DOE-FIU Cooperative Agreement Annual Research Review – FIU Year 3

Wednesday, August 23, 2023		
9:00 - 9:05 am EDT	Kick-Off /Welcoming Remarks (DOE-EM)	Rod Rimando (Acting Director, Technology Development) – DOE EM-3.2
9:05 - 9:10 am EDT	Welcoming Remarks (DOE-LM)	Ms. Jalena Dayvault (Site Manager) – DOE LM
9:10 - 10:40 am EDT	Project 2: Environmental Remediation Science & Technology	FIU, DOE HQ, SRNL, PNNL, ORNL, LANL, LBNL, CBFO
10:40 am - 12:10 pm EDT	Project 1: Chemical Process Alternatives for Radioactive Waste	FIU, DOE HQ, PNNL, WRPS, SRNL, SRS
LUNCH BREAK [12:10 – 1:30 pm]		
1:30 - 3:00 pm EDT	Project 3: Waste and D&D Engineering & Technology Development	FIU, DOE HQ, SRNL, PNNL, LBNL, INL, ANL
Thursday, August 24, 2023		
9:00 - 10:30 am EDT	Projects 4 & 5: STEM Workforce Development and Training	FIU, DOE HQ (EM & LM), SRNL, PNNL, WIPP, SRS, ORP, LBNL, WRPS, INL, Grand Junction
BREAK [10:30 – 10:35 am]		
10:35 - 12:00 pm EDT	Wrap Up (FIU Projects 1, 2, 3, 4 & 5)	FIU, DOE HQ (EM & LM)

PROJECT 2

Environmental Remediation Science & Technology

FIU Personnel and Collaborators

Principal Investigator: Leonel Lagos

Project Manager: Yelena Katsenovich

Faculty/Staff: Ravi Gudavalli, John Dickson, Vadym Drozd, Angelique Lawrence, Pieter Hazenberg

DOE Fellows/Students: *Angel Almaguer, *Kirsten Olson, Hannah Aziz, *Stevens Charles, Mariah Doughman, *Caridad Estrada, Aubrey Litzinzger, *Phuong Pham

DOE-EM: Genia McKinley, *Kurt Gerdes, Rod Rimando, Skip Chamberlain, Nick Machara, *Karen Skubal, Alexander Koenig

DOE-SRS: Phillip (Tony) Polk

SRNL: Brian Looney, Hansell Gonzalez-Raymat, Carol Eddy-Dilek, Mark Amidon, Bruce Wiersma, Connie Herman, Brady Lee

SREL: Daniel Kaplan

LBNL: Haruko Wainwright, Zexuan Xu

PNNL: Rob Mackley, Nik Qafoku, Jim Szecsody, Hilary Emerson, Matthew Asmussen

DOE-ORP: Erik Nelson

LANL: *Don Reed, Juliet Swanson, David Moulton, Jay Je-Hun Jang, Jean-Francois (Jef) Lucchini

DOE-CBFO: Anderson Ward

ORNL: Eric Pierce, Alexander Johs

*Former contributors

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Project Tasks and Scope

TASK 1: REMEDIATION RESEARCH AND TECHNICAL SUPPORT FOR THE HANFORD SITE	
Subtask 1.2	Re-oxidation of Redox Sensitive Contaminants Immobilized by Strong Reductants
Subtask 1.3	Eval. of Competing Attenuation Processes for Mobile Contaminants in Hanford Sediments
Subtask 1.4	Experimental Support of Lysimeter Testing
Subtask 1.5	Remediation Research on Combination of Reduction and Sequestration Treatment (NEW)
TASK 2: REMEDIATION RESEARCH AND TECHNICAL SUPPORT FOR THE SAVANNAH RIVER SITE	
Subtask 2.1	Investigate Environmental Factors Controlling the Attenuation and Release of Iodine in the Wetland Sediments at Savannah River Site
Subtask 2.2	Investigating the Effect of KW-30 (Humate Material) on the Removal of Comingled Contaminants
TASK 3: CONTAMINANT FATE AND TRANSPORT MODELING FOR THE SAVANNAH RIVER SITE	
Subtask 3.1	Calibration of the Tims Branch Watershed Model and Scenario Analysis
Subtask 3.2	Model Development for Fourmile Branch with Specific Focus on the F-Area Wetlands
TASK 5: RESEARCH AND TECHNICAL SUPPORT FOR WIPP	
Subtask 5.2	Fate of Actinides in the Presence of Ligands in High Ionic Strength Systems
TASK 6: HYDROLOGY MODELING OF BASIN 6 OF THE NASH DRAW NEAR THE WIPP	
Subtask 6.2	Model Development
Subtask 6.3	Fieldwork and Data Collection to Support Hydrological Model Calibration and Validation (NEW)
TASK 7: ENGINEERED MULTI-LAYER AMENDMENT TECHNOLOGY FOR HG REMEDIATION ON OAK RIDGE RESERVATION	

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Task 1

Remediation Research and Technical Support for the Hanford Site

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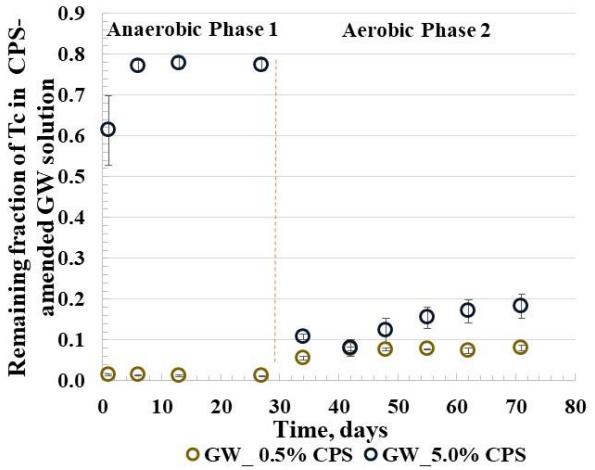
Subtask 1.2: Re-oxidation of Redox Sensitive Contaminants Immobilized by Strong Reductants

FIU Year 3 Research Highlights & Accomplishments:

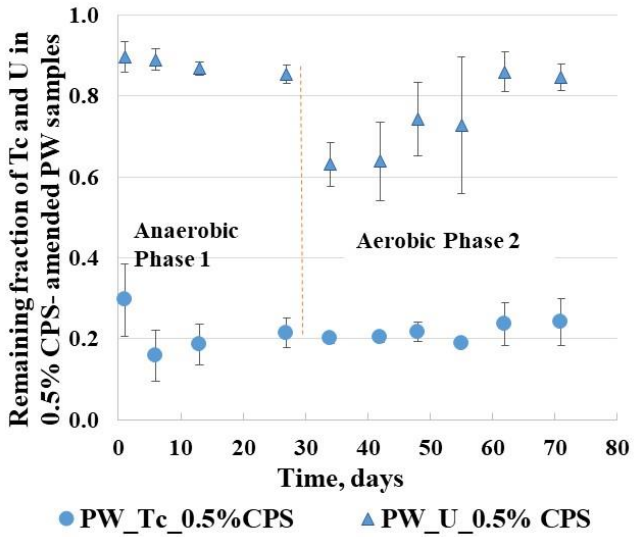
- Studied re-oxidation behavior of ^{99}Tc, U(VI), and NO_3^- after treatment with strong reductants:
  - 0.5 % calcium polysulfide (CPS)
  - 5 % CPS

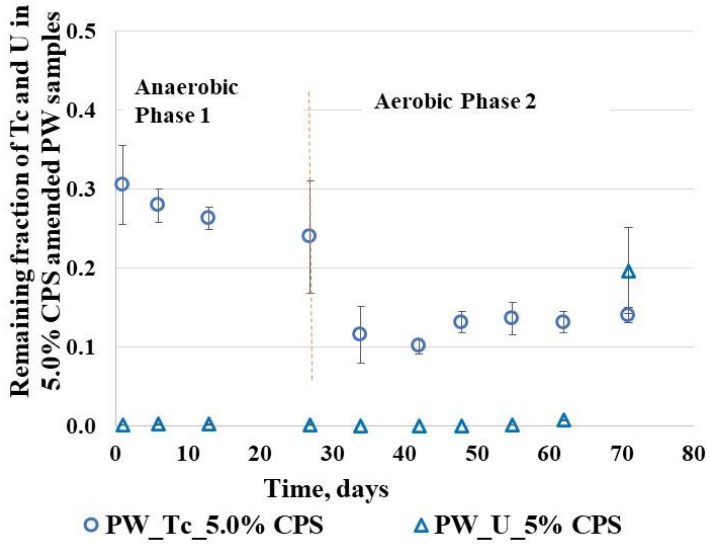
    CPS is a 29% solution.
  - PW: pH 8.2, 10 µg/L Tc, 150 mg/L U.
  - GW: pH 7.8, 420 µg/L Tc, 124 mg/L NO_3^-
- Ringold Formation sed<2 mm in triplicate samples
- Two phases of experiments for reduction of ^{99}Tc, U(VI), and NO_3^-:
  - In presence of CPS under anaerobic conditions for up to **30 days**
  - In aerobic conditions for up to **41 days with aeration 2x/week for 30 s**.

    Total testing = 71 days.



0.5% CPS samples contained smaller remaining fraction of Tc(VII) throughout both phases vs. 5.0% CPS.





Similar removal of Tc at both CPS conc, but Tc is higher compared to 1% ZVI.
U removal with 0.5% CPS – 0.85±0.03 remaining, 5.0% CPS improved to 0.13±0.06.

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Subtask 1.2: Re-oxidation of Redox Sensitive Contaminants Immobilized by Strong Reductants

FIU Year 4 Projected Scope

- Results indicate 1% ZVI or SMI were stronger reductants under anaerobic conditions and showed better resistance to re-oxidation in aerobic conditions, compared to 0.1% and 0.5% or 5%-CPS- amended PW and GW.

- Initiate preliminary experiments on coupling ZVI approach with ammonium hydroxide and investigate their effect on the re-oxidation behavior of comingled Tc, U and nitrate. Experiments will be conducted under the subtask 1.5.
    - Research will follow experimental matrix outlined in DV-1 Operable Unit treatability study ongoing at Hanford Site.

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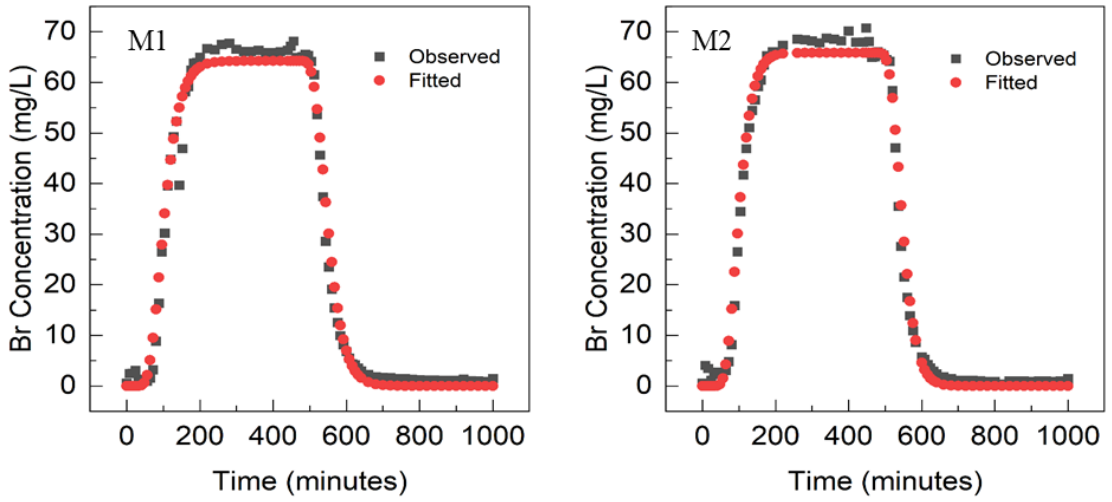
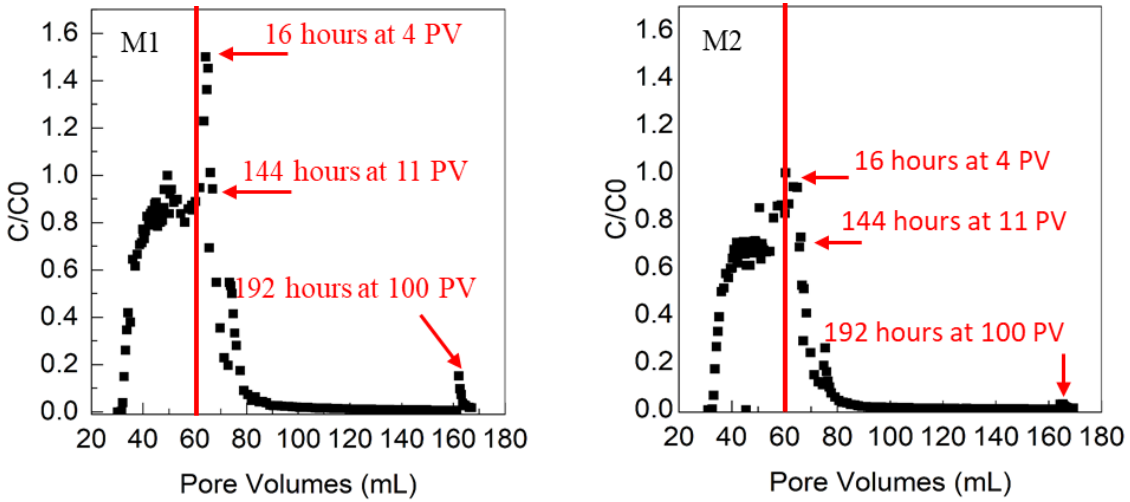
Subtask 1.3: Evaluation of Competing Attenuation Processes for Mobile Contaminants in Hanford Sediments

FIU Year 3 Research Highlights & Accomplishments:

Bromide Breakthrough Curves



Uranium Breakthrough Curves



Column	*D* (cm^2min^{-1})	*R*
M1	0.055	0.958
M2	0.044	0.953

$$K_d = \frac{[contaminant_{sediment}]}{[contaminant_{solution}]}$$

Column	K_d (L/kg)
M1	1.2
M2	1.5

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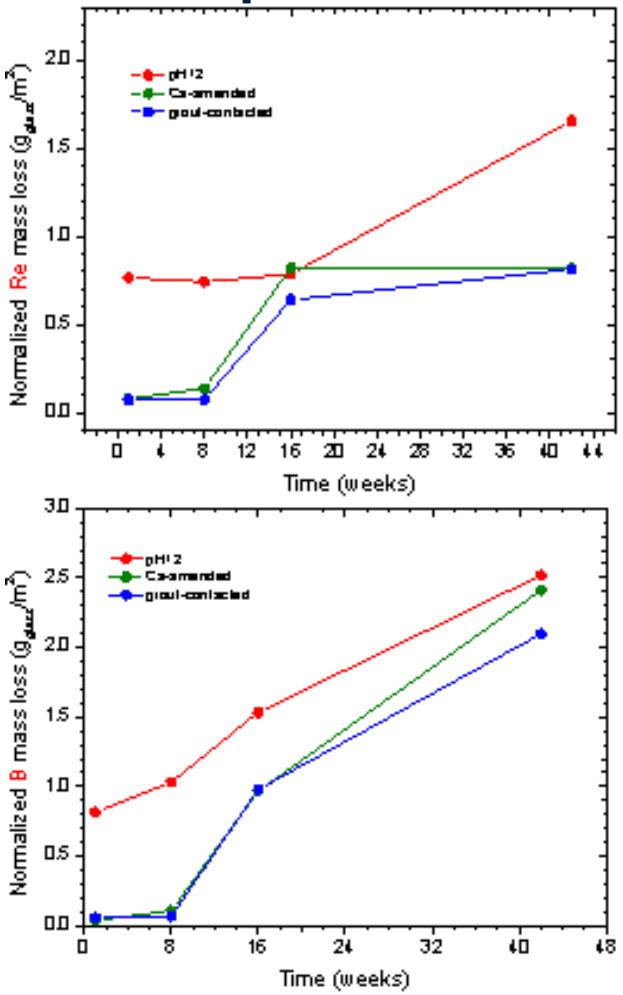
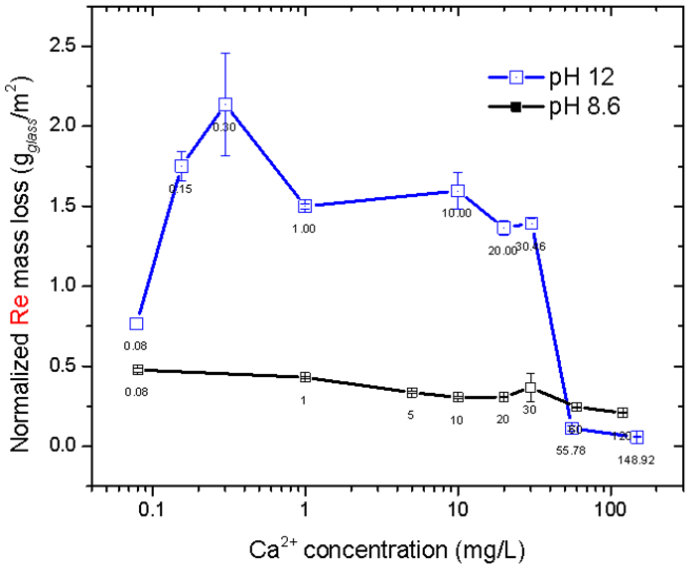
Subtask 1.4: Experimental Support of Lysimeter Testing

FIU Year 3 Research Highlights & Accomplishments:

- Investigated the impact of major elements on the dissolution behavior of borosilicate ORLEC28 glass
  - Test matrix in PCT: long-term experiment at 90°C in pH 12 buffer, grout-contacted and Ca-amended solutions (8, 16 and 42 weeks)
  - Test matrix in PCT: triplicated reactors, Ca-amended solutions at pH 12 and 8.6 (0-130 mg/L Ca^{2+})
- XRD and BET analyses of treated glass, SEM/EDS measurements in cross-sections of glass coupons to study glass erosion
- Manuscript *"The corrosion behavior of borosilicate glass in the presence of cementitious waste forms"* is ready for journal submission.



Normalized losses of Re and B by ORLEC28 glass in different solutions at 90°C in long-term PCT.



Ca^{2+} inhibits glass dissolution at concentration above 20 mg/L (pH 12 and 8.6)

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Subtask 1.4: Experimental Support of Lysimeter Testing

FIU Year 4 Projected Scope

- Investigate glass dissolution in contact with Al- and grout/sediment-contacted groundwater to investigate any common ion effect.
  - Al concentrations in the leachate will range from 0.3 ppm to 30 ppm at different pHs (7, 9, 10, 11).
- Support glass characterization studies via microscopy, spectroscopy, and X-ray diffraction techniques.

Major elements present in grout-, sediment-, and grout/sediment-contacted solutions are identified through analysis.

- **The impact of two major elements, Si and Ca, on glass dissolution has been systematically investigated.**

Element	Concentrations, µg/L		
	Grout-contacted	Sediment-contacted	Grout/sediment-contacted
pH	11.52	8.68	8.60
B	172(6)	2.8(6)	112.1(9)
Re	0.052(4)	0.006(3)	0.08(9)
Si	6397(83)	22711(443)	22719(199)
Al	423(118)	391(24)	18(5)
Ca	111506(2312)	2934(45)	48634(449)
Mg	33(2)	106(5)	24821(259)
K	3064(226)	6037(347)	18947(1084)
Fe	0	291(66)	0

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Subtask 1.5: Remediation Research on Combination of Reduction and Sequestration Treatment (NEW)

FIU Year 3 Research Highlights & Accomplishments:

- In a process of a developing a test plan, ordering necessary laboratory supplies for the experiments including a gas cylinder with 5% ammonia gas in 95% nitrogen as well as ammonia and sodium hydroxide.
- Preparing to conduct preliminary experiments to examine sequestration of redox sensitive contaminants via sequentially applied ZVI followed by ammonia gas treatment.
- Seeking an enthusiastic PhD student interested in pursuing this innovative research opportunity.

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Subtask 1.5: Remediation Research on Combination of Reduction and Sequestration Treatment (NEW)

FIU Year 4 Projected Scope

- Initiate experiments using sediments spiked with Tc(VII), U(VI) and nitrate.
  - This would allow the reduction of redox sensitive contaminants under site specific pH conditions followed by the application of ammonia gas to maximize contaminant sequestration by coating of aluminosilicates on top of reduced U and Tc phases (Szecsody et al., 2012; Szecsody et al., 2010).

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Task 2

Remediation Research and Technical Support for the Savannah River Site

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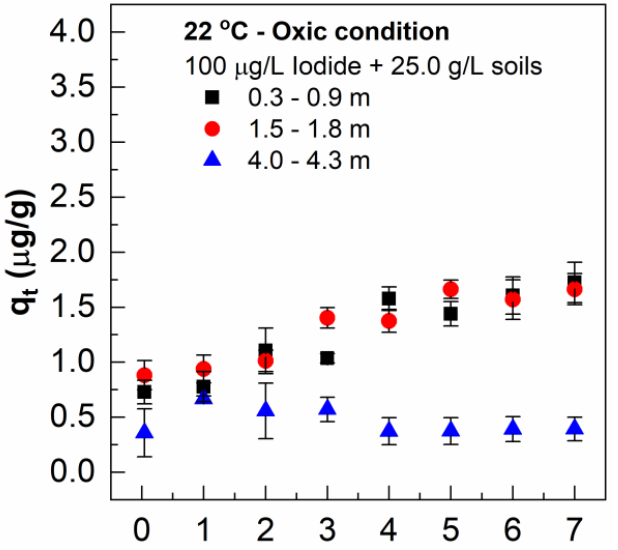
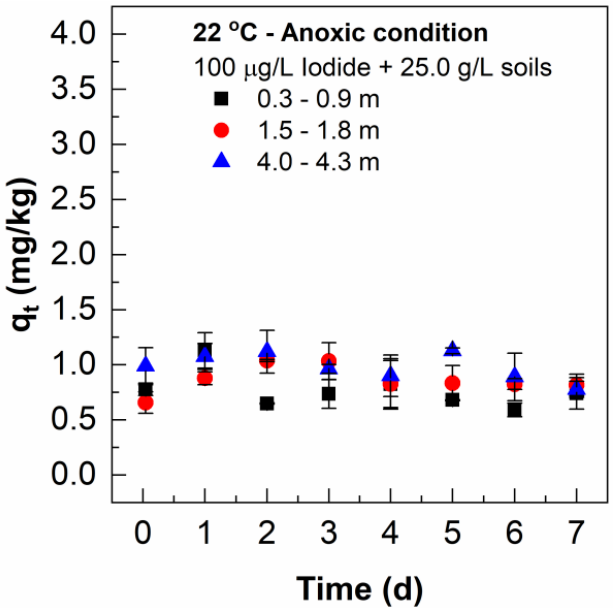
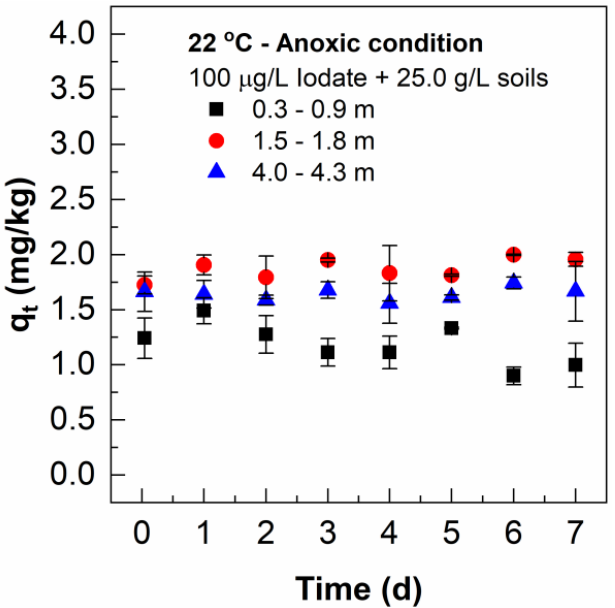
Subtask 2.1: Environmental Factors Controlling the Attenuation and Release of Contaminants in the Wetland Sediments at Savannah River Site

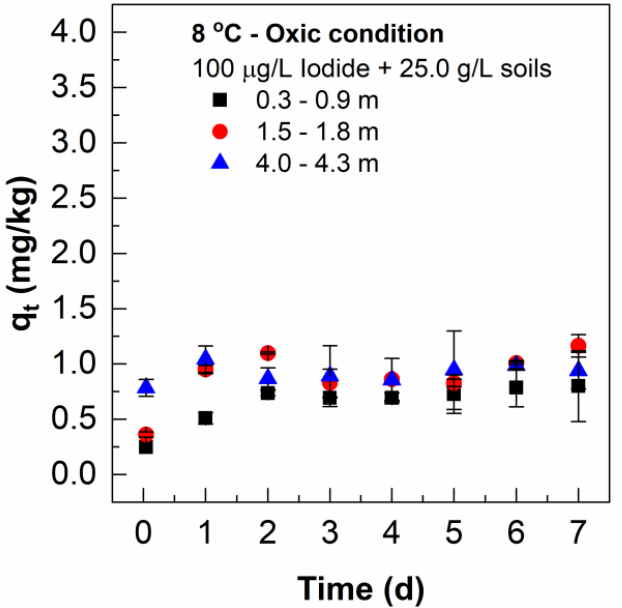
FIU Year 3 Research Highlights & Accomplishments:

- Investigated the attenuation of iodide and iodate by wetland soils at various depths @ pH 5.5, 22°C and 8°C
  - Solid to liquid ratio: 25 g/L
  - Concentration: 100 ppb
  - Depth intervals: 1-3 ft., 5-6 ft., and 13-14 ft.
  - Aerobic vs Anaerobic
- DOE Fellow Phuong Pham successfully completed Ph.D. and joined SRNL as Postdoctoral Scholar.









Subtask 2.1: Environmental Factors Controlling the Attenuation and Release of Contaminants in the Wetland Sediments at Savannah River Site

FIU Year 4 Projected Scope

- Continue to investigate the attenuation and release of iodine in the wetland sediments and the use of amendments such as organo clays (MRM, PM-199) to mitigate the release of iodine from wetland soils.
- Conduct experiments to understand the effect of:
  - Redox Conditions: perform experiments in anoxic conditions
  - pH on desorption: pH = 4 - 8
  - Competition of ions: nitrate and other relevant ions

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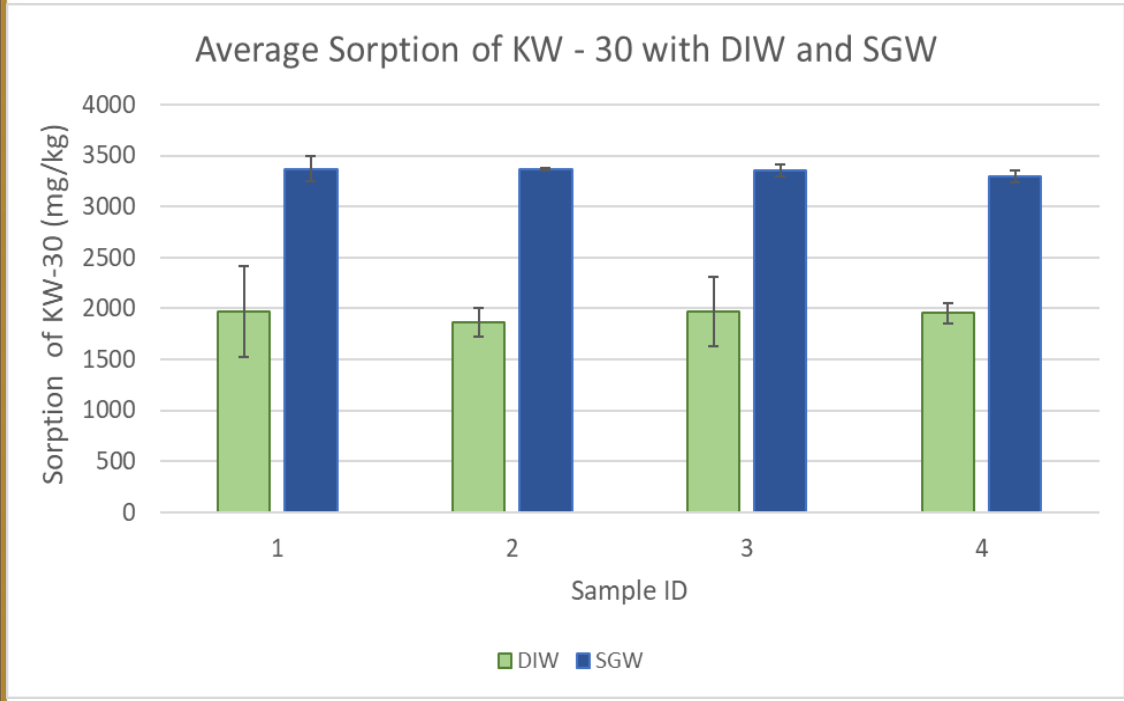
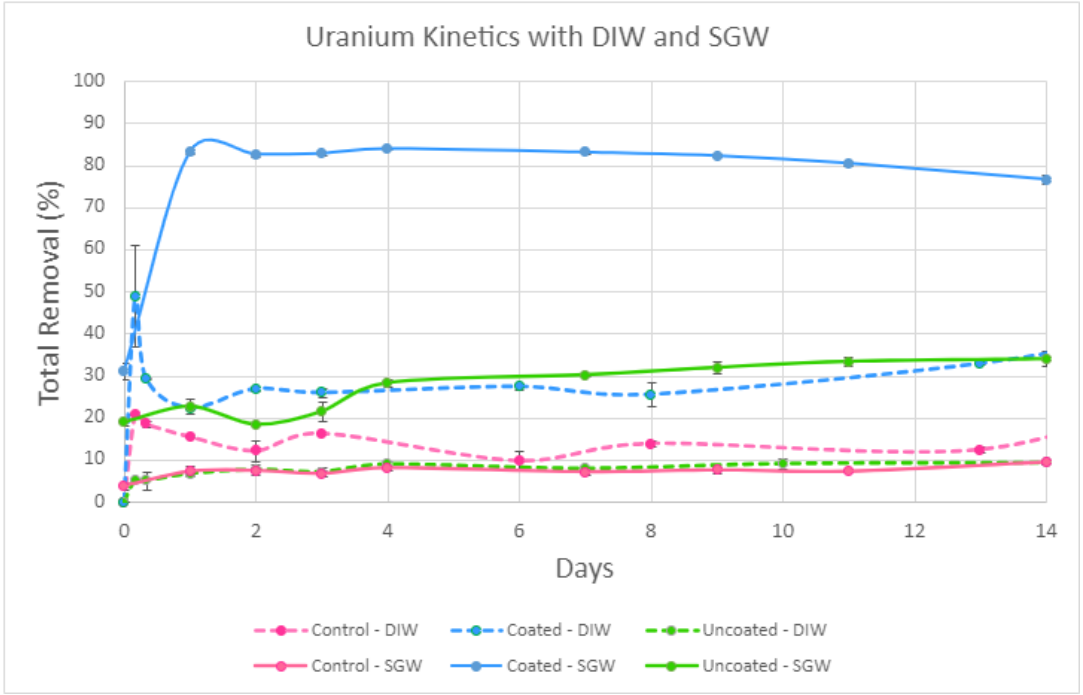
Subtask 2.2: Investigating the Effect of KW-30 (Humate Material) on the Contaminant Removal

FIU Year 3 Research Highlights & Accomplishments:

- Formulated synthetic groundwater (SGW) recipe
  - ➢ Used data FOB 21 and FOB20 wells from SRS F-Area (wells close to basins to represent untreated area)
- Studied the influence of humate and GW ions on kinetics of uranium removal
  - ➢ 200 mg of SRS sediment*
  - ➢ pH - 4 (0.1m HCl/NaOH)
  - ➢ Uranium: 700 ppb





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Subtask 2.2: Investigating the Effect of KW-30 (Humate Material) on the Contaminant Removal

FIU Year 4 Projected Scope

- Perform experiments with other contaminants and comingled contaminants
  - Effect pf pH
  - Effect of ORP
  - Effect of initial concentration (isotherms)
  - Desorption

	g/mol	K+	SO42-	Mg2+	Ca2+	Cl-	NO3-	Na+
CaCl2	110.98				0.041	0.082		
NaSO4	142.04		0.133					0.133
MgCl2	95.211			0.077		0.155		
NaCl	58.44					0.313		0.313
KCl	74.55	0.025				0.025		
NaNO3	84.99						1.845	1.845
TOTAL		0.025	0.133	0.077	0.041	0.574	1.845	2.290

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Task 3

Contaminant Fate and Transport Modeling for the Savannah River Site

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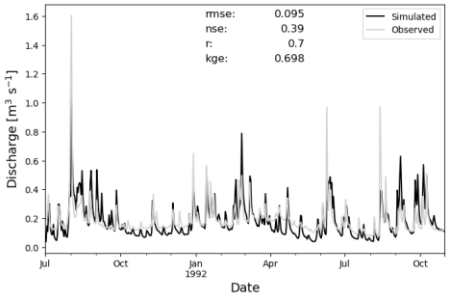
Task 3: Contaminant Fate and Transport Modeling for the Savannah River Site

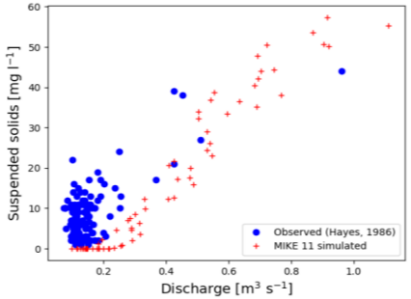
Overall problem and site needs

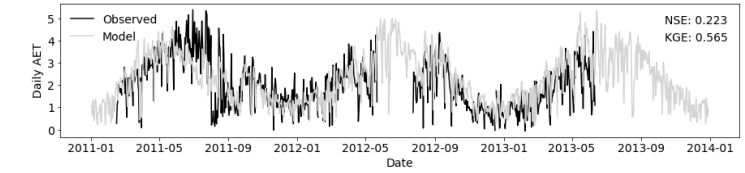
- SRS & other DOE sites challenged with heavy metal & radionuclide surface & subsurface contamination.
- Use numerical models to evaluate impact of extreme hydrological events & long-term hydrological changes on GW-SW interactions, and fate & transport of major contaminants of concern in SRS streams. Specifically focus on heavy precipitation, floods and drought.
- Establish long-term monitoring strategy.

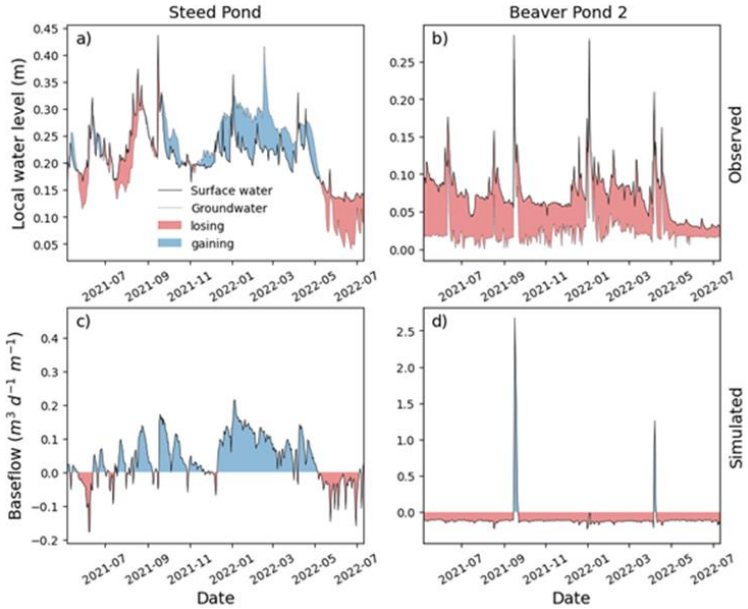
Focus domains:

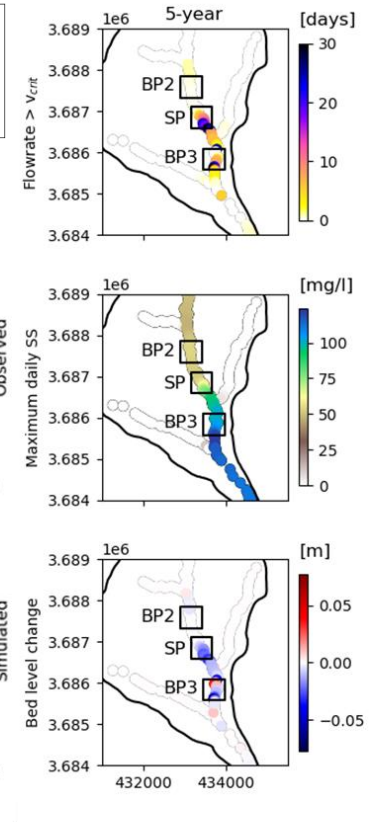
- Use Tims Branch as braided stream system test bed for model development (Subtask 3.1).
- Duplicate process for critically contaminated SRS watershed Fourmile Branch (Subtask 3.2).
- Completed optimization/calibration of Tims Branch (MIKE) **hydrological and sediment transport** model for long-term simulations (Milestone 2022-P2-M10) (Subtask 3.1).











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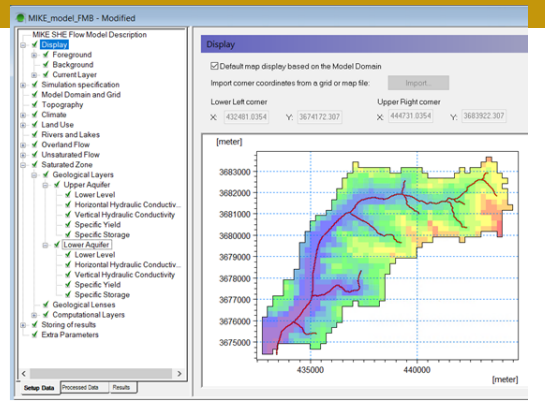
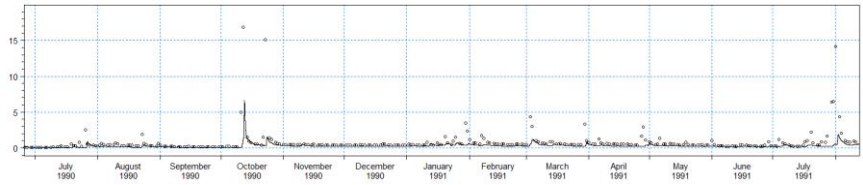
Task 3: Contaminant Fate and Transport Modeling for the Savannah River Site

Highlights & Accomplishments (Subtask 3.2):

- Developed MIKE SHE/MIKE 11 model for FMB watershed (250m res) using open-source data, GIS & Python scripts (Milestone 2022-P2-M2).
  - In the process of calibrating hydrological model parameters.
- Developed integrated surface-subsurface model of F-Area using ATS (Milestone 2022-P2-M7) and Python package Watershed Workflow, which enables quick generation of ATS model from publicly available data.
  - Python scripts developed to generate input files: high-res. mesh of F-Area
  - Automatic generation of XML file containing model parameters





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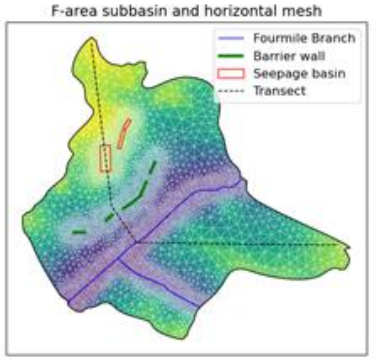
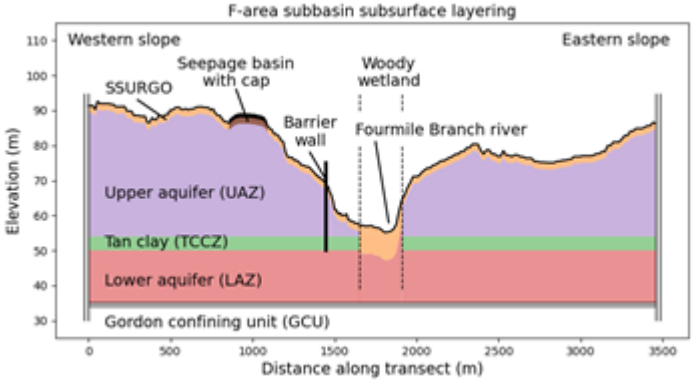
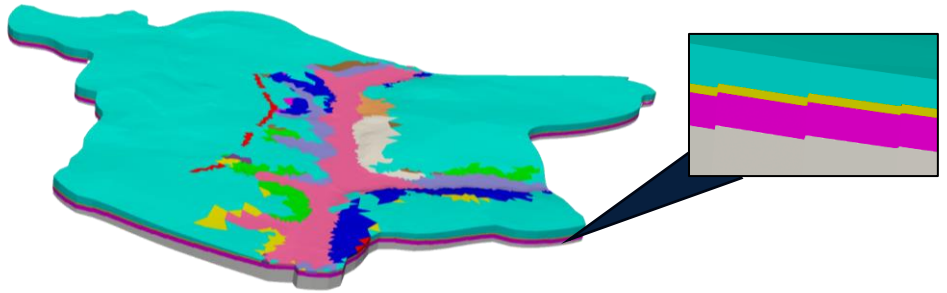
- Complete calibration of MIKE model and perform watershed-scale simulations (event-based & long-term).
- Calibrate ATS model using in-situ sensor network data and perform hydrology simulations (event-based & long-term) in F-Area seepage face and riparian zone.
- Train FIU graduate and/or undergraduate students (DOE Fellows) on model development using MIKE and ATS, as well as data generation and model evaluation using GIS and Python.
- Develop model code for U/I-129 transport in close collaboration with ALTEMIS project team members and perform preliminary simulations of flow and reactive transport of U/I-129 in SRS F-Area.







Task 5

Research and Technical Support for WIPP

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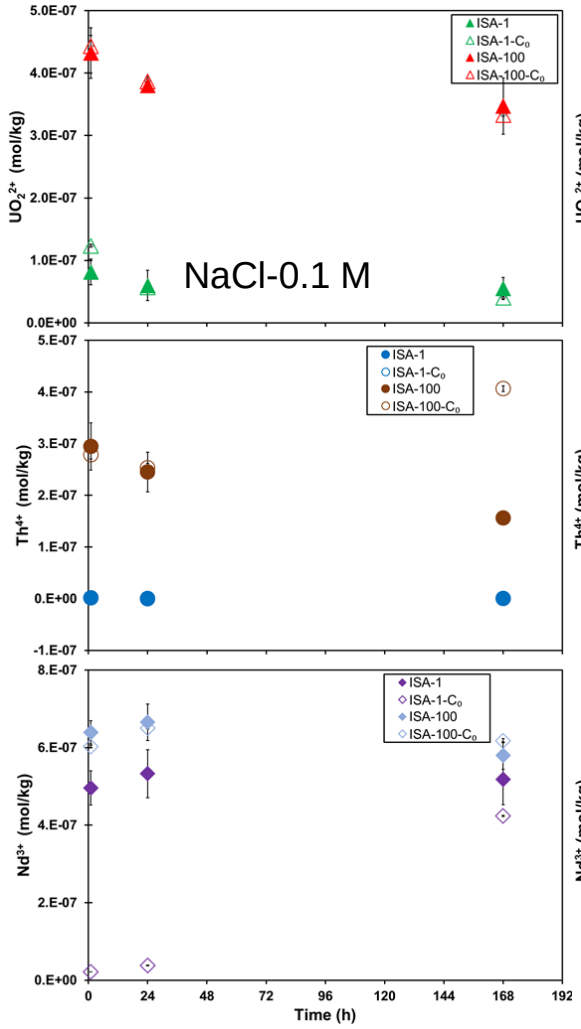
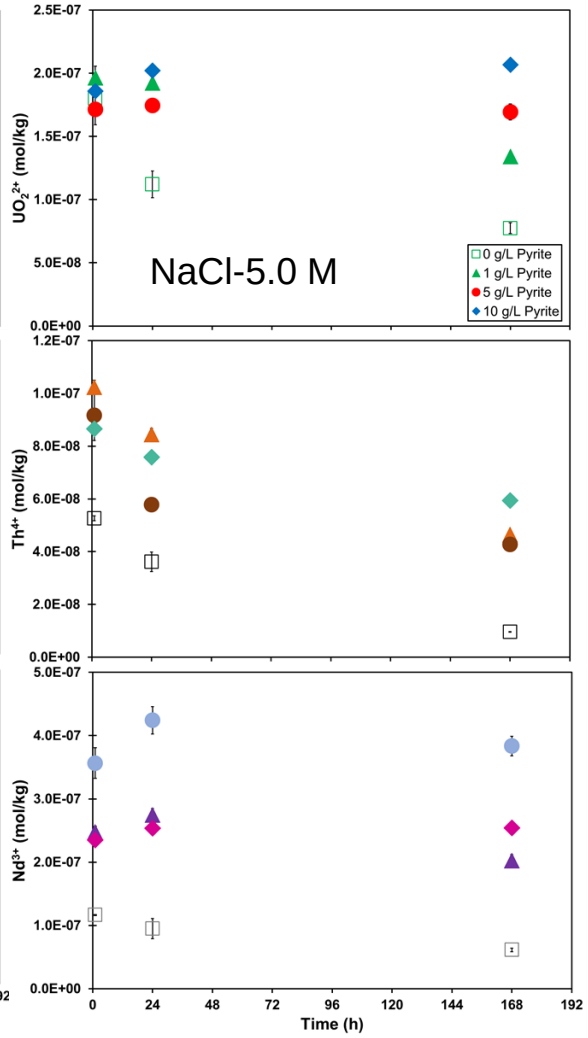
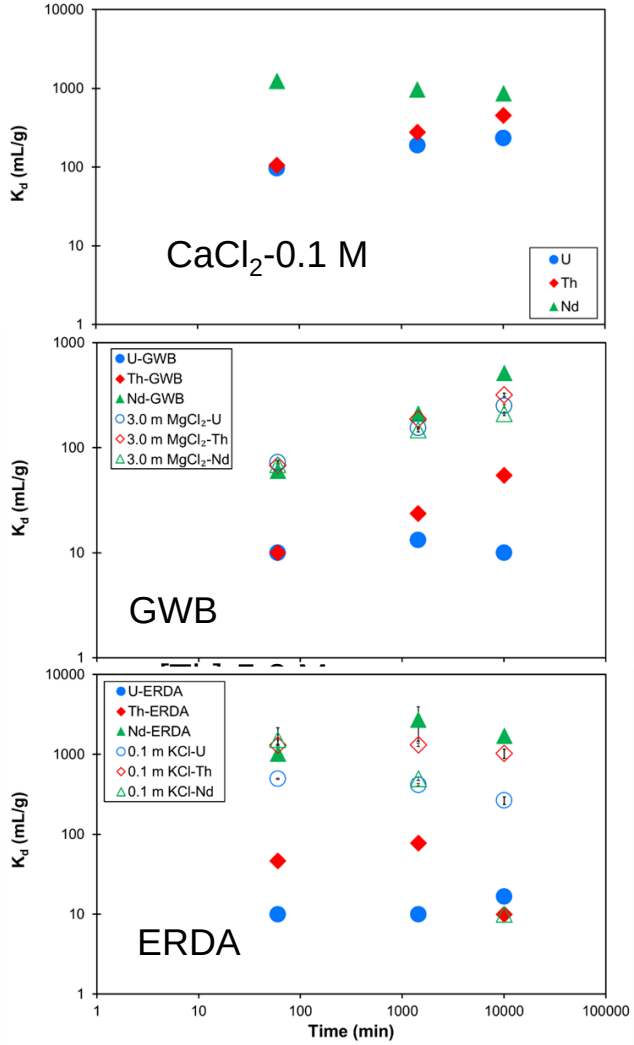
Subtask 5.1: The Fate of Actinide in the Presence of Ligands in High Ionic-Strength System

FIU Year 3 Research Highlights & Accomplishments:

- ISA addition enhanced actinide solubility
- Increasing pyrite dosage and ISA impacted solubility of UO_2^{2+} and, Nd^{3+}, Th^{4+} via changes to solution chemistry.
- Sorption of Nd^{3+} were typically larger compared to sorption of UO_2^{2+} and Th^{4+}







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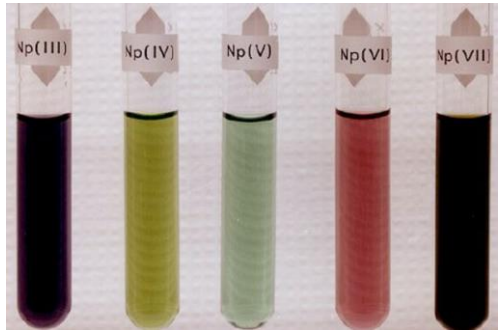
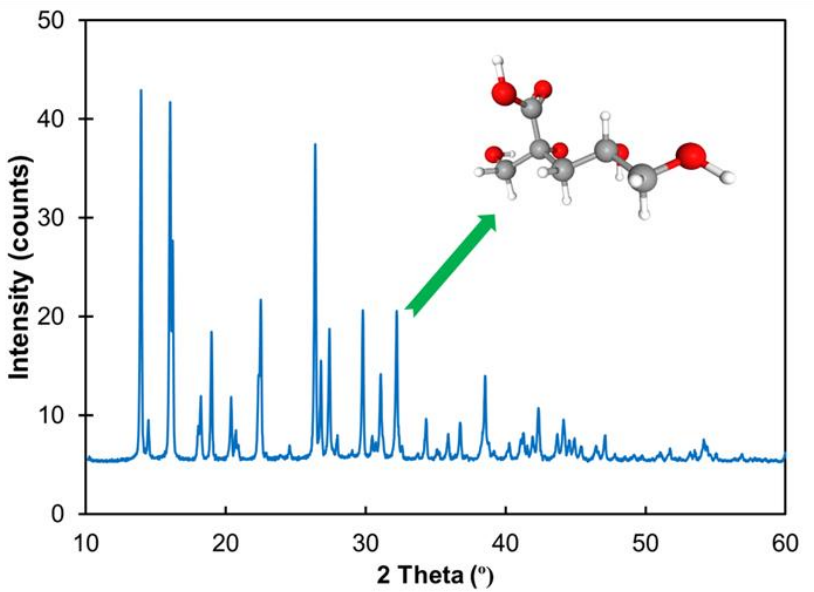
Subtask 5.1: The Fate of Actinide in the Presence of Ligands in High Ionic-Strength System

FIU Year 4 Projected Scope

- Employ EQ3/6 v8a speciation model to obtain pertinent Pitzer's interaction parameters (virial coefficients) for high salt systems.
- Study the impact of citrate, an important byproduct of alkaline degradation of cellulose on sorption of actinide onto iron oxide minerals in WIPP-relevant brines and conditions.
- Characterize treated solid phases employing microscopy, spectroscopy and x-ray diffraction techniques.





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Task 6

Hydrology Modeling of Basin 6 of the Nash Draw Near the WIPP

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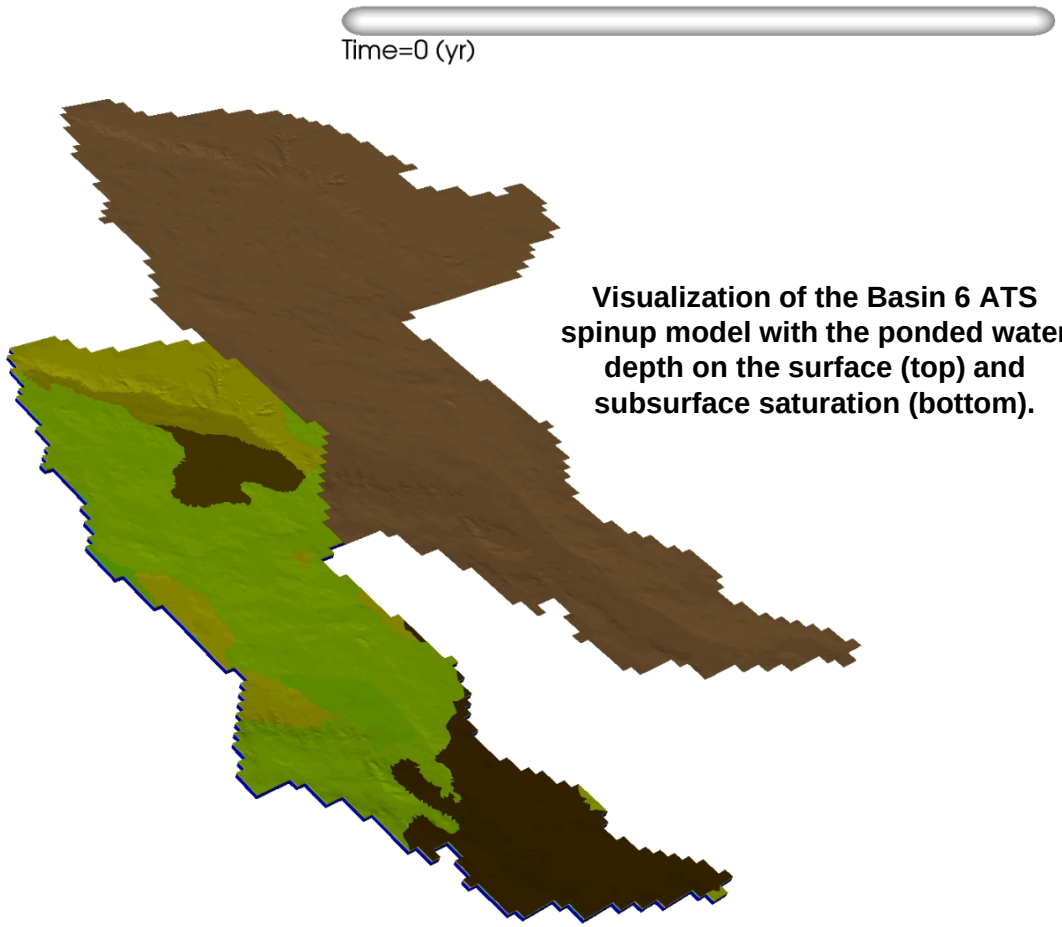
Task 6: Hydrology Modeling of Basin 6 of the Nash Draw Near the WIPP

Site Needs:

- Understanding of regional water balance near WIPP, particularly Culebra recharge, during intense, episodic precipitation events.
- Estimation of propagation rate of shallow dissolution front.
- Assessment of impact of land-use changes around WIPP on water levels in compliance-monitoring wells.
- A high-resolution DEM that can represent localized features (i.e., along river network, in gullies and sinkholes) where recharge anticipated to predominantly occur.

FIU Year 3 Highlights (Subtask 6.2):

- Implemented Python package, **Watershed Workflow** - enables quick generation of a site mesh from publicly available data.
  - High-res. mesh of Basin 6 (includes spatial variations in NLCD land cover types, SURRGO soil texture & subsurface information) with Daily atmospheric forcing using DayMet data.
- Developed a spinup and transient ATS model for Basin 6 by establishing an equilibrium state and then using that state to model multiple years of meteorological data.



Visualization of the Basin 6 ATS spinup model with the ponded water depth on the surface (top) and subsurface saturation (bottom).

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Subtask 6.3: Fieldwork and Data Collection to Support Hydrological Model Calibration and Validation (NEW)

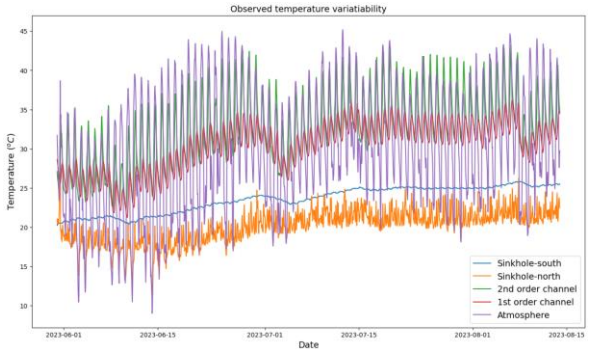
FIU Year 3 Highlights (Subtask 6.3):

- 5 HOBO U20L water level loggers (pressure transducers) deployed in Basin 6 in areas where surface flow likely to occur.
- 48 soil samples collected at depths from 0 - 10 feet below the ground surface.
- Performed GPR sensor measurements.

Setting up water level sensors and collecting soil samples.

FIU Year 4 Projected Scope

- Continue analysis of soil samples collected in Summer 2023.
- Collect and analyze additional soil samples at different depths throughout Basin 6.
- Collect additional water level measurements at sites previously monitored in FIU Year 3 and extend water level sensor network to include sinkholes along main river network and at basin exit near brine lakes.
- Take infiltration measurements at several locations throughout Basin 6.



HOBO water level data at all HOBO unit sites.

Using a GPR rover within Basin 6 (GPR rover developed in Project 5, Task 2).

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Task 7

Engineered Multi-Layer Amendment Technology for Hg Remediation on Oak Ridge Reservation

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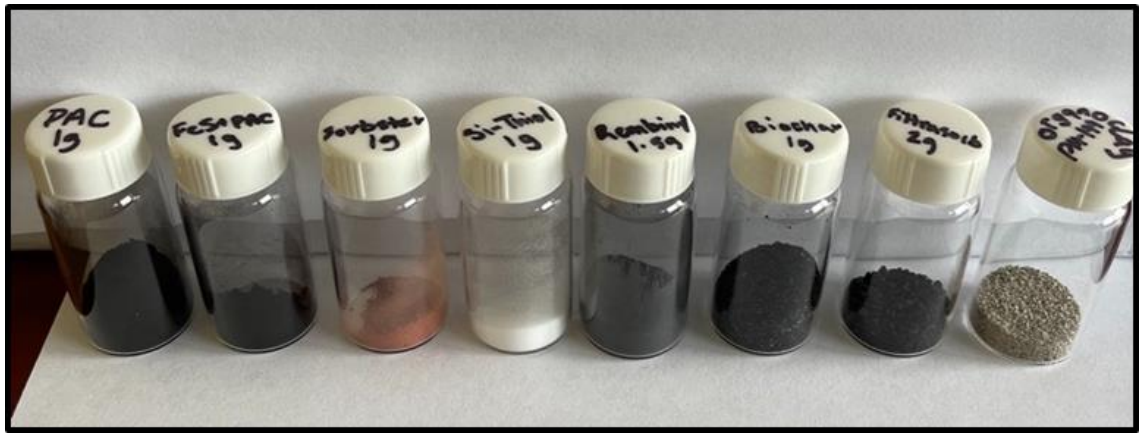
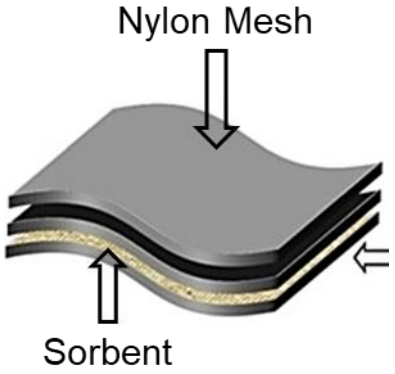
Task 7: Engineered Multi-Layer Amendment Technology for Hg Remediation on Oak Ridge Reservation

FIU Year 3 Research Highlights & Accomplishments:

- Performed stability evaluation of select sorbents via EPA's Toxicity Characteristic Leaching procedure (TCLP), SW-846 Test Method 1311 and leaching test to determine release of constituents from sorbent media.
- Eight sorbents were studied as potential sorbents for in-situ remediation of mercury species in EFPC ecosystem.
  - Sorbent media were mostly carbon-based materials or functionalized silica/clays.
  - All batch experiments were performed in using ACW and actual EFPC water
- Evaluated materials: *Biochar (PBC); Sorbster (eSorb); Si-thiol (Si-SH); Mackinawite blended powdered activated carbon (fsPAC); Powdered activated carbon (nsPAC); Organoclay PM-199 (Q-Clay); Filtrasorb 300 (F300) and RemBind (eBind).*





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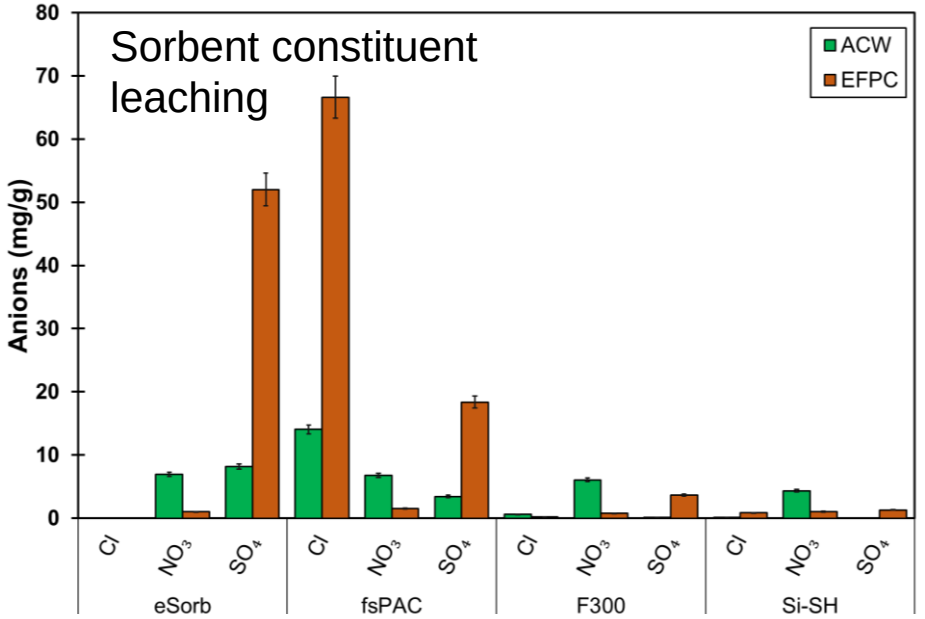
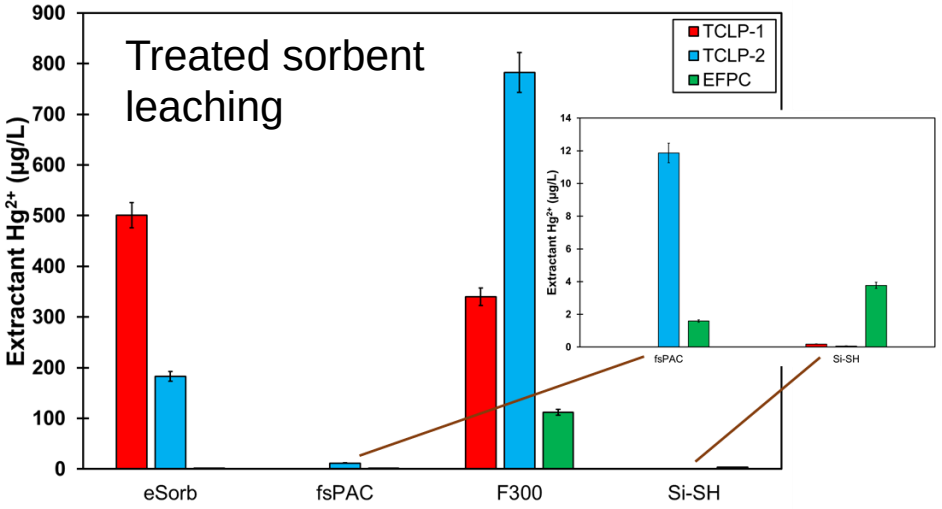
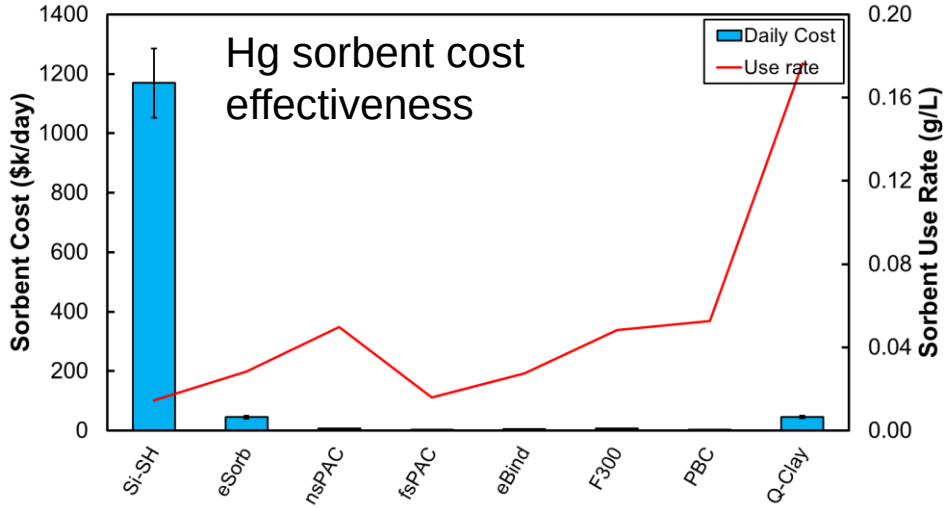
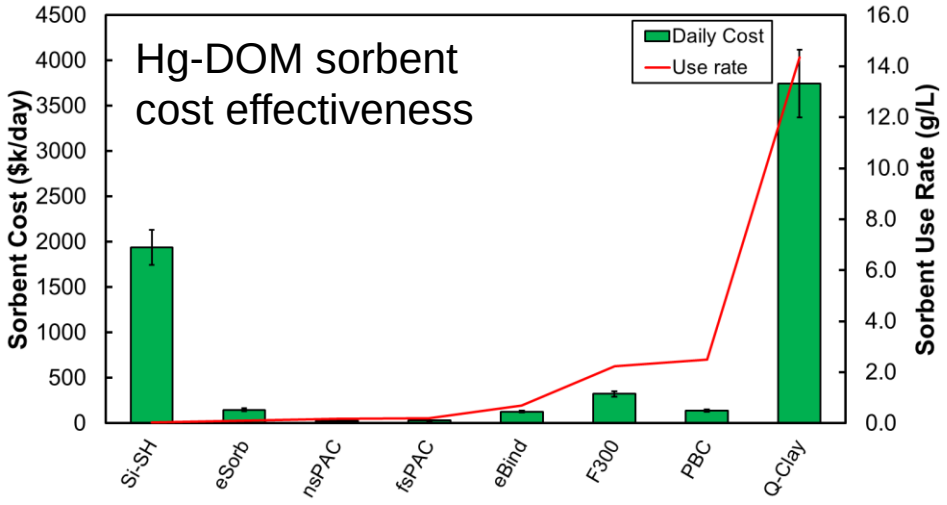
Task 7: Engineered Multi-Layer Amendment Technology for Hg Remediation on Oak Ridge Reservation

FIU Year 3 Research Highlights & Accomplishments:









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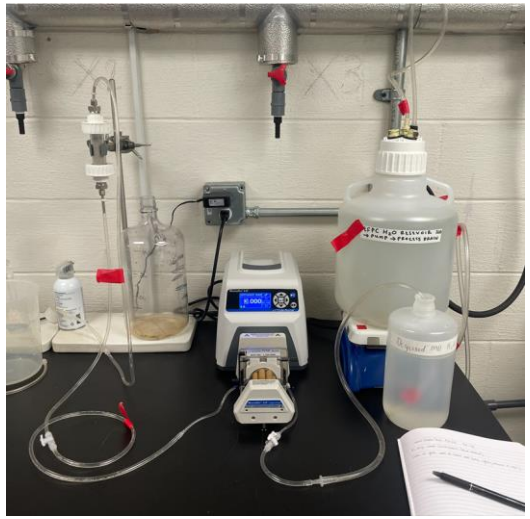
Task 7: Engineered Multi-Layer Amendment Technology for Hg Remediation on Oak Ridge Reservation

FIU Year 3 Research Highlights & Accomplishments:

- Poster presentations at the 2023 Waste management symposium.
- Caridad Estrada, the DOE Fellow supporting this subtask gained admission to Princeton University to conduct doctoral studies in Environmental engineering.

FIU Year 4 Projected Scope

- Determine physiochemical properties of Hg sorbents upon contact with environmental media (DI Water, ACW, EFPC water) and characterize pristine and treated sorbents using microscopy and spectroscopy techniques.
- Conduct comprehensive column studies with select sorbent media to elucidate mercury sorption under conditions representative of EFPC site.



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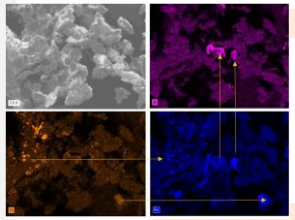
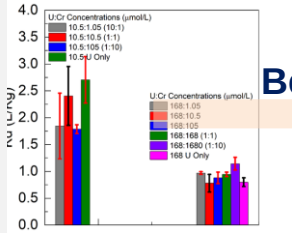
Technology Development and Deployment Road Map

Hanford

Technology	2020	2021	2022	2023	2024
In Situ Sequestration	 Science to Support Technology Deployment				Knowledge transfer to support eval. of treatment options of select zones in Central Plateau Subsurface (200-DV-1 Treatability Studies)
	Bench-scale testing	Bench-scale testing	Bench-scale testing	Bench-scale testing	
Natural Attenuation					Knowledge transfer to support Modeling for No-Treatment or MNA Scenario across multiple locations on Central Plateau
	Bench-scale testing	Bench-scale testing	Bench-scale testing	Bench-scale testing	
Glass Waste Forms					Bench-scale testing via PCT Knowledge transfer to support PA of WRPS Field Lysimeter Test Facility and ILAW glass testing programs; DOE-EM R&D Roadmap
	Bench-scale testing via SPFT	Bench-scale testing via SPFT	Bench-scale testing via SPFT/ PCT	Bench-scale testing via SPFT/PCT	

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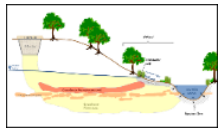
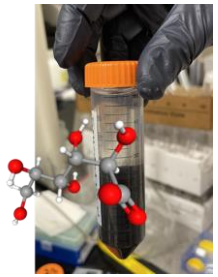
Technology Development and Deployment Road Map

Technology	2020	2021	2022	2023	2024
SRS/SRNL	Environmental factors controlling contaminants at SRS 	Fundamental Science to Support Technology Deployment for SRS Area Completion Project			
	Bench-scale experiments	Bench-scale experiments	Bench-scale experiments	Bench-scale experiments	Knowledge transfer for long term monitoring
	Evaluation of Humic Acid for uranium sequestration 				
	Bench-scale experiments	Bench-scale experiments	Bench-scale experiments	Bench-scale experiments	Knowledge transfer for long term monitoring
Mercury ORNL					
	Conceptual Design of Technology	Bench-Scale Testing	Bench-Scale Testing	Bench-Scale Testing	Knowledge transfer to support tech. selection for Hg remediation at ORR
WIPP					
	Bench-Scale Testing	Bench-Scale Testing	Bench-Scale Testing	Bench-Scale Testing	Knowledge Transfer to support site safety & performance asses sment for WIPP

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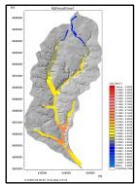
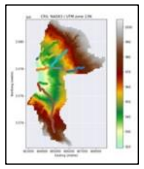
Technology Development and Deployment Road Map

Technology	2020	2021	2022	2023	2024
SRS		Tims Branch Model: Integrated SW/GW Model Calibration	Sed. Transport Model Calibration/ Validation	Contaminant Transport Model Calib./Validation	Model Implementation/ Hand-over	
		Fourmile Branch Model: Conceptual Model Development	Conceptual Model Development	Integrated SW/GW Model Dev. & Calib.	Sed./Contam. Transport Model Calib./Validation	Model Implementation/ Hand-over
WIPP (Basin 6)		Basin 6 DEM: Aerial Survey & DEM Dev. Methods Assessment	Basin 6 Aerial Survey & DEM Development	Basin 6 DEM Hand-over & Implementation in ATS Model		
			Implementation in model			
		WIPP (Basin 6) Model: Numerical Model Assessment	ATS Basin 6 SW/GW Model Training & Development	ATS Basin 6 SW/GW Model Training & Development	ATS Model Calib./Validation	Model Implementation/ Hand-over
				Implementation in model		
				WIPP (Basin 6): Field observation & data analysis	Field observation & data analysis	Hand-over

*DEM: Digital Elevation Model, SW: Surface water, GW: Groundwater

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Thank You. Questions?