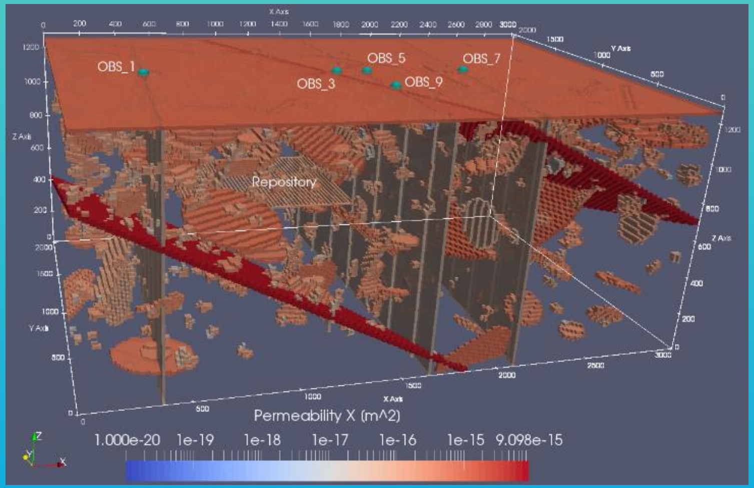

The Smart K_d Approach: Integrating Coupled THC Processes for Radionuclide Transport into GDSA

U.S. Nuclear Waste Technical Review Board
Fact Finding Meeting
July 19, 2022

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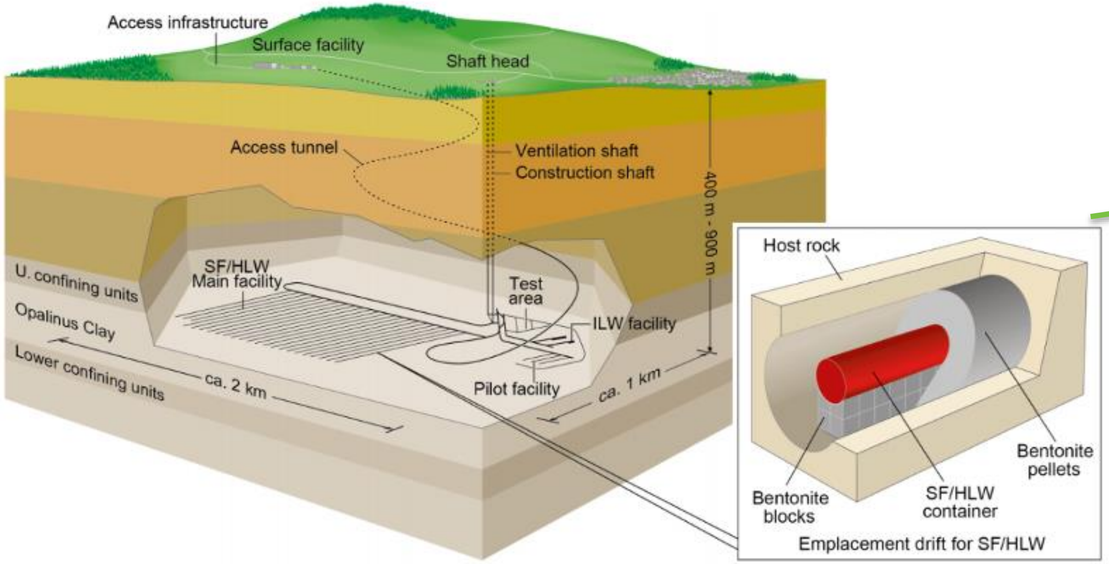
Agenda

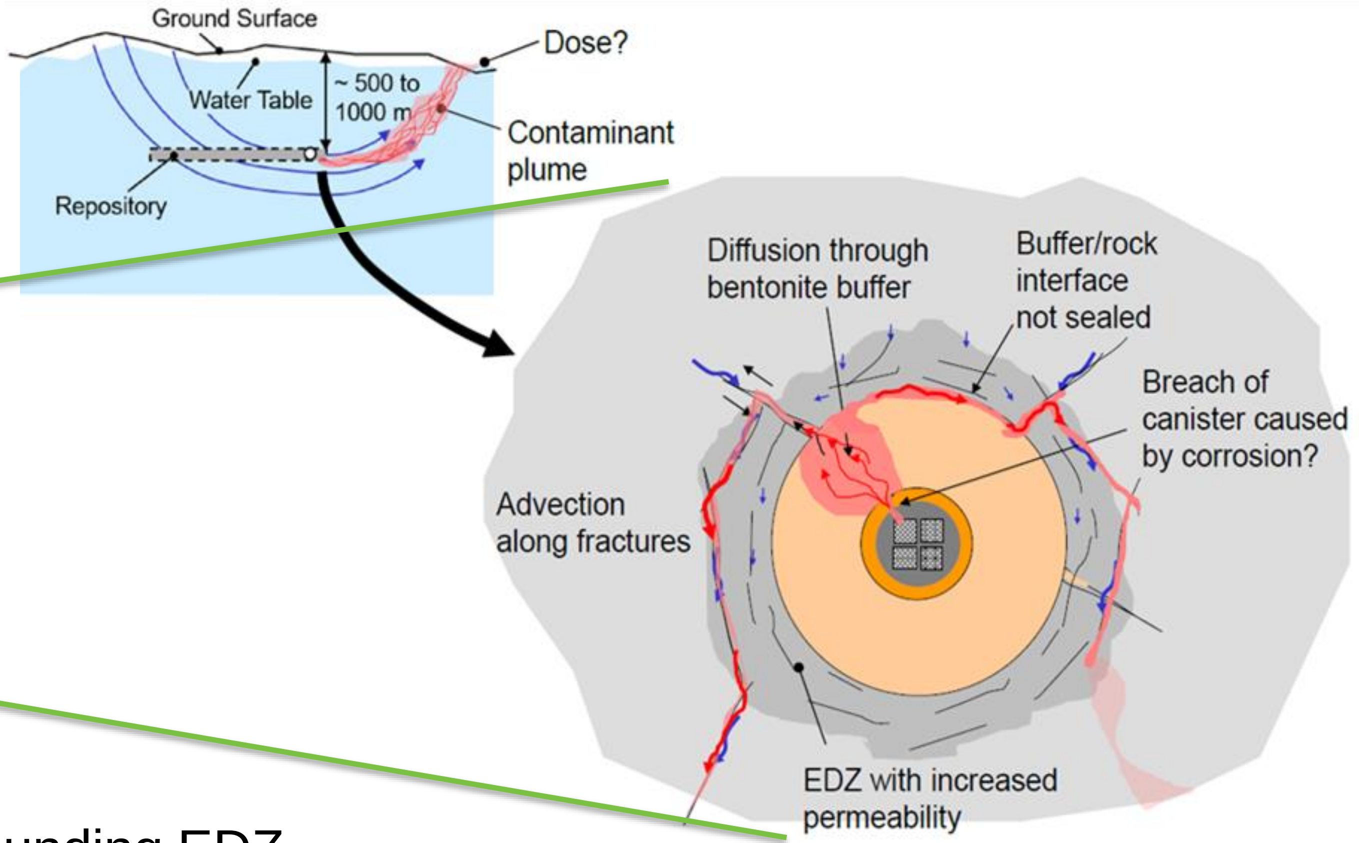
- Background/Motivation
- The Process Model
- ROM with "Smart" $\boldsymbol{K_d}$
- Integrating "Smart" $\boldsymbol{K_d}$ into GDSA
- Summary

ROM – Reduced-Order Modeling
GDSA – Geologic Disposal Safety Assessment
$\boldsymbol{K_d}$ = Distribution coefficient

Background and Motivation

Engineered Barrier System (EBS)





- EBS Integrity:
  - Breach of canister
  - Cracks within bentonite buffer and surrounding EDZ
  - Imperfect seal at buffer-rock interface
- Containment of radionuclides
- Contamination of ground water

SF - Spent Fuel
HLW - High-Level Radioactive Waste
ILW - Intermediate-Level Radioactive Waste
EBS - Engineered Barrier System
EDZ - Excavation Damaged Zone

Background and Motivation – cont'd

Buffer-Averaged
K_d

Thermal-Hydrological-Chemical (THC) model: *TOUGHREACT*

Performance Assessment (PA) model: *PFLOTRAN*

- Models critical THC processes that drive the integrity of EBS containment
- Computationally expensive to model a single waste package

THC - Thermal-Hydrological-Chemical
EBS - Engineered Barrier System
PA - Performance assessment

- K_d = Distribution coefficient
- A measure of contaminant partitioning between the solid and aqueous phases

$$K_d = \frac{\textbf{Mass of Adsorbate Sorbed}}{\textbf{Mass of Adsorbate in Solution}}$$

- Large-scale flow and radionuclide transport model
- Traditionally uses K_d to model mobility of radionuclides
- Capable of modeling multiple waste packages
- Need to include all the detailed THC processes around EBS in the PA model
- Computationally not feasible

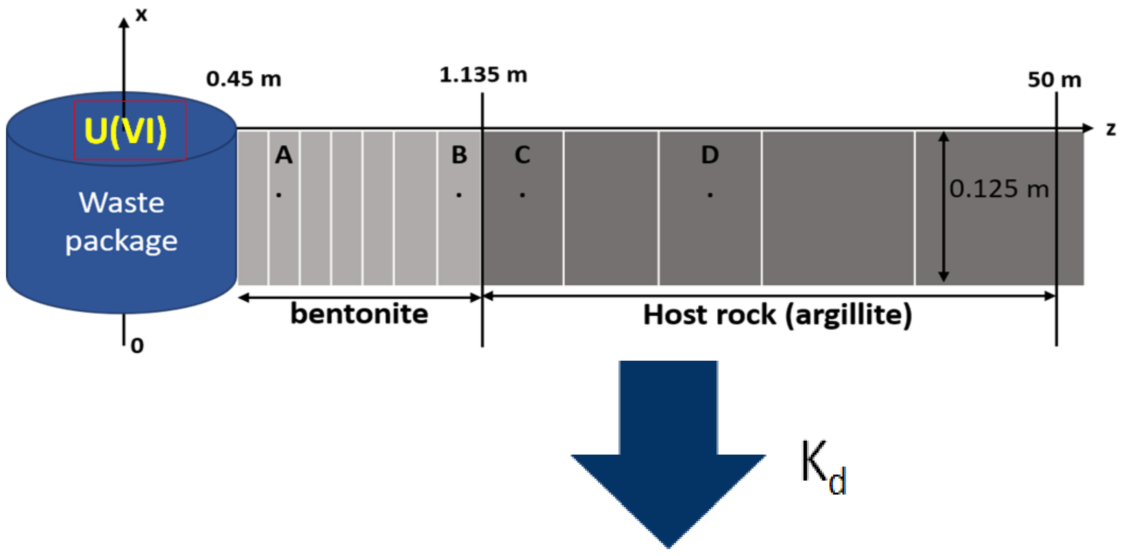
Objective

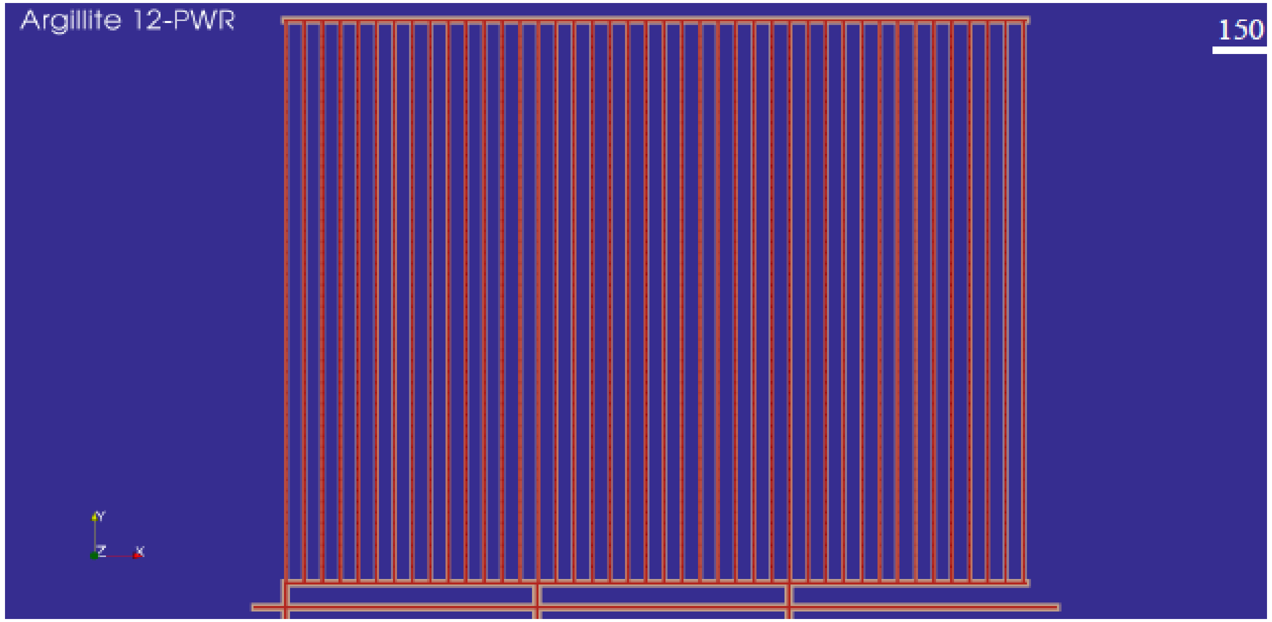
- Develop the methodology to:
  - Bridge the scale
  - Incorporate THC processes in PA
  - Represent buffer changes in PA
  - Transfer the uncertainty from THC models to PA

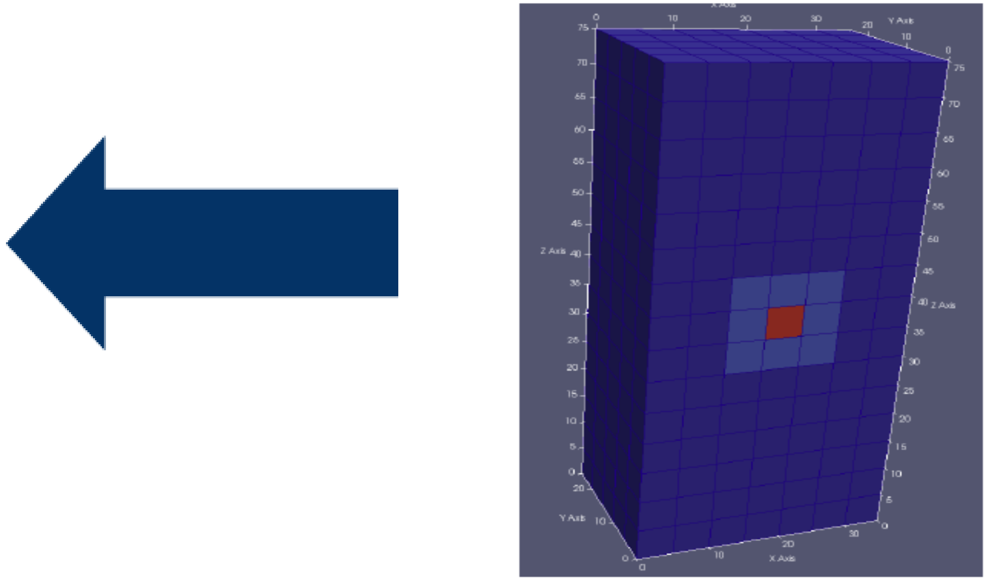
THC - Thermal-Hydrological-Chemical
PA - Performance assessment

Bridging the Scales: Surrogate Models/Emulators

- Spatially integrated K_d (mass preserved)
- Compute time-varying K_d
- Propagate the uncertainty of geochemical parameters to K_d
  - Develop an emulator for $K_d(t)$ ~ f(Geochemical parameters)







K_d = Distribution coefficient

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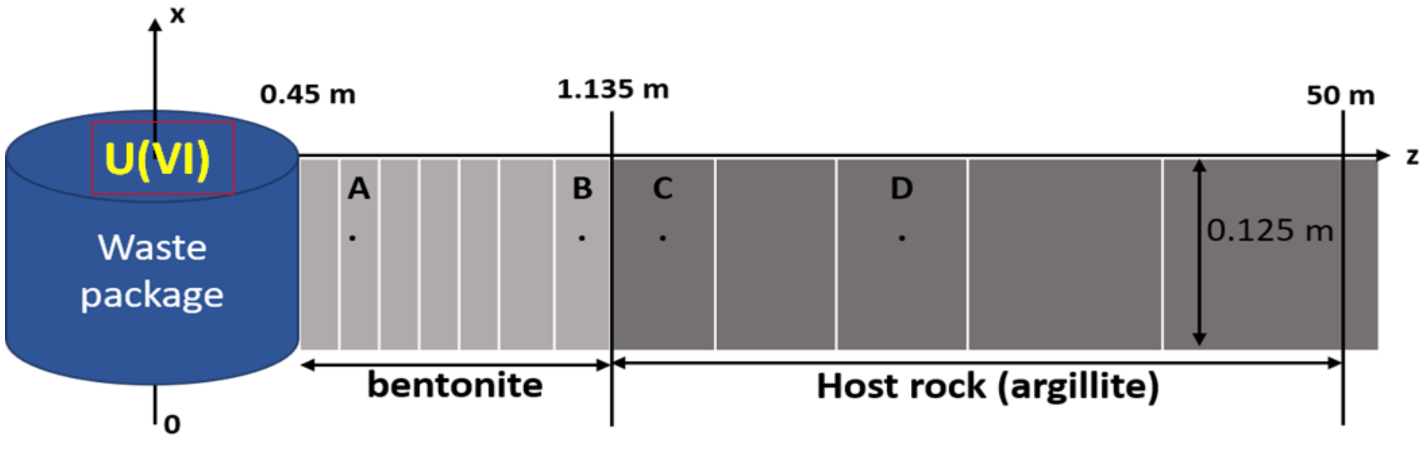
The Process Model

- Buffer-Argillite system (Zheng et al., 2019; Cao et al., 2019)
- Two-Site Protolysis Non-Electrostatic Surface Complexation and Cation Exchange sorption model (2 SPNE SC/CE) (Bradbury and Baeyens, 2011)
- Start from the unsaturated condition at 0 years
- Uranium transport after 1000 years (fully-saturated buffer)
    - Dissolution of Schoepite
- Adsorption, cation exchange, surface complexation of U
- Kinetically controlled mineral dissolution and precipitation

SPNE SC/CE - Site Protolysis Non-Electrostatic Surface Complexation and Cation Exchange sorption model

U - Uranium



Parameter Range

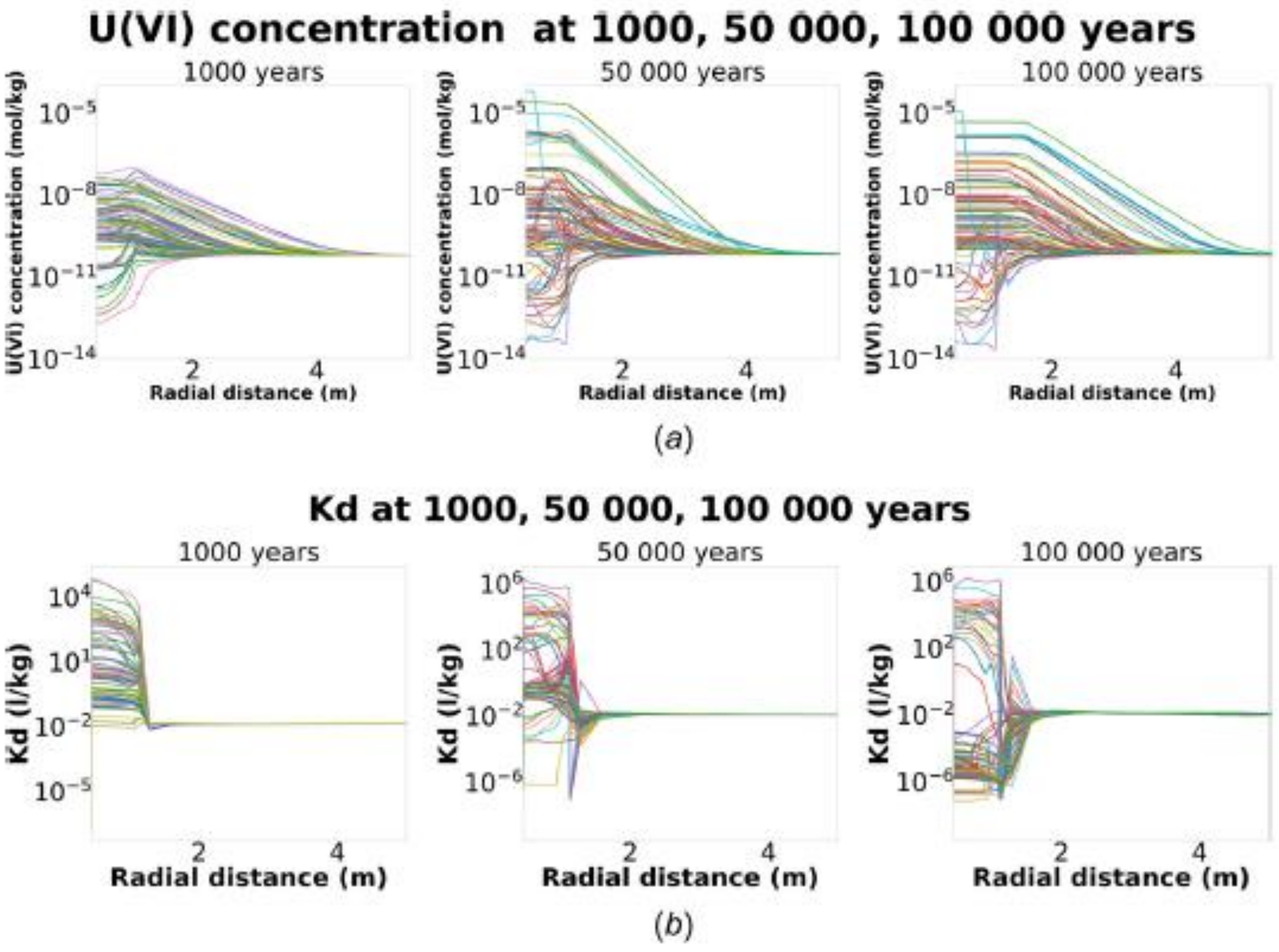
7 Parameters: $\{x_i | \ i = 1,2\ldots,k\}, k=7$

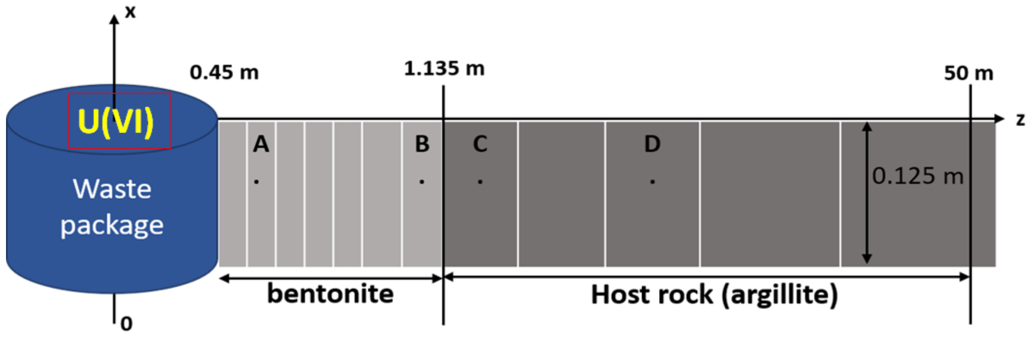
Parameters	Range	Base value
Site density (cm^2/g) of illite	10^3-10^6	10^5
Site density (cm^2/g) of smectite	10^3-10^6	10^5
Volume fraction: calcite	0.01-0.03	0.01
Volume fraction: smectite	0.3-0.95	0.92
Volume fraction: illite	0.01-0.2	0.0001
Initial pore water composition: pH or H^+	10^{-9}-10^{-7}	$1.91 * 10^{-8}$
Initial pore water composition: Ca^{2+}	10^{-3}-10^{-1}	0.022

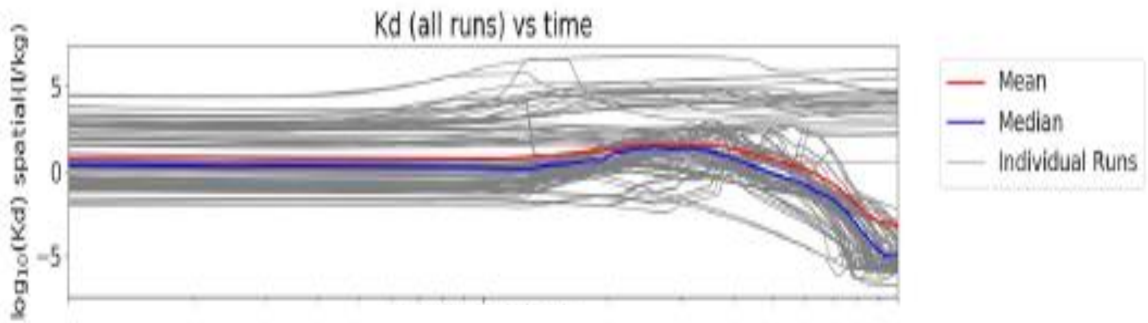
- The key point of this THC model is the mineral composition change (illitization: smectite --> illite), which alters $\boldsymbol{K_d}$ and affects safety functions.

Ermakova, D., Wainwright, H.M., Li, H., Zheng, L., “Global Sensitivity Analysis for Coupled Thermal-Hydorological-Chemical Simulations in Generic Nuclear Waste Repositories”, Journal of Nuclear Engineering and Radiation Science, 7(4), 041902.

Integrating “smart” K_d into GDSA: U & K_d Distribution Across the Buffer







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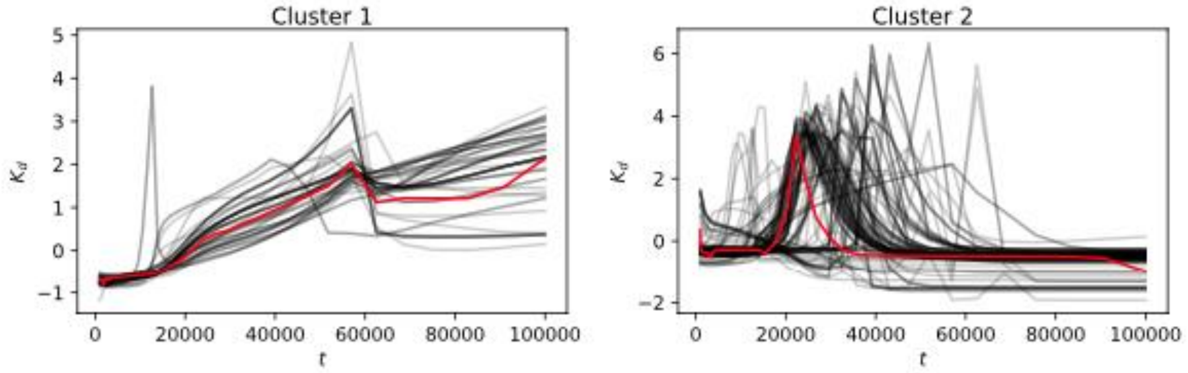
ROM for "smart" K_d: Development of Surrogate Models (Emulators)

- Establish the relationship between input parameters and $\boldsymbol{K_d}$

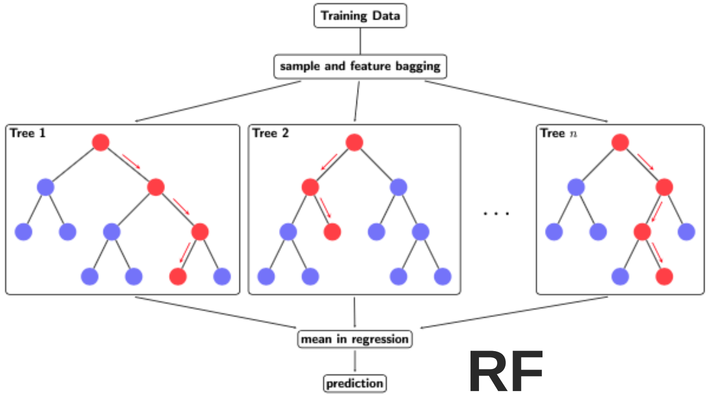
$$\mathrm{K_d} = f(t, \mathbf{p})$$

- Surrogate models (emulators) are statistical representations:
  - Trained on a set of the ($\boldsymbol{K_d}$, $\boldsymbol{p}$) combination
  - Predict $\boldsymbol{K_d}$ at any $\boldsymbol{p}$ values in the range
  - Once established, we don't have to run THC models.
- Regression: Random Forest (**RF**), Neural Network (**NN**)

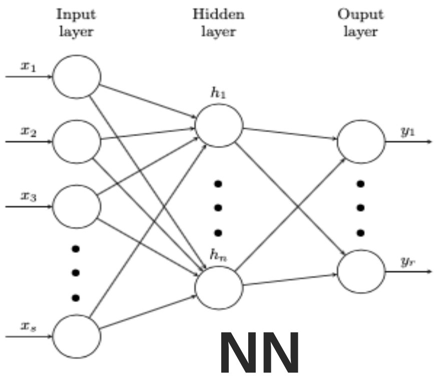
Domain splitting



- Clustering of training sets: k-means with dynamic time warping



RF



NN

Lu, H., Ermakova, D., Wainwright, H. M., Zheng, L., & Tartakovsky, D. M. (2021). Data-informed Emulators for Multi-Physics Simulations. Journal of Machine Learning for Modeling and Computing, 2(2).

Development of Surrogate Models (Emulators)

- Establish the relationship between input parameters and K_d

$$\mathrm{K_d} = f(t, \mathbf{p})$$

- Surrogate models (emulators) are statistical representations:
  - Trained on a set of the (K_d, p) combination
  - Predict K_d at any p values in the range
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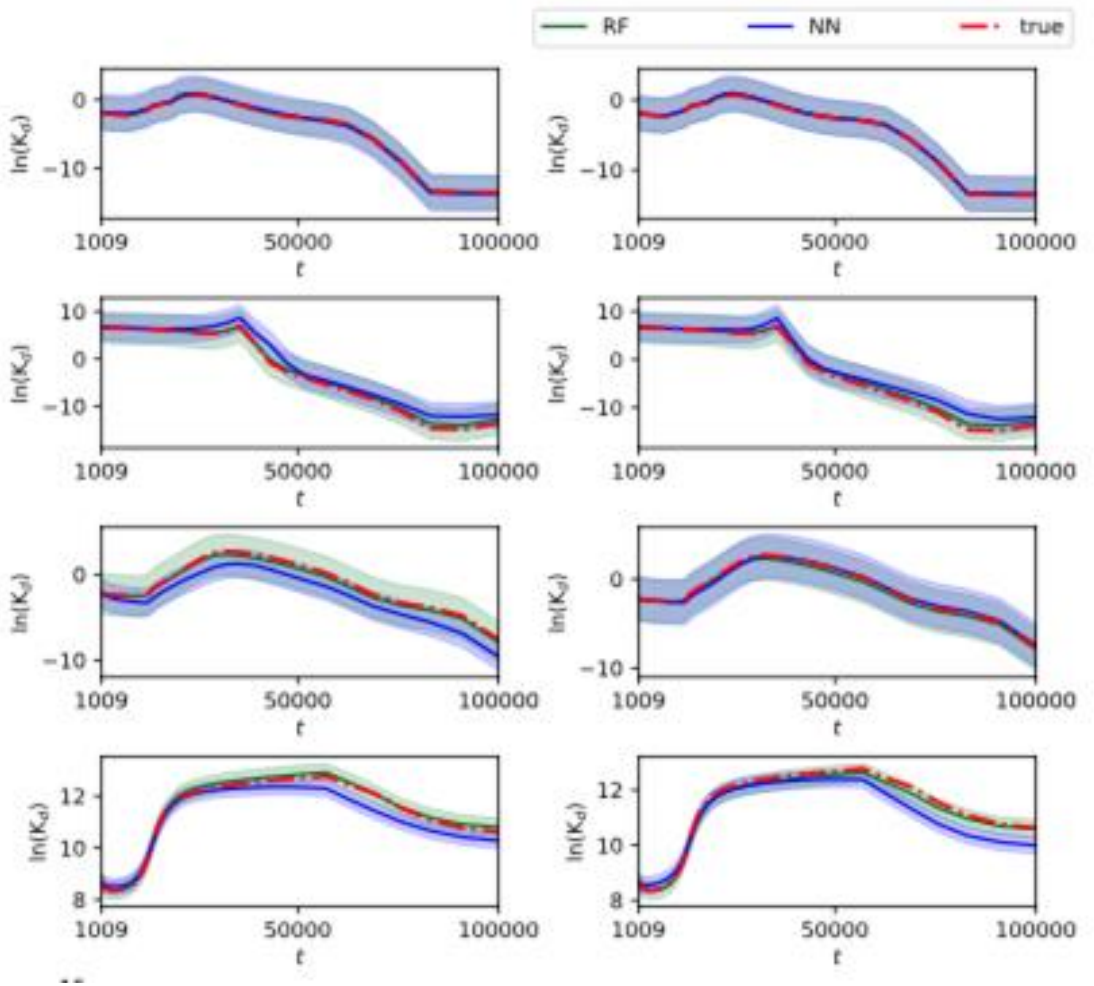
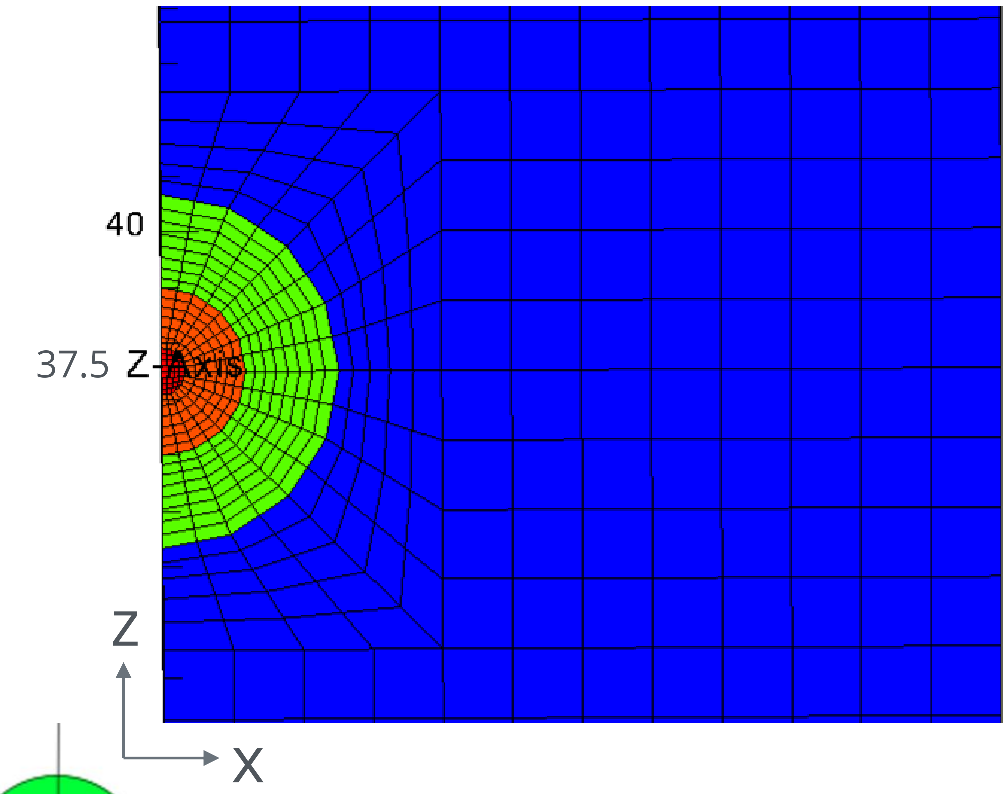


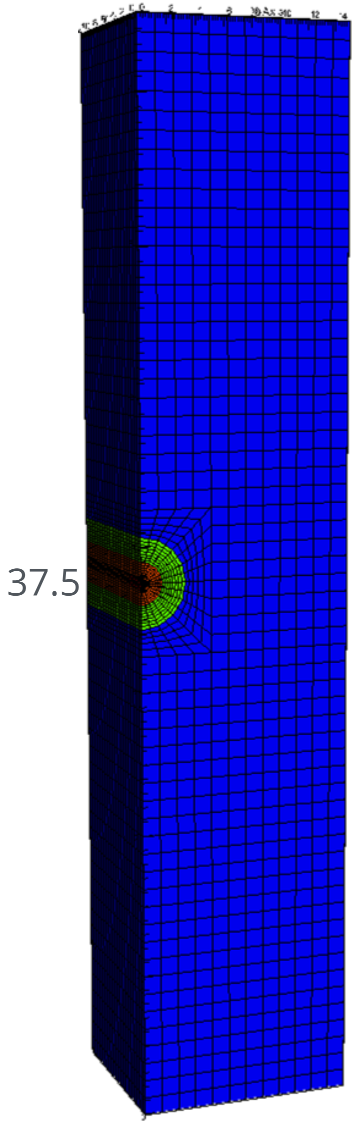
Without Clustering With Clustering

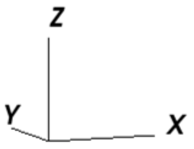
Lu, H., Ermakova, D., Wainwright, H. M., Zheng, L., & Tartakovsky, D. M. (2021). Data-informed Emulators for Multi-Physics Simulations. Journal of Machine Learning for Modeling and Computing, 2(2).

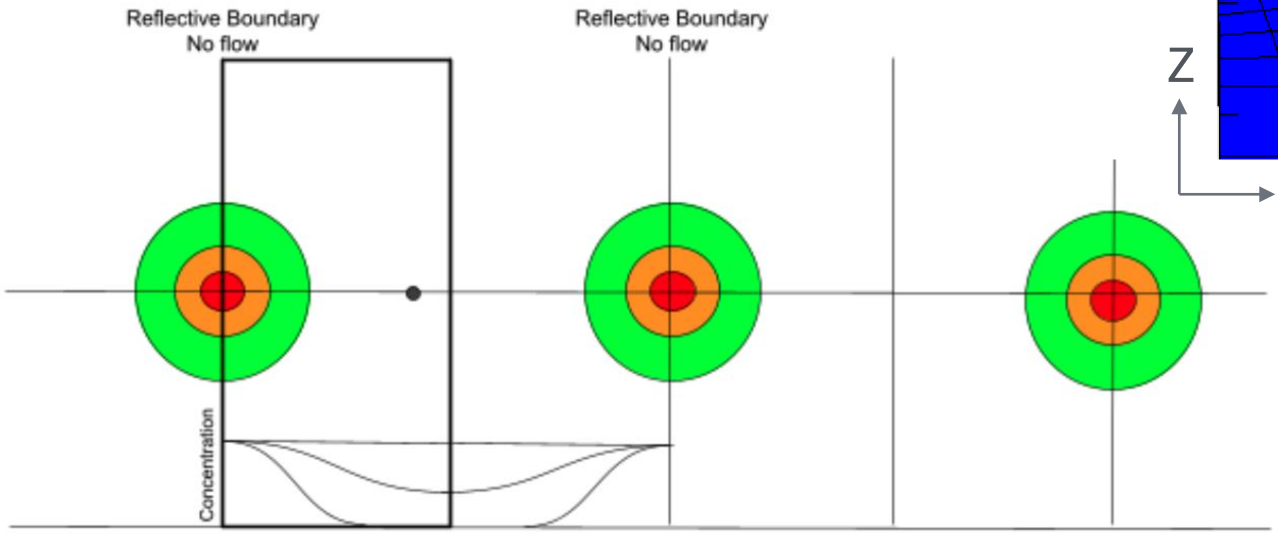
PA (*PFLOTRAN*) Model

- Canister-Buffer-EDZ-Rock System
- Reflective boundaries
- Uranium transport
- Constant K_d



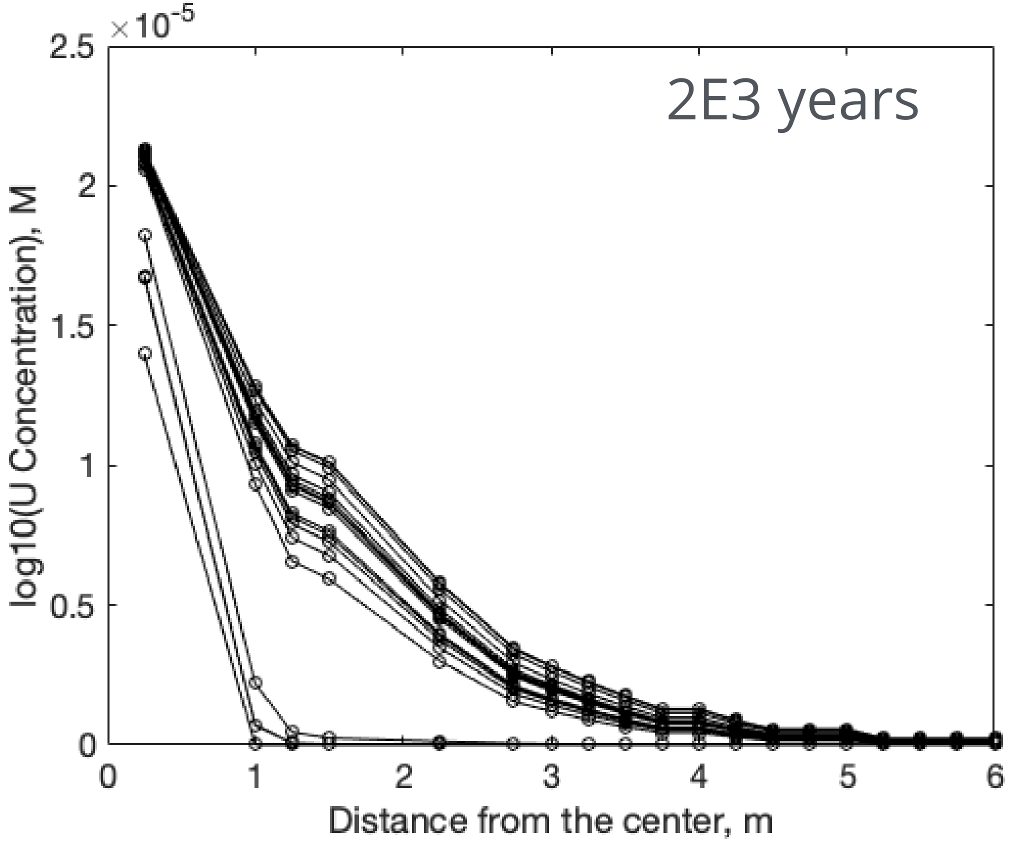




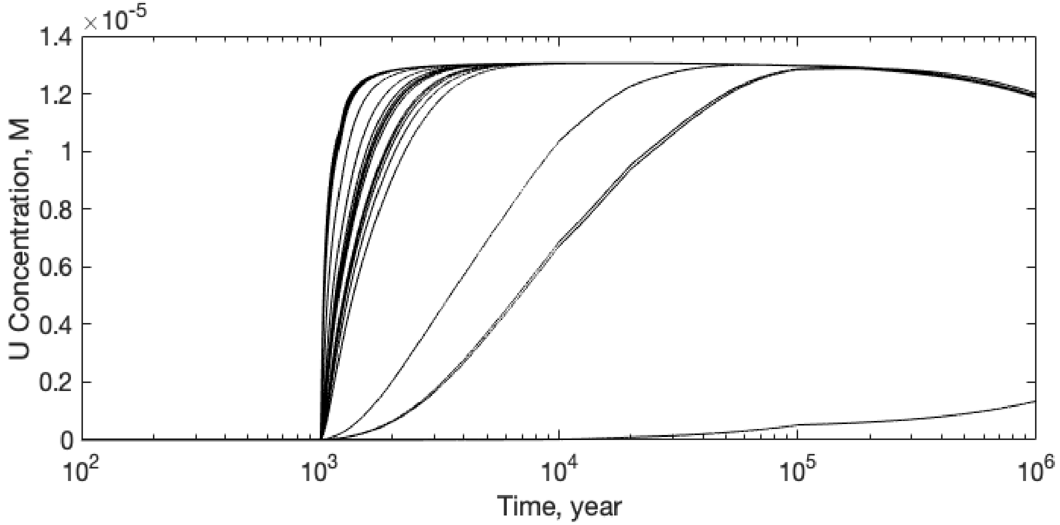


EDZ - Excavation Damaged Zone

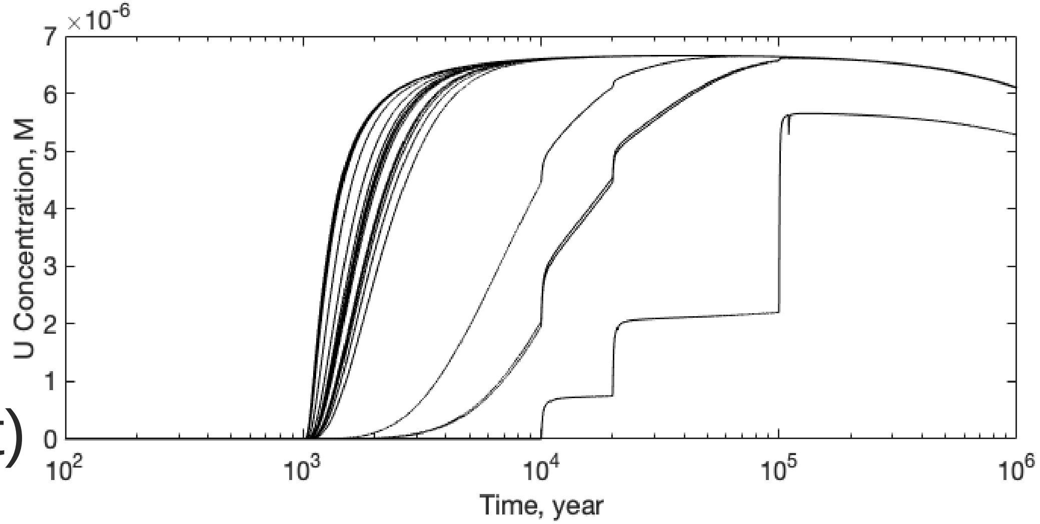
PA (*PFLOTRAN*) Model with THC-derived K_d



Buffer



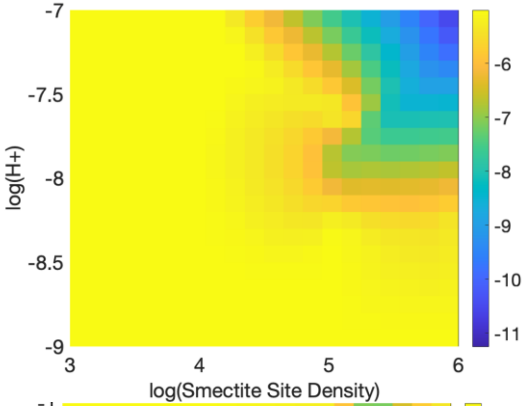
Rock (4.5 m)



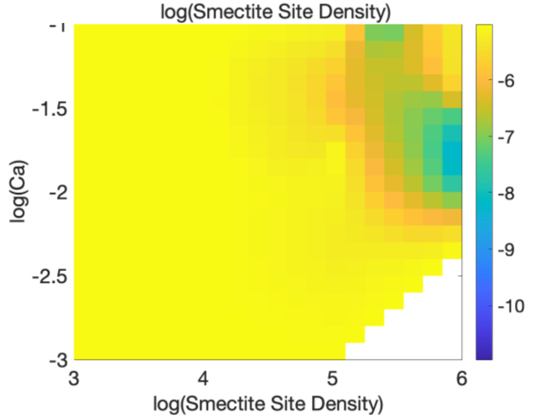
- K_d values from the emulator
- *PFLOTRAN* simulations (temporally constant)

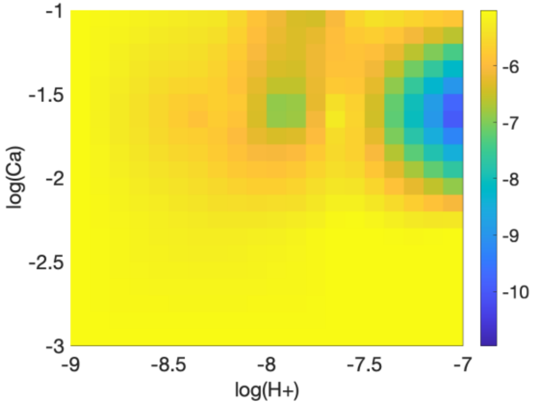
PA (*PFLOTRAN*) Model with THC-derived K_d

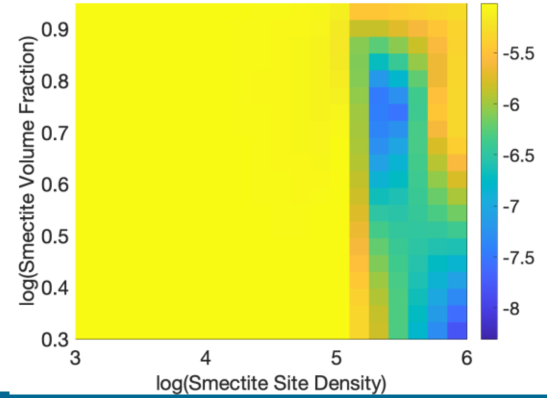
Geochemical parameters → TOUGHREACT (THC) → K_d → PFLOTRAN (PA)

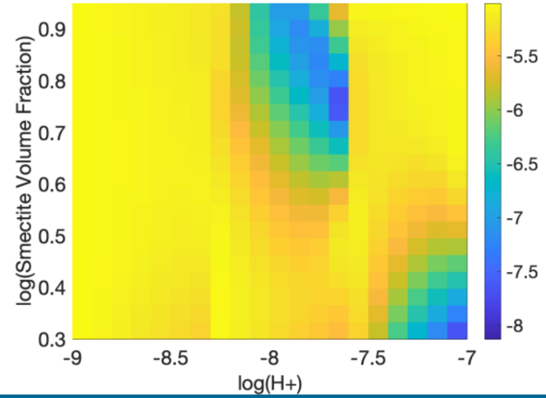


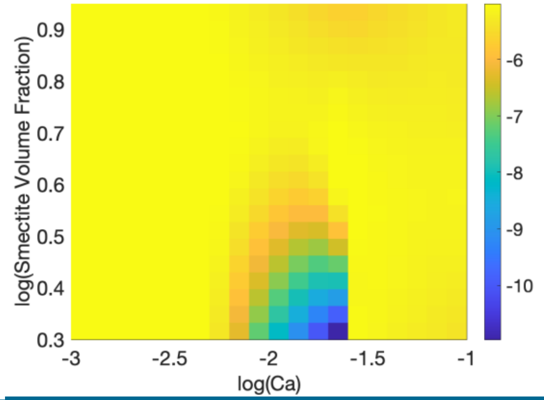
- U concentration (log) at the buffer-rock interface
- Transport simulation with *PFLOTRAN*
- Function of geochemical parameters







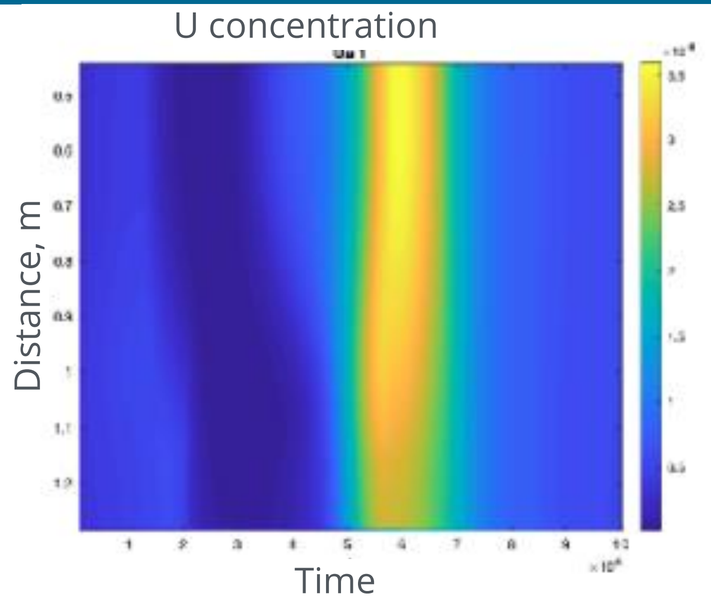


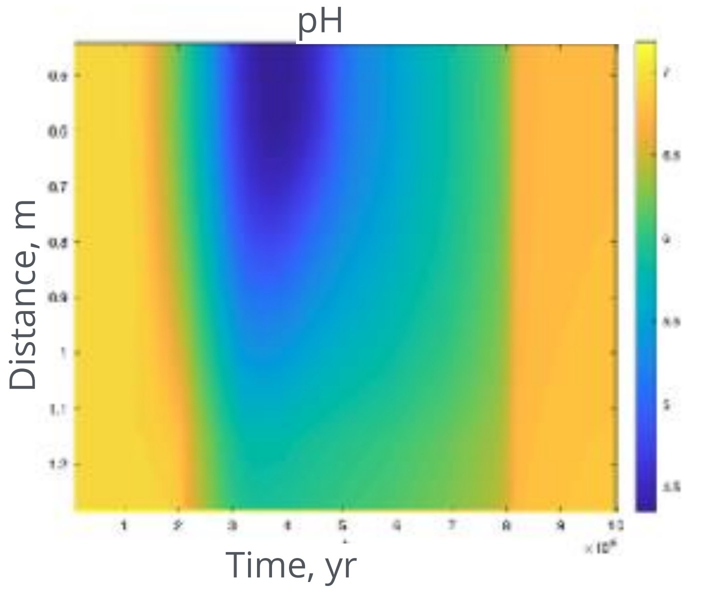


THC - Thermal-Hydrological-Chemical
PA - Performance assessment
U - Uranium

Reduced-Order Modeling (ROM) for SpatioTemporal Variability of U Concentration

- Surrogate models to capture the spatiotemporal variability
- PA Connection:
  - Spatiotemporal evolution of U concentrations within the buffer
  - Temporal evolution of flux from the buffer → Flux preserved $\boldsymbol{K_d}$





ROM - Reduced-Order Modeling
U - Uranium
PA - Performance Assessment

Physics-coupled Reduced-Order Modeling (ROM) with Dynamic Mode Decomposition (DMD)

- Dynamic Mode Decomposition
  - A dimensionality reduction algorithm developed for time-series datasets

High-Fidelity-Model (HFM) of interest:

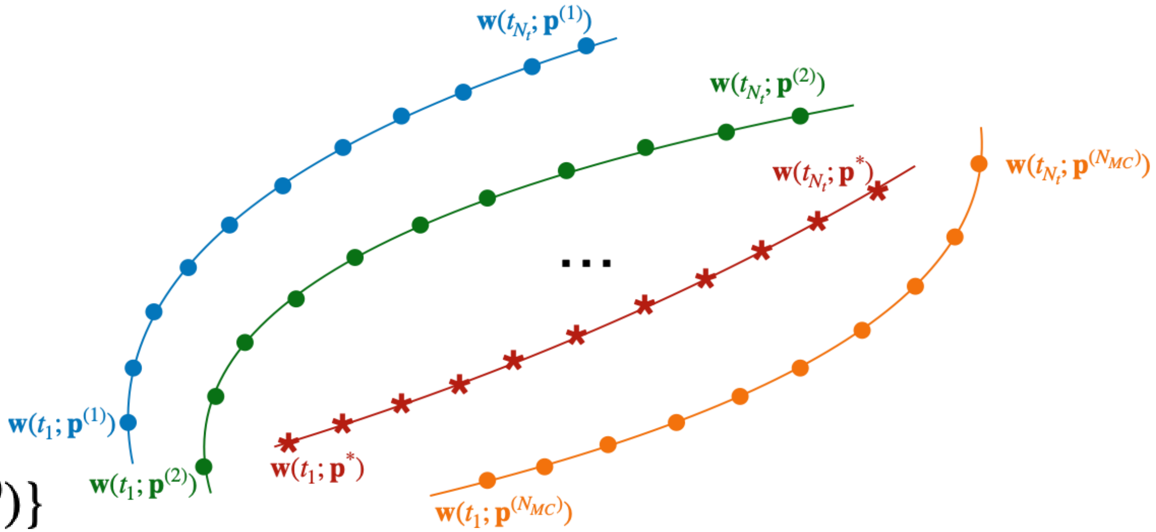
$$\frac{d\mathbf{y}}{dt}(t;\mathbf{p}) = \underset{\substack{\uparrow \\ \text{Unknown}}}{\mathbf{f}}(\mathbf{y}(t;\mathbf{p}), t; \mathbf{p})$$

Question: can we use the pre-computed snapshots

$$\{\underbrace{\mathbf{w}(t_1;\mathbf{p}^{(1)}), \cdots, \mathbf{w}(t_{N_t};\mathbf{p}^{(1)})}_{\text{one sample trajectory}}, \cdots, \underbrace{\mathbf{w}(t_1;\mathbf{p}^{(N_{MC})}), \cdots, \mathbf{w}(t_{N_t};\mathbf{p}^{(N_{MC})})}_{\text{one sample trajectory}}\}$$

to reconstruct a solution for a queried but unsampled parameter point

$$\mathbf{p}^* \notin \{\mathbf{p}^{(1)}, \cdots, \mathbf{p}^{(N_{MC})}\} \ ?$$



ROM – Reduced-Order Modeling
DMD - Dynamic Mode Decomposition

Lu and Tartakovsky, in prep

Physics-coupled Reduced-Order Modeling (ROM) with Dynamic Mode Decomposition (DMD)

- Dynamic Mode Decomposition
  - A dimensionality reduction algorithm developed for time-series datasets

High-Fidelity-Model (HFM) of interest:

$$\frac{d\mathbf{y}}{dt}(t;\mathbf{p}) = \mathbf{f}(\mathbf{y}(t;\mathbf{p}),t;\mathbf{p})$$

Unknown

DMD surrogate linear model:

$$\frac{d\mathbf{y}}{dt}(t;\mathbf{p}) = \mathbf{K}(\mathbf{p})\mathbf{y}(t;\mathbf{p}) + \mathbf{B}(\mathbf{p})$$

$$\mathbf{K}(\mathbf{p}) \in \mathbb{R}^{N_y \times N_y}, \mathbf{B}(\mathbf{p}) \in \mathbb{R}^{N_y}$$

Compute K(p) and B(p) based on the ensemble data

$$\mathbf{K}(\mathbf{p}^{(i)}), \mathbf{B}(\mathbf{p}^{(i)}) = \arg\min_{\hat{\mathbf{K}},\hat{\mathbf{B}}} \|\mathbf{Y}_2^{(i)} - \hat{\mathbf{K}}\mathbf{Y}_1^{(i)} - \hat{\mathbf{B}}\|_F, \mathbf{Y}_1^{(i)} = \begin{bmatrix} | & & | \\ \mathbf{y}(t_1;\mathbf{p}^{(i)}) & \cdots & \mathbf{y}(t_{N_t-1};\mathbf{p}^{(i)}) \\ | & & | \end{bmatrix}, \mathbf{Y}_2^{(i)} = \begin{bmatrix} | & & | \\ \mathbf{y}(t_2;\mathbf{p}^{(i)}) & \cdots & \mathbf{y}(t_{N_t};\mathbf{p}^{(i)}) \\ | & & | \end{bmatrix}$$

Define ROM

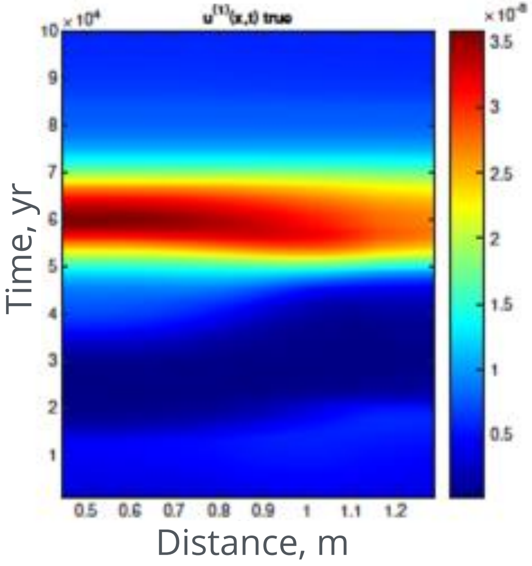
$$\mathbf{q}(t^{k+1};\mathbf{p}^{(i)}) \approx \tilde{\mathbf{K}}(\mathbf{p}^{(i)})\mathbf{q}(t^k;\mathbf{p}^{(i)}) + \tilde{\mathbf{B}}(\mathbf{p}^{(i)}),\ \mathbf{y}(t;\mathbf{p}^{(i)}) = \mathbf{V}(\mathbf{p}^{(i)})\mathbf{q}(t;\mathbf{p}^{(i)}),\ i = 1,\cdots,N_{MC}$$

Lu and Tartakovsky, in prep

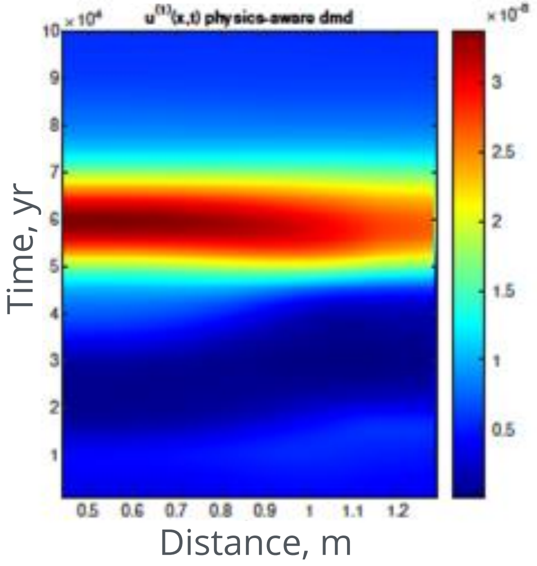
Physics-coupled ROM:DMD

- Preliminary Results
  - Co-estimation of {U, Ca, pH}

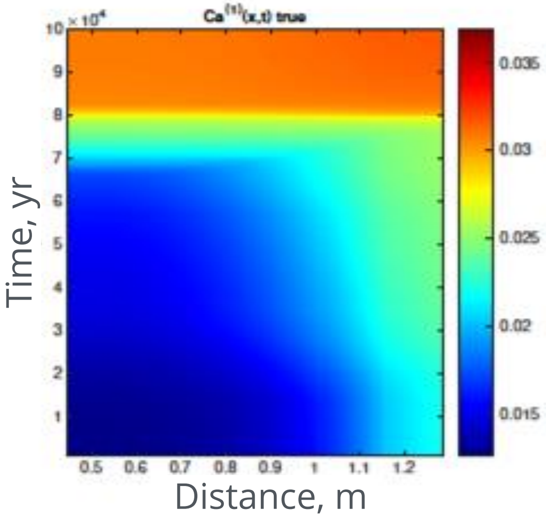
U concentration
(True)

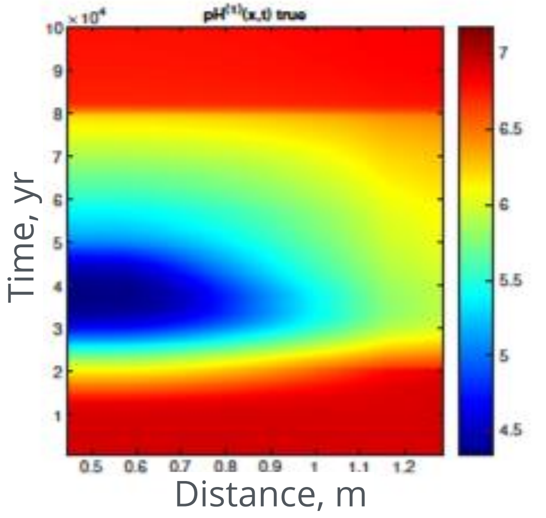


U concentration
(DMD Prediction)

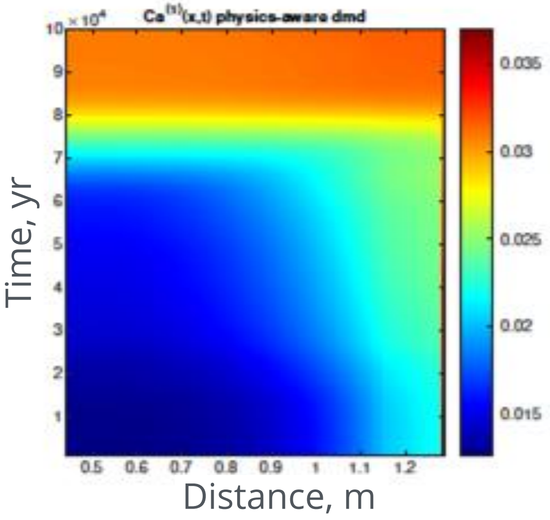


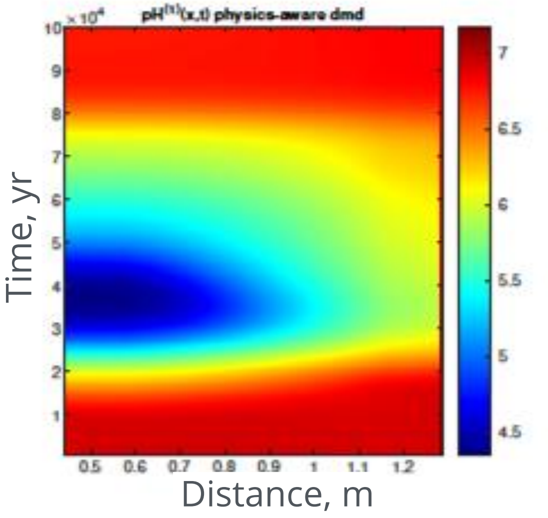
U concentration
(True)





U concentration
(DMD Prediction)





Summary

- Demonstrated a workflow to connect THC to PA
  - Temporal evolution of buffer-averaged K_d (mass-preserved K_d)
  - Emulator for buffer-averaged K_d
    - ✓ NN/RF comparison
    - ✓ Parameter domain splitting for capturing non-linearity
  - UQ with K_d from THC
    - ✓ Geochemical parameters → *PFLOTRAN*-predicted U concentrations
- Developing new surrogate models for THC
  - Emulator based on Dynamics Mode Decomposition
  - Spatially/temporally resolved U concentrations within the buffer (--> flux-preserved K_d)

THC - Thermal-Hydrological-Chemical
PA - Performance assessment
K_d = Distribution coefficient

References

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Wainwright, H.M., Ermakova, D., Lu, H., Wainwright, H. M., Zheng, L., & Tartakovsky, D. M. “Report: LBNL FY20 research in GDSA Modeling andIntegration”, August 21st, LBNL report.