PNNL-SA-59826



Multi-Scale Mass Transfer Processes Controlling Natural Attenuation and Engineered Remediation: An IFC Focused on Hanford's 300 Area Uranium Plume

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http://ifchanford.pnl.gov

ERSP PI Meeting April 7-9, 2008 Pacific Northwest National Laboratory U.S. Department of Energy

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Hanford IFC

Science Theme ~ Multiscale mass transfer processes influencing sorbed contaminant migration





Associated Practical Issues

- 1. Accurate projection of dissipation times for groundwater plumes of sorbing contaminants
 - Sorbing solutes not equal
 - Concentrations at different scales
- 1. Optimal delivery of remediation reactants
 - Access
 - Kinetic formation and reaction
 - Persistence
- 2. Practicality and effectiveness of remediation



Hanford 300 Area in 1962





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300 A Waste Streams

- Sodium aluminate (to ~1956)
 - Dissolved AI cladding from rejected fuel assemblies
 - 15% NaOH, Density of 1.5
- Effluents from REDOX and PUREX process development (1944 – 1954)
 - Nitric acid solutions containing uranyl nitrate
- N-reactor fuels fabrication wastes (1978 1986)
 - Nitric acid solutions containing U and Cu
- Different grades of enriched U as well as natural and depleted U
- Primary chemical inventory in NPP and SPP
 - 37,000 65,000 kg of U; 265,000 kg of Cu



The 300-FF-5 Operable Unit

Locations of CERCLA monitoring wells (open circles) and wells instrumented for automated hourly measurements (purple dots)

- water levels
- temperature
- EC

IFC site selection strongly dependent on polyphosphate plume trajectory

- Model projections performed
- Continuous monitoring initiated around SPP to improve the hydrologic model



Field Site Location in WMA 300-FF-5



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Primary Objectives

- Quantify the role of mass transfer in controlling U(VI) distribution under various geochemical, hydrologic, and remedial conditions
 - Vadose zone
 - Saturated zone
- Investigate in-situ microbiologic processes that couple with mass transfer to control phosphate barrier performance and longevity
- Create enduring field experimental data sets for model and fieldscale hypothesis evaluation
- Test and improve existing models of multi-reaction chemistry and multi-scale mass transfer by comparison to new, robust experimental field data
- Proactively transfer results to site for decision making and remediation



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Approaches

- Robust 3-D geostatistical characterization of the experimental domain
 - Borehole samples and geophysics
 - geo-, hydro-, chemo-, bio-, and U(VI)-facies
 - Correlative transfer functions with key process-specific parameters
- Field experimental campaigns based on 3 hypothesis at an integrated vadose zone-saturated zone site
 - Well field sufficient to sample heterogeneities
 - Infiltration experiments in vadose zone
 - Passive river stage experiments in capillary fringe
 - Injection experiments in saturated zone
 - Collaborative experiments with EM-20

Modeling of different types

- Stochastic-deterministic
- STOMP, MODFLOW, and FLOTRAN by code originators
- STOMP as the integrative project code
- Leverage broad data base and other site activities
 - ERSD
 - EM-30, EM-20



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Geological Cross Section





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Seasonal Dynamics of 300 A Uranium Plume



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Vadose Zone Release Model





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FY08 Activities

Website development Website operation						
Permits and permissions	Carl Contes Tay and Contes Tay and Co					
Experimental site-selection, set-up, and infrast	tructure					
Design of well field and monitoring system	Well drilling					
	In-situ characterization					
Ex-situ characterization						
Field experi	iment infrastructure					
Evaluations and	testing of new characterization methods					
IFC- Data baae; Historic data, 5708, new data	(website linkage) new data					
Interpretational stochastic data (UCB)	(modeling plan) implementation					
Improvements in microscopic models and param	neter (w/SFA, EM-40, ERSP)					
	Rent Northwe					

Site Management Status

- Received approval from DOE Richland Operations for use of the 300 Area for the IFC May 2007
 - Cultural and ecological reviews completed August 2007
 - NEPA categorical exclusion granted September 2007
- Underground Injection Control permit submitted to Washington State Department of Ecology (Ecology) February 2008
- Controlling documents completed and available on web site (<u>http://ifchanford.pnl.gov/documents</u>)
 - Field Site Management, Health and Safety, QA/QC, Communications Plans
- Processing requests for sample materials submitted by ERSP investigators
- Washington State Waste Discharge Permit ST4511 in place for injections
 - Work with Ecology staff to ensure compliance of injections with permit (e.g. tracer concentrations



300 A IFC Infrastructure

- Site selection completed October 2007
- Drilling specifications document completed December 2007 and provided to Fluor Hanford (FH)
 - IFC wells located and staked
- Well drilling contract in Fluor Hanford Procurement
 - Excavation permit issued
 - Contract award by March 31, start date in April
 - Drilling pad constructed (March 31)
- Field office trailer and sample storage containers in place
- Power for field site in process



Criteria for Site Selection

- Seasonal changes in [U] ~ 2; [U]max > 50 ppb; no organic contaminants
- Proximate to previous excavations for which significant laboratory and characterization data exists
- Maximal amount of fines in saturated zone
- Near but out of the zone of influence of the EM-20 polyphosphate injection experiment
- Saturated zone thickness (Hanford formation) of 2-3 m
- Relatively flat Hanford-Ringold contact to minimize vertical gradients
- Located within coverage domain of existing groundwater monitoring domain
- Site location (and experiment timing) to allow relatively slow and predictable travel times



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Surface Geophysical Surveys

- Involved data collection using range of instruments —hand carried or in contact with surface
- Quantitative spatial information to characterize features controlling transport
 - Sedimentary (and other) facies
 - Soil type (surface charge, CEC)
 - Pore size distribution
 - Hydraulic characteristics
- Quantitative temporal information to characterize flow and transport processes
 - Pore-water conductance
 - Temperature







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Geophysical Lines for Initial Characterization and Site Selection





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IFC Experimental Site- South Process Pond



Hanford -line2

Relatively Uniform Hanford-Ringold Contact





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Shaded Relief of Hanford Ringold Contact





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Seasonal Changes in Groundwater Flow Vectors



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Layout of Hanford 300 Area IFC Well Array





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Surveyed and Staked IFC Well-Field Awaiting Drilling





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Drilling, Sampling, and Well-Completion Information for 35 New IFC Wells

New IFC Wells

Туре	# Wells	Preferred Drill Method	Total Depth (ft)	Borehol Diamete	e Screen r	Screen Interval (ft)	Proposed Sampling	Total # samples/well	Total # samples	Comments
ERT- instrumented/gw monitoring <i>without</i> core collection	18	cable tool or sonic	58	8"	4" PVC	31-56	Grab samples every 2 ft	29 grab samples'	f 464 grab samples*	sandpack below 10' depth; one well to be used as saturated- zone injection well
ERT- instrumented/gw monitoring <i>with</i> core collection	7	sonic	58	8"	4" PVC	31-56	4 holes continuous core in 0.5 ft lexan liners within 5-ft long, min. 4-in ID split spoor;	12 split spoons of 58, 1-ft lexan liners;	84 split spoons or 406, 1-ft lexan liners;	s continuous core (min 4" OD); sandpack below 10' depth
							3 holes collected in 2-ft long sections of lexan, approximately the same diameter as the core barrel OD, at surface (i.e. grab samples)	29, 2-ft core samples	203, 2-ft core samples	
3-well cluster (multi-level) gw monitoring	3 clusters	cable tool or sonic	37, 46, 57	8"	4" PVC	30-35, 42-44, 53-55	Grab samples every 2 ft	66 grab samples/cluster*	198 grab samples*	ERT on deep (56 ft) well only (sandpack below 10' depth)
Deep characterization; gw monitoring	1	sonic	~180	8"	4" PVC	60-140	~60 ft of core collected in lexan liners, from five intervals (30-35', 50-70 ', 95- 100', 122'-132 ft', and 170 ft to TOB); grab samples every 2 ft between core runs		Up to 12, 5-ft split-spoon segments; 60 grab samples	Continuous screen to test groundwater across redox boundaries in Ringold Formation
Total wells	35						*collected at surface by emptying core barrel into 5 gal. buckets or capped lexan liners (if collected by the sonic method use 2-ft long sections of lexan, approximately the same diameter as the core barrel OD)			



As-Built Diagram for ERT-Instrumented, Groundwater-Monitoring Wells (25*)





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As-Built Diagram for Multi-Level, Groundwater Monitoring Well Clusters (3X3)





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Schematic of ERT / Monitoring Wells



- Electrodes spaced at 60 cm (2 ft)
- Electrode length approx 10 cm (4 in)
- Electrode material 316 stainless steel
- Single wire connections to electrodes
- Wires run on outside of PVC well pipe
- Thermistors placed between electrodes
- Wire wrap PVC from 106-98 m elevation
- Tube capped at bottom
- Well head ~0.6 m (2 ft) above ground
- Central connector/DAQ box at top of wellhead
- Heat dissipation unit (HDU), timedomain reflectrometry (TDR) probe and porous cup solution sampler at multiple depths on 5 wells around infiltration site



Monitoring Instrument Calibration and Well Assembly has Begun to Support April-May Installation



Thermistor calibration

Templates established for each well type defining spacings and locations for thermistors and ERT electrodes



- QA/QC plan defines calibration needs, testing requests, and documentation.
- All monitoring equipment has been received. Testing and assembly underway for ~1 month.
- Thermistor and ERT electrode string, and their inwell locations require pre-assembly for rapid field deployment.



Installation of Thermistors and ERT Electrodes on Well String





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Issues of Vertical Resolution in Groundwater/Plume Monitoring

- Vertically resolved GW sampling difficult because of travel times and water volumes involved
- Given regulations and sediment properties, individual wells completed over distinct depth intervals are required for ML sampling. Three of these are included in the IFC well field.
- Degree/impacts of vertical heterogeneity (K, properties, etc.) will be assessed in characterization activities by seasonal
 - EBF measurements (all wells)
 - MLS geochemical sampling (~6 wells or more if necessary) (poster)
 - Cold/warm river heat tracer studies (poster)

Large water quality variations monitored by ERT



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Multi-Level Sampler (MLS) for Depth-Discrete Groundwater Sampling





- The MLS for 4" wells uses tiers of diffusion cells to collect depthdiscrete water samples every 2"
- Baffles "seal" well bore every 12"
- 8 h required for equilibration
- Sorbents and other materials can be added



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MLS Results for Hyporeic Zone Near 100D





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Defining Heterogeneities in Hydrologic Properties and Flow



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The Hanford IFC Experimental Domain



- A geostatistical model that correlates U concentration, reactivity, facies properties, and permeability will be established from geophysical logging and direct measurements.
- Injection experiments of ~1.0 x 10⁵ gallon will be performed in the 6 m saturated zone under different seasonal gradients.
- Passive experiments will exploit natural gradients
- Continual water level monitoring at 12 locations to provide necessary hydrologic linkages.



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IFC Experiments

U(VI) fluxes from the vadose zone

- Scale-dependent mass transfer, geochemical kinetics (adsorption/desorption) and water pathway effects on U(VI) fluxes to groundwater
 - Infiltration experiments with varying water application rates, volumes, and composition (pH, HCO₃, Na/Ca)
 - Passive experiments to explore rising and falling water table effects on U(VI) solubilization and release from lower vadose zone
- U(VI) concentration dynamics within the groundwater plume
 - Scale-dependent mass transfer involved in forward (adsorption), backward (desorption), and steady-state (isotopic exchange) reaction processes in flow paths with different trajectories and residence times
 - Injection experiments with varying HCO₃ and U(VI) concentrations, and U(VI) isotopic ratios
 - Passive experiments follow vadose zone pulses, or inland riverwater groundwater gradients
- Optimized and sustained remediation strategies
 - Evaluate role of mass transfer and microbiological processes on different forms of phosphate used to precipitate and immobilize U
 - Injection experiments with polyphosphate, Ca-citrate/PO₄³⁻, organic P with HCO₃
 - In collaboration with EM-22 and team



Characterization ~ At Least a 2 Yr Effort

Objective is a 3-D hydrogeostatistical model of domain and identification of critical scales

- Tiered approach through first 4 injection experiments
 - Described through plan
 - Maximize useful knowledge gain
 - Support a field-scale reactive transport model
- Based on experimental and interpretational need
 - Obvious beginnings; augmented as knowledge expands
 - Focused on formation, facies, and pore scales

<u>Types</u>

- Physical, hydrologic, geochemical, microbiologic
 - In-situ (borehole, aquifer)
 - Ex-situ (laboratory)



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Basis for Characterization – A Field Scale Reactive Transport Model





Initial Characterization (Tier 1)

Vadose zone characterization interval (13'-25')

- 5 sampling points/well location for Tier 1 lab properties (~30 samples)
- 15 samples for θ/K relationships

Saturated zone characterization interval (25'-56')

- 6 sampling points/well location for Tier 1 lab properties (~174 samples)
- Electromagnetic borehole flowmeter (EBF) measurements every 30 cm
- Limited constant rate injection tests (~8 wells)
- 8 h passive MLS deployments in 6-8 wells (@ 2 seasons)
- Non-reactive tracer experiments (salt, temperature)

Tier 1 lab properties

 $\rm U_{T},\, \rm P_{E},\, \rm P_{M},\, psd,\, pvd,\, \rm K_{sat},\, \rm K_{\Theta}$



Weak-Acid Extractable U(VI) from C5708

Formation	Notes	Depth (ft)	U(μg/g)
BF		4.5-7	3.4
HF	vadose zone injection depth	15-17	1.03
	, , , , , , , , , , , , , , , , , , ,	20-22	1.24
		23.5-26	3.09
	smear zone	28-31.5	5.17
	water table	32-33	3.29
	upper screen 🛓	33.8-36.8	0.99
	middle screen	40.8-42.8	0.93
	÷	45-47	0.64
	lower screen	49.5-51.5	0.56
RF	C.W. upper screen	55-56	1.43
	C.W. lower screen	73-75	0.58
		74-76	0.57
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U(VI) Adsorption Isotherms on Uncontaminated 300 A Vadose Zone and Aquifer Materials from LFI Cores



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Well Logs and Correlations

Gamma logs

- Spectral gamma (K-40) and total gamma correlate with fines (%silt+clay, grain size.). Can be used to estimate hydraulic properties. Ratios of K-U-T may correlate with mineralogy and lithology.
- Neutron moisture logs
 - Correlated with water content and texture in the vadose zone (not useable for saturated zone). Used with pedo-transfer functions and scaling methods to estimate hydraulic properties and zones of fine and coarse-textured sediment.
- Lithodensity/Porosity logs
 - Correlate with wet density, which depends on porosity or water content. If water content is known, density can be used to estimate porosity. Variations in lithodensity below water table correlate with porosity..

Induction and Resistivity logs

 Borehole resistivity logs provide "point" measurements for vertical control and improved use of cross-hole ERT measurements

Combination logs

- Easier to delineate facies with multiple logs.
- Contributions of fluids and solids to bulk resistivity can be determined if resistivity and lithodensity/porosity logs are available.
- Combination of resistivity, neutron moisture, lithodensity/porosity measurements can allow for unambiguous determination of porosity.



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Observed and Predicted (from gamma logs) Porosity and Hydraulic Conductivity Profiles



Evolving Project Data Base

- Uses Google Map API (v2) to provide lightweight GIS access to data. Has aerial imagery of 300-FF-5 as optional underlay.
- Allows rapid oversight and access to data
- Currently being coupled to back end (HEIS, water level data) and new project-specific databases
- Primary data sets have been organized in relational databases (borehole data, geophysics, and chemical monitoring data).
- Prototype (but operational) interfaces exist allowing user to access some data.





Deep Characterization Borehole

- Approximately 150' in depth to sample water-saturated Ringold Formation
- Major focus is microbiological characterization (aspectic sampling)
- To be drilled and analyzed in collaboration with (shared support from) evolving PNNL SFA focused on microenvironments and transition zones
- Located along east margin of IFC experimental site
- Will allow passive biogeochemical and mass transfer studies within Ringold transition zones
- Possible evaluation of diffusive mass transfer from the Ringold as a U source



As-Built Diagram for the Deep Characterization Well





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Microbiological Characterization of IFC Sediments

- IFC to support core collection, logging, and well installation. PNNL SFA to support microbiologic characterization and study.
- Enable development of microbial component of IFC research

Objectives

- Determine biomass and phylogenetic diversity
- Metal reduction/oxidation behavior and chemoheterotrophic catabolism
- Numbers and functional diversity of key organism groups that influence U or Tc
- Functional potential of key microbial isolates to transform U and Tc and involved mechanisms
- Basis for understanding biogeochemical processes across transition zones that collect contaminants



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Transition Zones in the 300 A Unconfined Aquifer



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Microbiologic Characterization of Deep Borehole Sediments

Microscopic

- Direct counts (w SYTO and PI) and active cells (TDR)
- Culture independent
 - Phylogenetic diversity/richness
 - Real time PCR for specific functional groups
 - Phylo & functional gene arrays (Geochip)
- Culture dependent
 - Liquid MPN's with various TEA's
 - Filter based cultivation
 - Isolation & characterization of representative cultures
- Activity analysis
 - Targeted incubations w/ radiolabeled substrates



Major IFC Research Objectives in FY 09

- Complete and implement multi-investigator IFC modeling plan
- Perform two temperature tracer studies
- Complete MLS well characterization, and initiate passive biogeochemical experimentation during periods of head change
- Integrate geologic, geophysical, and hydrophysical characterization measurements of various kinds into site geostatistical and hydrologic models
- Begin assembly of site U(VI) reaction model based on IFC, SFA, ERSD, and RACS/EM40 research
- Perform two U(VI) injection tracer experiments (+U, -U, high/low water)
- Begin development of collaborative IFC/SFA microbiologic research plan based on results from the HRDCB
- Initiate publication program



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Materials Available for ERSD Researchers

Existing

- Historic, highly contaminated sediments from process ponds (small masses available)
- Excavation samples from the vadose zone at two locations in the North Process Pond (NPP) and two locations in the South Process Pond (SPP). Samples contain various U(VI) speciation states, including adsorbed, precipitated, and surface complexed phases (variable sample masses are available).
- Uncontaminated vadose zone and aquifer sediments from the EM-40 Limited Field Investigation (LFI)
- Low-level contaminated samples from SPP C5708

To be Collected in Jan - Feb

- Becker-hammer grab samples from monitoring well installation in the Hanford formation screened to < 2 cm (~ 100 samples saved for ERSD researchers)</p>
- 4" continuous sonic core samples from 7 characterization boreholes in Hanford sediment (~ 75-100' saved for ERSD researchers)
- 4" continuous sonic, aseptic core samples from one -150' characterization borehole through the Hanford and Ringold formations (select sample aliquots and undisturbed cores will be saved for ERSD researchers)

<u>Other</u>

An excavation will be opened below backfill (~15 ') to allow bulk sample collection and in-situ structural analysis



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Example Opportunities for Collaborative Research

- In-situ adsorption/desorption experiments of various types
- Laboratory to field comparisons
- Evaluation of geophysical methods and inversion techniques
- Mass transfer processes of different types at different scales
- Microbiology of linked groundwater-river systems of low to high transmissivity
- Geologic, hydrologic, geochemical, and biogeochemical modeling of different types
- Microbiology and geochemistry of phosphate amended systems



EM-20 Polyphosphate Treatability Test in the 300 A Uraium Plume

Objectives

- Evaluate the use of phosphate amendments for immobilization of U
- Identify implementation challenges
- Evaluate feasibility of full-scale
- deployment
- Activities
 - Bench-scale studies
 - Amendment formulations finalized
 - Phased treatment approach selected
 - Site specific characterization
 - Installation of well network
 - Hydrogeologic characterization
 - Hydraulic/tracer injection testing
 - Polyphosphate injection design
 - Development of local-scale flow and transport model
 - linjection volumes, rates, and chemical mass requirements
 - Polyphosphate injection test performed in June 07





Vermeul and collaborators (PNNL) Mike Thompson (DOE-RL)

General Findings

Initial groundwater performance monitoring data show mixed results

- Initial reduction in U concentrations to below MCL in most wells within a radial distance of 75 ft
- Limited Ca/PO₄ sorption/mixing and U concentration rebound indicates (apatite) mineral formation was limited
- Performance monitoring is ongoing
- Complex hydrogeologic conditions may limit technology
 - Excessive groundwater velocities (50 ft/d or more)
 - High permeability, coarse-grained formation
 - Unfavorable geochemical conditions (elevated pH and carbonate)
 - In situ technologies must be robust to account for field-scale heterogeneities



Future Plans for EM-20 Treatability Studies for the 300 A U(VI)-Plume

- ► Future research focus → development of a direct treatment approach for source zone contamination (infiltration through vadose zone)
- Schedule:
 - FY08 → Bench- and Intermediate-scale laboratory experiments
 - FY09 → Field-scale demonstration



Collaborations

<u>EM-40</u>

Remediation and Closure Science Project (RACS, Freshley) - Conceptual and numeric reactive transport models to support remediation

<u>EM-20</u>

Polyphosphate Treatability Studies (Vermeul) - Field scale remediation concept to reduce groundwater U(VI)<MCL

<u>ERSD</u>

"Role of Microenvironments and Transition Zones in Subsurface Reactive Contaminant Transport ". PNNL Scientific Focus Area (Bolton)

"Microscale Metabolic, Redox, and Abiotic Reactions in Hanford 300A Subsurface Sediments." (Beyenal)

"Geophysical Characterization and Monitoring Strategies for Quantifying Hydrologic Transport processes in the Hanford Hyporheic Corridor". (Slater)

<u>SBIR</u>

"A New High-Resolution Method for the Characterization of Heterogeneous Subsurface Environments: Providing Flow and Transport Parameters via the Integration of Multi-Scale HydroGeophysical Data." (Bussod)



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