



Pacific Northwest
NATIONAL LABORATORY

*Proudly Operated by **Battelle** Since 1965*

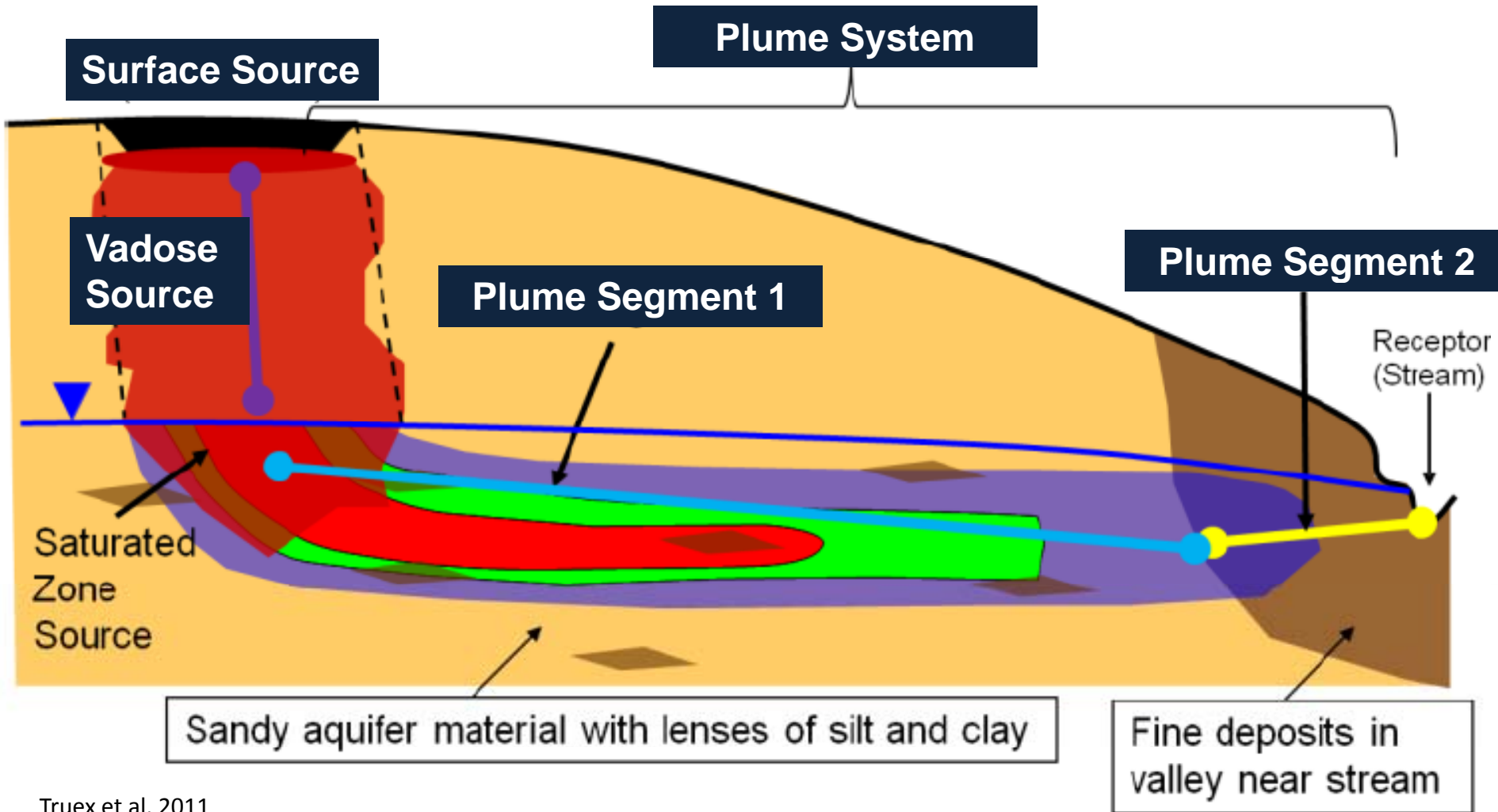
Characterization Approaches for Radionuclide-Contaminated Subsurface Sites

MICHAEL TRUEX

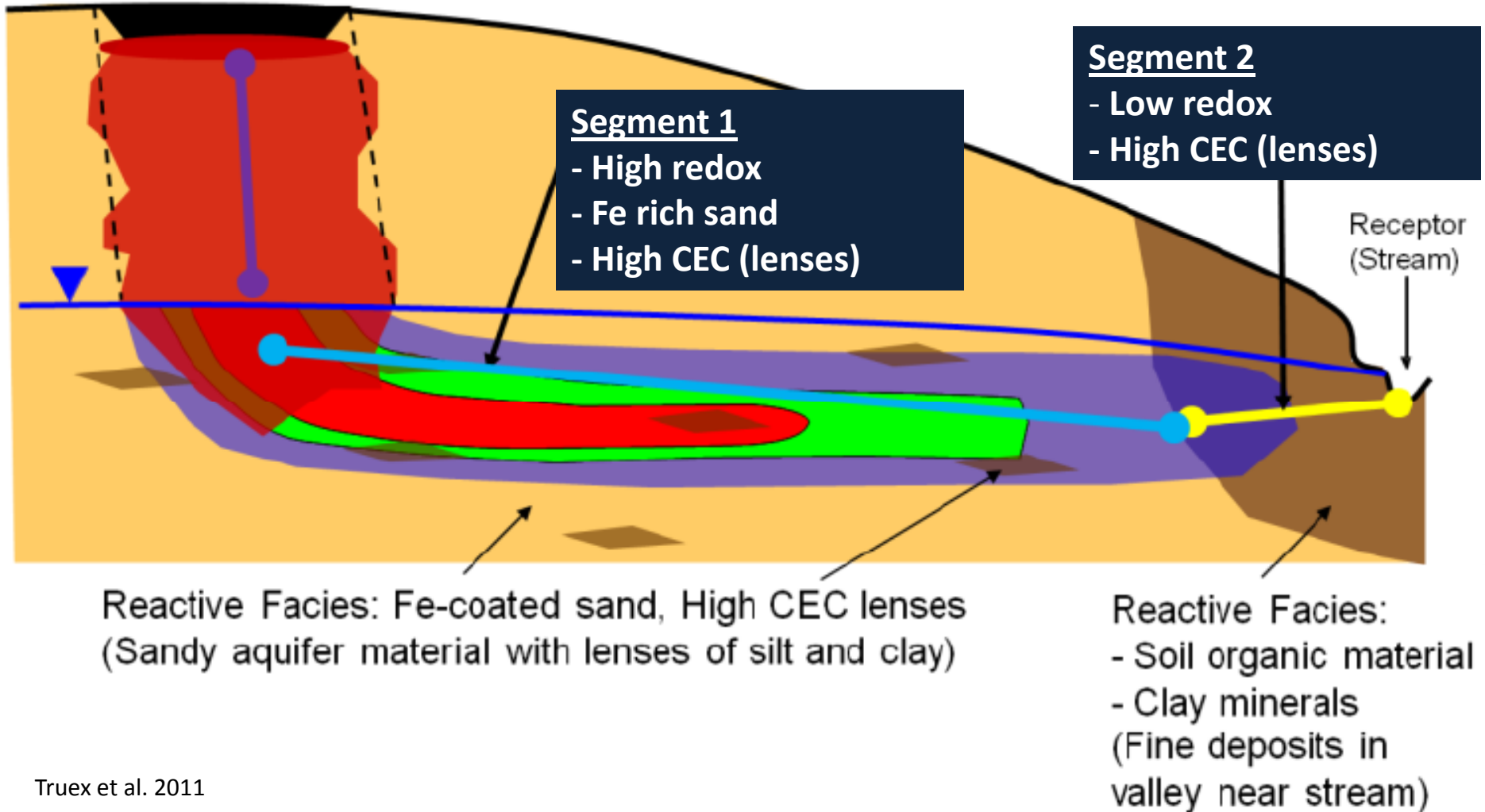
Pacific Northwest National Laboratory

- ▶ Conceptual model framework
- ▶ Characterization for factors controlling fate and transport and remedy selection
 - Vadose Zone
 - Disposal chemistry affects
 - Transport factors
 - Groundwater
 - Speciation and biogeochemistry
 - Secondary sources
 - Groundwater dynamics
 - Natural attenuation processes
- ▶ Conclusions

Conceptual Model Framework



Conceptual Model Framework



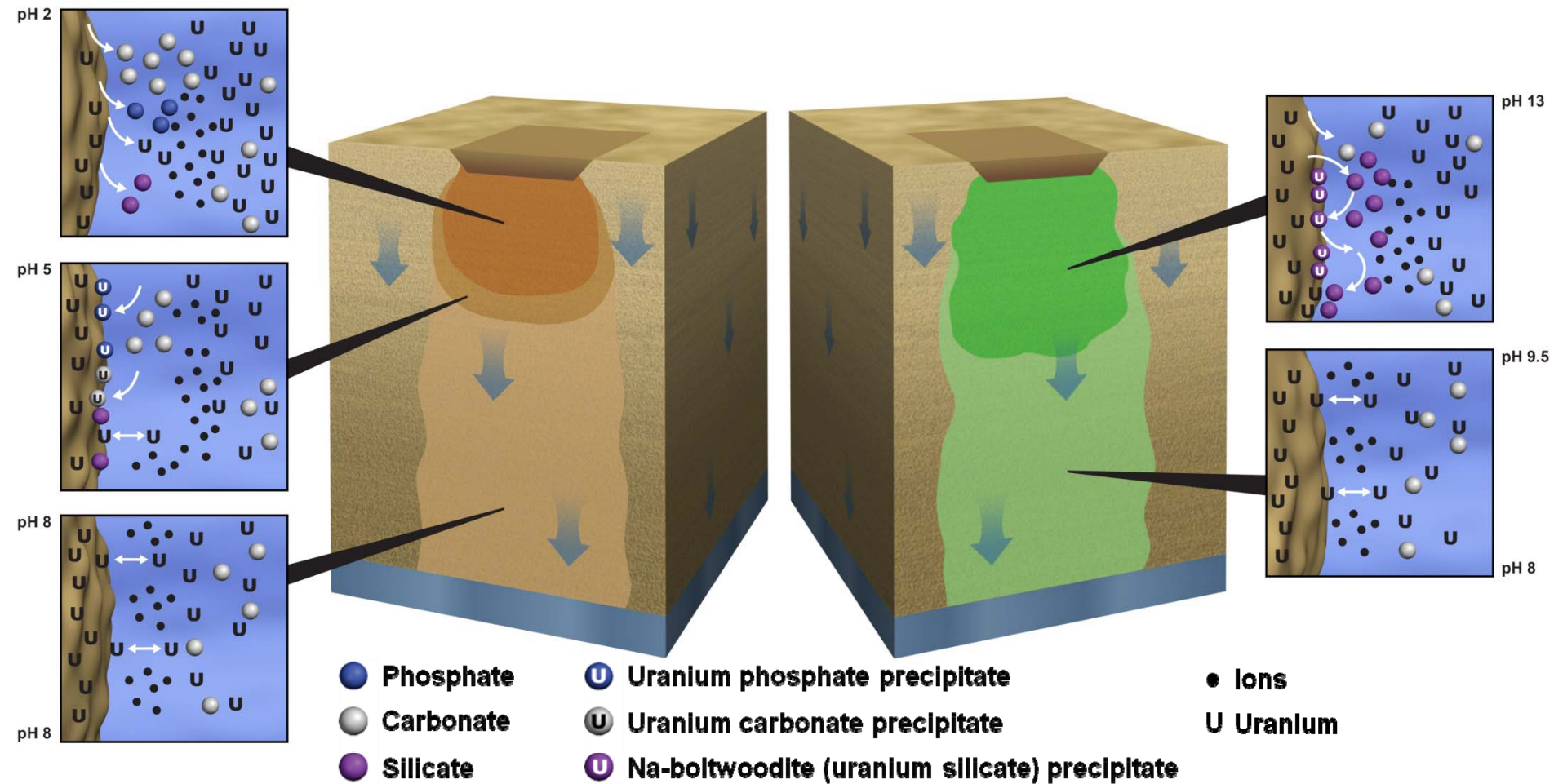
▶ Disposal Chemistry

- Co-contaminants and other characteristics of the disposed waste may impact transport for the contaminant of interest. These effects may be most intense near the disposal location (vadose zone).






▶ Transport Factors

- For surface waste disposal, transport of contaminants through the vadose zone affects the nature of the source to groundwater.

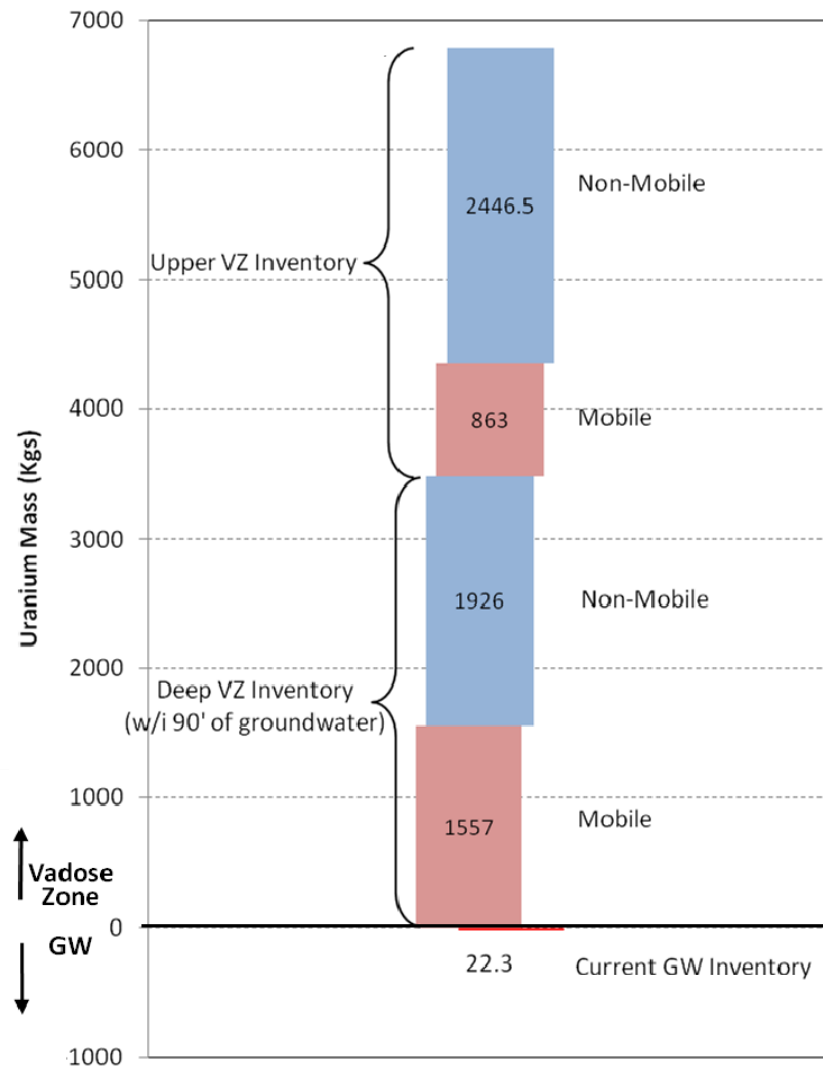
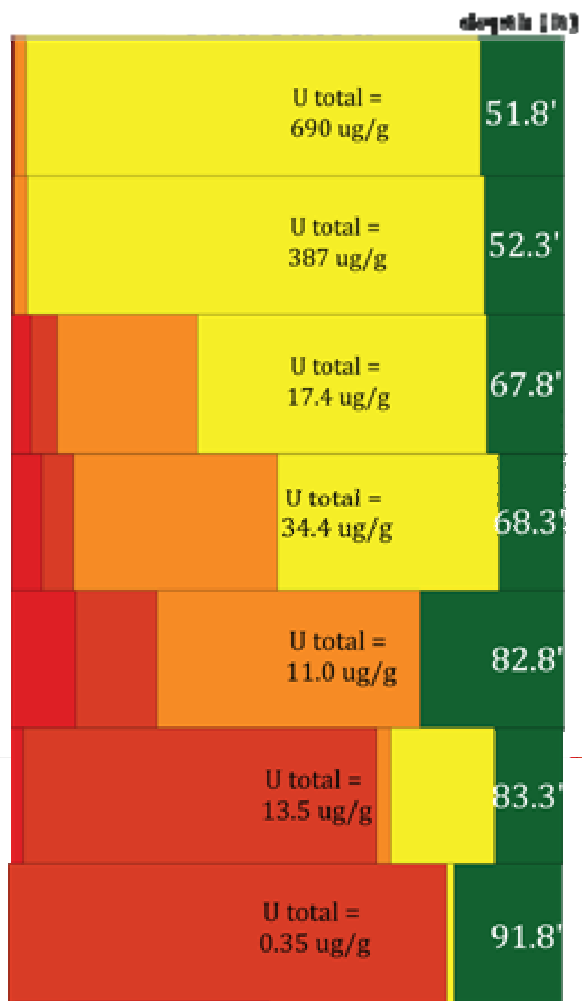
Disposal Chemistry



Disposal Chemistry

Extraction Solution	Hypothesized targeted sediment components	Interpreted uranium mobility of extracted fraction	Color Code
1. Aqueous: uncontaminated Hanford groundwater	Uranium in pore water and a portion of sorbed uranium	Mobile phase	
2. Ion Exch.: 1M Mg-nitrate	Readily desorbed uranium	Readily mobile through equilibrium partitioning	
3. Acetate pH5: 1 hour in pH 5 sodium acetate solution	Uranium associated with surface exposed carbonate precipitates, including uranium carbonates, or other readily dissolved precipitates	Moderately mobile through rapid dissolution processes	
4. Acetate pH 2.3: 1 week in pH 2.3 acetic acid	Dissolution of most carbonate compounds, including uranium carbonates, and sodium boltwoodite	Slow dissolution processes are associated with uranium release from this fraction such that uranium mobility is low with respect to impacting groundwater	
5. 8M HNO ₃ : 2 hours in 8M nitric acid at 95°C	Dissolution of most minerals expected to contain uranium, considered to represent total uranium extraction for this study ¹	Very slow dissolution processes are associated with uranium release from this fraction such that uranium mobility is very low with respect to impacting groundwater	

Disposal Chemistry



Serne et al. 2010

Evaluation of VZ Transport



Pacific Northwest
NATIONAL LABORATORY

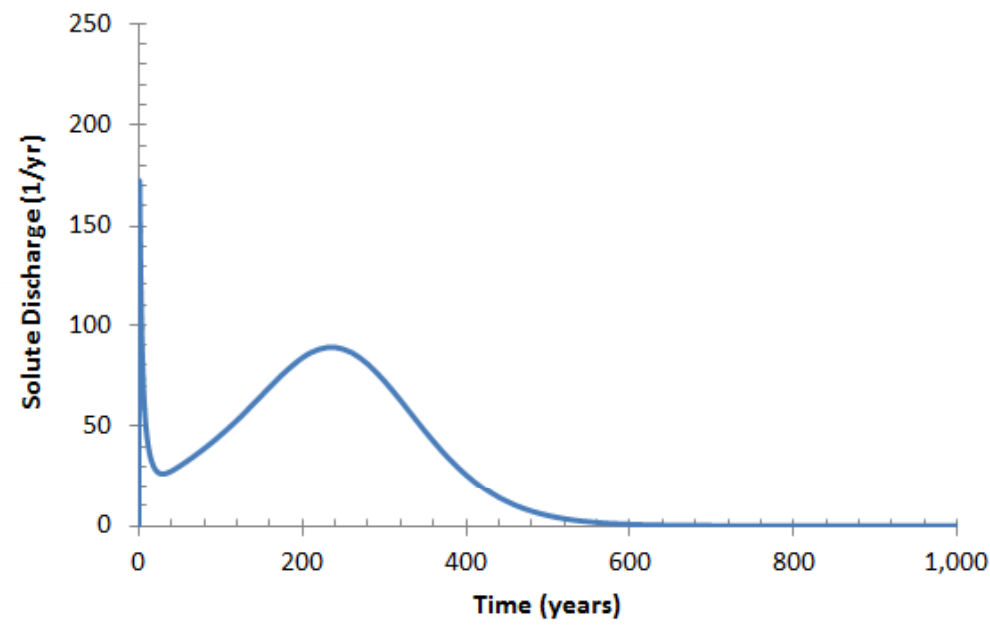
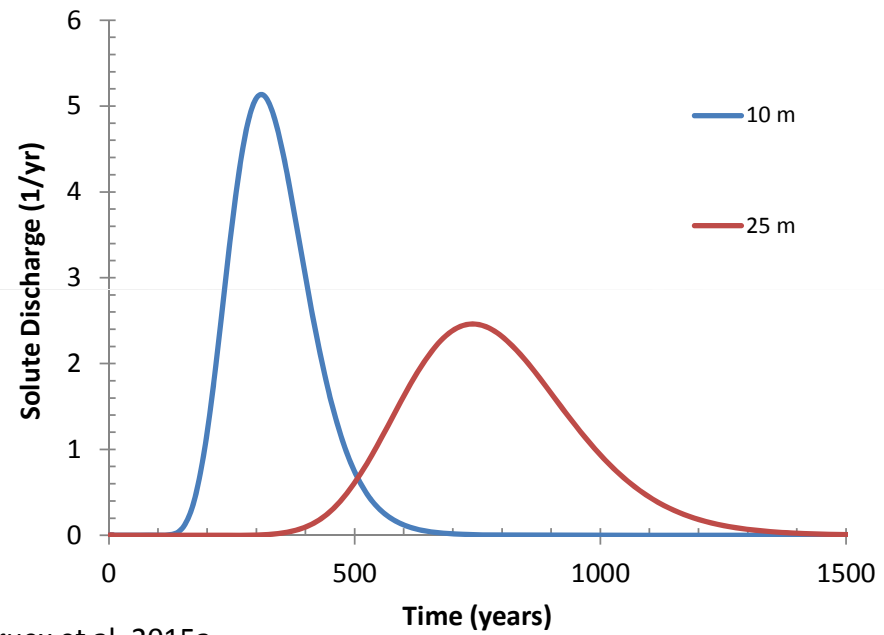
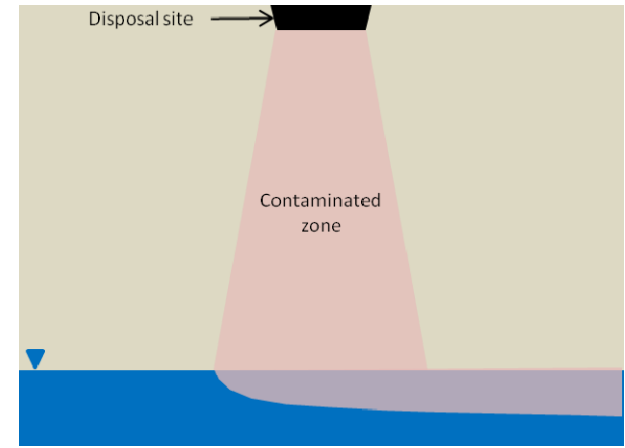
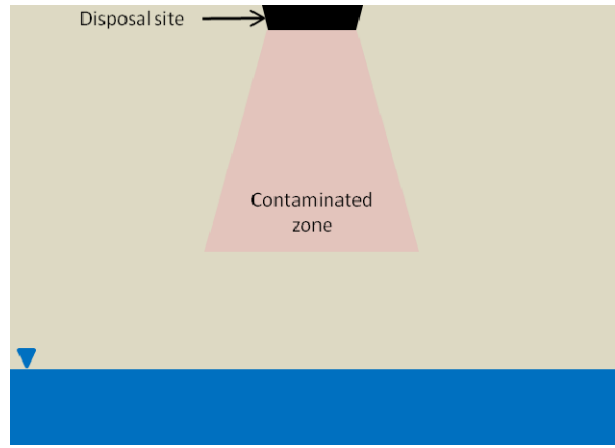
Proudly Operated by **Battelle** Since 1965

- ▶ There are characteristic behaviors that are useful in assessing the nature of contaminant transport from aqueous waste disposal/leaks to the vadose zone.
- ▶ There are two primary categories of transport behavior
 - Category I: small volume disposed compared to vadose zone thickness
 - Category II: large volume disposed compared to vadose zone thickness

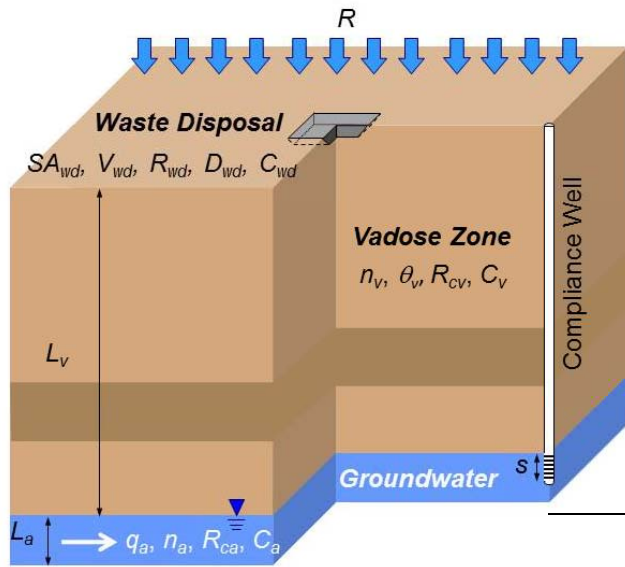


Category I

Category II



Analysis/Characterization Framework

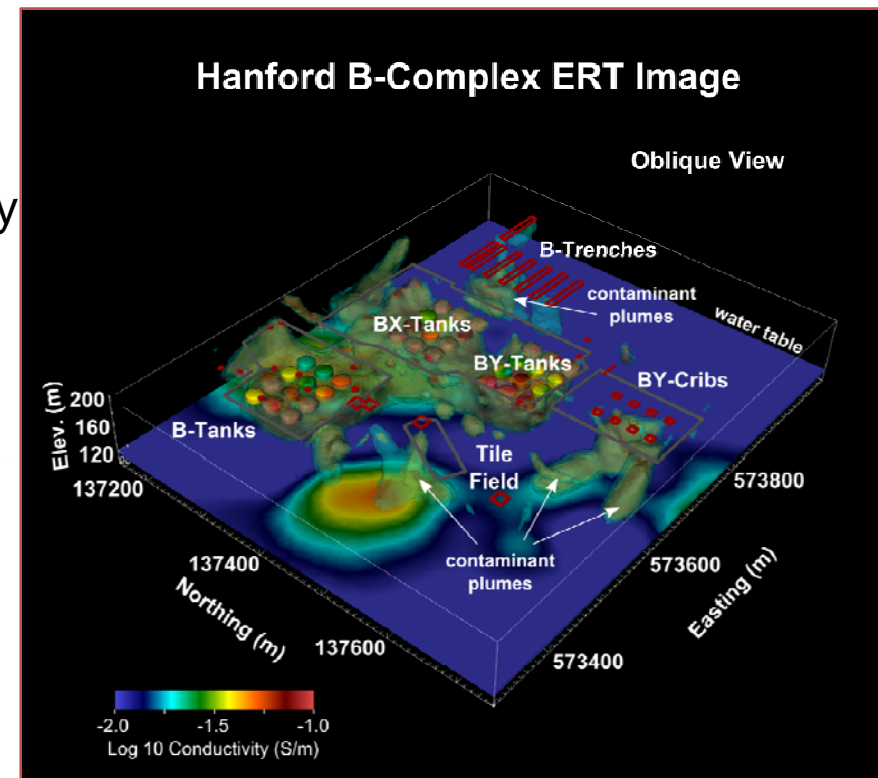


Vadose Zone Parameters	Waste Disposal Parameters	Groundwater Parameters
Thickness (L_v)	Aqueous volume (V_{wd})	Groundwater Darcy flux (q_a)
Recharge rate (historical, current, and estimated future rates) (R)	Disposed mass (M_{wd})	Contaminant mixing thickness in aquifer (L_a)
Porosity (n_v)	Rate of waste disposal (R_{wd})	Monitoring well screen length for compliance (s)
Contaminant retardation coefficient (R_{cv})	Contaminant concentration (C_{wd})	Porosity (n_a)
Current vertical distribution of contamination	Surface area of aqueous disposal (SA_{wd})	Contaminant retardation coefficient (R_{ca})
Moisture content profile (θ_v)	Acidity or alkalinity of the waste	
	Ionic strength and co-contaminants/species in the waste	
	Timing of waste disposal	

Evaluation of VZ Transport

▶ Contaminant Distribution Characterization Tools

- Geophysical logging
 - Spectral gamma log
 - Neutron moisture log
- Borehole sediment samples
- Geophysics
 - Electrical Resistivity Tomography



- ▶ Speciation and biogeochemistry
 - Characterization of a plume as ^{129}I can be augmented with speciation information to provide insight into mobility
 - Iodide and iodate have different transport characteristics

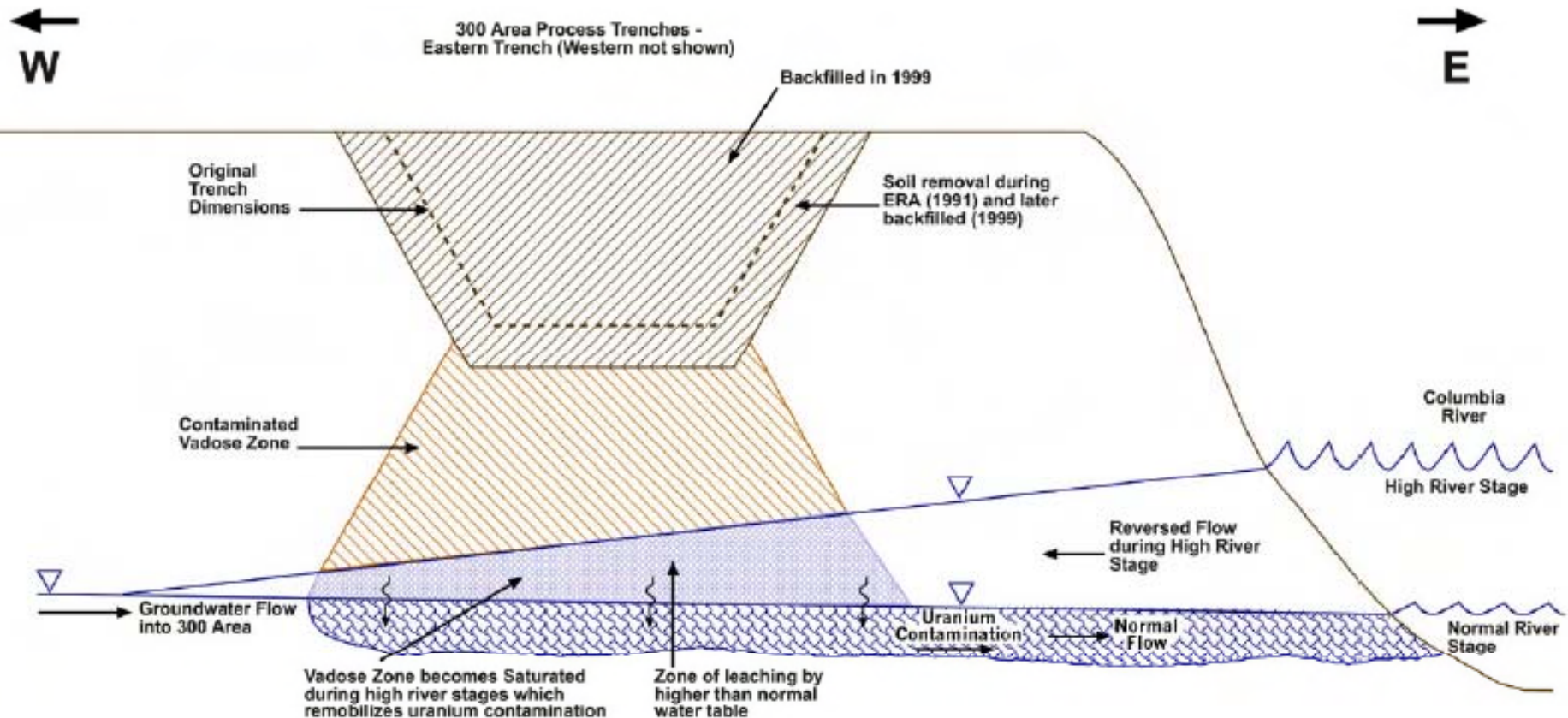
Composite Sediment ^(a)	Organic Carbon (%)	Inorganic Carbon (%)	Total Sediment Iodine ($\mu\text{g/g}$)	Total DOC ^(b) (μM)	Iodide-spiked K_d ^(c) (mL/g)	Iodate-spiked K_d ^(c) (mL/g)
H1	0.12	0.92	4.79	284 ± 33	0.08	1.78
H2	0.04	0.01	0.68	0	0.00	0.83
H3	0.15	0.18	2.10	94 ± 17	3.38	3.94

Xu et al. 2015

Truex et al. 2015b

Groundwater Elements

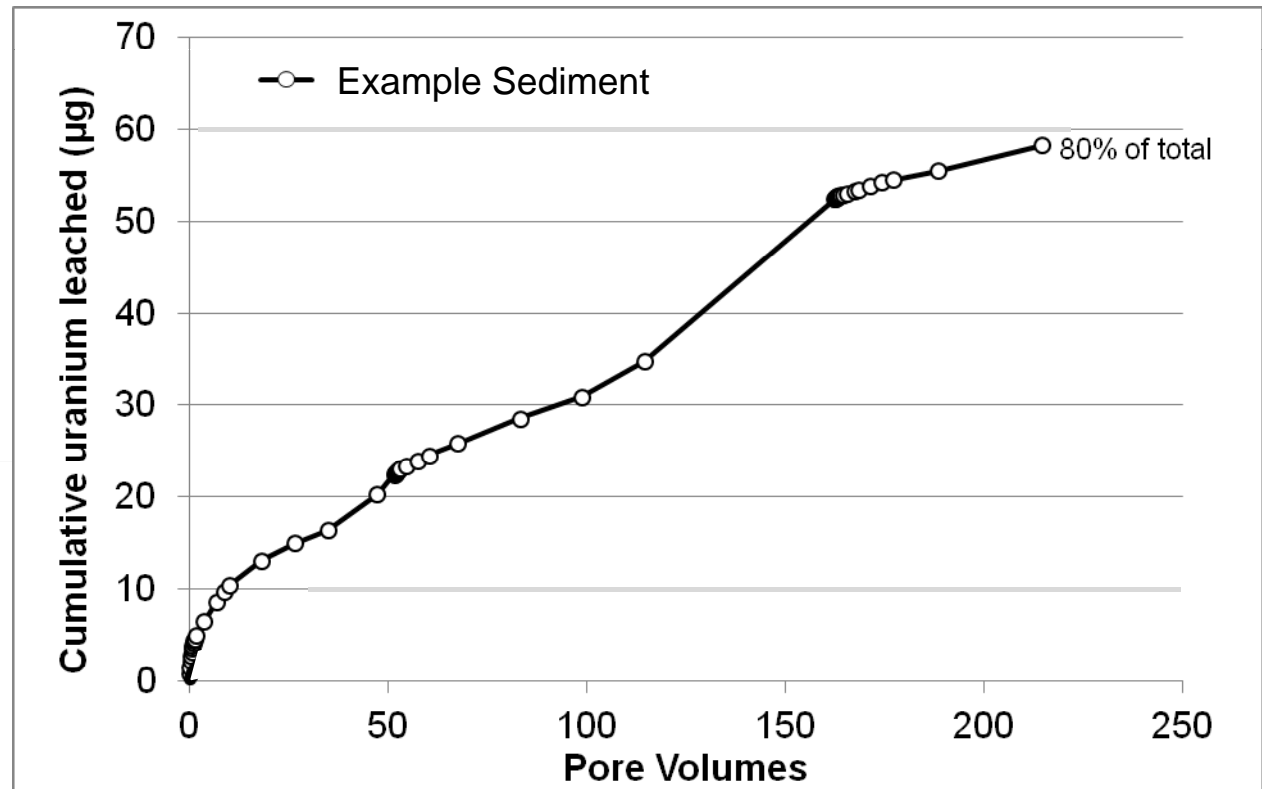
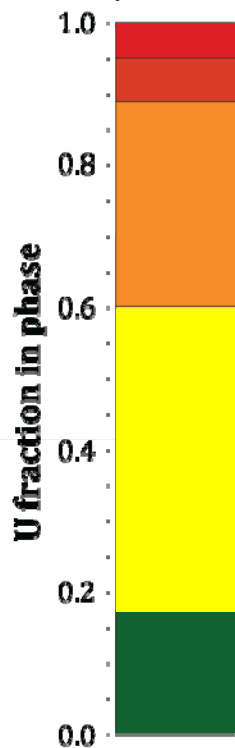
- ▶ Secondary sources (leaching)
 - Uranium sources related to periodic rewetting and leaching



▶ Secondary sources (leaching)

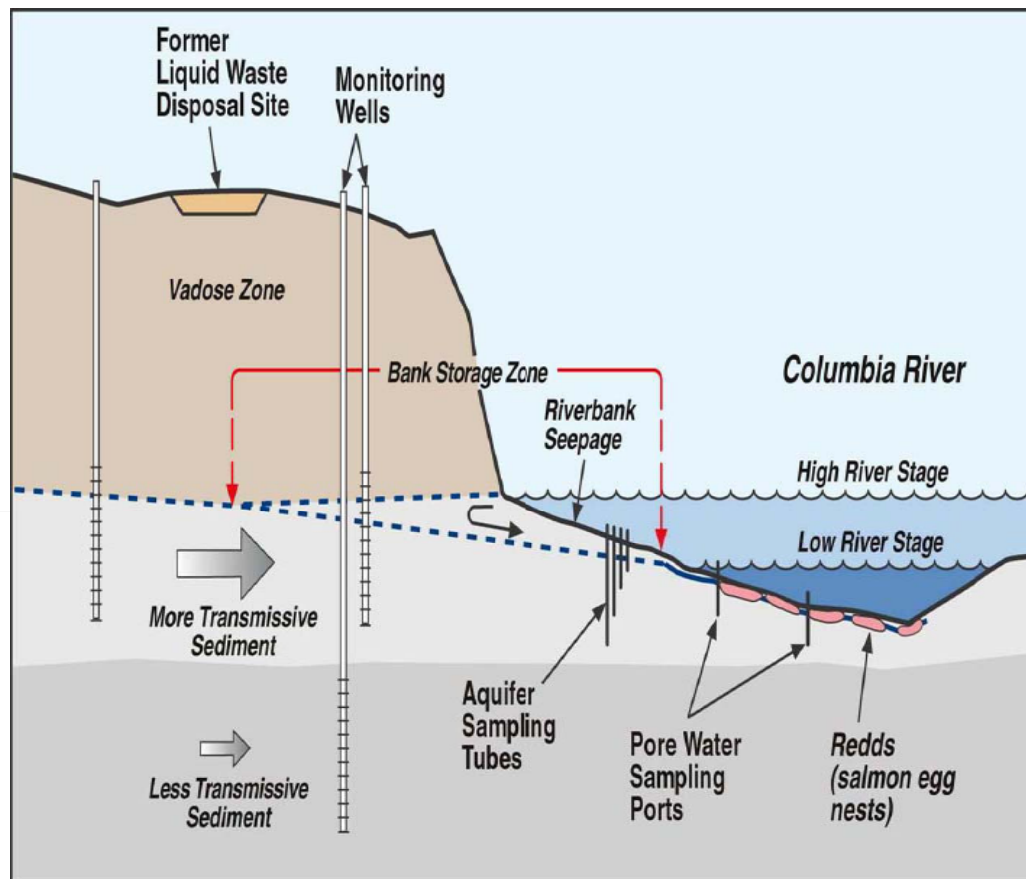
- Uranium sources related to periodic rewetting and leaching
- Relevant for other inorganic contaminants (e.g., I, Cr)

Example Sediment



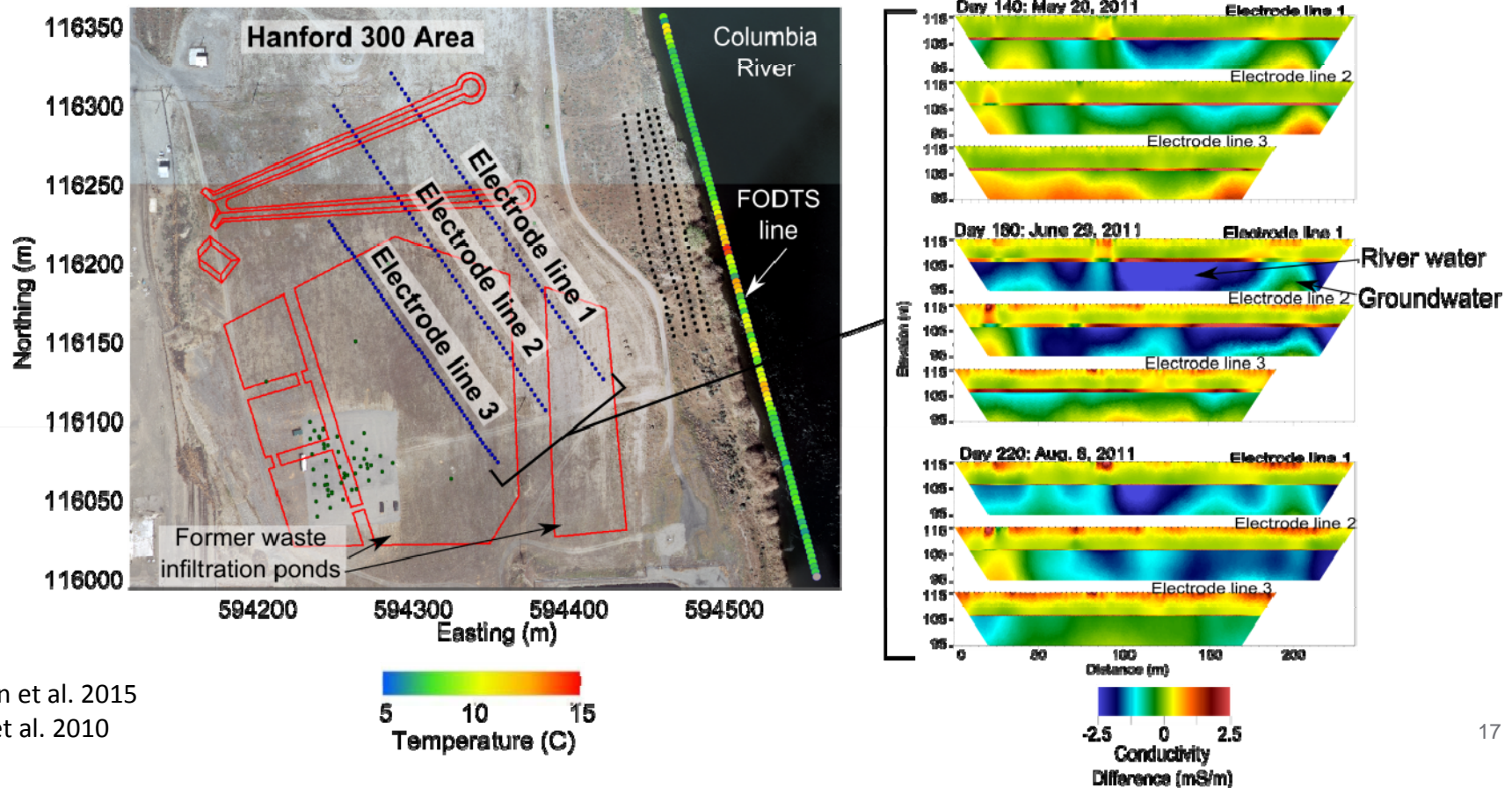
Groundwater Elements

- ▶ Groundwater dynamics
 - Hydrologic information can be augmented with geophysical techniques such as Electrical Resistivity Tomography



Groundwater Elements

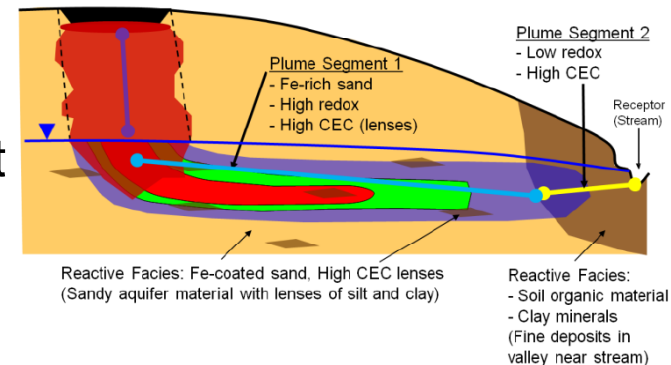
- ▶ Groundwater dynamics
 - Hydrologic information can be augmented with geophysical techniques such as Electrical Resistivity Tomography



▶ Natural attenuation processes

■ EPA MNA Technical Protocol

- “Scenarios” conceptual model document
- Sediment biogeochemical factors



■ Monitoring data

- Temporal data provides insights not possible with static data
- Natural or induced perturbations aid the interpretation of temporal data

Conclusions

- ▶ Conceptual model framework
 - Technical basis and organization of efforts
 - Communication
- ▶ Assess controlling factors
 - Fate and transport assessment
 - Technical focus
 - Characterization
 - Monitoring
 - Remediation

References

- ▶ Johnson, T., et al. 2015. Four-dimensional electrical conductivity monitoring of stage-driven river water intrusion: Accounting for water table effects using a transient mesh boundary and conditional inversion constraints, *Water Resour Res*, 51:6177-6196.
- ▶ Johnson TC, and DM Wellman. 2013. Re-Inversion of Surface Electrical Resistivity Tomography Data from the Hanford Site B-Complex . PNNL-22520; RPT-DVZ-AFRI-014, Pacific Northwest National Laboratory, Richland, WA
- ▶ Peterson RE, ML Rockhold, RJ Serne, PD Thorne, and MD Williams. 2008. Uranium Contamination in the Subsurface Beneath the 300 Area, Hanford Site, Washington . PNNL-17034, Pacific Northwest National Laboratory, Richland, WA.
- ▶ Peterson RE, and MP Connelly. 2001. Zone of Interaction Between Hanford Site Groundwater and Adjacent Columbia River. PNNL-13674, Pacific Northwest National Laboratory, Richland, WA.
- ▶ Serne R, et al. 2010. *Conceptual Models for Migration of Key Groundwater Contaminants Through the Vadose Zone and Into the Upper Unconfined Aquifer Below the B-Complex*. PNNL-19277, Pacific Northwest National Laboratory, Richland, WA.
- ▶ Slater, L. D., et al. 2010. Use of electrical imaging and distributed temperature sensing methods to characterize surface water-groundwater exchange regulating uranium transport at the Hanford 300 Area, Washington, *Water Resour Res*, 46
- ▶ Szecsody, J.E., M.J. Truex, N. Qafoku, D.M. Wellman, T. Resch, and L. Zhong. 2013. Influence of acidic and alkaline waste solution properties on uranium migration in subsurface sediments. *J. Contam. Hydrol.* 151:155-175.
- ▶ Szecsody, J.E., et al. 2012. Geochemical and Geophysical Changes During NH₃ Gas Treatment of Vadose Zone Sediments for Uranium Remediation. *Vadose Zone J.* 11(4) doi: 10.2136/vzj2011.0158.
- ▶ Szecsody, JE, et al. 2010. Remediation of Uranium in the Hanford Vadose Zone Using Ammonia Gas: FY10 Laboratory-Scale Experiments. PNNL-20004, Pacific Northwest National Laboratory, Richland, WA.
- ▶ Truex, MJ, M Oostrom, and GD Tartakovsky. 2015a. Evaluating Transport and Attenuation of Inorganic Contaminants in the Vadose Zone for Aqueous Waste Disposal Sites. PNNL-24731, Pacific Northwest National Laboratory, Richland, WA.
- ▶ Truex, MJ, BD Lee, CD Johnson, NP Qafoku, GV Last, MH Lee, and DI Kaplan. 2015b. Conceptual Model of Iodine Behavior in the Subsurface at the Hanford Site. PNNL-24709, Pacific Northwest National Laboratory, Richland, WA.
- ▶ Truex, M.J., et al. 2014. Conceptual Model of Uranium in the Vadose Zone for Acidic and Alkaline Wastes Discharged at the Hanford Site Central Plateau. PNNL-23666, Pacific Northwest National Laboratory, Richland, WA.
- ▶ Truex, MJ, PV Brady, CJ Newell, M Rysz, M Denham, and K Vangelas. 2011. The Scenarios Approach to Attenuation Based Remedies for Inorganic and Radionuclide Contaminants. SRNL-STI-2011-00459, Savannah River National Laboratory, Aiken, SC. Available at www.osti.gov, OSTI ID 1023615, doi: 10.2172/1023615.
- ▶ Xu, C., et al. 2015. "Radioiodine Sorption/Desorption and Speciation Transformation by Subsurface Sediments from the Hanford Site." *J. Environ. Radioact.*, 139:43-55.