrix is monitored by changes in matric potential, water content, and thermal conductivity of the lysimeter materials. Migration of brine salts associated with percolation of water followed by formation of leachate may be indicated by changes in electrical properties of the material. The sensors provide valuable information regarding water movement; however, the chemical and physical properties of the matrix vary significantly from the natural soils for which the sensors are designed. The sensor output is therefore considered qualitative rather than quantitative. In particular, laboratory studies conducted by DBS&A prior to the field installations indicate that the magnitude of the volumetric water content readings would be skewed due to the salt content, conductivity, and dielectric properties of the surrounding fly ash and brine. Results of laboratory testing using the 5TE sensors placed in a fly ash and brine paste indicate that the actual volumetric moisture content may be expected to be in the range of approximately 50 to 90% of the field-reported values.

Additionally, each lysimeter installation is equipped with a weather station that records hourly temperature, precipitation, barometric pressure, relative humidity, wind speed, wind direction, and solar radiation data. A pan evaporation station provides an estimate of the potential evaporation at the site. Together these data can be used to estimate the potential evaporative losses from the lysimeter surfaces.

Water Balance

Rainwater incident on the lysimeter surfaces either infiltrates the material, evaporates, or leaves the lysimeter as surface runoff. An objective of the long-term study is to determine a runoff coefficient for the various lysimeter materials that can be used to inform the design of large-scale test cells and, eventually, full-scale disposal facilities. Surface runoff is collected and quantified at each of the lysimeter stations, while rainwater volumes are calculated using weather station data. Rainwater that infiltrates may be stored in the material matrix, eventually leave as evaporation from the material surface, or pass through the lysimeter to be collected as leachate from the basal drainage layer. Water may be stored within the primary porosity of the lysimeter materials or secondary porosity developed during curing (e.g., vugs or cracks), or may be chemically incorporated into the structure of hydrated minerals that form during curing.

Leachate Characterization

Studies have shown that, in addition to reducing the overall volume of water that passes through the waste matrix, encapsulation of brine wastewater within a cementitious CCB paste matrix may serve to effectively sequester leachable constituents (Oza et al., 2015; Renew et al., 2017; EPRI, 2018), particularly compared to conventional low-moisture compaction disposal methods. Runoff and leachate samples have been collected and retained for laboratory analysis. The long-term nature of the field simulations allows for an assessment of the temporal variability in leachate characteristics, as the simulated waste cell ages under environmental conditions.

Lysimeter Installations

Site A Field Lysimeter Installations

Between October 23 and November 16, 2017, two field-scale lysimeters were installed using materials derived from wastewater volume reduction pilot testing and fly ash generated from a nearby facility. Both lysimeters were installed in repurposed reinforced concrete vaults available at the site, with interior dimensions of approximately 6 ft (1.8 m) wide by 4 ft (1.2 m) deep by 12 ft (3.7 m) long. Lysimeter vaults were placed on a gravel ring foundation with a 2% slope to promote leachate drainage via a single 2-inch (5.1-cm) penetration that was grout-sealed prior to material placement. Within each lysimeter, a nominal 6-inch (15.2-cm) layer of commercial playground sand was placed as a drainage layer, and fly-ash-based materials were placed in three 8-inch (20.3-cm) lifts over the drainage layer.

One lysimeter was constructed of an engineered paste-encapsulated CCB consisting of approximately 60% fly ash and 40% concentrated wastewater reject from a reverse osmosis volume reduction system. The concentrated wastewater was amended with approximately 1.1% (by mass) commercial 1/8” minus quicklime prior to mixing with the ash to increase the system pH. The fly ash used was a Class C (ASTM C 618) ash collected from the electrostatic precipitator of a pulverized coal power plant fueled with sub-bituminous coal from the Powder River Basin. Paste materials were mixed on-site using a horizontal single-shaft continuous flow ploughshare mixer and pumped to the lysimeter using a purpose-built grout plant. The second lysimeter was composed of approximately 88 to 90% fly ash and 10 to 12% unamended concentrated wastewater, placed via in situ manual mixing and compaction with a hand-guided vibratory plate compactor.

An annotated picture of the field installation is provided in Figure 1. Eight sensor arrays were installed in each lysimeter during emplacement of the fly ash materials in two vertical profiles. Each vertical profile consisted of four arrays, with one array installed in each lift of fly-ash-based material and the underlying sand drainage layer. The profiles were located in relative downslope and upslope positions within the vault, as shown in Figure 2. Sensor and weather station data were collected via a remote wireless connection to an on-site datalogger. Field observations by on-site personnel included recording the volume of captured surface runoff water and leachate.

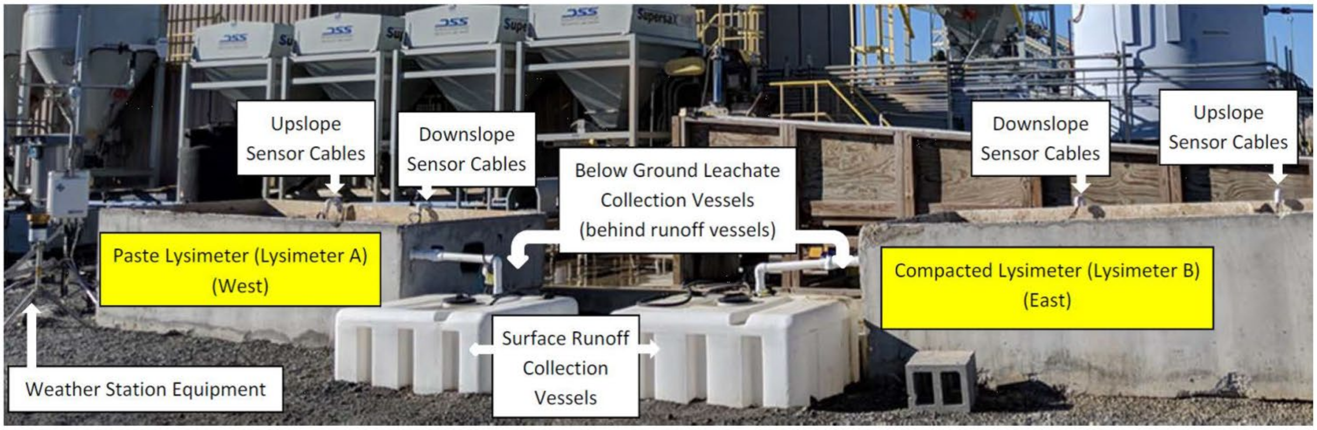
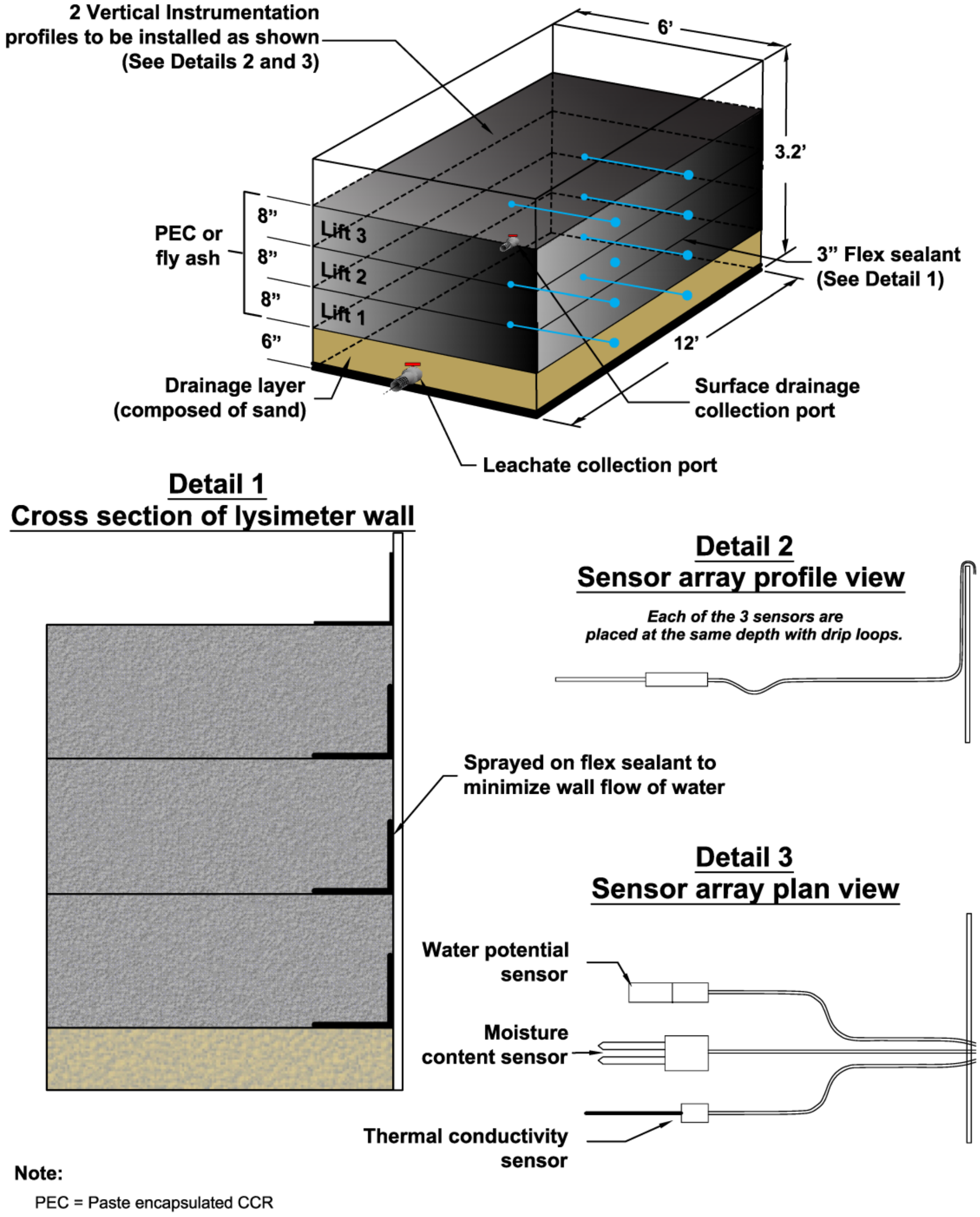


Figure 1. Site A field lysimeter photograph



Note:
PEC = Paste encapsulated CCR

Figure 2. Site A field lysimeter schematic

Site B Field Lysimeter Installations

Between August 27 and August 30, 2018, two field-scale lysimeters were installed at an electric utility host site. Custom-designed and purpose-built 250-gallon (946-L) cylindrical polypropylene containers were used as the lysimeter vessels; each is mounted on a spill containment pallet. The top of each lysimeter is furnished with a penetrative annulus ring to preclude flow of runoff water toward the outer wall, where unwanted vertical wall flow may occur. The 6-inch (15.2-cm) basal sand drainage layer is subdivided by a 7-inch- (17.8-cm-) high raised concentric ring on the bottom of the vessel (vessel internal diameter of 42 inches [1.1 m], concentric base ring outer diameter of 36 inches [0.9 m]), to segregate and capture fluid entering the drainage layer due to wall flow within the outer annulus (Figure 3).

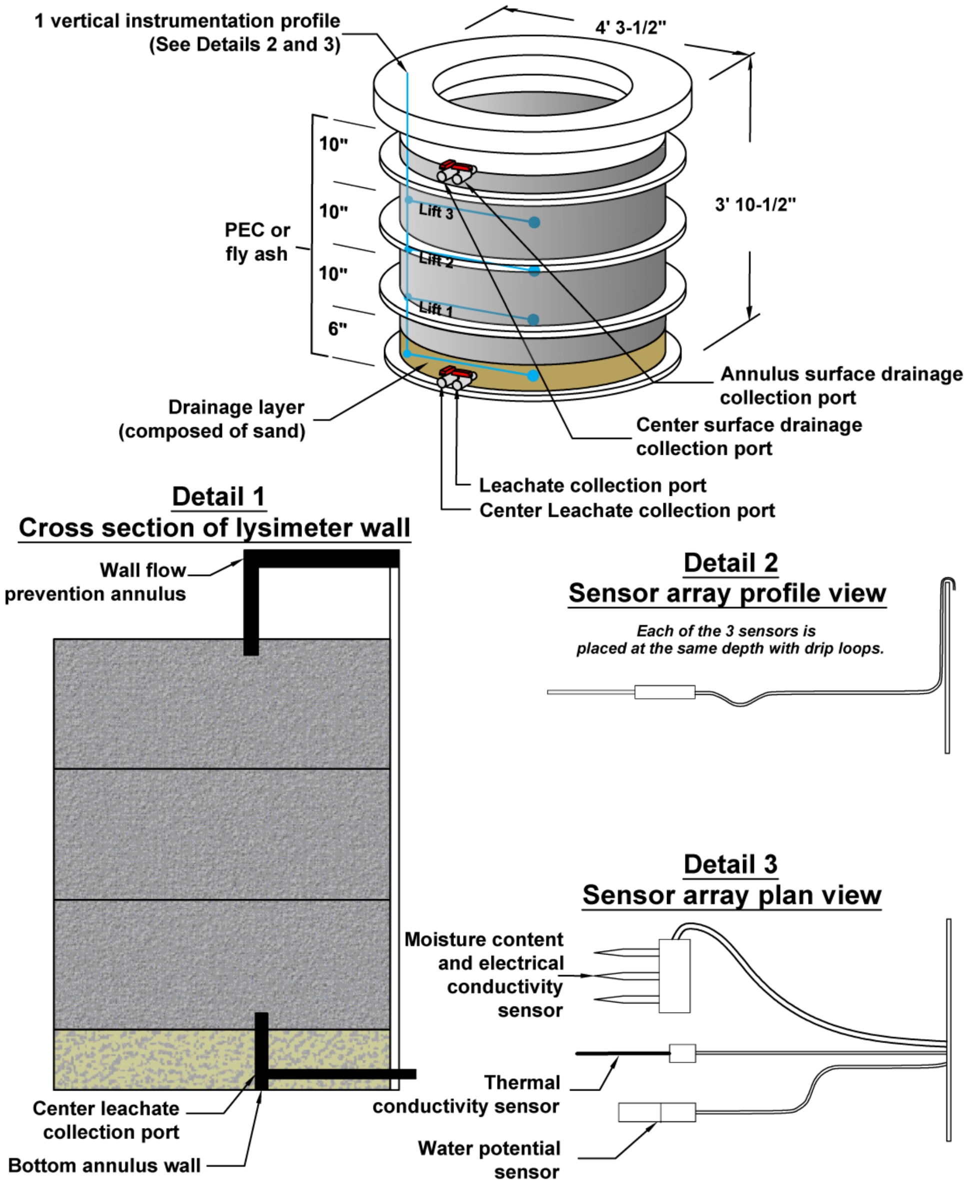


Figure 3. Site B lysimeter schematic

Similar to the Site A installations, one lysimeter was composed of a high-fluid-content pumpable paste, mixed on-site using a purpose-built mobile plant, while the second lysimeter was constructed using a relatively low-moisture, hand-compacted fly ash/brine mixture. The materials were installed in three lifts over a basal sand drainage layer, with minimal curing time allowed between lift placements. The fly ash and brine used in the study were both produced on-site. The brine is a highly concentrated FGD WW brine, and the fly ash meets Class F specifications. The paste lysimeter mix consisted of approximately 35% brine, 59% fly ash, and 6% quicklime. The compacted lysimeter mix consisted of approximately 18% brine and 82% fly ash. The bases and top surfaces of the lysimeter materials were sloped toward the drainage ports with an approximate 2% grade to facilitate drainage. The lysimeters were covered for the first four weeks to allow the materials to cure without influence from precipitation (an atypical tropical weather system impacted the site during this time as well, therefore also requiring the use of the cover). The covers were removed on September 28, 2018.

Sensor arrays similar to those used at Site A were installed in a single vertical profile in each lysimeter, as shown in Figure 3. Decagon 5TE sensors were replaced by METER Teros 12 sensors for a wider range of output and improved tolerance of highly saline conditions.

Surface runoff from each lysimeter is collected in external tanks; the volume of captured runoff is measured in real time by monitoring the liquid level in a calibrated tank with a pressure transducer. For each lysimeter, leachate is captured and stored separately from the center and annulus of the sand drainage layer. Center leachate is quantified using a tipping bucket gauge, while annulus leachate is measured in a calibrated tank using a transducer, as above. Samples are collected from all three collection tanks for laboratory analyses.

Site C Field Lysimeter Installations

A smaller-scale lysimeter study is underway at a third utility site. Between September 25 and 26, 2018, two field-scale lysimeters were installed in 55-gallon (208-L) black HDPE drums. Whereas previous installations compared lysimeters constructed using high-moisture paste and conventionally conditioned and compacted fly-ash materials, the Site C installation consists of side-by-side identical paste-based lysimeters to assess the reproducibility of results between duplicate systems.

The lysimeter vessels are mounted on separate spill containment pallets, and are shimmed to facilitate drainage toward the runoff and leachate ports. Both lysimeters were constructed using pumpable paste containing approximately 64% fly ash, 33% concentrated wastewater, and 3% quicklime reject from a reverse osmosis volume reduction system. The fly ash used was a Class F ash collected from the electrostatic precipitator of a pulverized coal power plant fueled with bituminous coal from Columbia, and was about 150°F (66°C) when mixed. The concentrated wastewater was the reject from reverse osmosis system processing FGD blowdown, and was about 80°F (27°C) when mixed. The quicklime was a commercial 1/8” minus product. Paste materials were mixed on-site using a vertical single-shaft batch flow paddle mixer and pumped to the lysimeter using a purpose-built grout plant. A nominal 6-inch- (15.2-cm-) thick sand drainage layer was placed at the base of each lysimeter around a pre-installed ½-inch-

(1.3-cm-) diameter slotted PVC screen and sealed leachate port. A majority of the paste was placed in small batches during a single day. Three sensor arrays identical to those employed at Site B were installed in each of the lysimeters. One sensor array was placed in the center of the sand drainage layer, and the other two were placed in the paste at nominal heights of 8 inches (20.3 cm) and 16 inches (40.6 cm) above the sand layer (Figure 4). Surface runoff and leachate from each lysimeter are directed to tipping-bucket gauges and recorded in real time.

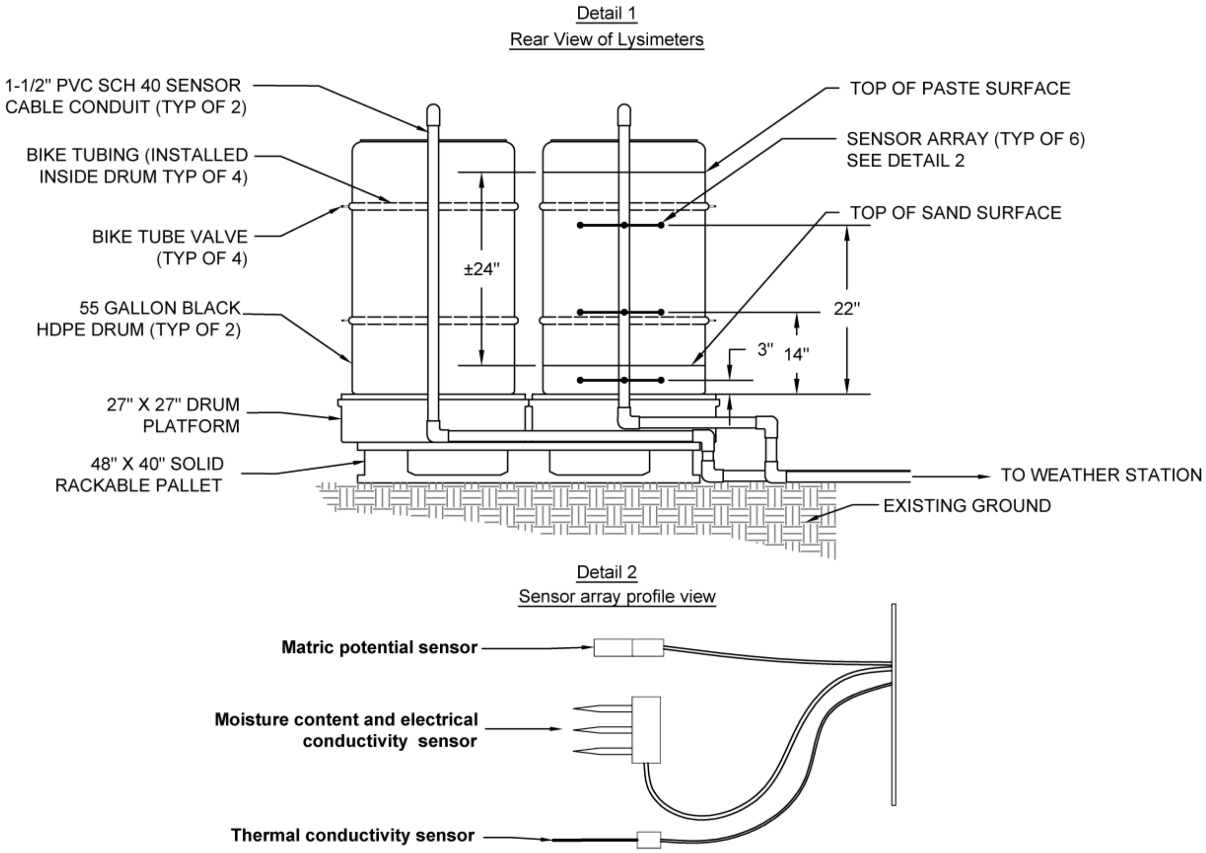


Figure 4. Site C lysimeter schematic

Key Results

Monitoring of the lysimeter installations has shown that the sensor arrays are largely robust and functional within the fly-ash-based lysimeter materials and continue to return interpretable data over a period of at least several months and possibly much longer. Calculation of runoff coefficients and detailed assessment of the paste materials under long-term exposure to environmental conditions are currently in preliminary stages. Notable observations and findings thus far are summarized in the following subsections.

Site A

The Site A lysimeter study provided insight into the ability of the selected sensors to monitor changes in moisture content, temperature, electrical conductivity, and matric potential conditions in the fly-ash-based lysimeter materials. As noted above, the properties of the lysimeter materials differ significantly from natural soils; however, sensor responses following precipitation events indicate that the reported values can be used to infer relative changes in moisture content over time (Figure 5). Matric potential data show clear sensor responses to precipitation events and dry periods, particularly in the upper lifts of the lysimeter, as would be expected (Figure 6), and generally indicated that the lysimeter materials remained relatively wet throughout the study period. Thermal conductivity data from the TR-1 sensors were consistent with trends observed in the electrical conductivity and matric potential datasets.

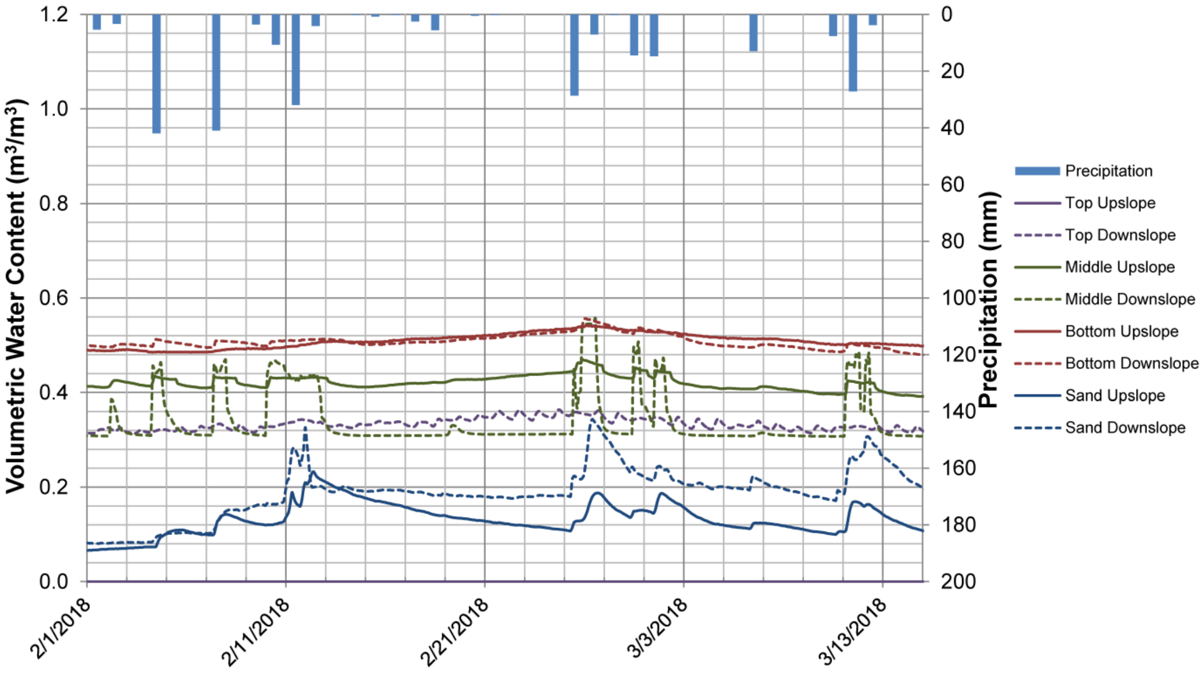


Figure 5. Volumetric water content, Site A compacted lysimeter

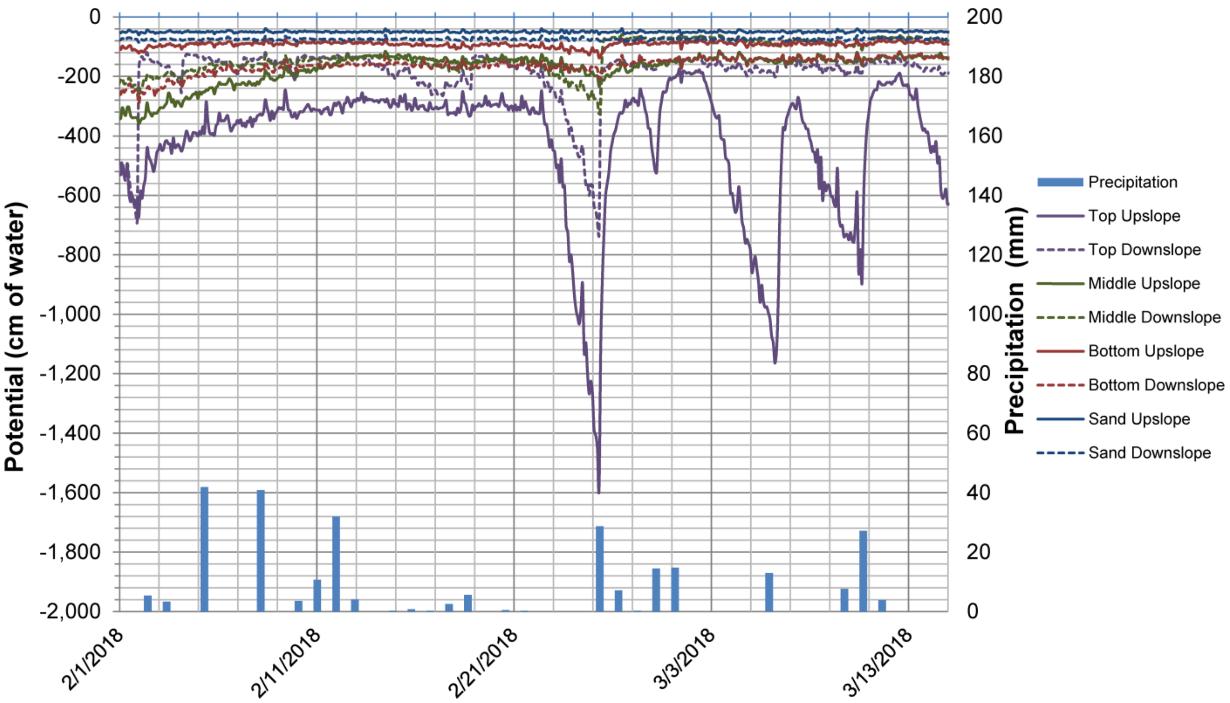


Figure 6. Matric potential, Site A compacted lysimeter

Later in the study, nearly instantaneous responses to precipitation recorded in lower material lifts and/or the sand drainage layer are interpreted to indicate development of preferential flow paths, likely related to observed structural degradation of the lysimeter vaults. The observed degradation included propagation of vault cracking, which continued to grow through the end of the study. Ponded infiltration testing with dye tracer on the paste lysimeter at the end of the test period revealed that water flow was largely restricted to lateral movement through the fractures and voids (vugs) within the upper surface of the solidified paste monolith, with little downward movement.

Temperature data collected from all 24 sensors installed in the paste lysimeter captured a secondary heating event four and a half months after installation (Figure 7), with a peak difference between the temperature recorded by internal material sensors and the external air temperature of 47°F (8°C). The maximum observed temperature inside the lysimeter was 90°F (32°C) during the secondary event, which is only slightly lower than the peak temperature of 39°C (102°F) reached during the initial curing phase. This secondary heating may be the result of a second curing event due to residual unreacted fly ash and lime coming in contact with percolating water. Alternatively, the secondary heating event may be a result of precipitation of calcium sulfate, which is an exothermic reaction. During the reaction of calcium with sulfate, water is removed from the system to form gypsum, a hydrated mineral. Physical consequences of this reaction may include a decrease in hydraulic conductivity of the material and a corollary increase in compressive strength; the reaction may also remove conductive fluids from pore spaces. Evidence to support this hypothesis is the decrease in electrical conductivity observed in data from each of the three lifts, which occurs at roughly the same time as the increase in temperature (Figure 8).

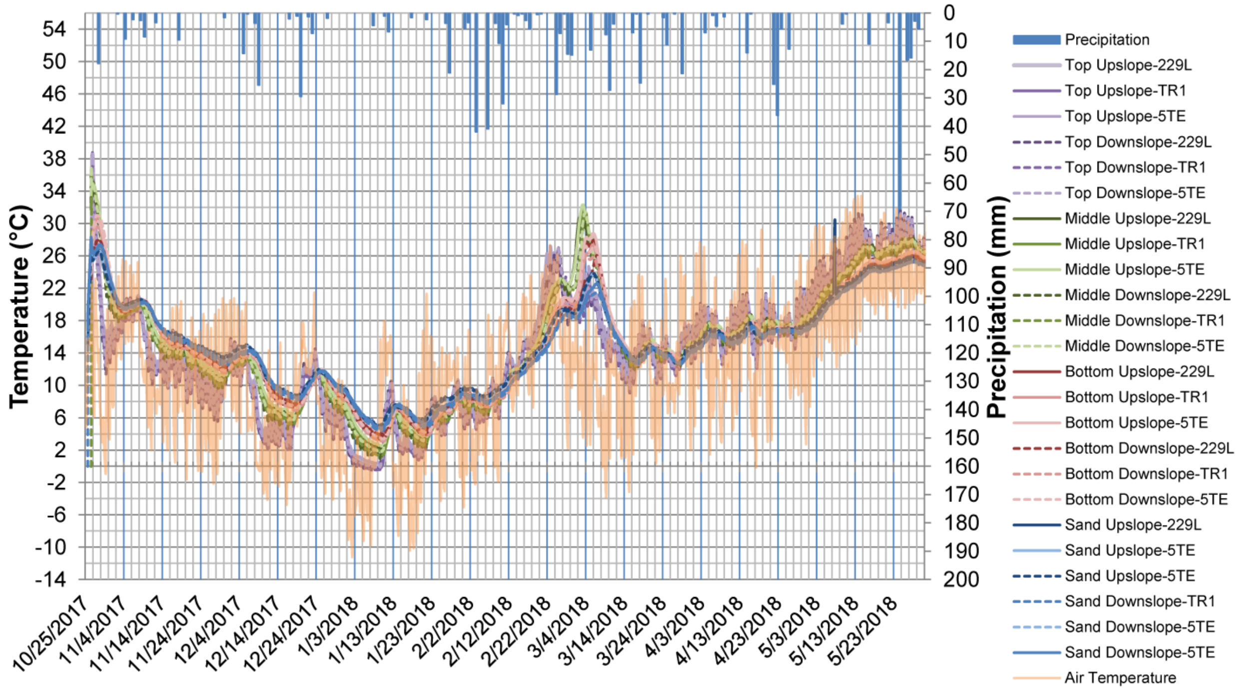


Figure 7. Temperature data for all sensors, Site A paste lysimeter

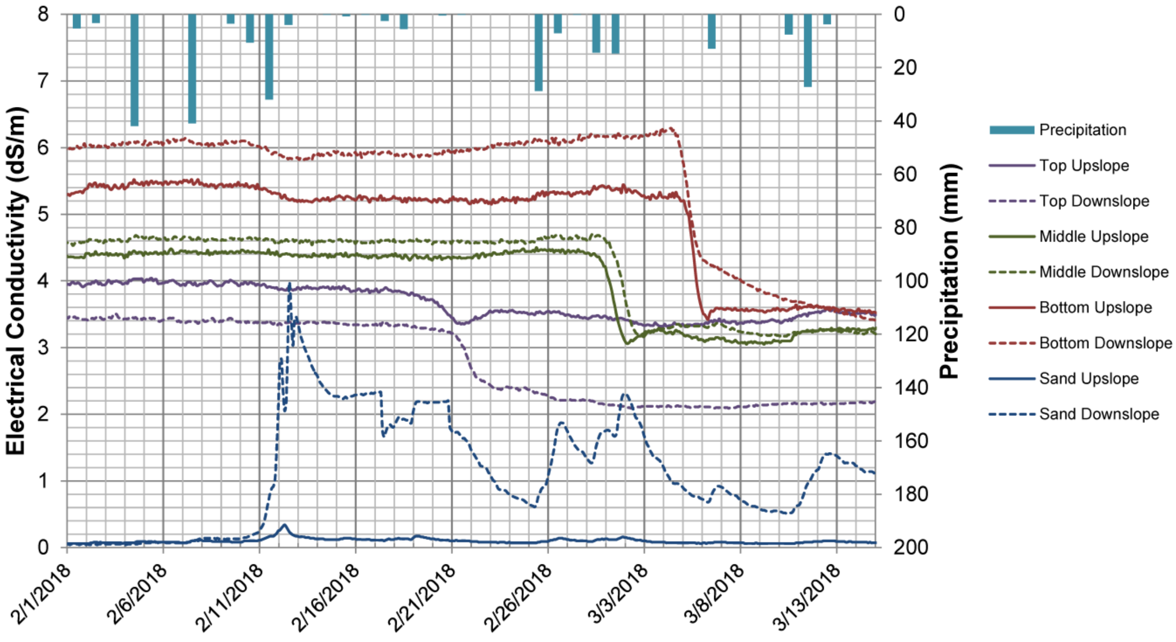


Figure 8. Electrical conductivity, Site A paste lysimeter

Structural degradation of the concrete lysimeter vaults, exacerbated by the winter freeze-thaw cycle, resulted in water loss through leakage and vertical fracture flow, and precluded determination of a useful water balance over the duration of this study. However, analysis of data collected prior to the deterioration of the vaults indicates that the paste material has a greater capacity to resist infiltration and shed water. In a one-month period during December and January, 83 gallons (314 L) of precipitation fell on each of the lysimeter surfaces; 78 gallons (295 L) (94%) was collected in the runoff tank for the paste lysimeter, as opposed to 56 gallons (212 L) (69%) for the compacted lysimeter. The difference in runoff characteristics is attributed to relatively lower initial moisture content and greater permeability of the compacted material. Following repair of the vaults in May 2018, captured runoff represented 83% of total precipitation for the paste lysimeter and 68% of total precipitation for the compacted lysimeter.

Site B

The surface of the compacted material rose approximately 1 inch (2.5 cm) following removal of the cover, which resulted in plugging of the ¾-inch (1.9-cm) surface runoff port. Conversely, the paste surface subsided by nearly 1 inch (2.5 cm) during curing, which resulted in ponded water on the lysimeter surface until the port was lowered during a December 2018 site visit. Due to the blockage of the runoff ports, both lysimeters initially produced significant leachate during this period. Significant surface-level changes during curing or following exposure to rainfall were not observed at the previous Site A installation, likely due to differences in the initial chemistry, curing, and climate conditions of the material mixtures.

Little leachate has been produced following the adjustment of the level of the surface drain ports, and preliminary results clearly indicate that the paste material has a much higher propensity to produce runoff. Between December 13, 2018 and March 12, 2019, 44 gallons (167 L) of precipitation fell on the lysimeter surfaces, 41.5 gallons (157 L) (94%) of which was collected as runoff from the paste lysimeter, as opposed to only 7.5 gallons (29 L) (17%) for the compacted lysimeter. During the same period, 1 gallon (3.8 L) and 25 gallons (95 L) of leachate was collected from the paste and compacted lysimeters, respectively.

Matric potential data for the cementitious paste lysimeter at Site B are indicative of a material that is undergoing some drying from the surface down, with periodic wetting due to precipitation (Figure 9). Matric potential data for the hardened compacted lysimeter are indicative of a material that was placed dry and has since wetted thoroughly. Precipitation on October 11 and 26, 2018 and subsequent surface ponding served to wet the entire lysimeter profile, and continued rainfall and limited wintertime evaporative potential have not allowed the material to dry (Figure 10).

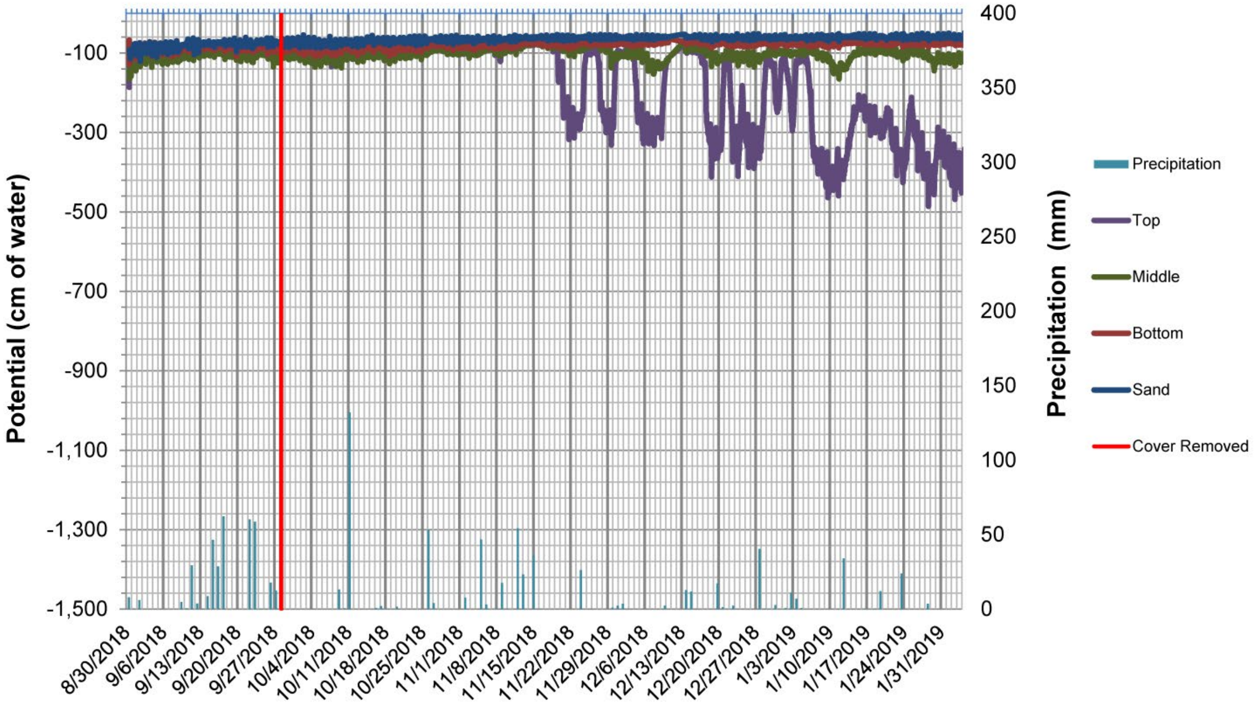


Figure 9. Matric potential, Site B paste lysimeter

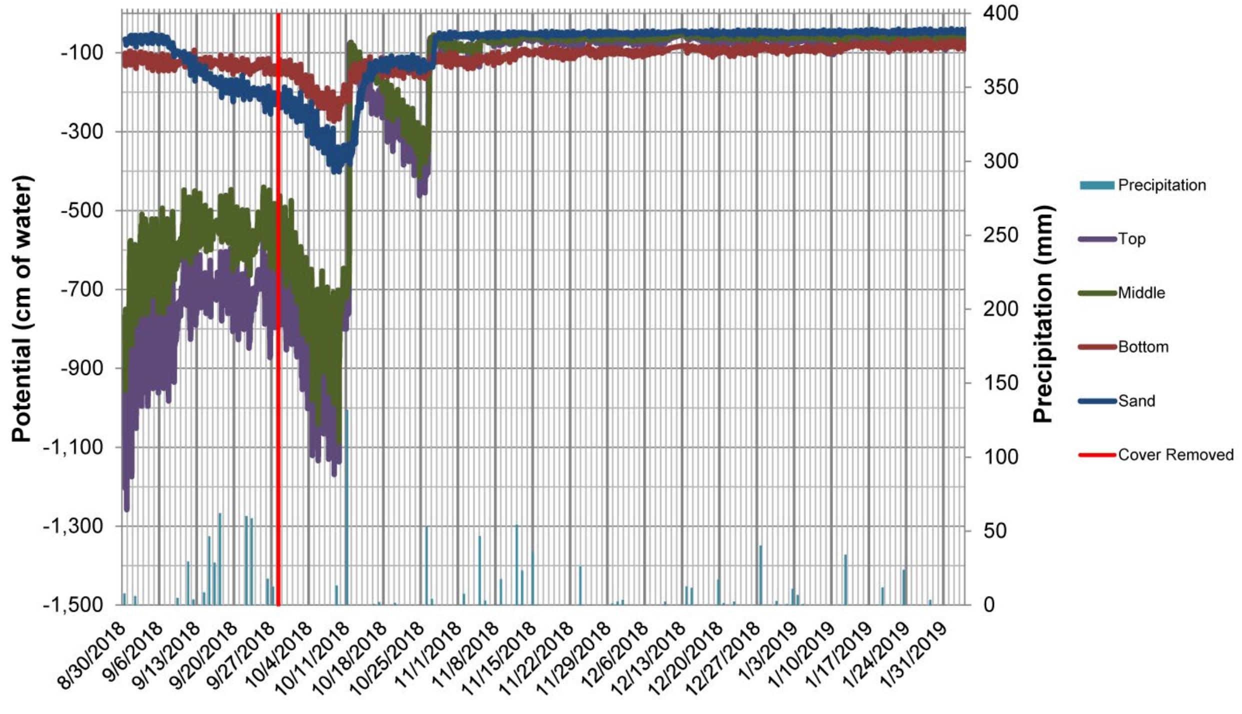


Figure 10. Matric potential, Site B compacted lysimeter

Decreasing electrical conductivity recorded by each of the three fly ash sensors, along with an increased value recorded by the sand sensor, may be representative of downward water movement from the compacted fly ash matrix during this period

(Figure 11). Electrical conductivity of the compacted lysimeter materials has continued to slowly decrease, indicating potential continued water movement through the system. Similar trends have not been observed in the paste lysimeter, which has produced little leachate since correction of the surface drainage port.

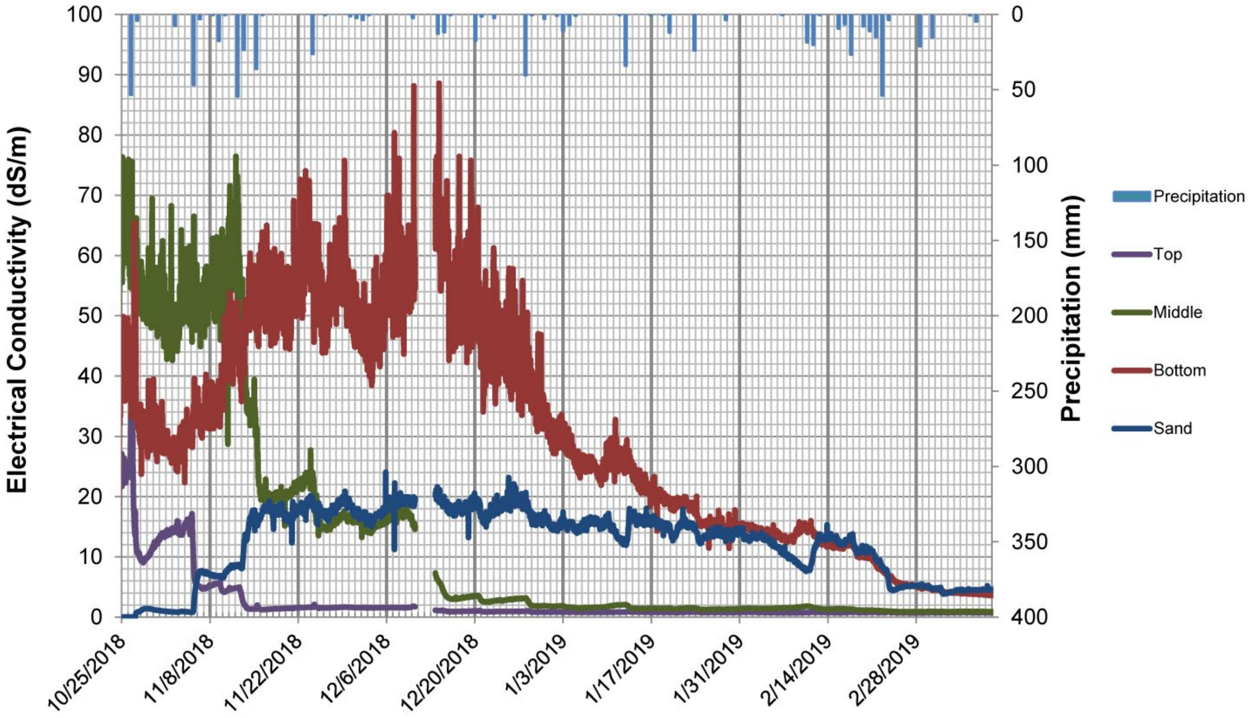


Figure 11. Electrical conductivity, Site B compacted lysimeter

Site C

Matric potential measurements in the Site C lysimeters also exhibit a clear response to precipitation and moisture profile changes (Figure 12). Similar to the Site B installation, the paste material sensors recorded wet conditions for a period after material placement. After approximately two weeks, the sensors began exhibiting a drying response. After this initial period of drying, matric potential of the lower paste sensor has oscillated within a consistent range, whereas the upper sensor has generally trended toward a drier state (Figure 12).

The electrical conductivity readings are following a similar trend as observed at the Site B paste lysimeter, although at a much lower magnitude—presumably due to the difference in brine properties between sites. Electrical conductivity readings decreased sharply following material placement and are currently exhibiting a slowly decreasing trend (Figure 13). Electrical conductivity around the sensors in the sand drainage layer has increased during the same period.

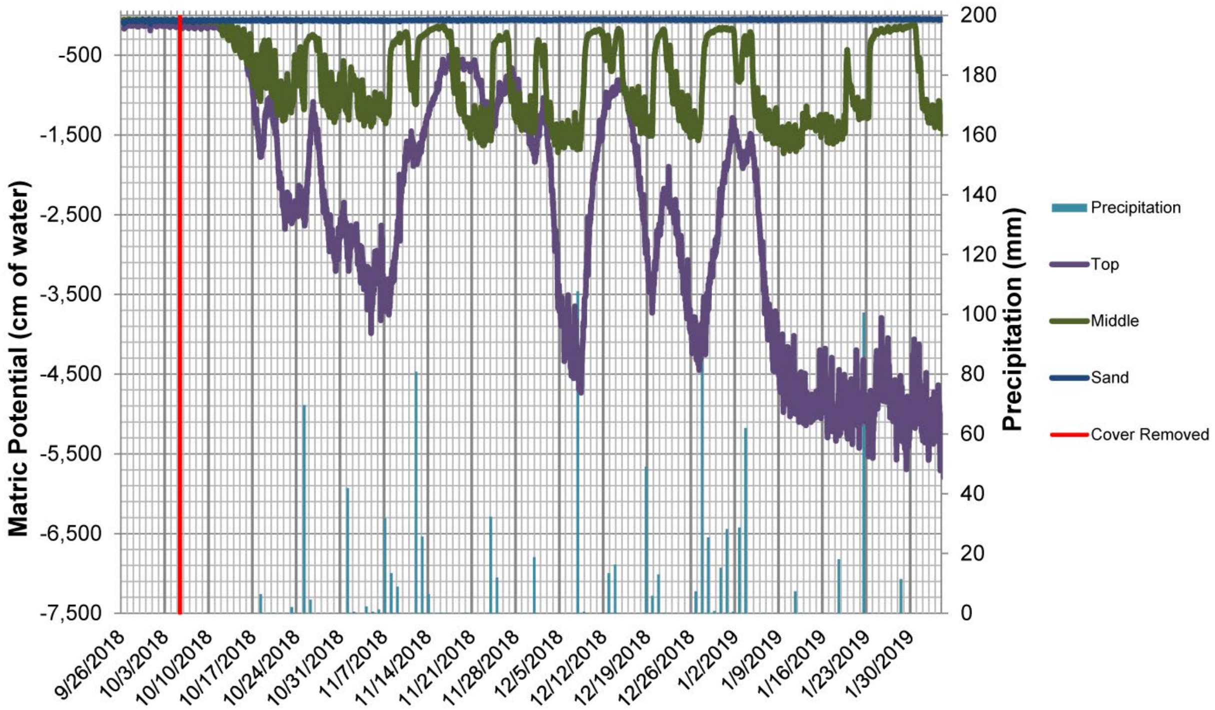


Figure 12. Matric potential, Site C east paste lysimeter

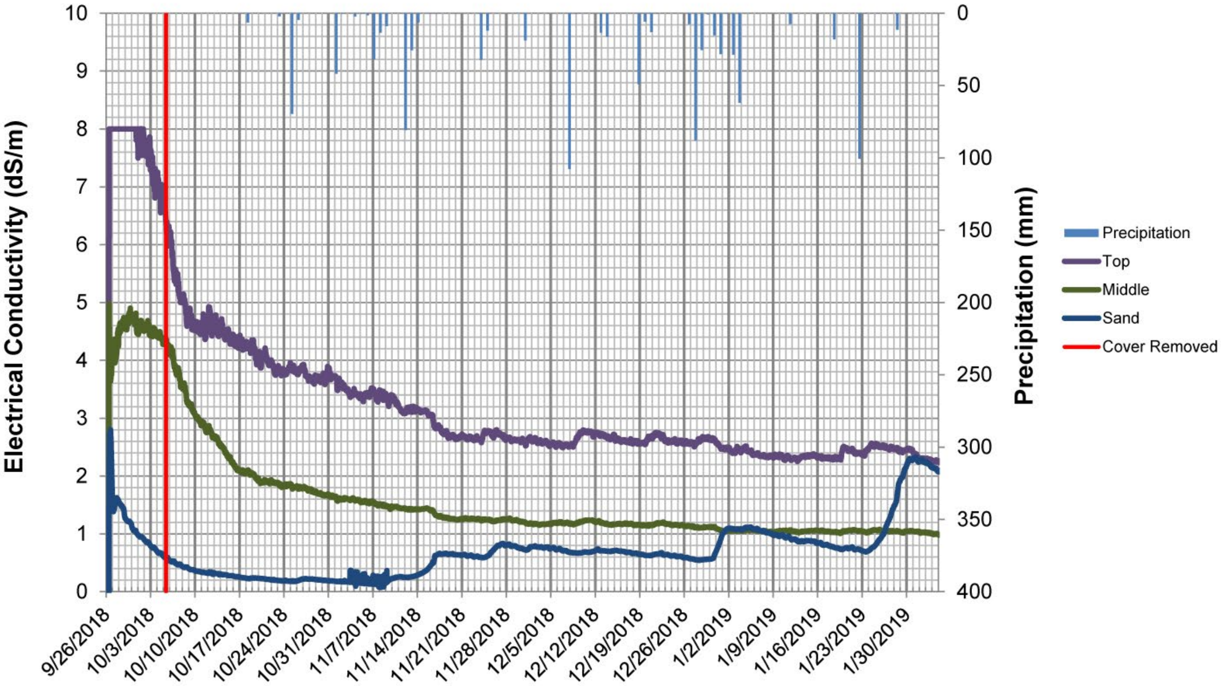


Figure 13. Electrical conductivity, Site C east paste lysimeter

The water budget calculation for Site C is currently ongoing; however, preliminary results indicate a high degree of similarity in the behavior between the two lysimeters, and each has produced less than 0.75 gallon (2.8 L) of total leachate.

Next Steps

The three field lysimeters installed at coal-fired power plants in 2017 and 2018 represent the preliminary phases of a long-term investigative effort to determine the potential for achieving disposal of coal combustion by-products by encapsulation of FGD brine residuals in a solidified fly-ash-based cementitious paste. These efforts have established that commercially available sensors can be useful for detecting and monitoring movement of moisture and dissolved salts within the solidified paste monolith, and data collected from these sensors may be useful to infer other events, such as secondary curing or reactions resulting in physical changes to the material matrix. Preliminary results suggest that solidified paste monoliths may demonstrate significantly less infiltration, leachate production, and migration of dissolved constituents than alternative disposal methods.

To assess the physical and chemical properties of an encapsulation monolith through time and provide a design basis for potential future operation-scale field trials, proposed or currently ongoing tasks include the following:

- Chemical analysis of captured leachate and runoff. These data can be used to establish temporal trends, determine constituent mass movement, and evaluate the sequestration efficacy of the paste monolith compared to both conventional disposal methods and regulatory thresholds.
- Chemical and hydraulic property analysis of material cores from decommissioned lysimeters. Analysis of material cores from field installations can be used to determine properties of solidified materials cured under environmental conditions, as well as changes in material properties over an extended period of environmental exposure, including changes in mineralogy, compressive strength, primary and secondary porosity, and hydraulic conductivity.
- Long-term monitoring of runoff, leachate production, and evaporative potential should continue, and will generate data to support calculation of a robust water budget and runoff coefficients for both solidified paste monoliths and conventionally conditioned and compacted materials.
- Large-scale field studies should be conducted that assess the performance of paste encapsulated materials in realistic waste disposal scenarios and settings.

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