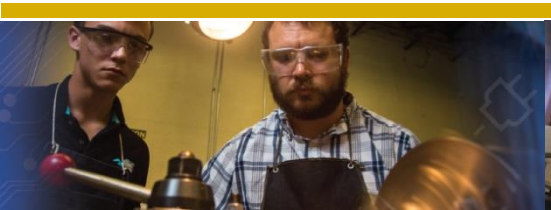
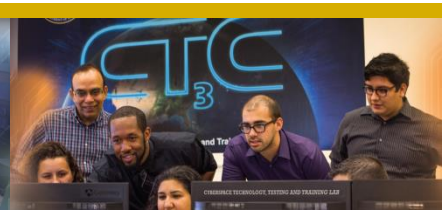
FIU PROJECT 2:
YELENA KATSENOVICH

ENVIRONMENTAL REMEDIATION SCIENCE & TECHNOLOGY

FLORIDA INTERNATIONAL UNIVERSITY





FIU Personnel and Collaborators

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Project Tasks and Scope

Task 1: Remediation Research and Technical Support for the Hanford Site

- Laboratory-scale experiments to investigate the effect of environmental factors on contaminants behavior including Tc-99, U(VI), Cr, and iodine (I) at Hanford subsurface.
- Experimental support of long-term field-scale lysimeter testing via single-pass flow-through laboratory experiments to validate model predictions of long-term waste form behavior upon disposal in the Hanford site's integrated disposal facility.

Task 2: Remediation Research and Technical Support for Savannah River Site

- Laboratory-scale experiments to study contaminant fate and the impacts of potential *in situ* remediation techniques on the subsurface at SRS.

Task 3: Contaminant Fate and Transport Modeling in the Tims Branch Watershed

- Development of a numerical modeling tool to simulate flow and contaminant fate and transport under various hydrological conditions in Tims Branch watershed at SRS.

Task 5: Research and Technical Support for WIPP

- Laboratory-scale experiments to study the fate of actinides and lanthanides at the WIPP site.

Task 6: Hydrology Modeling for WIPP(NEW)

- Development of a GWM for the WIPP site using ASCEM modeling toolset to improve the current understanding of regional and local groundwater flow at the WIPP site.

Task 1 – Remediation Research and Technical Support for the Hanford Site

FIU Applied Research Center

Site Needs:

DOE-EM has a critical need to understand the biogeochemical processes influencing the behavior and fate of contaminants (U(VI), Tc-99 and iodine in Hanford Site's deep vadose zone that can impact groundwater. Research to address environmental risks and remediation challenges involving Tc-99 is a one of the high-priority activities for the DOE-EM complex (*Technetium Management Program Plan (DOE EM, 2016*)). In addition, the DOE has no approved treatment technologies to control iodine mobility in the vadose zone (VZ) and groundwater. Alkaline pH manipulation is a potential remediation technology that can lead to incorporation of U(VI) into the sediments. This research also supports the Field Lysimeter Test Facility (FLTF) by generating data on the corrosion of various waste forms to confirm the ability of laboratory data to model dissolution behavior in a field environment.

Year 9 Objectives:

- Identify physicochemical mechanisms controlling immobilization of U via $NH_3(g)$ injection in Hanford vadose zone.
- Investigate pertechnetate reduction by ferrous iron minerals and Hanford soil in the absence and presence of bicarbonate.
- Investigate co-precipitation of iodine and iodine comingled with chromium with calcium carbonate and study the effect of silica at different pH conditions.
- Investigate the effect of grout-contacted groundwater on glass dissolution behavior at varying pH (9-12) and temperature (25°C, 40°C, 70°C) using single-pass flow-through (SPFT) and static experiments.

Present (Year 9) Subtasks:

- **1.1** Determine the long-term stability of U-solid phases after NH3(g) injection and conduct solid phases characterization during treatment pH12 & post treatment pH 8 to identify dominant mineral phases controlling U behavior.
- **1.2** Investigate Tc(VII) reduction by ferrous iron minerals with and without HCO_3^- at pH 8 to compare the reduction rate by Hanford sediment and magnetite
- **1.3** Study the effect of pH and different silica concentrations (0-20mM) on iodate and iodine comingled with chromium co-precipitation with calcium carbonate.
- **1.4** Determine experimental conditions necessary for forward rate of glass dissolution and investigate the effect of grout-contacted groundwater on dissolution behavior

Task 1 – Remediation Research and Technical Support for the Hanford Site

FIU Applied Research Center

Accomplishments Year 9:

Ammonia Gas for Uranium Remediation

- Finalized 3-months mineral dissolution of phyllosilicates exposed to aqueous NH4OH
- SEM-EDS shows potential secondary precipitates upon aeration and strong correlation between U & Fe via EMPA
- *Ongoing* – Long-term (~6 months) aging montmorillonite mineral to high U loading for Kd analysis and solid characterization for treated samples pH12 & post treated pH8

Technetium Fate and Transport

- Obtained and characterized two biotite and two ilmenite minerals
- Observed significant reduction of Tc-99 by ilmenite 2 that featured higher iron content.
- *Ongoing* – Study Tc(VII) reduction in the presence of HCO3- and characterize solids to identify factors responsible for mineral transformation.

Iodine incorporation in calcium carbonate

- Finalized experiments on iodine incorporation in calcium carbonate in the presence of comingled chromium.
- EPMA analysis showed good alignment of Ca and iodine.
- *Ongoing* – Solids analysis via XRD and complete "late spike" experiments

Experimental Support of Lysimeter Testing

- Completed q/S sweep to determine conditions for forward rate at 25°C and 70°C
- Initiated SPFT experiments utilizing grout-contacted groundwater as the leachate
- *Ongoing* – Analyze results for grout experiments at 40°C and complete grout experiments at 70°C.

5% ammonia/95% nitrogen injection for montmorillonite long term U-aging experiment

- **Publication in J. Env. Manage., "Potential for U sequestration with select minerals and sediments via base treatment".**
- **Presented results at WM2019 and ACS.**
- **Manuscript in preparation "Phyllosilicate Mineral Dissolution following Alkaline Treatment"**
- **Completed draft literature review on glass dissolution**

Task 1 - Remediation Research and Technical Support for the Hanford Site

Proposed Scope for Year 10

Site Needs:

DOE-EM faces a number of environmental challenges that are technically complex and unique to EM with tremendous associated cleanup costs. This project is focusing on basic science to fill knowledge gaps and validate potential remediation technologies to assist with environmental cleanup of Hanford Site's contaminated vadose zone and groundwater. This investigation will assist Hanford Site and other DOE EM sites in their efforts to better understand Tc-99 behavior in the presence of Fe(II)-bearing minerals and the fate and transport mechanisms of comingled contaminants including Tc-99, U(VI), Cr, and iodine.

Objectives:

- **1.1** Continue to characterize impacts of base treatment on long-term fate of U and evaluate physical and mineralogical changes due to dissolution and precipitation of muscovite, illite, and Hanford sediments.
- **1.2** Investigate re-oxidation rates of reduced Tc in the presence of ferrous-iron-containing minerals with and without bicarbonate at pH 8.
- **1.3** Initiate a new subtask on competing attenuation processes for mobile contaminants using Hanford sediments to determine the mechanisms affecting the behavior and fate of contaminants.
- **1.4** Continue evaluating effect of grout on glass dissolution behavior and determine if contacting the grout solution with sediment leads to a buffering of dissolution behavior

Task 2 - Remediation Research and Technical Support for Savannah River Site

FIU Applied Research Center

Site Needs:

DOE EM's current mission places emphasis on innovative approaches and novel technologies which help to address the significant challenges associated with the remaining cleanup of contaminated sites (Innovation and Technology Program (*DOE EM) - EM Test Bed Capability at SRS F area*). This study will supplement ongoing activities at SRS pertaining to the Area Completion Project and associated permitting strategies to evaluate and meet standards for contaminants in the Four Mile Branch Wetland. Low cost modified humic substances are potential amendments for treatment of uranium in groundwater associated with F-Area Seepage Basins plume. This research assists in the design and validation of novel in situ remediation technologies that support EM test bed demonstrations and benefit SRS cleanup initiatives.

Objectives:

- Understand I, Tc, and U interactions with reactive oxygen species (ROS) and natural organic matter (NOM) impacted by ROS due to nitrate and UV interactions
- Determine if the modified humic acid (KW15 modified Humics) can be used to control the mobility of uranium in groundwater and study the sorption/desorption of modified HA on SRS sediment at various pH via batch experiments.

Present (Year 9) Subtasks:

- **2.1** Impact of NO_3^- and reactive oxygen species on the fate of Tc, I, and U in wetlands at Savannah River Site. Investigate a synergetic effect between ROS and organic matter degradation on radionuclide fate.
- **2.2** Batch sorption experiments using a modified humic acid to simulate the creation of a sorbed humate treatment zone in acidic groundwater contaminated with U. Evaluate the effect of contact time, pH and initial uranium concentrations.

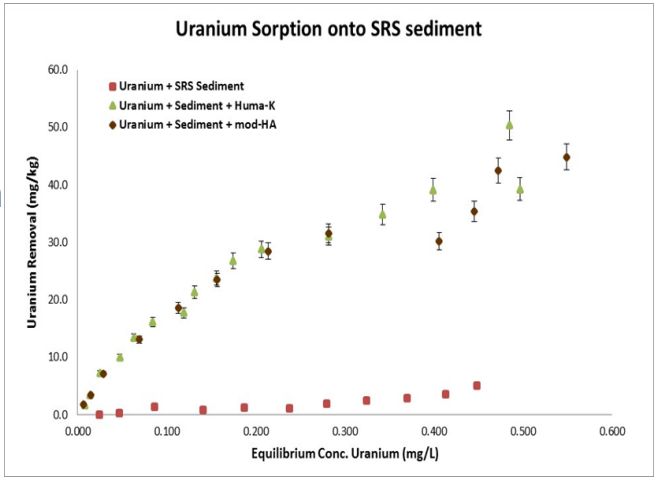
Task 2 – Remediation Research and Technical Support for Savannah River Site

FIU Applied Research Center

Accomplishments Year 9:

Impact of reactive oxygen species on the fate of Tc, I, and U in SRS Wetlands

- Observed similar removal of Tc and I from the aqueous phase occurred under light and dark conditions; significant difference for aqueous U with light versus dark treatment (27±3 and 15.1±0.4 µg/L).
- Significant removal of U occurs in the presence of both Everglades and SRS sediments (~ 20% remaining in solution).
- *Ongoing*:
  - Characterize NOM via UV-vis, TOC, ATR-FTIR, FT-ICR-MS, and NMR; complete task in FIU Year 9.



Batch Experiments with Modified Huma-K

- Completed uranium batch sorption experiments with modified humic acid to study the effect of pH (3-8).
- Evaluated the effect of contact time, pH and initial uranium concentrations on U sorption.
- *Ongoing*: Complete mod-HA sorption experiments and initiate desorption experiments.

- Research presented at WM2019 Symposia.
- DOE Fellows Ripley Raubenolt & Silvina Di Pietro won 3rd place in WM2018 Student Poster Contest.
- Prepared a manuscript, *"In situ sequestration of uranium from contaminated groundwater using a low-cost unrefined humic substances"*.

Task 2 – Remediation Research and Technical Support for Savannah River Site

Proposed Scope for Year 10

Site Needs:

This study will supplement ongoing activities at SRS pertaining to the Area Completion Project and associated permitting strategies to evaluate and meet standards for contaminants in the Four Mile Branch Wetland. Significant data gaps still exist regarding the behavior and chemistry of radionuclides of concern such as Tc, I and uranium, as well as the co-mingling of these contaminants that affect groundwater remediation strategies at SRS and other DOE EM sites. Evaluation of the role of different environmental factors on the fate and transport of contaminants and co-contaminants in soil and groundwater will assist in the design and validation of novel in situ remediation technologies that support EM test bed demonstrations and benefit SRS cleanup initiatives.

Objectives:

- **2.1** Finalize mod-HA sorption studies and to initiate desorption experiments and study the effect of pH on uranium sorption on SRS sediment coated with mod-HA
- **2.2** Initiate a new task focusing on the natural attenuation and the geochemical factors controlling the release process of iodine from wetland sediments.

Task 3: Contaminant Fate And Transport Modeling in the Tims Branch Watershed

FIU Applied Research Center

Site Needs:

- Heavy metal and radionuclide contamination (e.g. Hg, Ni, U) at SRS and other DOE sites still exists*. Prediction of the fate and transport of these contaminants during severe rainfall/storm events is required as well as long-term monitoring to evaluate the effectiveness of implemented remediation technologies.

Objectives:

- Develop numerical modeling tool to evaluate impact of extreme hydrological events on fate and transport of major contaminants of concern in Tims Branch.
- Develop this tool as a transferable technology potentially applicable in other contaminated stream systems at SRS/other DOE EM sites.
- Collect *in-situ* field data (e.g., flow depth & velocity, suspended particle conc. and other water quality parameters) to support model calibration and validation via in-person sampling and data collection as well as deployment of remote monitoring devices.

Present Subtasks:

3.1 Modeling of SW Flow & Contaminant Transport in Tims Branch

3.2 Application of Geospatial Tech. for Long-Term Env. Monitoring

3.3 Data Collection, Sampling & Analysis in Tims Branch Watershed

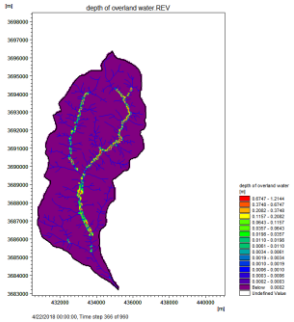
**DOE EM's Technology Plan to Address EM Mercury Challenge & DOE EM's Innovation & Technology Program*

Task 3: Contaminant Fate And Transport Modeling in Tims Branch
Research Highlights

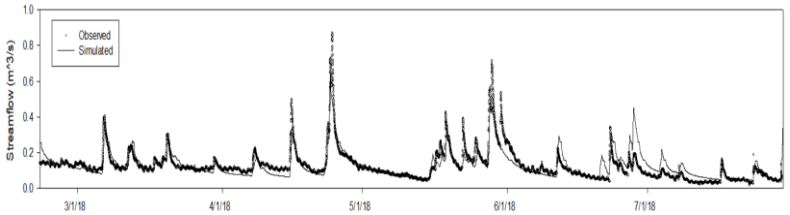
FIU Applied Research Center

Tims Branch Hydrology Model Optimization

- FY 2017-2018: Coupled MIKE SHE/MIKE 11 hydrology model of Tims Branch watershed developed.
  - 2D land surface/3D subsurface model and 1D stream flow model.
- FY 2018-2019: Enhanced model performance and model representativeness:
  - Monitored streamflow TS prepared to evaluate predicted TS
  - Model inputs updated/diversified (PCP, ET, Veg, Soil, NWK, etc.)
  - Input data properly resampled to model grid (Veg, Soil, DEM, etc.)
  - Databases updated: Veg (LAI, RD, K_c), Soil (ρ_b, K_{sat}, θ_r, θ_s)
  - Parameter optimizations with Autocal tool



Optimized model performance:
Root mean squared error (RMSE): 0.050 cms
Nash-Sutcliffe efficacy coefficient (NSE): 0.847



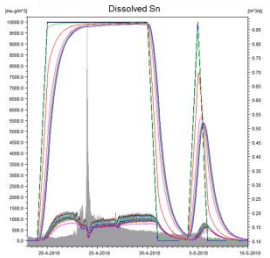
Current model calibration result contrasting predicted streamflow (solid line) with observed streamflow (circle) for the calibration period of 02/20/2018 to 07/30/2018.

Coupled Hydrology & Contaminant Transport Model

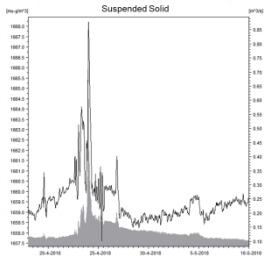
- FY 2018-2019: MIKE 11 AD & ECO Lab module activated.
  - Solute transport: advection, dispersion
  - Sediment transport: sedimentation, resuspension
  - Interactions: adsorption, desorption

Continued Remote Data Collection

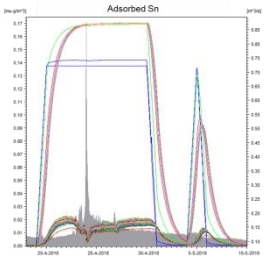
- FY 2018-2019: Water level/discharge TS data from remote monitoring devices deployed in Tims Branch collected and used for model calibration and validation.



Dissolved Sn



Suspended Solids



Adsorbed Sn

Task 3: Contaminant Fate And Transport Modeling in Tims Branch

Accomplishments

- Developed GW model using MODFLOW as part of DOE Fellow Ron Hariprashad's Master's thesis:
  - *"Groundwater/surface water Interaction Along the Tims Branch Watershed, Savannah River Site, SC".*
- WM19 paper & oral presentation:
  - Mahmoudi, M., A. Lawrence. *"An Integrated Hydrological Model for Long-Term Monitoring in Tims Branch Watershed, SC"*, Proceedings of the Waste Management Symposia 2019, Phoenix, AZ, March 2019.
- DOE Fellow Amanda Yancoskie completed 2019 10-wk summer internship with SRNL's Atmospheric Technologies Group under the mentorship of Dr. Grace Maze.
  - *"2D Dam-Break Analysis of L Lake and PAR Pond Dams Using HEC-RAS"*
- DOE Fellow Juan Morales completed 2019 10-wk summer internship at Argonne National Lab with the DOE Office of Science - Office of Biological and Environmental Research's Genomic Science Program under mentorship of Dr. Pamela Weisenhorn.
  - *Investigation of the use of metagenomics to investigate the composition, structure and diversity of microbes in soil contaminated with different levels of heavy metals from Tims Branch system.*

Task 3: Contaminant Fate And Transport Modeling in the Tims Branch Watershed

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FIU Year 9 Ongoing:

- MIKE 11 ECO Lab heavy metal template dev't for major contaminants of concern (U, Hg & Ni).
- Preliminary simulations of contaminant transport for the various contaminants, troubleshooting of errors and optimization of input parameters to improve model performance.
- Fine tuning of model time step settings to improve reliability of coupled model and runtime efficiency.

Proposed Scope for Year 10

- Sensitivity analysis, calibration and validation of fully coupled contaminant transport model.
- Scenario analysis under extreme hydrological conditions that provide information related to inter-compartmental transfers, stormflow impacts and downstream transport of priority contaminants of concern (e.g., Hg, U, Ni, and other heavy metals and radionuclides).
- Data collection limited to monitoring and download of water level data from existing remote monitoring devices deployed in Tims Branch watershed for model calibration and validation.
- Travel to SRS to perform routine maintenance and calibration of remote monitoring devices.

Task 5 – Research and Technical Support for WIPP

FIU Applied Research Center

Site Needs:

This research strives to help the LANL ACRSP team to better understand the long-term fate of the actinide elements in the Waste Isolation Pilot Plant (WIPP). Specifically, the effects of ligands in the waste stream (e.g. EDTA and oxalate) on near field mobility of actinides is still unknown (Dunagan, 2007; Brush, 1990). Complexation constants have been measured for most actinides and lanthanides (Thakur *et al*., 2014; 2015; Borkowski *et al*., 2001). However, their long-term stability and sorption are not yet understood in high ionic strength systems. EDTA is a significant risk factor as it is present in significant amounts in waste and could reach up to 0.3 mM in the repository (Roach *et al.,* 2008).

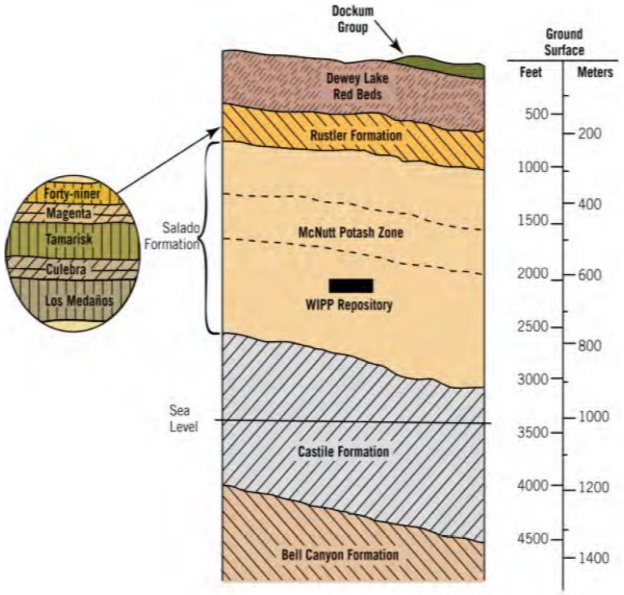
Year 9 Objectives:

To understand the ternary interactions between actinides and WIPP-relevant ligands and minerals and their potential fate in the subsurface.

- Measure sorption parameters for Nd(III), Th(IV), and U(VI) to dolomite in variable ionic strength with and without EDTA
- Measure sorption parameters for Nd(III), Th(IV), and U(VI) to dolomite in WIPP-relevant brines, GWB and ERDA-6

Present (Year 9) Tasks:

- Batch experiments with Nd(III), Th(IV), and U(VI) in variable ionic strength systems (0.1, 0.5, 1.0, and 5.0 M NaCl) with dolomite and with/without EDTA
- Batch experiments with Nd(III), Th(IV), and U(VI) in GWB and ERDA-6



Oxidation State Distribution of Key Actinides in WIPP Performance Assessment

Actinide	Oxidation State				Speciation Data used in Model Predictions
	III	IV	V	VI	
Uranium		50%		50%	Thorium for U(IV), 1 mM fixed value for U(VI)
Plutonium	50%	50%			Am/Nd for Pu(III) and thorium for Pu(IV)
Americium	100%				Americium/neodymium

Task 5 – Research and Technical Support for WIPP

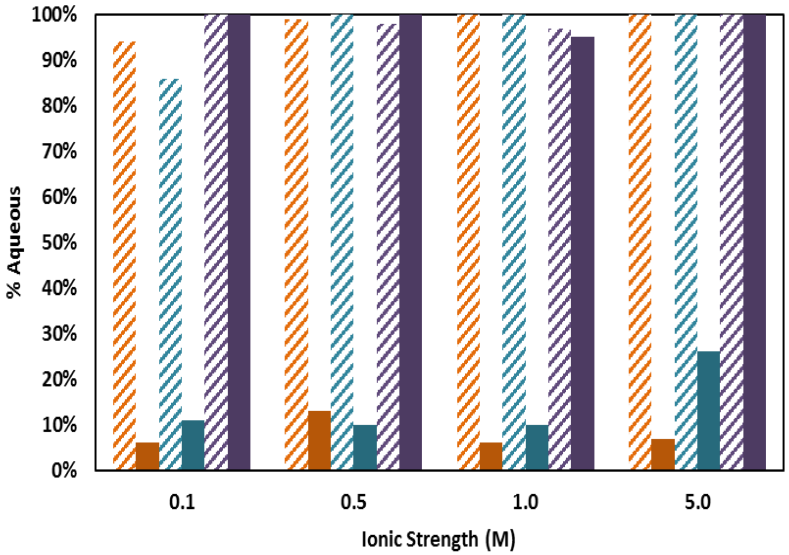
FIU Applied Research Center

Research Highlights Year 9:

- Finalized batch experiments to investigate effect of ionic strength on sorption of Nd(III), Th(IV), and U(VI) to dolomite (0.1, 0.5, 1.0, and 5.0 M NaCl)
- Conducted batch experiments in two WIPP-relevant brines, ERDA-6 and GWB (Borkowski *et al*., 2009)

Accomplishments Year 9:

- Publication titled "*Potential for transport of Cesium as a biocolloid in high ionic strength systems*" was accepted to Chemosphere (May 2019)
- DOE Fellow Frances Zengotita was accepted into the Seaborg Institute Nuclear Science and Security Summer Internship Program at Lawrence Livermore National Laboratory (Summer 2019)
- DOE Fellow Alexis Vento is interning at Sandia National Laboratory with Andy Ward (Summer 2019)
- DOE Fellow Alexis Vento presented on dolomite dissolution in high ionic strength systems at the Mirion Connect 19 Conference (July 2019)



Aqueous fractions of Nd^{3+} (orange), Th^{4+} (blue), and UO_2^{2+} (purple) in variable ionic strength (NaCl) with **(striped)** and without **(solid)** EDTA after 48 hours

Task 5 – Research and Technical Support for WIPP

FIU Applied Research Center

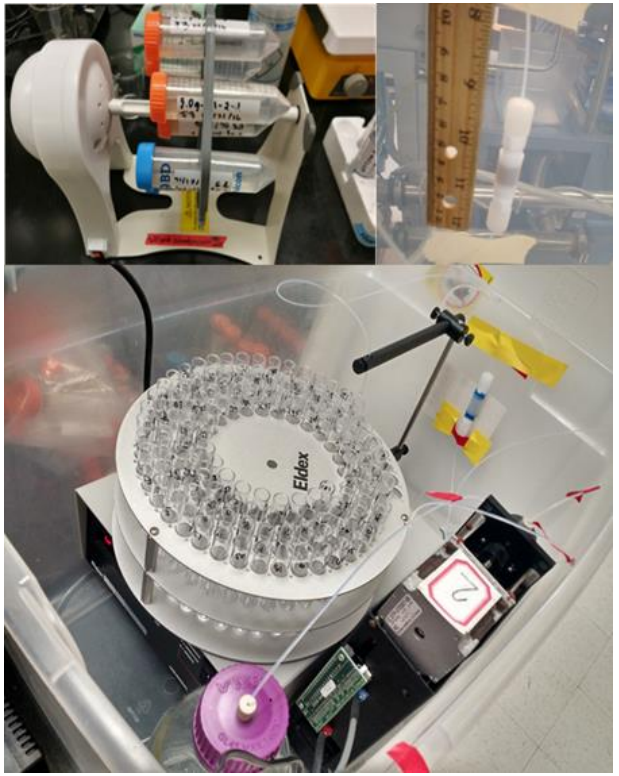
Upcoming for Year 9:

- Present results on ionic strength effects on sorption of Nd(III), Th(IV), and U(VI) to dolomite at the Fall ACS national meeting, August 2019
- Complete batch experiments with WIPP-relevant brines, GWB and ERDA-6

Objectives for Year 10:

To understand the ternary interactions between actinides and WIPP-relevant ligands and minerals and their potential fate in the subsurface.

- Expand work scope to investigate the impact of citrate and oxalate on the sorption of contaminants to dolomite in high ionic strength systems via batch and column experiments
- Continue the work with EDTA with mini-column experiments to further investigate the removal mechanisms of Nd(III), Th(IV), and U(VI) in the presence of ligands and high ionic strength systems



Task 6: Hydrology Modeling For WIPP(New)

Site Needs:

- Significant changes within the last several years, including increased water withdrawals outside the LWA boundary that have impacted water levels and chemistry in compliance monitoring wells on site.
- There is a need for an improved understanding of the regional water balance, particularly the relationship between Culebra recharge and the intense, episodic precipitation events typical of the monsoon.
- This relationship is essential for understanding the rate of propagation of the shallow dissolution front, and the impact of land-use changes around the WIPP facility on water levels in compliance-monitoring wells.
- These types of analyses require a revision of the current site conceptual model to couple surface water and groundwater processes, which both require a high resolution DEM including channels and sink holes to account for surface water routing and return flow.

Objectives:

- Development of a GWM for the WIPP site using the DOE-developed Advanced Simulation Capability for Environmental Management (ASCEM) modeling toolset to improve the current understanding of regional and local groundwater flow at the WIPP site.
- An open source LSM will also be used to provide surface process parameters for input into the ASCEM model (e.g. infiltration rate) to compute the surface water balance, across multiple scales and reduce uncertainties in recharge estimates and propagation of the shallow dissolution front.

Task 6: Hydrology Modeling For WIPP(New)

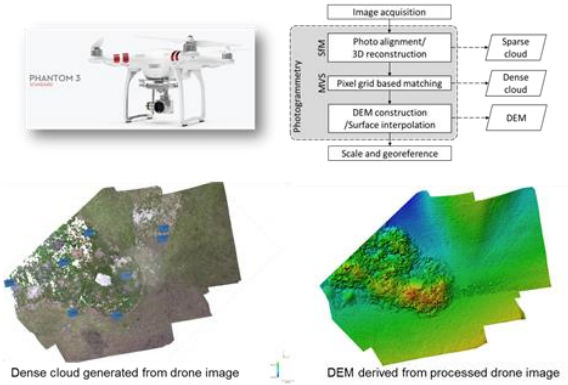
Research Highlights

FIU Applied Research Center

Subtask 6.1: DEM and Hydrologic Network

High-res DEM needed to delineate/extract topo. features (drainage basins, hydrolines, brine lakes, sink holes, discharge pts.) for input into LSM to simulate overland flow, channel routing, and subsurface flow processes.

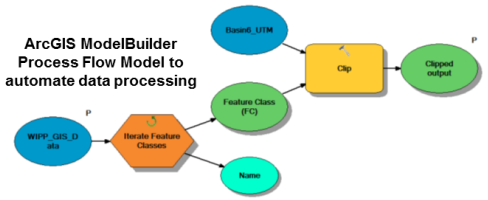
- Lit. review of methods for development of a high-res. DEM. and for extracting surface features (sink holes, hydrology network, etc.)
- Photogrammetry test – aerial images of small field beside ARC collected using drone with 12 MP digital camera and processed using photogrammetry software to generate a high-res DEM (5 mm/pixel). Method can generate sub-meter accuracy DEMs.t.
- Training of DOE Fellows on GIS and image processing techniques.



Subtask 6.2: WIPP Hydrologic Database

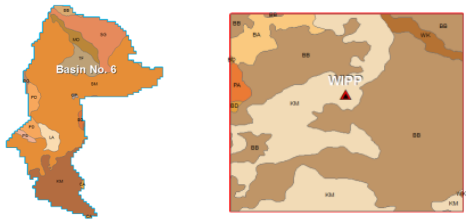
Creation of central data repository & use of geospatial tools for processing model input data.

- Data mining/download of spatial/temporal data (DEMs, hydrolines, water bodies, basins, geology, soils, veg./land cover, roads, admin. boundaries, etc.) from federal/state/local online databases (USDA, USGS, NM State, CFO).
- Training of students to use Python scripts and ArcGIS process flow models to automate download of large datasets and repetitive geoprocessing tasks.



Subtask 6.3: ASCEM GWM and LSM Training

- Delayed – ASCEM team awaiting funding to prepare short training course.
- Training of FIU personnel/students on ASCEM modeling toolset and understanding the requirements for coupling LSM with ASCEM GWM - projected for Aug/Sept 2019 or FIU Performance Year 10.



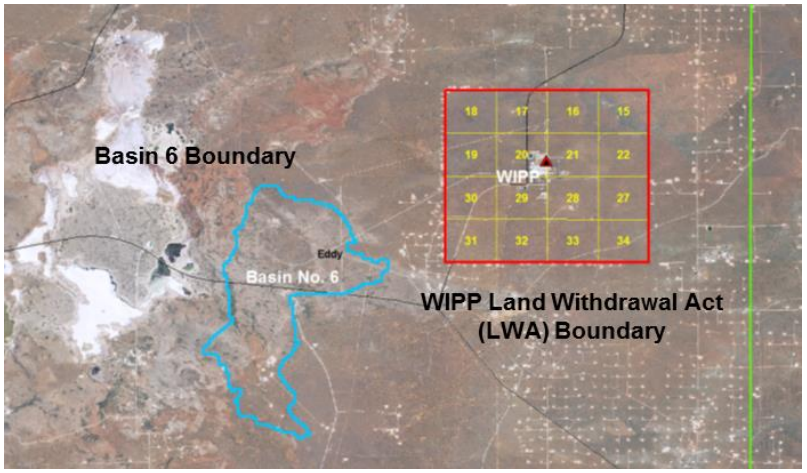
Soil types in Basin 6 *(left)* and within the WIPP LWA boundary *(right)*

Task 6: Hydrology Modeling For WIPP(New)

FIU Applied Research Center

FIU Year 9 Ongoing:

- Trip to WIPP/Basin 6 planned in August 2019 to survey study areas, meet with collaborators, present project progress and discuss plans for next year.



Proposed Scope for Year 10

- Pilot study to capture high-res imagery of representative basin at WIPP in Nash Draw area using UAV (drone).
- Image processing using state-of-the-art photogrammetric techniques to build a high-res DEM for accurate delineation/extraction of topographical and hydrologic features.
- Utilize selected open-source LSM, delineated features and other relevant hydrological data collected to initiate development of a LSM of Basin 6, to be used in future to force ASCEM GWMs to predict GW flow patterns.
- Spatial distribution of recharge, and GW flow rates and directions will be used to estimate rate of halite dissolution and rate of propagation of the shallow dissolution front, which both have potential to affect post-closure repository performance.
- Pilot study will serve as proof of concept that the proposed methodology is feasible and has practical applications at WIPP to generate high-res imagery for development of a DEM, which is essential for detailed delineation of hydrologic basins within and surrounding the WIPP Land Withdrawal Act (LWA) boundary.
- Training of FIU's research personnel and students on selected LSM and ASCEM to carry out proposed scope.