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Spent Fuel and Waste Science and Technology (SFWST)

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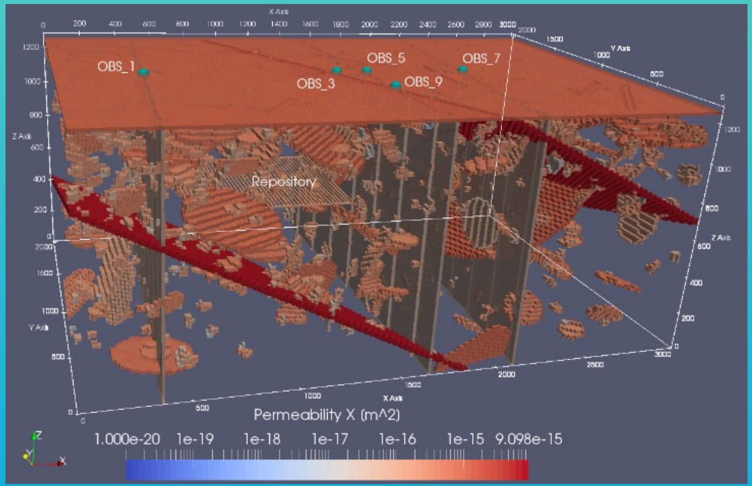

Machine Learning Surrogates for Fuel Degradation Processes in Nuclear Waste Repository Simulations

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Sandia National Laboratories

Sandia ML DL Workshop

Jul 18, 2023

SAND2023-ZZZZC

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Acknowledgements and Disclaimers

- Acknowledgements:
  - Funding by the Department of Energy (DOE), Office of Nuclear Energy, Spent Fuel and Waste Science and Technology

- Disclaimer:
  - This presentation describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the presentation do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

Overview

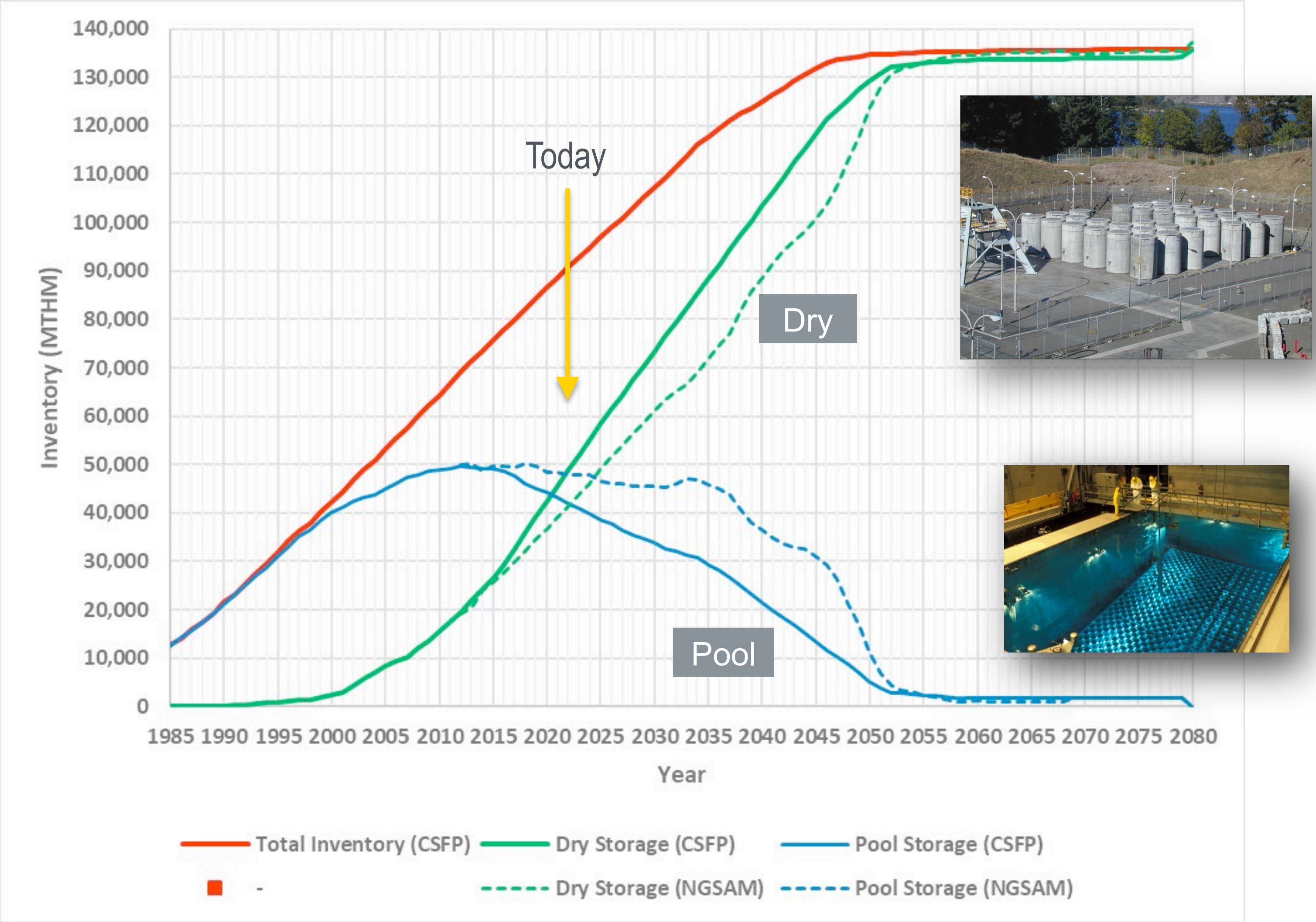
- Computational Challenges in Handling Spent Nuclear Fuel
- Training Data for Machine Learning Surrogates
- ML Surrogates
  - kNNr
  - ANN
  - Neural ODEs
- Conclusions

Computational Challenges in Handling Spent Nuclear Fuel

The US inventory of spent nuclear fuel is rapidly increasing

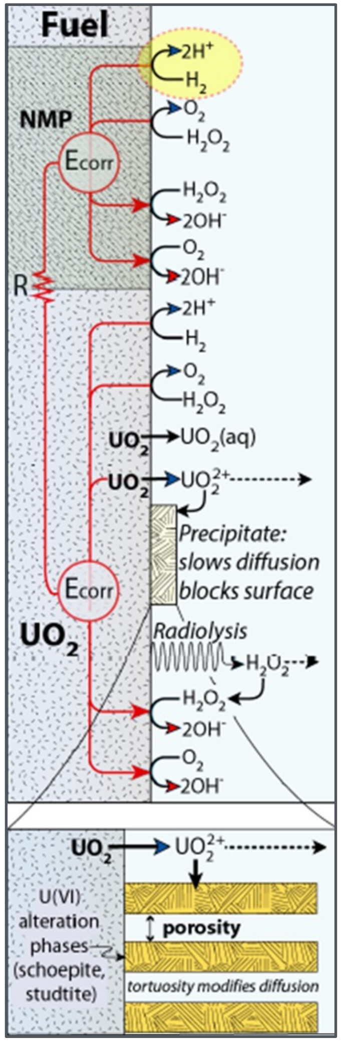
- 90,000 MTHM and increasing
- Pools have reached capacity limits
- Utilities have implemented dry storage
- Where facilities have shut down, some “stranded” fuel remains at independent spent fuel storage installations

Storage Projections (2 models)



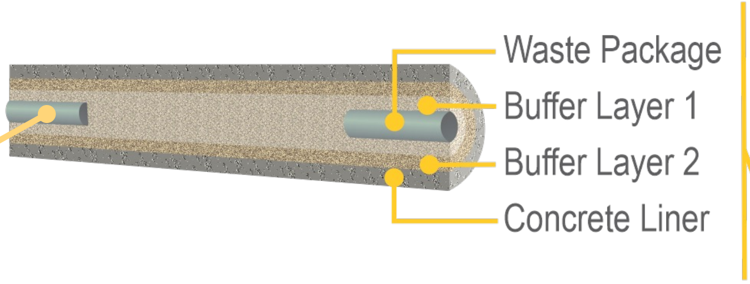
Adapted from Freeze et al. (2021, Figure 2-3)

Our challenge is to provide realistic UO_2 degradation rates in underground nuclear waste repository simulations



Fuel Matrix Degradation Model (FMDM)
adapted from Jerden et al. (2015)

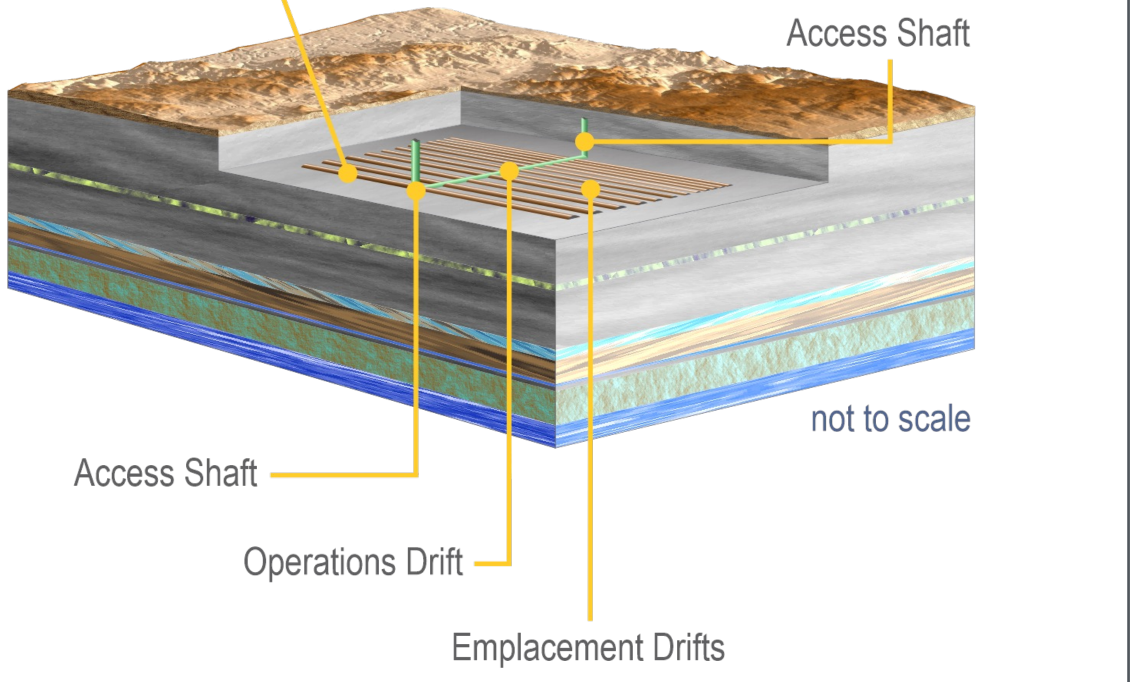
Waste Package Model



Fuel Matrix Degradation Model (L)

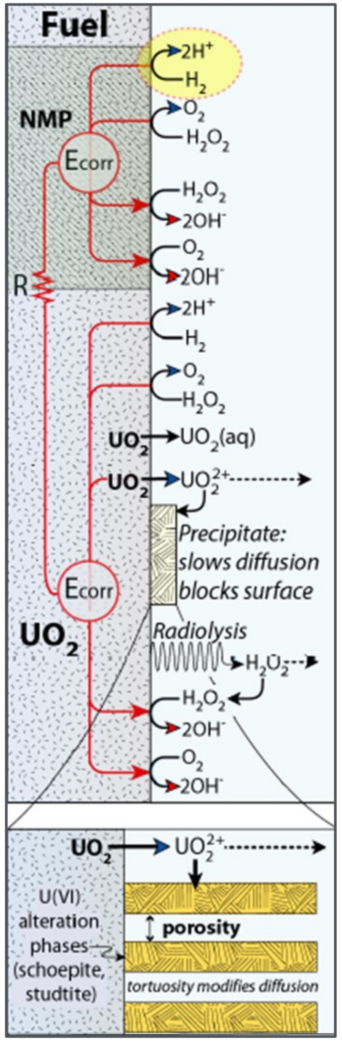
- **Needed for each breached package at each time step**
- **Computationally intensive**
- **Coupling to repository scale model is challenging**

Nuclear Waste Repository



Nuclear Waste Repository

Surrogate models provide a cheap-to-evaluate mapping between the model inputs and its outputs



- Inputs are the environmental conditions along with the internal state at any point in time
  - Environmental Concentrations of CO_3^{2-}, O_2, Fe^{2+}, and H_2
  - Temperature T
  - Dose Rate, which is f(time, burnup)
  - Corrosion Layer Thickness
  - Internal concentration profiles
- Relevant output is the UO_2 degradation rate (expressed as a flux)

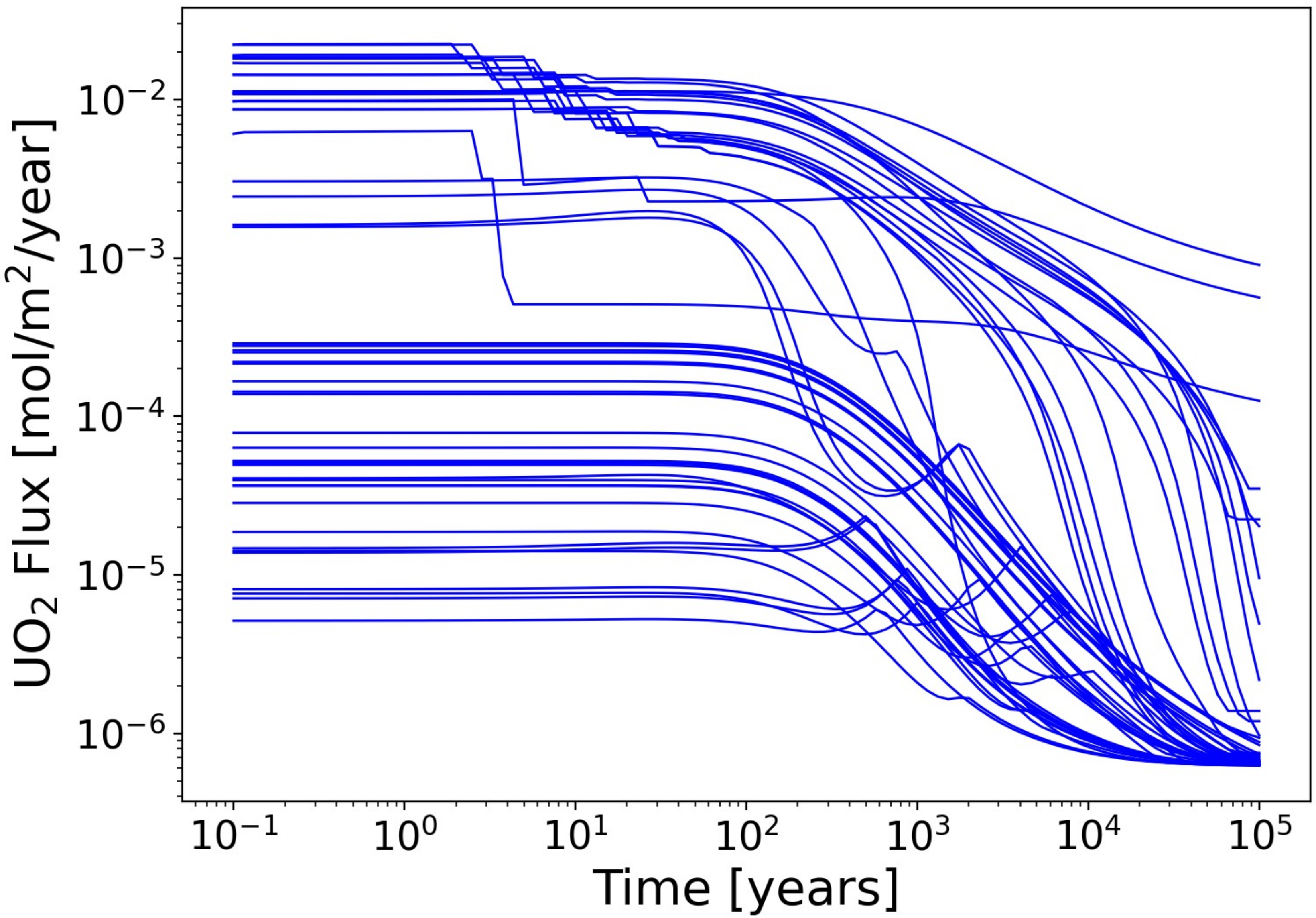
Training Data for Machine Learning Surrogates

Process model input parameters were sampled from expected ranges in reservoir simulations to generate training data

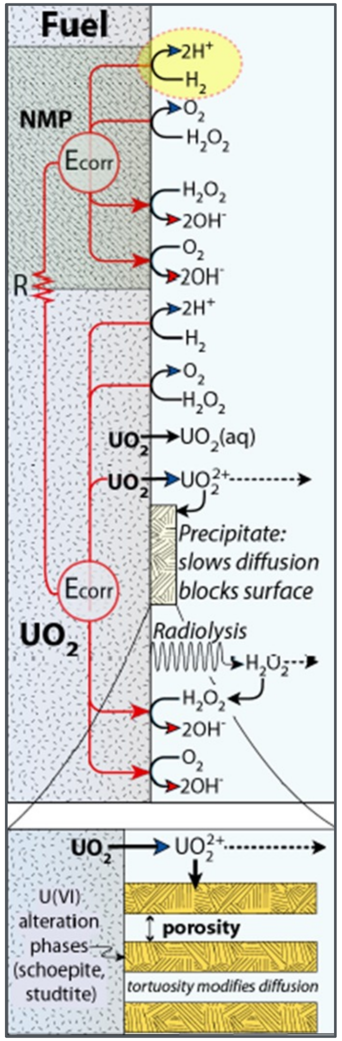
Parameter	Distribution	Min.	Max.
Init. Temp. (K)	Uniform	300	400
Burnup (Gwd/MTU)	Uniform	40	65
Env. CO_3^{2-} (mol/m^3)	Log-uniform	10^{-3}	$2\text{x}10^{-2}$
Env. O_2 (mol/m^3)	Log-uniform	10^{-7}	10^{-5}
Env. Fe^{2+} (mol/m^3)	Log-uniform	10^{-3}	10^{-2}
Env. H_2 (mol/m^3)	Log-uniform	10^{-5}	$2\text{x}10^{-2}$

- Same ranges used for training, validation, and testing data
- Ranges that span multiple orders of magnitude sampled with log-uniform distribution

Training data is pulled from FMD Process model UO_2 Flux trajectories for randomly sampled initial conditions



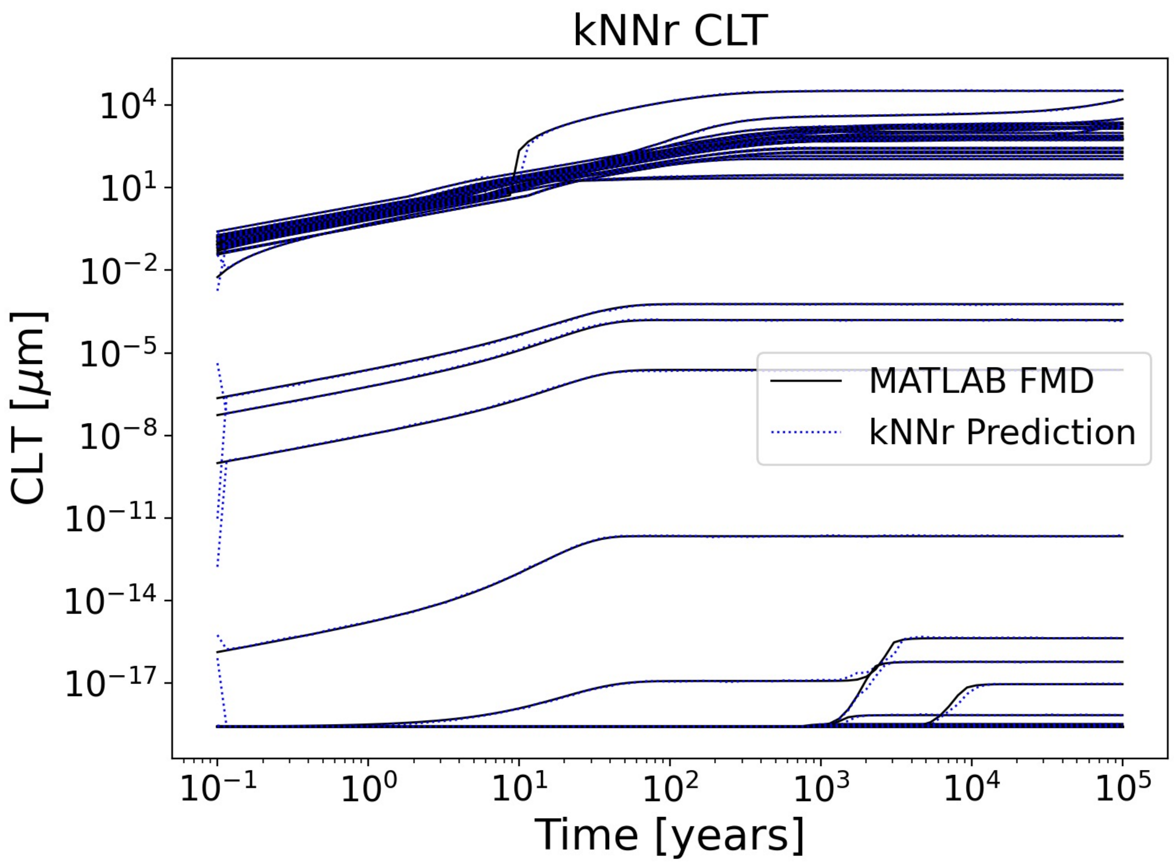
FMD surrogate model inputs aim to track the internal fuel cask state

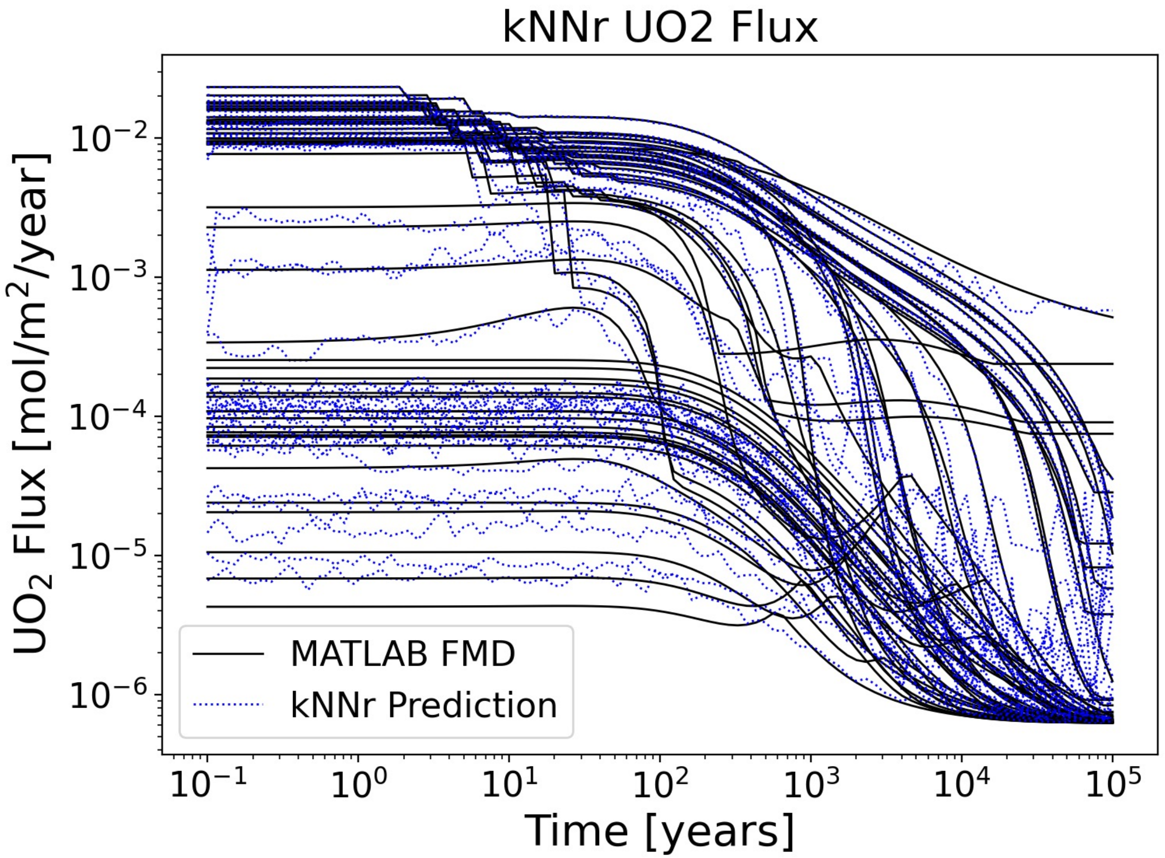


- Inputs that do not require detailed knowledge of the fuel cask state
  - Environmental Concentrations of CO_3^{2-}, O_2, Fe^{2+}, and H_2
  - Temperature T
  - Dose Rate, which is f(time, burnup)
- Inputs that require detailed knowledge of the internal fuel cask state
  - Corrosion Layer Thickness
  - Internal concentration profiles

Prior results: kNNr and ANN

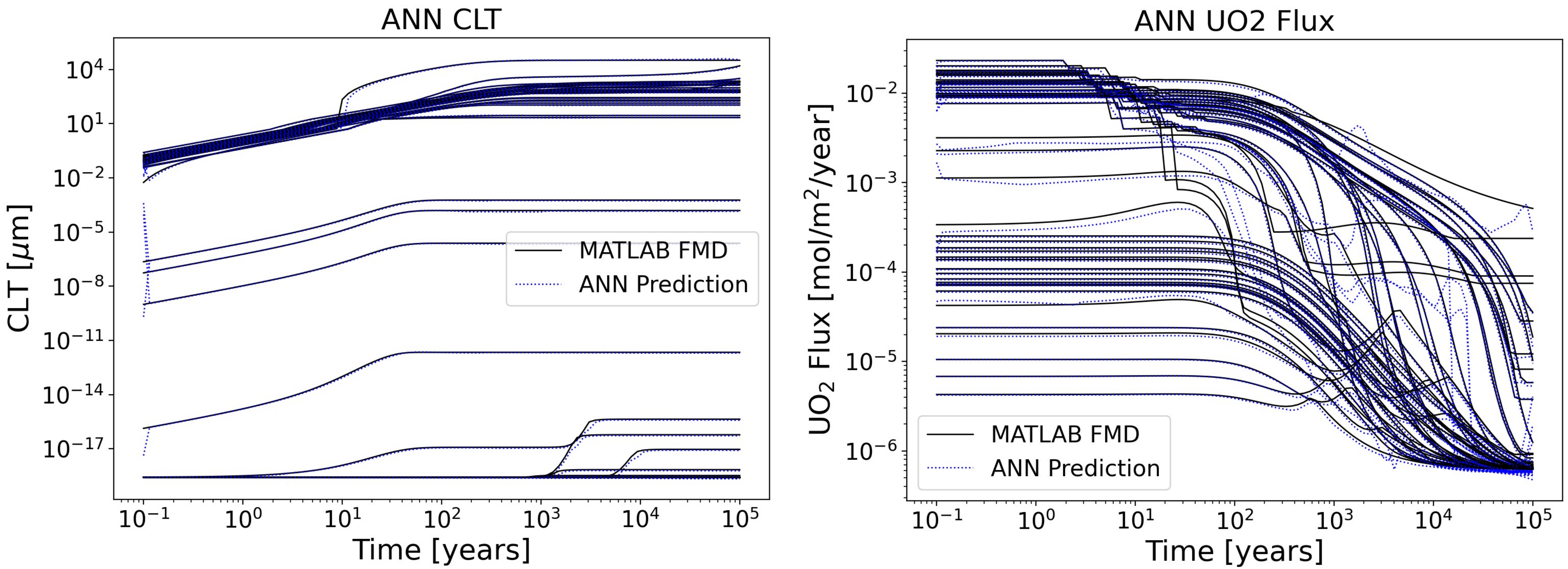
kNNr has good but noisy predictions on the test data due to the local character of the representation





- The inputs for each prediction are taken from test data (rather than from previously predicted points)
- More details in Debusschere et al. 2022 & 2023

ANN has smoother predictions as it is a global functional approximation



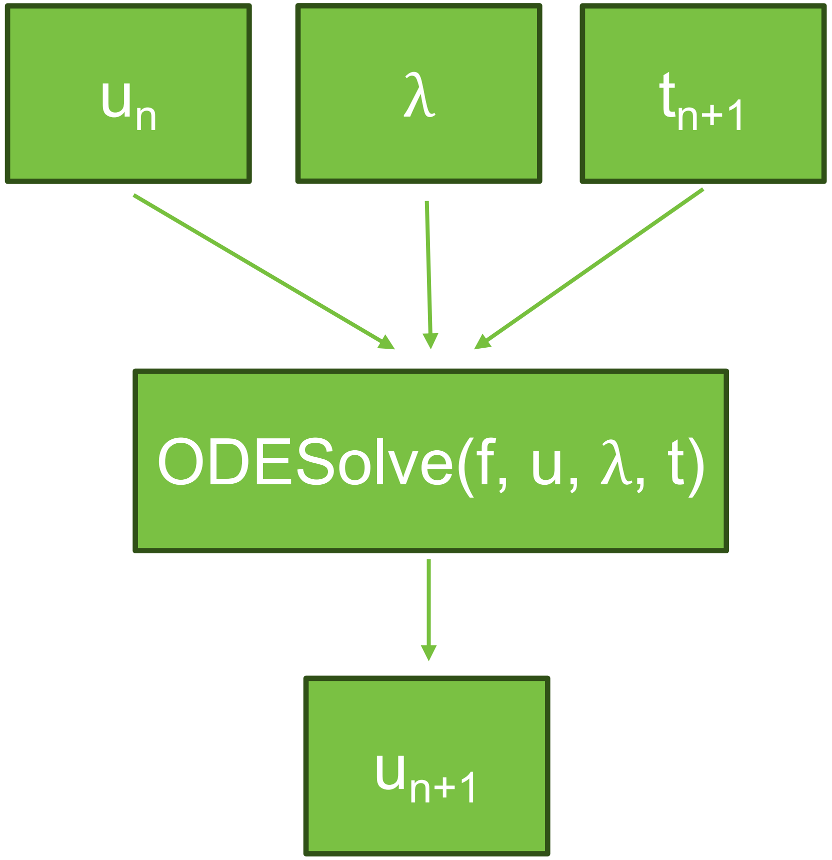
- The inputs for each prediction are taken from test data (rather than from previously predicted points)
- More details in Debusschere et al. 2022 & 2023

Neural ODE

Neural ODEs approximate the derivative of the system state as a Neural Network

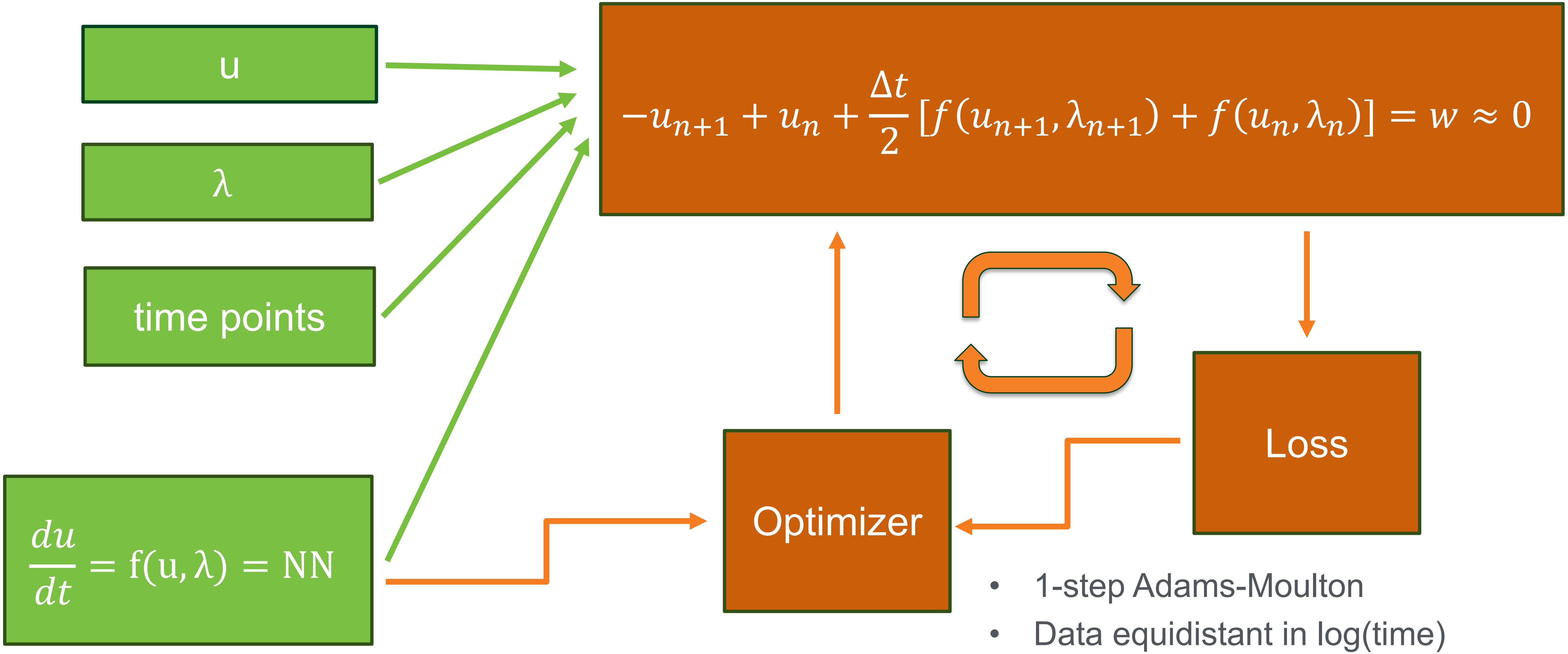
$$\frac{du}{dt} = \mathrm{f}(\mathrm{u}, \lambda) = \mathrm{NN}$$

- Train NN based on data at equidistant timesteps[1]
- Predict with ODE Solver
- Hyperparameters to tune:
  - Number of layers
  - Number of nodes (neurons) per layer
  - Amount of training data



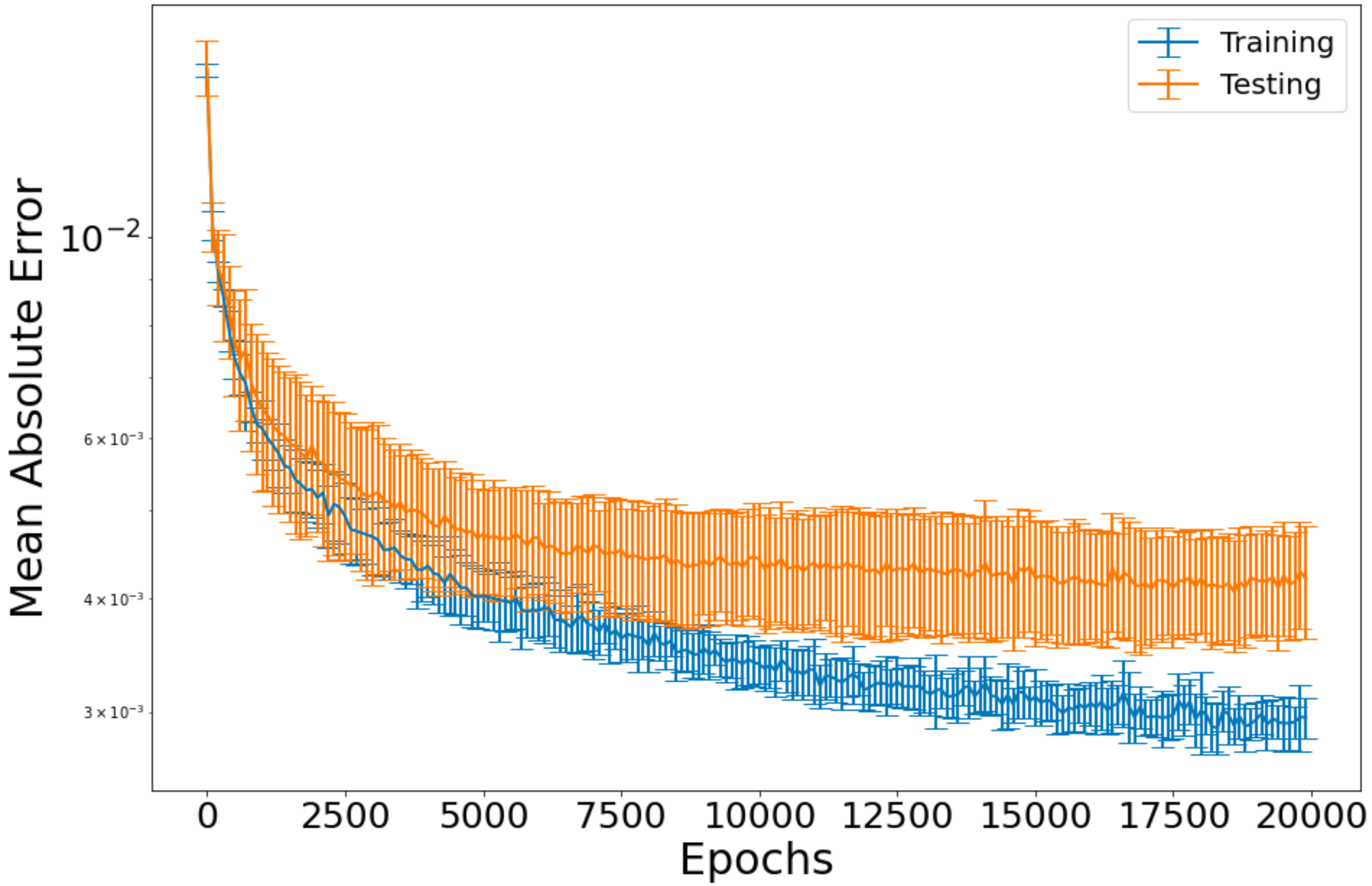
[1] For more details, see Raissi et al. 2018

Data at regular time intervals is used to train a multi-step method

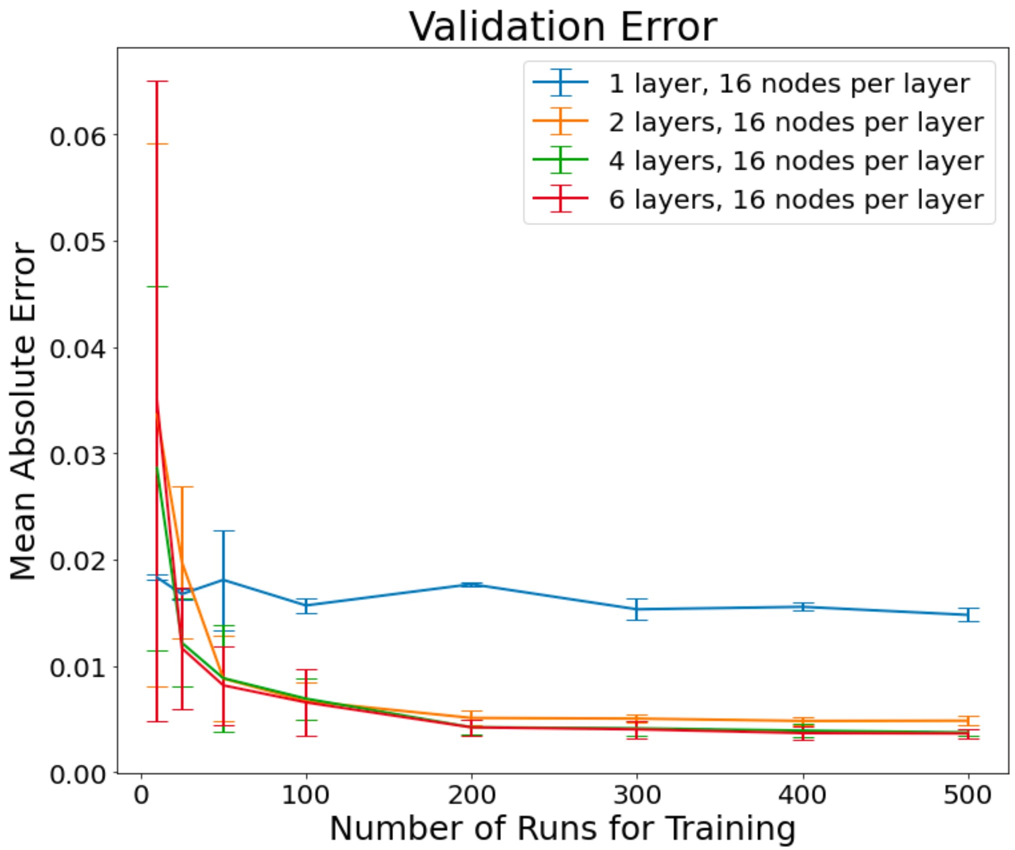


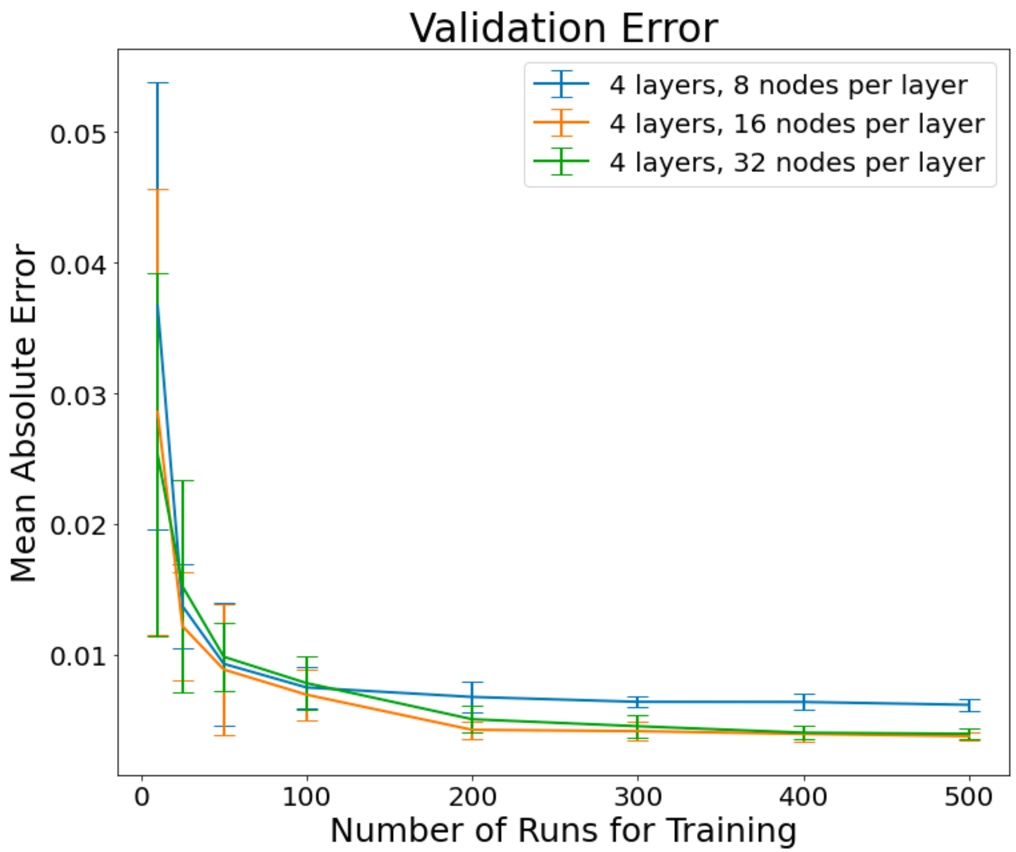
- 1-step Adams-Moulton
- Data equidistant in log(time)
- For more details, see Raissi 2018

Testing error plateaus by 20,000 epochs



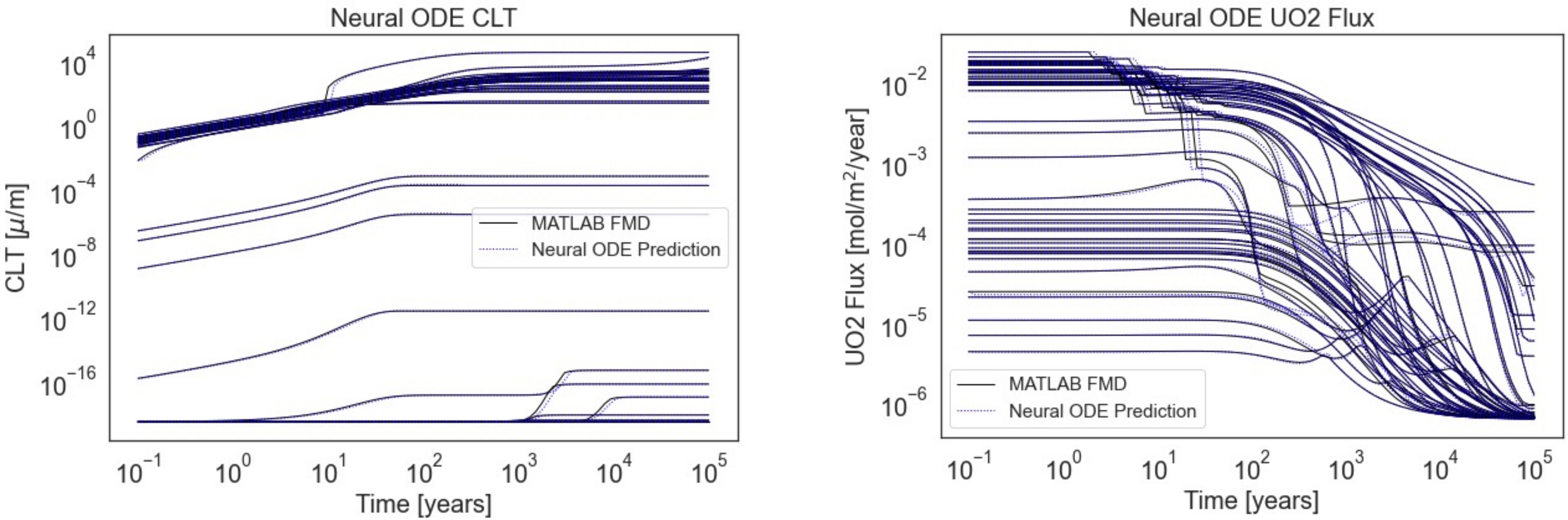
Best results are obtained with 4 layers and 16 nodes per layer





- Inputs: [CO_3^{2-}], [H_2], T, Dose Rate, CLT and UO2 Flux @ previous time, time
- Better accuracy when using more training data, but levels off at 500 runs
- Optimal accuracy for 4 layers, 16 nodes

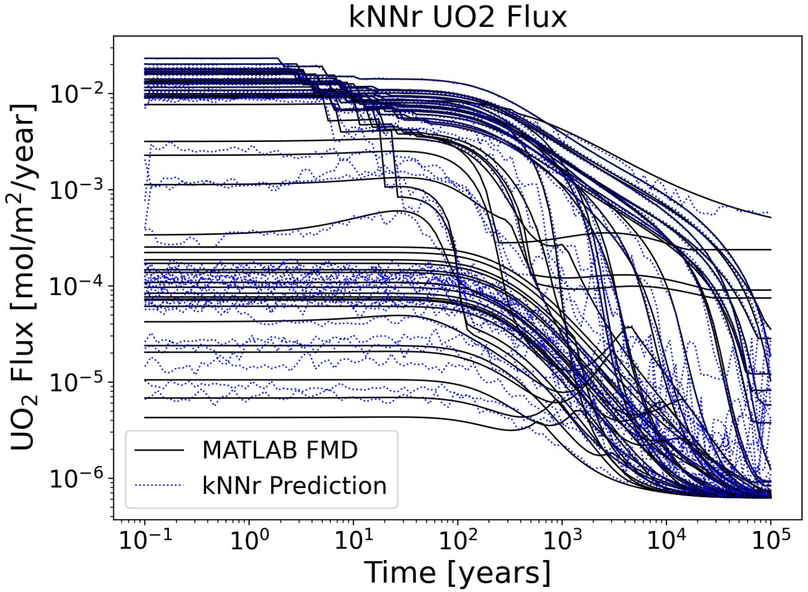
Neural ODE predicts the test data very well, and has fewer outliers than the regular ANN approach

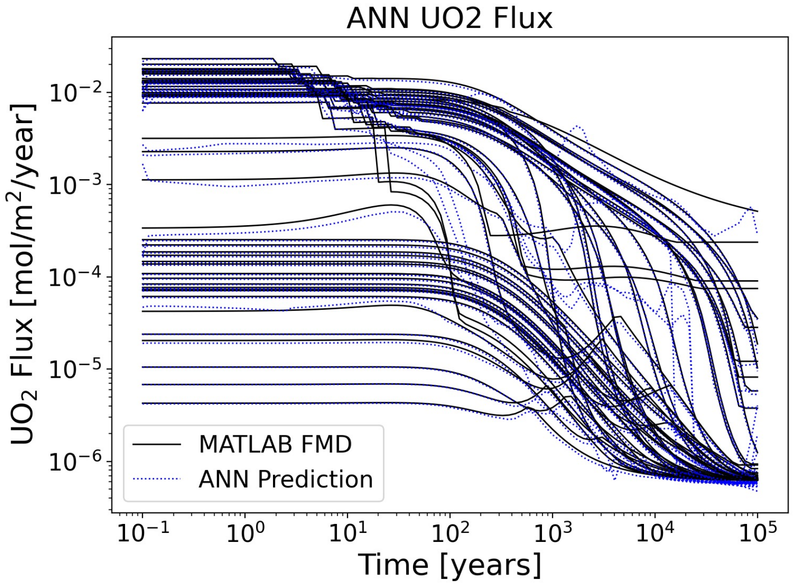


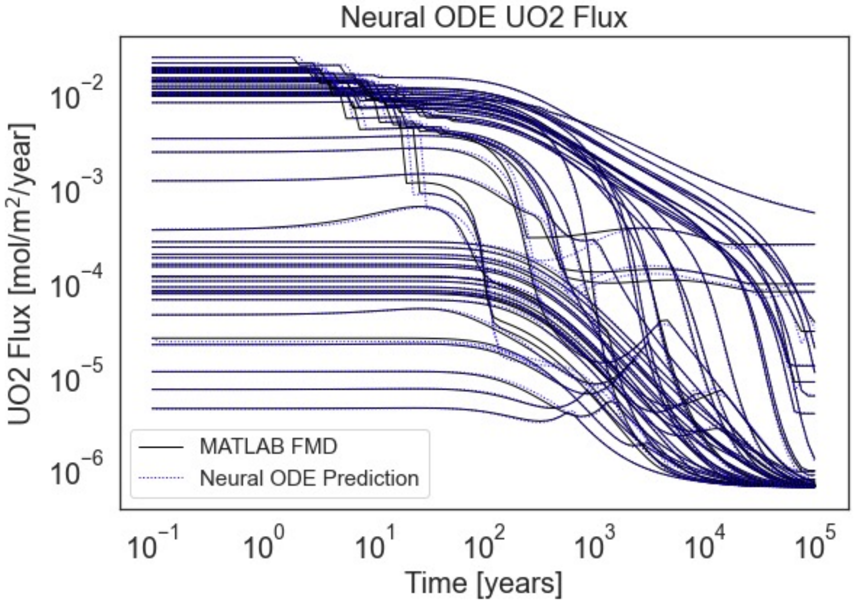
- Data integrated over a single time step only (all inputs taken from test data), to be consistent with kNNr and ANN results

Comparison kNNr – ANN – Neural ODE

The neural ODE approach gives the lowest errors on the testing data







CLT

Surrogate	nrmse	mape_f
kNNr	0.26	1.4%
ANN	0.37	2.4%
Neural ODE	0.18	0.80%

Flux

Surrogate	nrmse	mape
kNNr	0.11	29%
ANN	0.12	14%
Neural ODE	0.086	1.9%

- Neural ODEs use the UO_2 flux at the current time step as input, whereas kNNr and ANN do not

Conclusions

Conclusions and Ongoing Work

- Machine Learning offers powerful ways to approximate the FMD process model outputs
- The Neural ODE formulation lends itself well to time advancement and gives very accurate results
- Adding more internal fuel surface state information may further improve accuracy
  - But will require additional surrogate predictions at each time step
- Surrogate models enable more detailed FMD dynamics in repository simulations
- Ongoing work focuses on determining an appropriate description of internal fuel cask state to balance accuracy and complexity

Relevant References

1. J. Jerden, K. Frey, and W. Ebert, “A Multiphase Interfacial Model for the Dissolution of Spent Nuclear Fuel,” Journal of Nuclear Materials, 462, 135, https://doi.org/10.1016/j.jnucmat.2015.03.036 (2015)
2. S. D. Sevougian et al., GDSA Repository Systems Analysis FY19 Update. SAND2019-11942R. Sandia National Laboratories, Albuquerque, New Mexico (2019).
3. Freeze, G., E.J. Bonano, P. Swift, E. Kalinina, E. Hardin, L. Price, S. Durbin, R. Rechard, and K. Gupta, “Integration of the Back End of the Nuclear Fuel Cycle.” SAND2021-10444. Sandia National Laboratories, Albuquerque, New Mexico (2021)
4. Bert J. Debusschere, D.T. Seidl, T.M. Berg, K.W. Chang, R.C. Leone, L.P. Swiler, P.E. Mariner, “Machine Learning Surrogates of a Fuel Matrix Degradation Process Model for Performance Assessment of a Nuclear Waste Repository,” Nuclear Technology (2023)
5. Bert J. Debusschere, D. Thomas Seidl, Timothy M. Berg, Kyung Won Chang, Rosemary C. Leone, Laura P. Swiler, and Paul E. Mariner, “Machine Learning Surrogate Process Models for Efficient Performance Assessment of a Nuclear Waste Repository,” IHLRWM 22 (2022)
6. J. Harvey et al.: Development of an Efficient Version of the Fuel Matrix Degradation Model, IHLRWM 22 (2022)
7. Maziar Raissi, Paris Perdikaris, George Em Karniadakis, “Multistep Neural Networks for Data-driven Discovery of Nonlinear Dynamical Systems,” https://doi.org/10.48550/arXiv.1801.01236 (2018).

Additional Materials

US Department of Energy (DOE) Office of Nuclear Energy

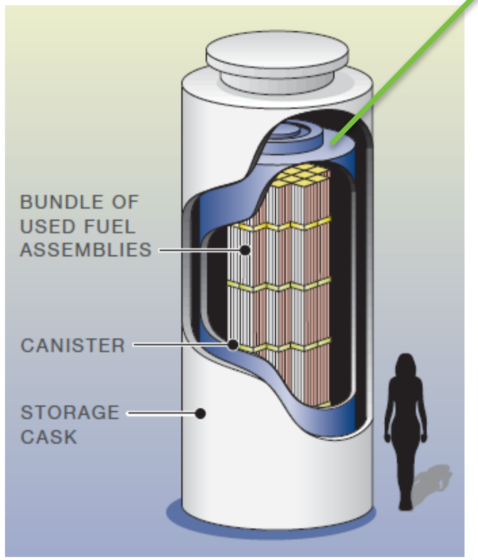
- Spent Fuel and Waste Science and Technology (SFWST)
  - Research and Development (R&D) Campaign (2010 – current)
- Mission
  - To identify alternatives and conduct scientific research and technology development to enable storage, transportation and disposal of used nuclear fuel and wastes generated by existing and future nuclear fuel cycles
- Mission work
  - Storage and transportation R&D
  - Disposal R&D

"Geological disposal remains the only long-term solution available."

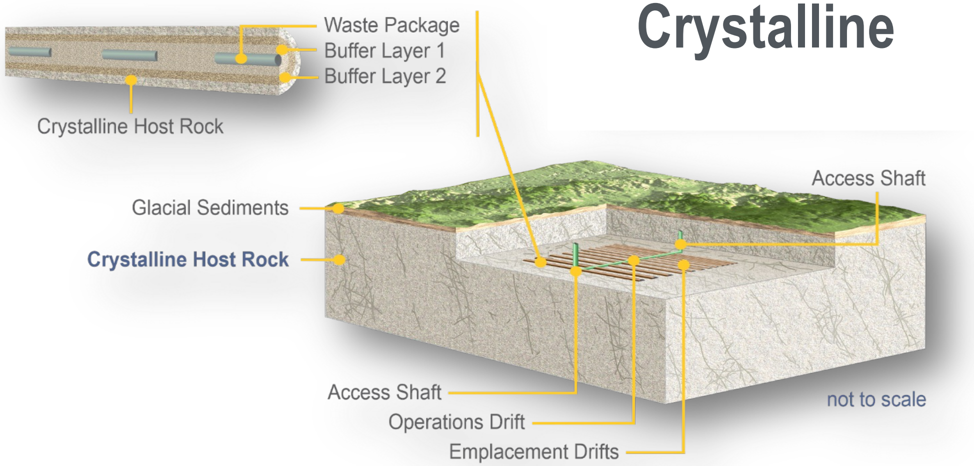
National Research Council, 2001

Deep Geologic Disposal

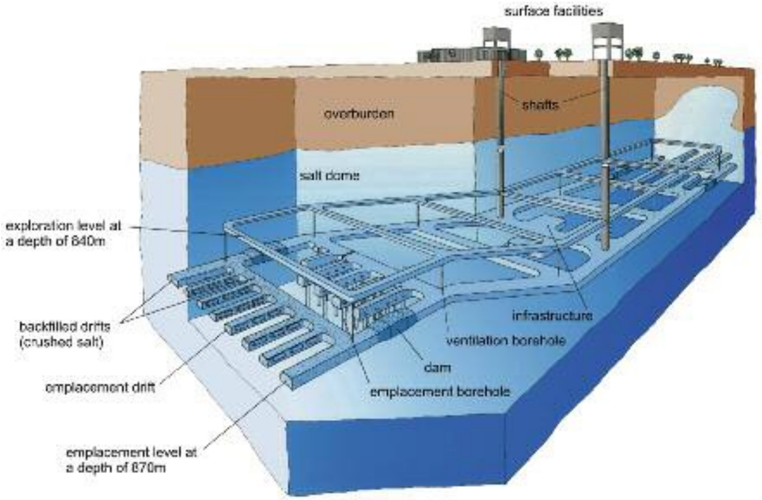
- Several possible host rocks in US
- Investigating direct disposal of dual-purpose canisters (DPCs)



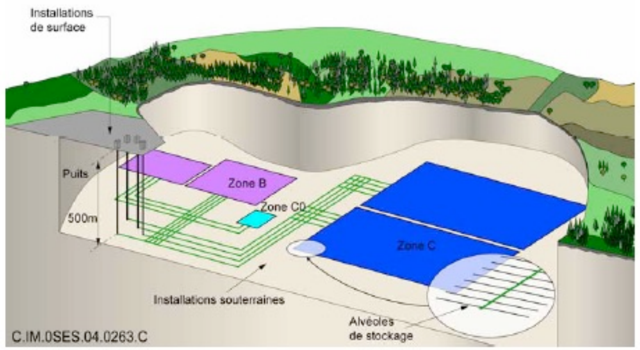
(BRC 2012, Figure 4)



Crystalline



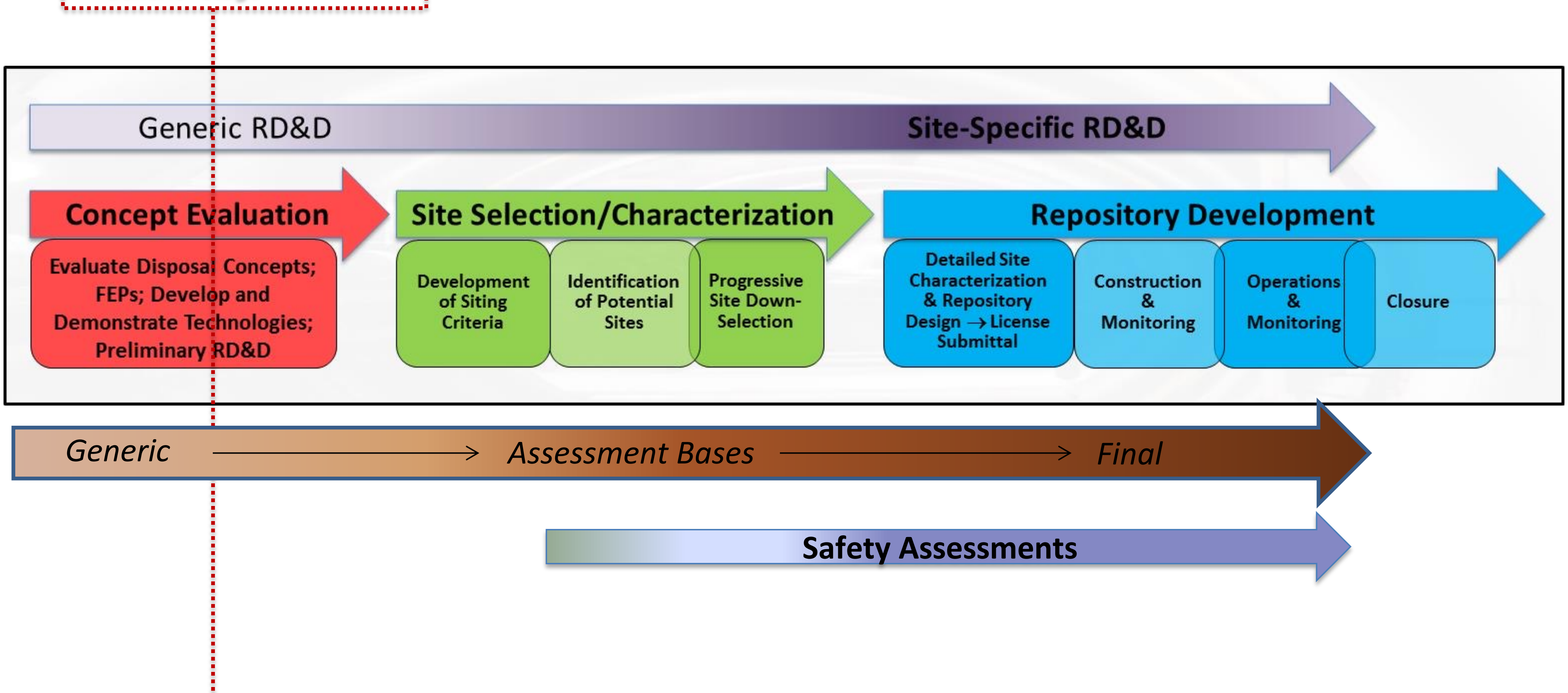
Salt



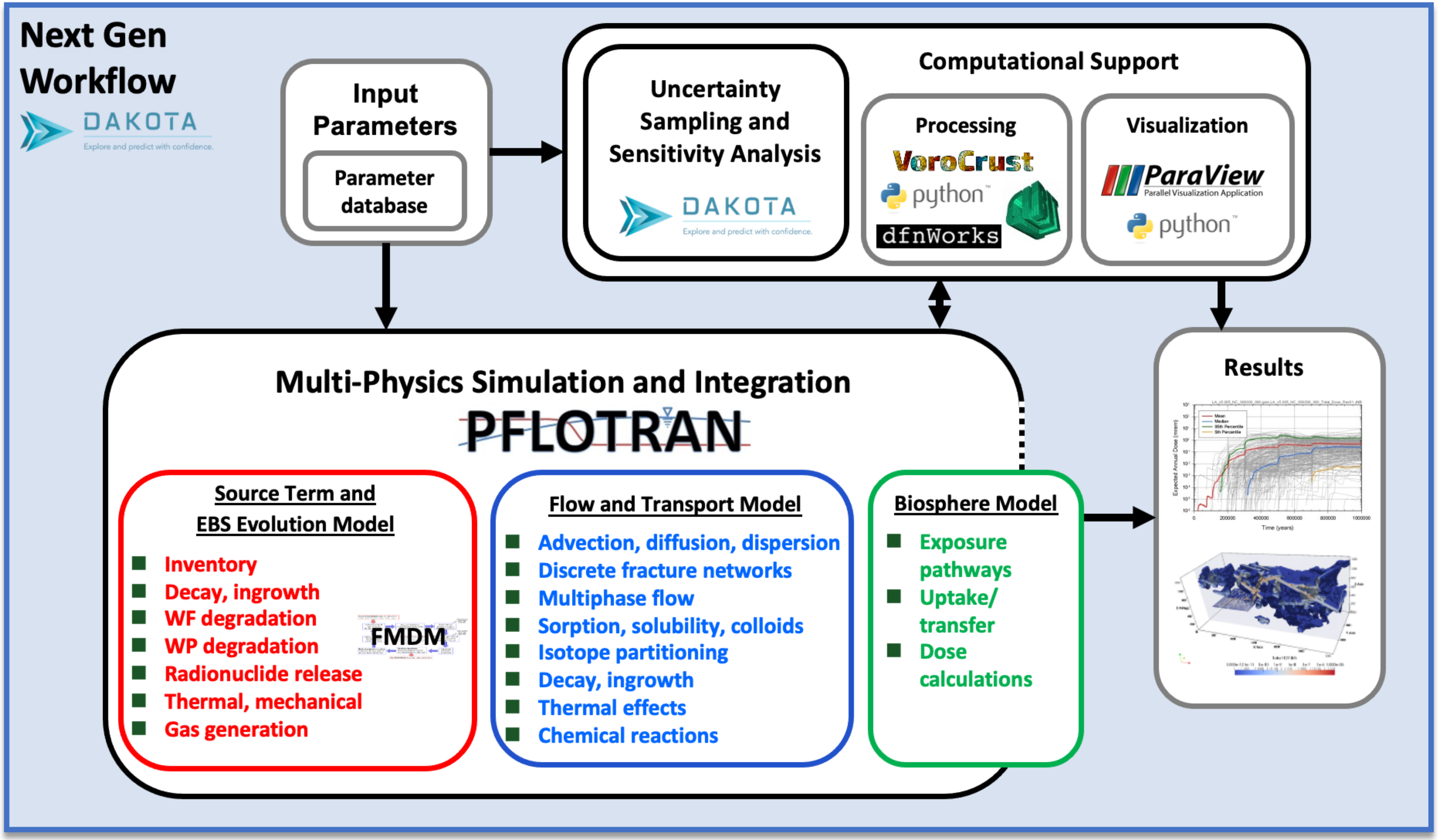
Shale/Argillite

Stages of a Deep Geologic Disposal Program

U.S. Program Currently:
- *Concept Evaluation stage*
- *"Generic" stage*



Performance Assessment R&D



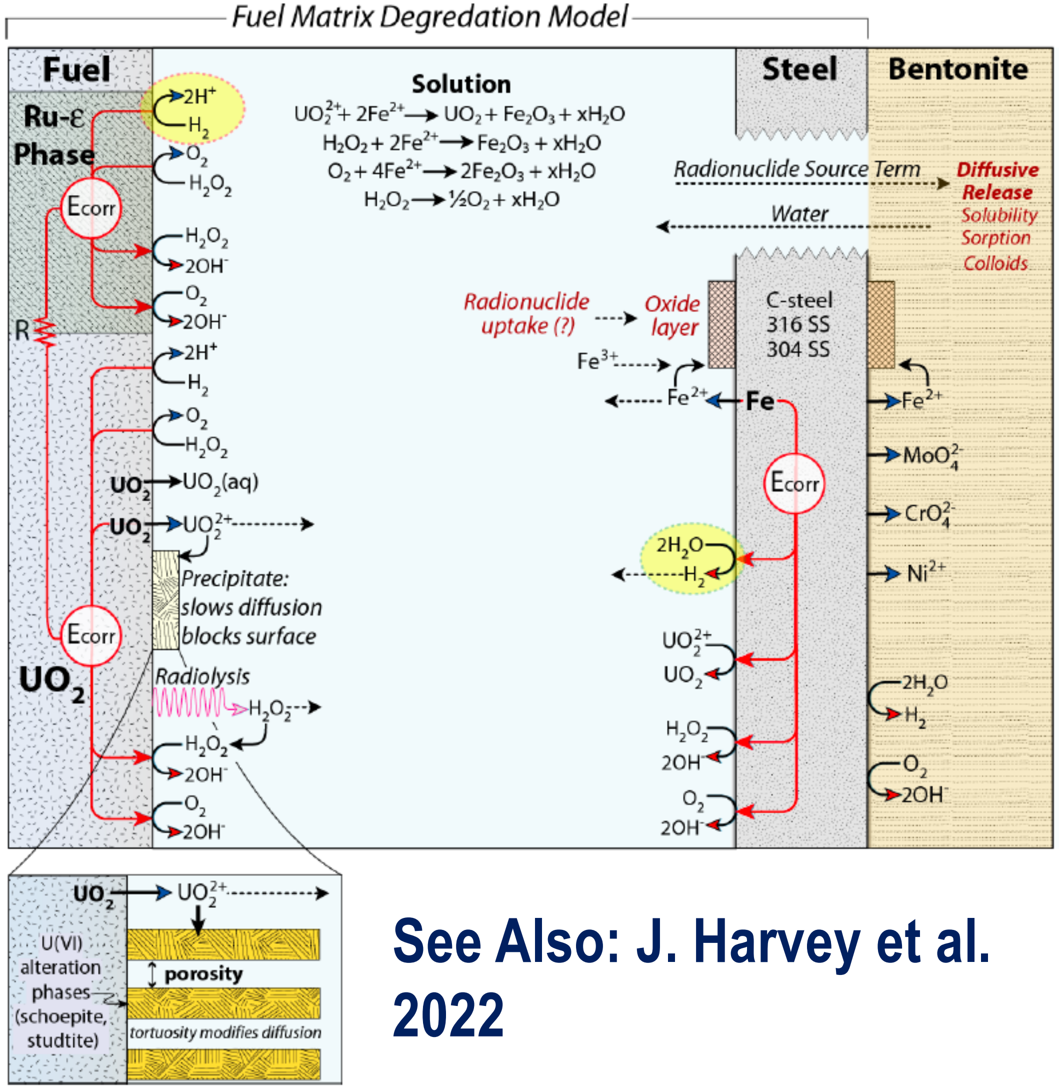
GDSA Framework

- Geologic disposal safety assessment (GDSA) framework
- PFLOTRAN for multi-physics simulation
- Dakota for probabilistic performance assessment (PA)
- dfnWorks for DFN tools
- Open source
- Massively parallel
- Freely available (pa.sandia.gov)

The Need for Surrogate Models

The Fuel Matrix Degradation (FMD) process model computes the degradation rate of spent nuclear fuel

- 1D reactive-transport model (diffusion only)
- Chemical (slow) and oxidative (fast) dissolution of UO_2 matrix
- Hydrogen peroxide production via alpha-radiolysis
- Precipitation and dissolution of U(VI) (i.e., schoepite) corrosion layer at the fuel surface
- Arrhenius temperature dependence
- Complexation of uranium with carbonates
- Hydrogen as an oxidation sink (focused on fuel interface)
- Logarithmic spatial discretization for enhanced accuracy near the solid interfaces



See Also: J. Harvey et al. 2022

Surrogate FMD models can alleviate cost of UO_2 flux computation in probabilistic repository assessments

Input Parameters

Parameter database

Computational Support

Uncertainty Sampling and Sensitivity Analysis

DAKOTA
Explore and predict with confidence.

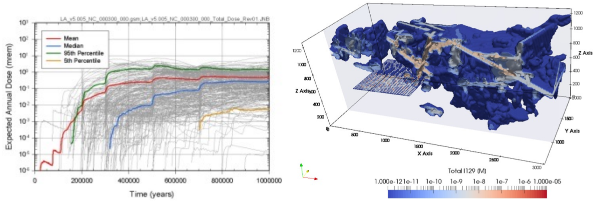
Processing

VoroCrust
python
dfnWorks

Visualization

ParaView
Parallel Visualization Application
VisIt

Results



Fuel Matrix Degradation Model (FMDM)

Offline, MATLAB

FMDM Surrogates:
- ✓ PFLOTRAN Coupled
- ✓ HPC Speed and Scale
- ✓ Fortran

Multi-Physics Simulation and Integration

PFLOTRAN

Models

Source Term and Engineered Barrier System Evolution Model

Inventory
Decay, ingrowth
Waste form degradation
Waste package degradation
Radionuclide release
Thermal, mechanical
Gas generation

Biosphere Model

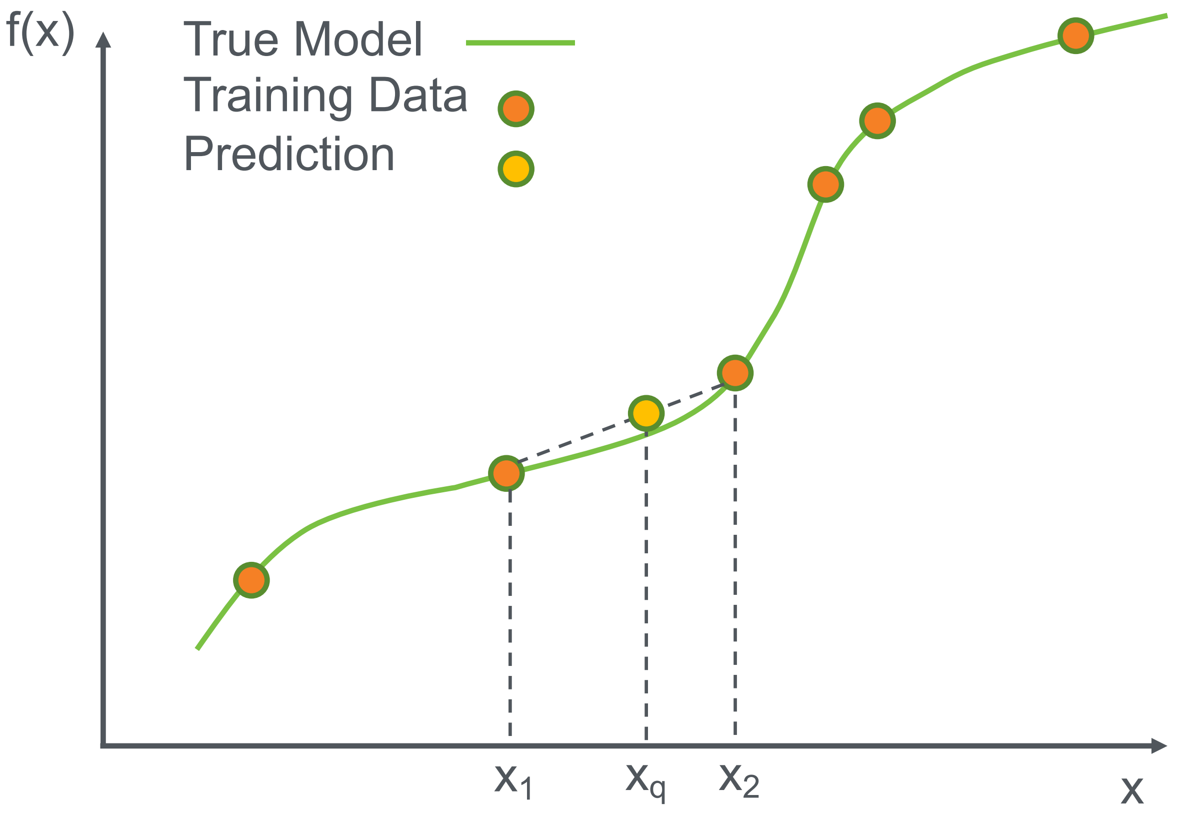
Exposure pathways
Uptake/ transfer
Dose calculations

Flow and Transport Model

Advection, diffusion, dispersion
Discrete fracture networks
Multiphase flow
Sorption, solubility, colloids
Isotope partitioning
Decay, ingrowth
Thermal effects
Chemical reactions

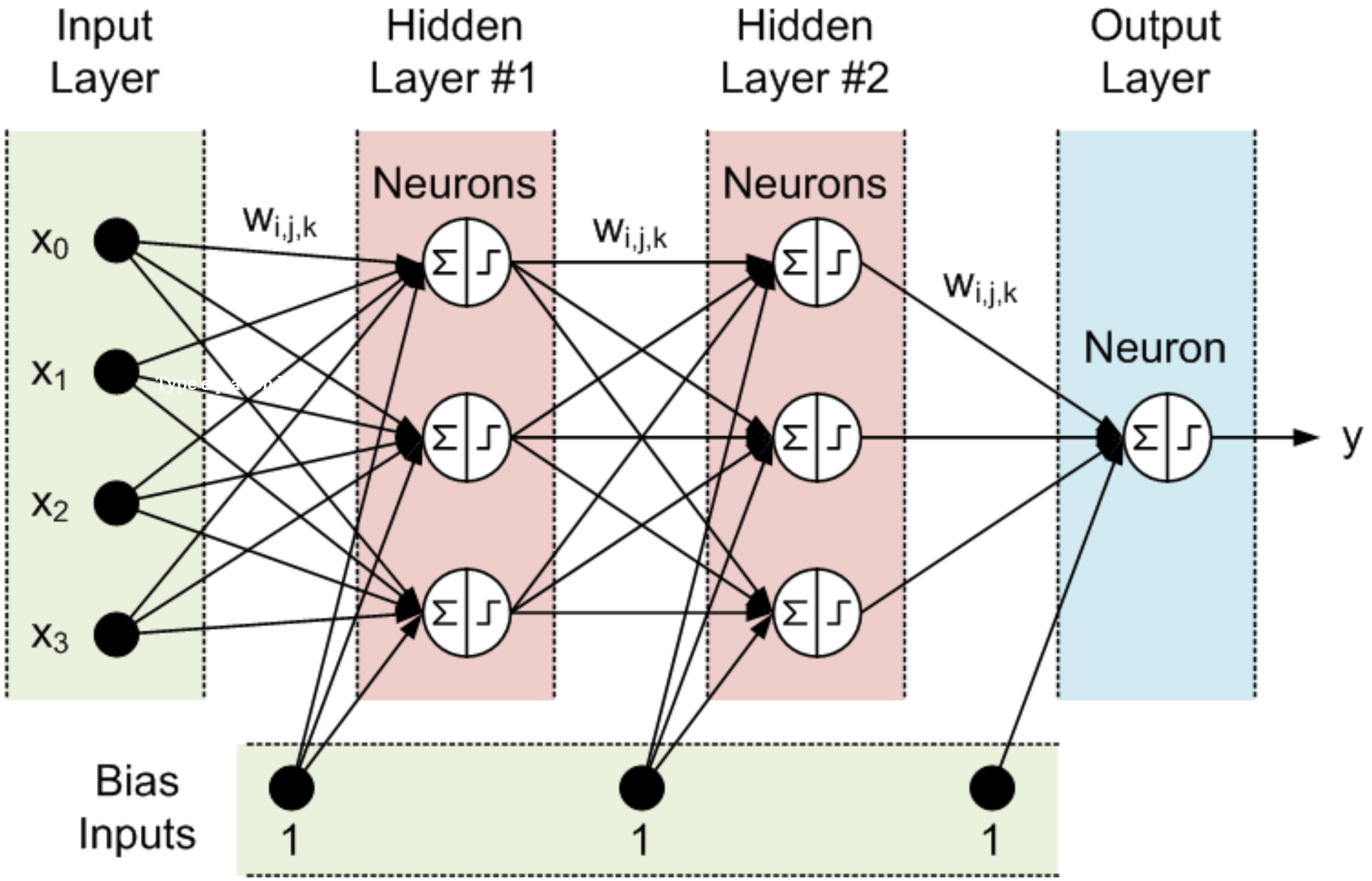
k-Nearest Neighbor regression (kNNr) interpolates between points in the training data closest to the query point

- Generalization of table lookup in higher dimensional setting
- Local approximation
- Inverse distance weighting means no training error
- Kd-Tree structure offers efficient table search
- Hyperparameters to tune:
  - Amount of training data
  - Number of nearest neighbors to use in interpolation



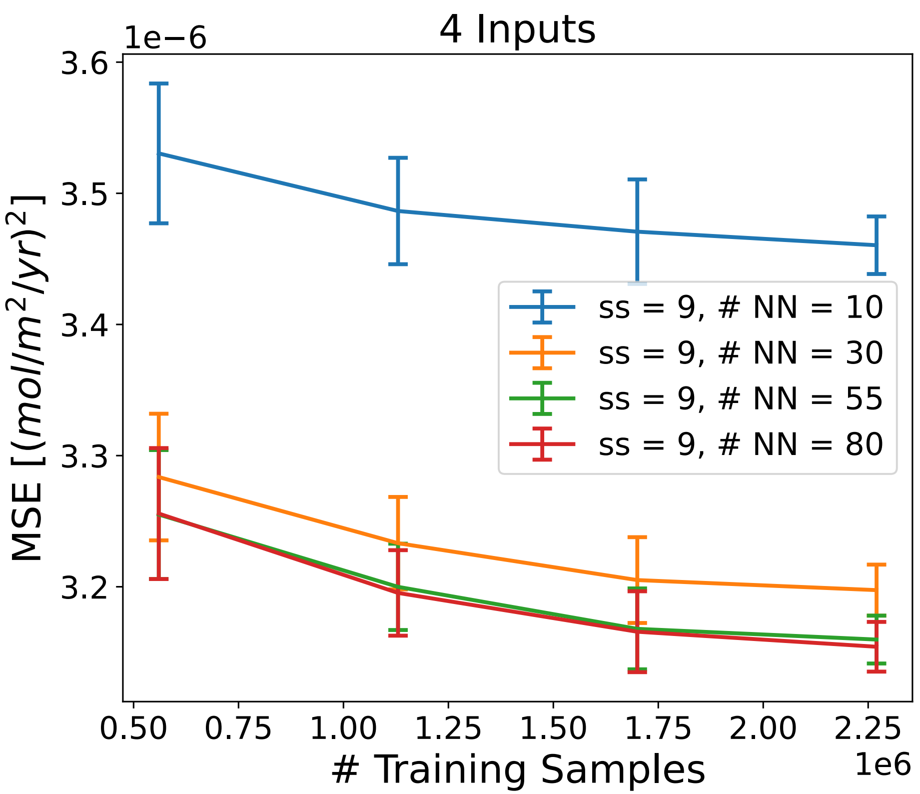
Artificial Neural Networks (ANN) approximate a function as a weighted combination of nonlinear functions

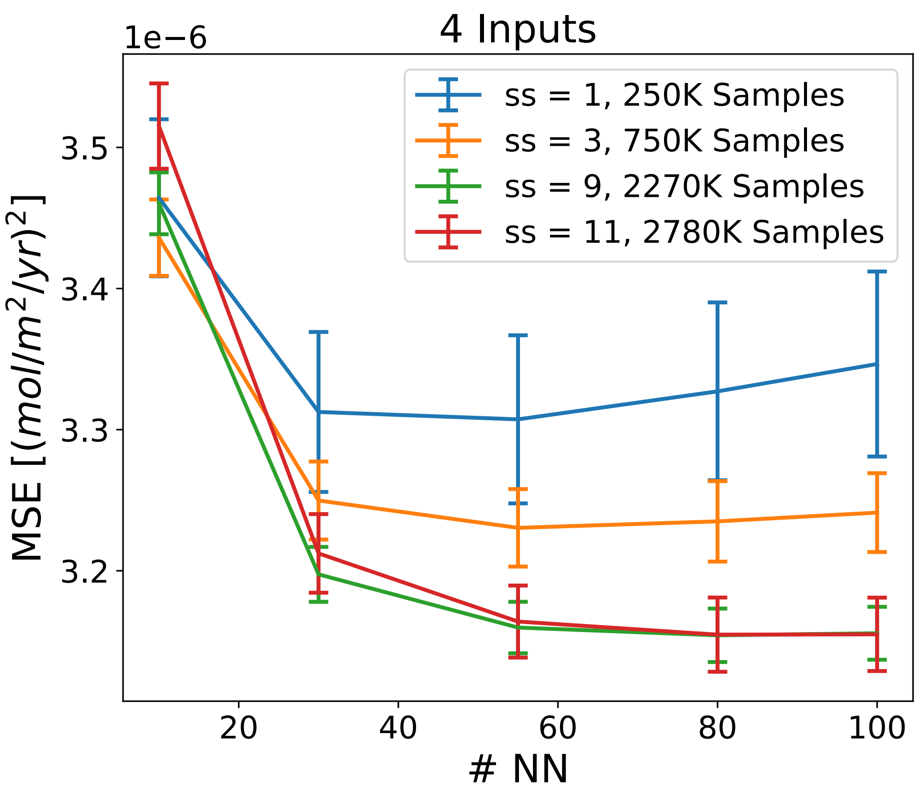
- Global functional approximation
- In each layer:
  - $y_i = f\left(b_i + \sum w_{i,j} x_j\right)$
  - ReLU activation function
- Prediction cost does not depend on amount of training data
- Hyperparameters to tune:
  - Number of layers
  - Number of nodes (neurons) per layer



Surrogate Models Based on Environmental and Global Inputs Only

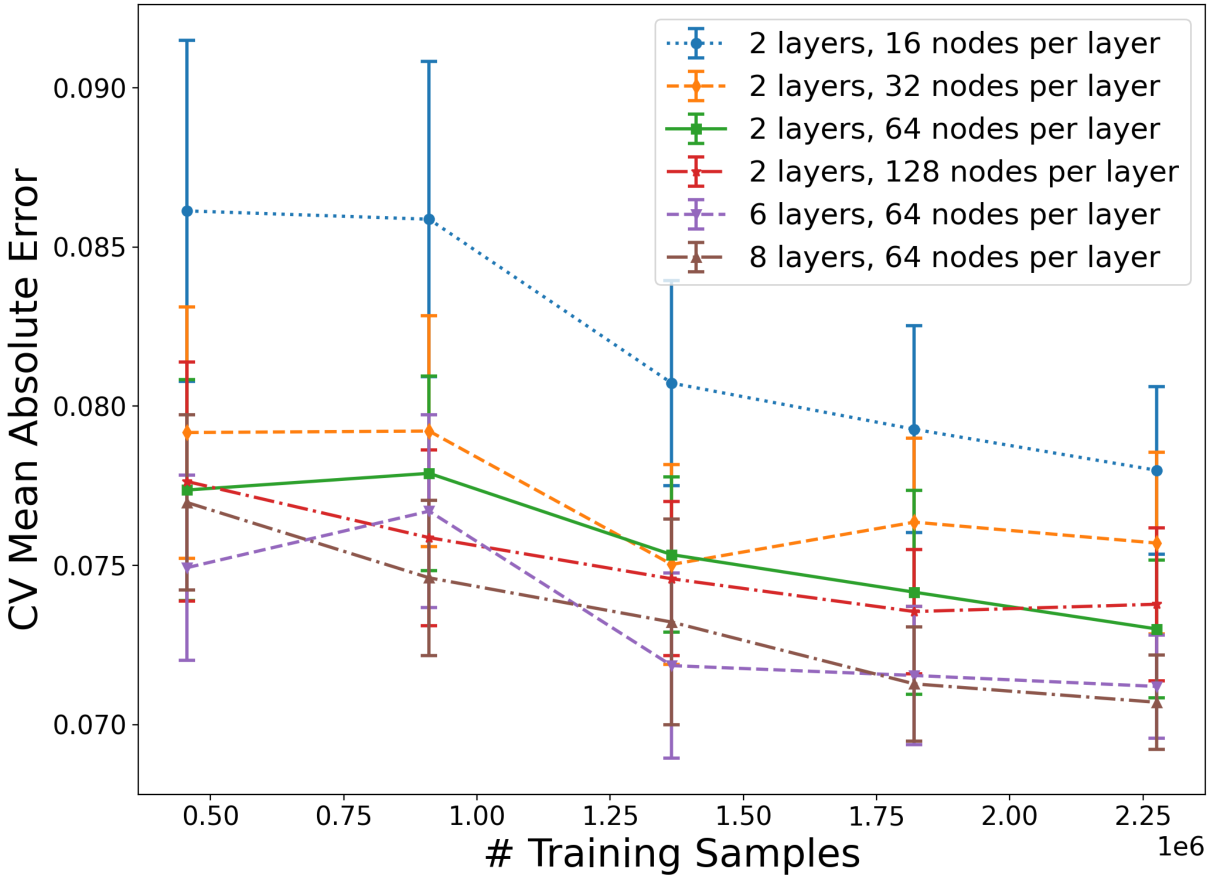
Hyperparameter tuning improves the performance for kNNr





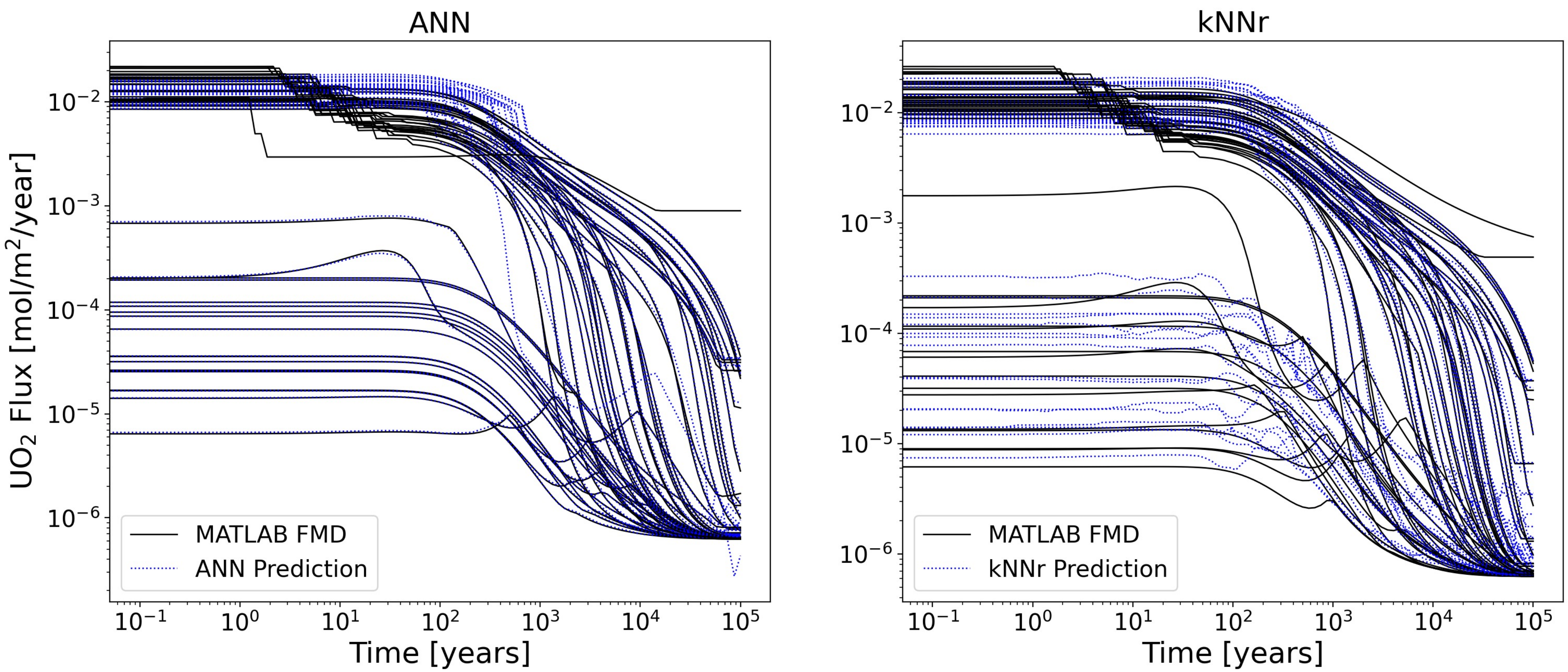
- Inputs: [CO_3^{2-}], [H_2], T, Dose Rate
- Accuracy improves with more training data
- Best accuracy with 9 subsamples per FMD process model run and 80 nearest neighbors

ANN gives optimal results with 2 layers and 64 nodes per layer



- Inputs: $[CO_3^{2-}]$, $[O_2]$, $[Fe^{2+}]$, $[H_2]$, T, Dose Rate
- Adding more layers or more nodes per layer does not significantly improve accuracy

Despite minimal input, surrogates based on only environmental inputs approximate the actual UO_2 flux quite well

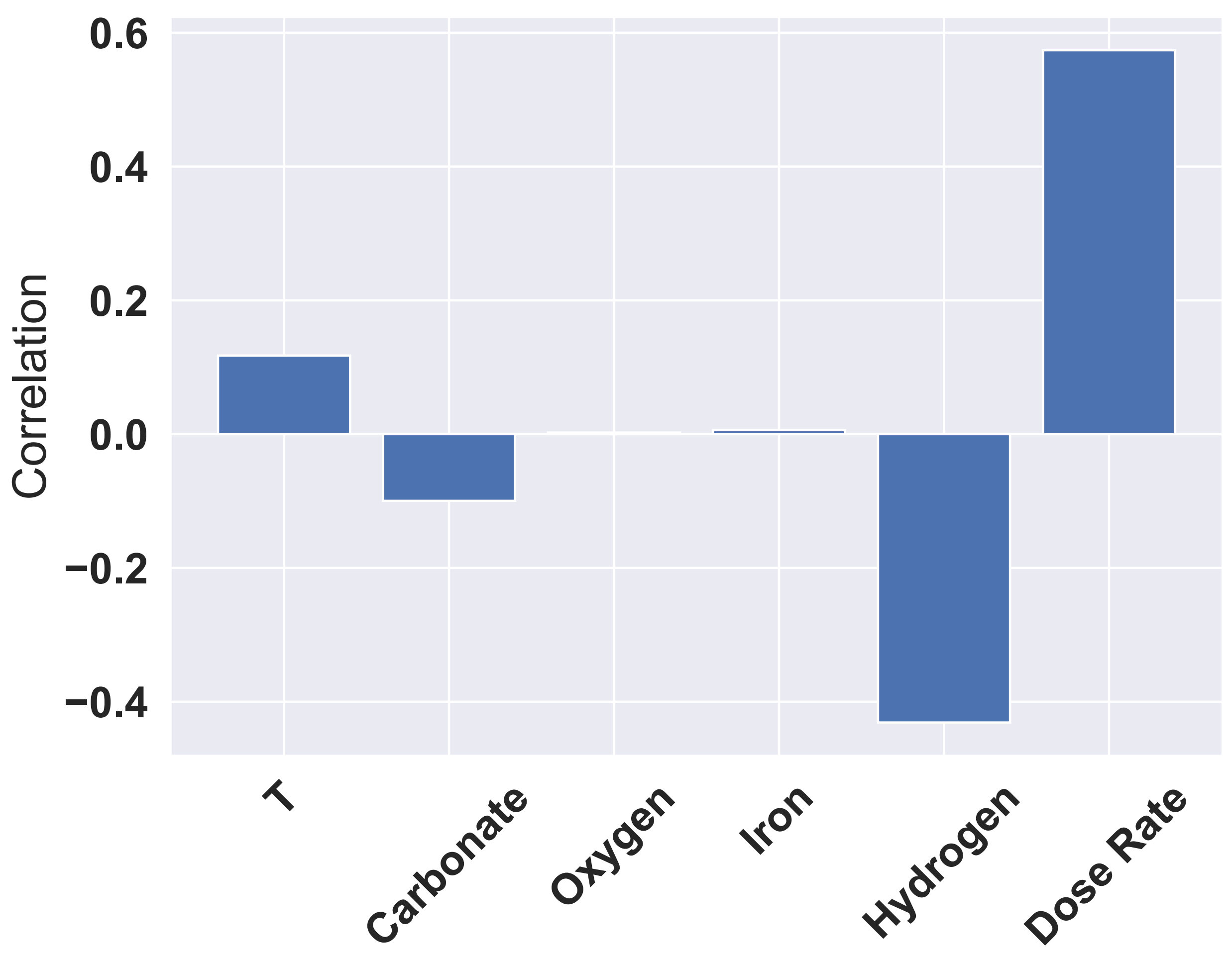


- Overprediction between 10 and 1000 years
- kNNr approximations are noisier due to the nature of the local approximation

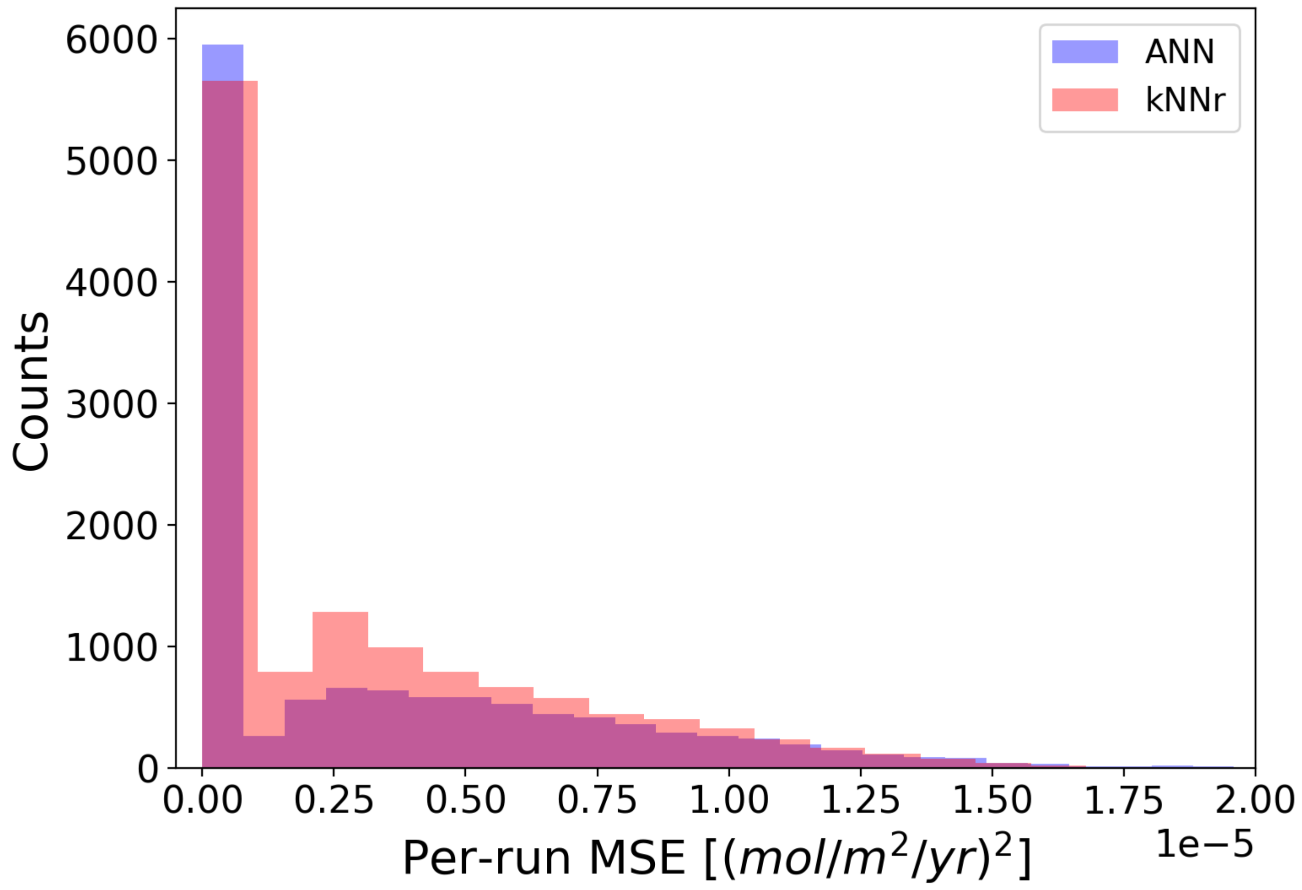
Surrogate	nrmse	mape
kNNr	0.48	44%
ANN	0.52	25%

Model inputs that do not impact the fuel degradation rate much can be dropped

- Correlation between fuel degradation rate and O_2, Fe^{2+} is very small
- Training kNNr without these species gave better accuracy
- Fewer inputs also speeds up table lookup
- ANN not as impacted by extra inputs

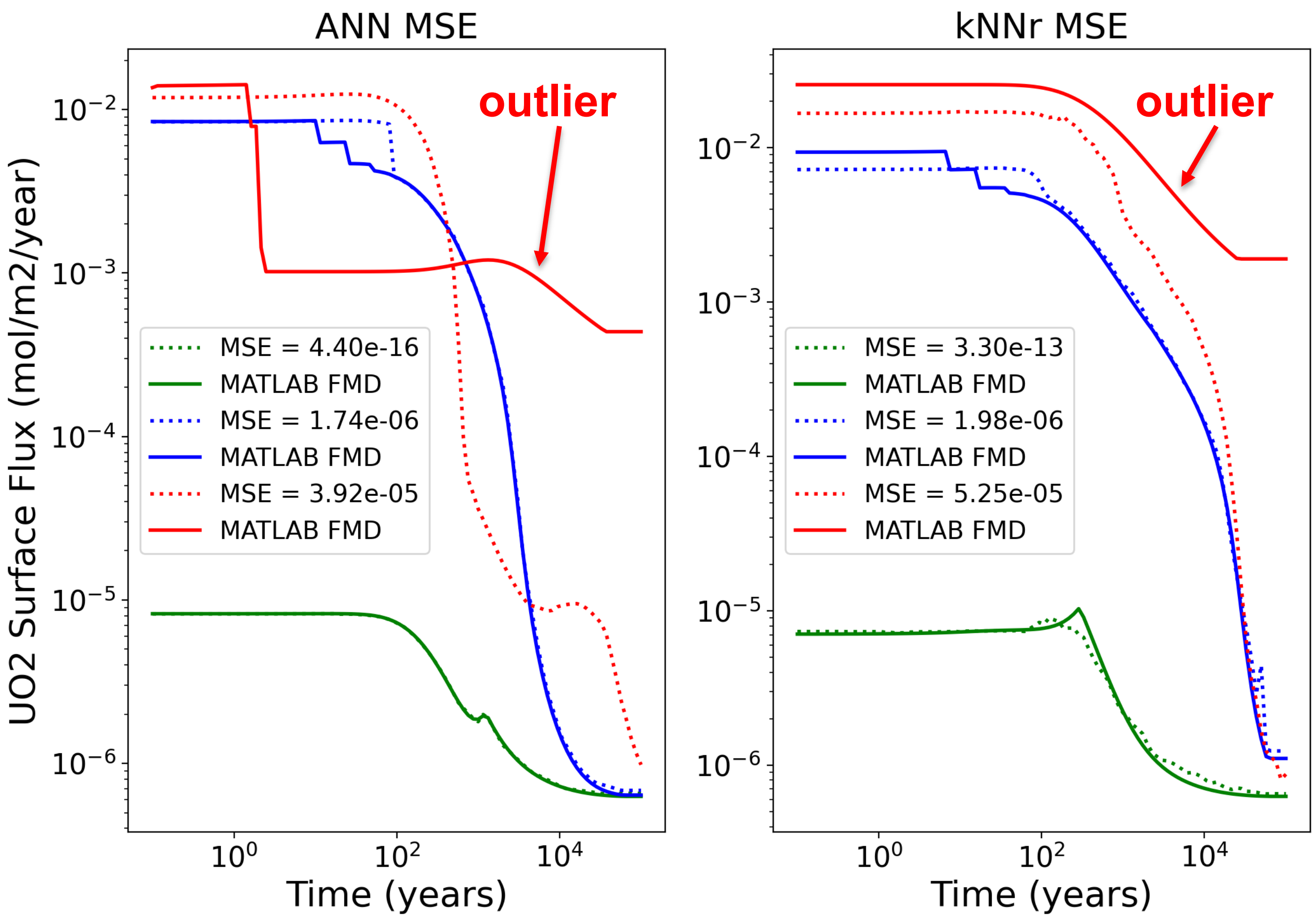


Most of the errors are very small



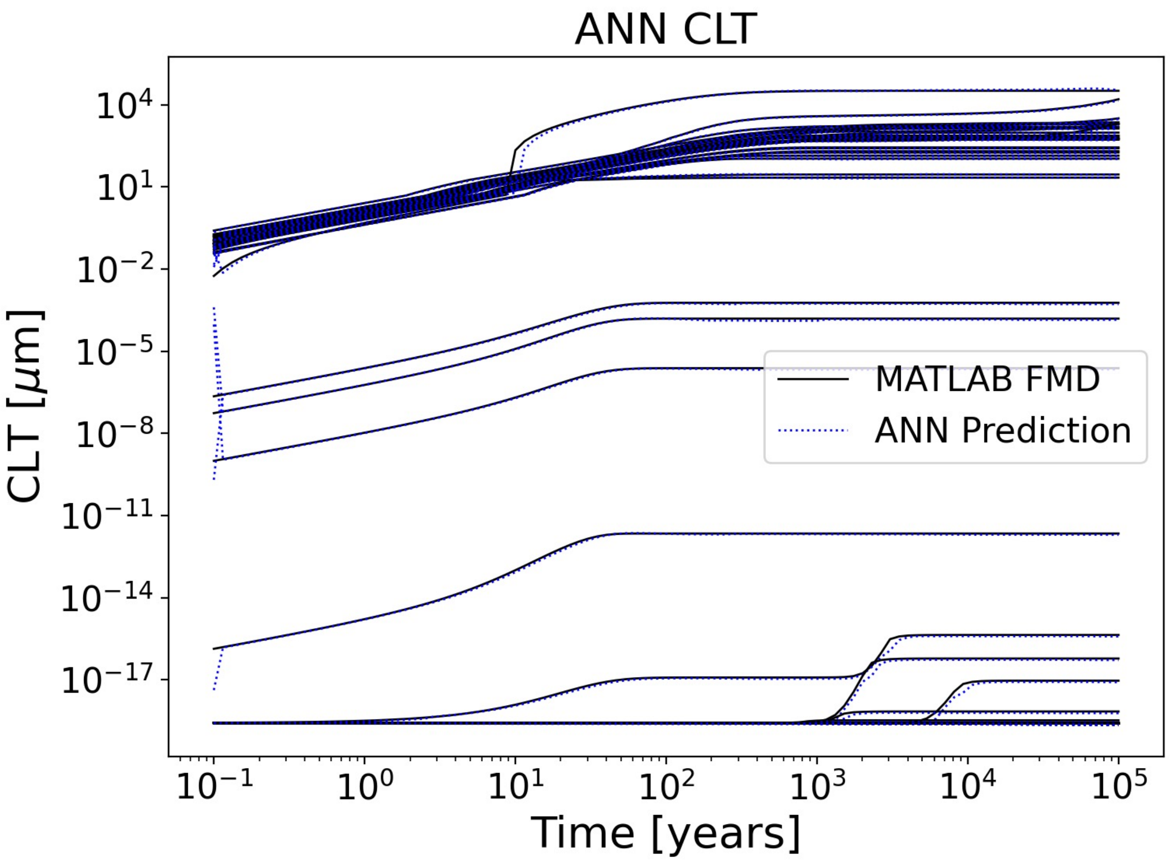
- Histogram shows MSE averaged over each FMD process model run
- Some outliers with very low probability have MSE greater than $2\ 10^{-5}$ $(mol/m^2/yr)^2$

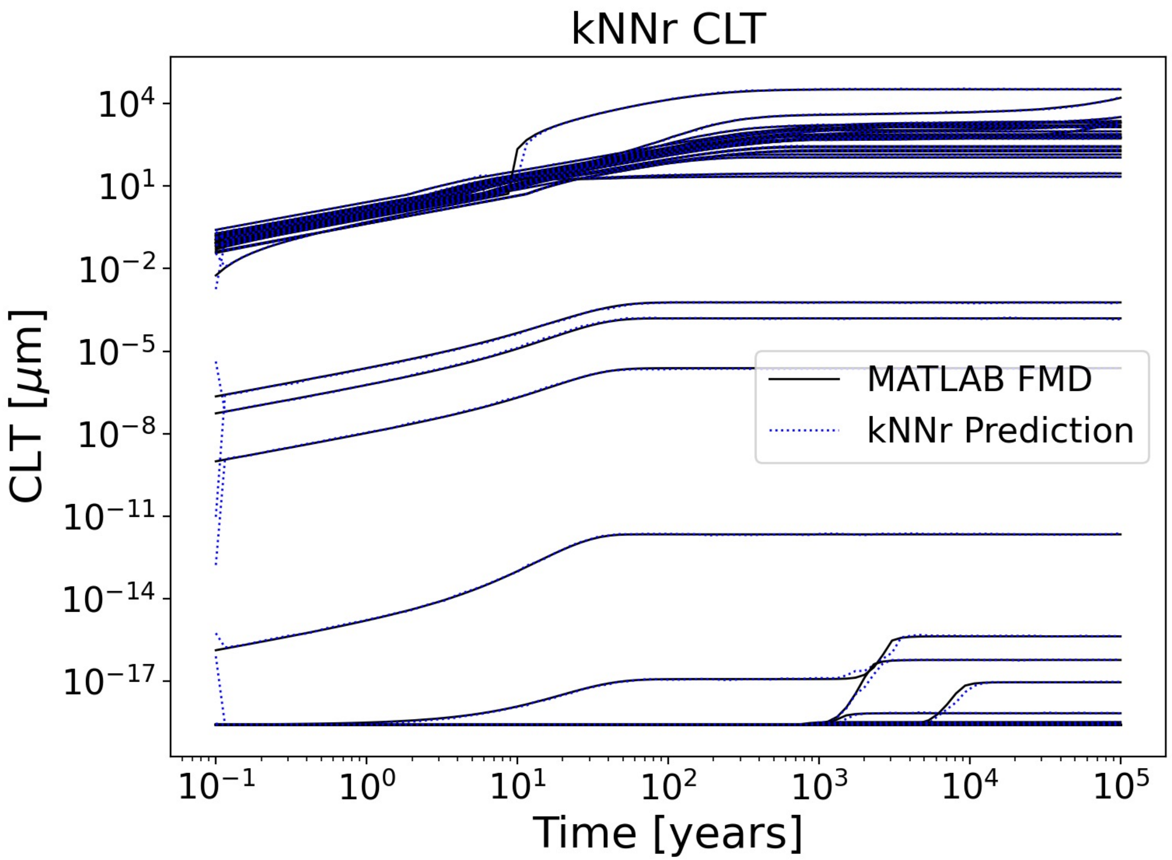
Except for outliers, the agreement with test data is adequate



Surrogate Models with CLT added

The corrosion layer thickness can be predicted with high accuracy

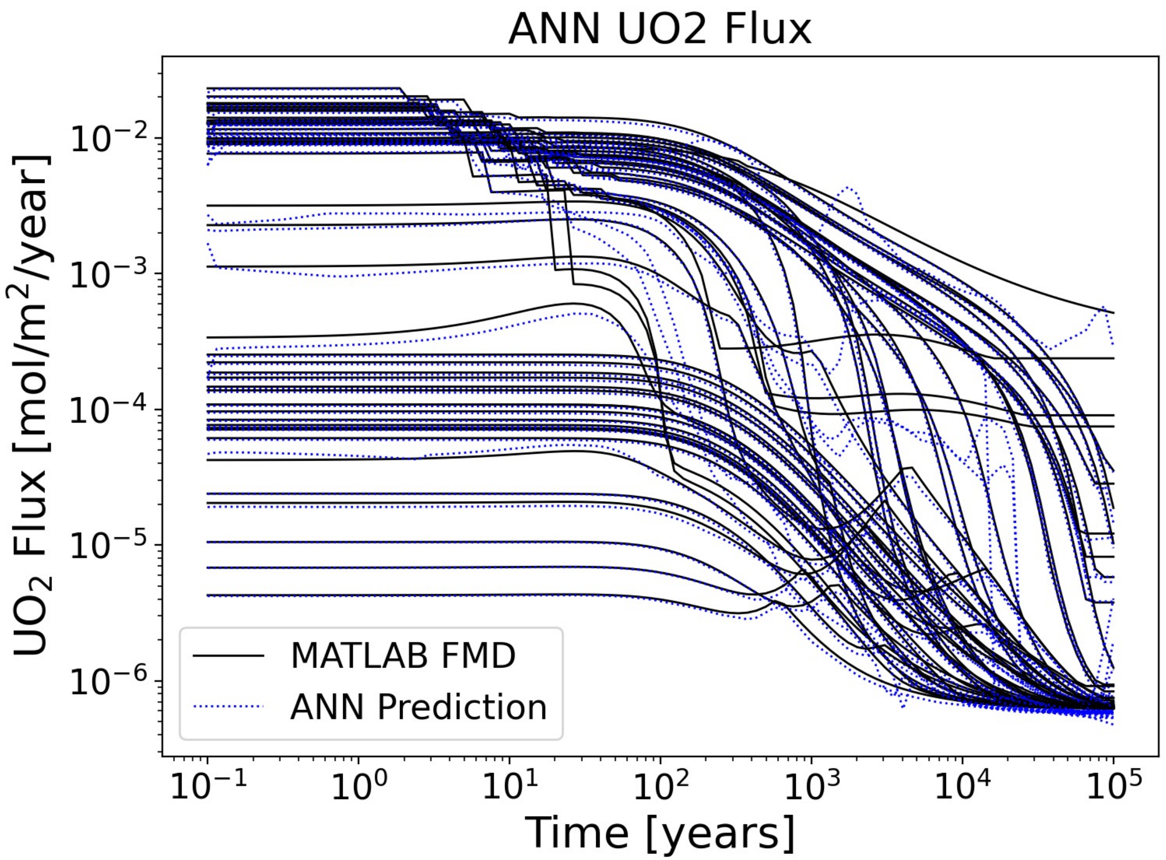


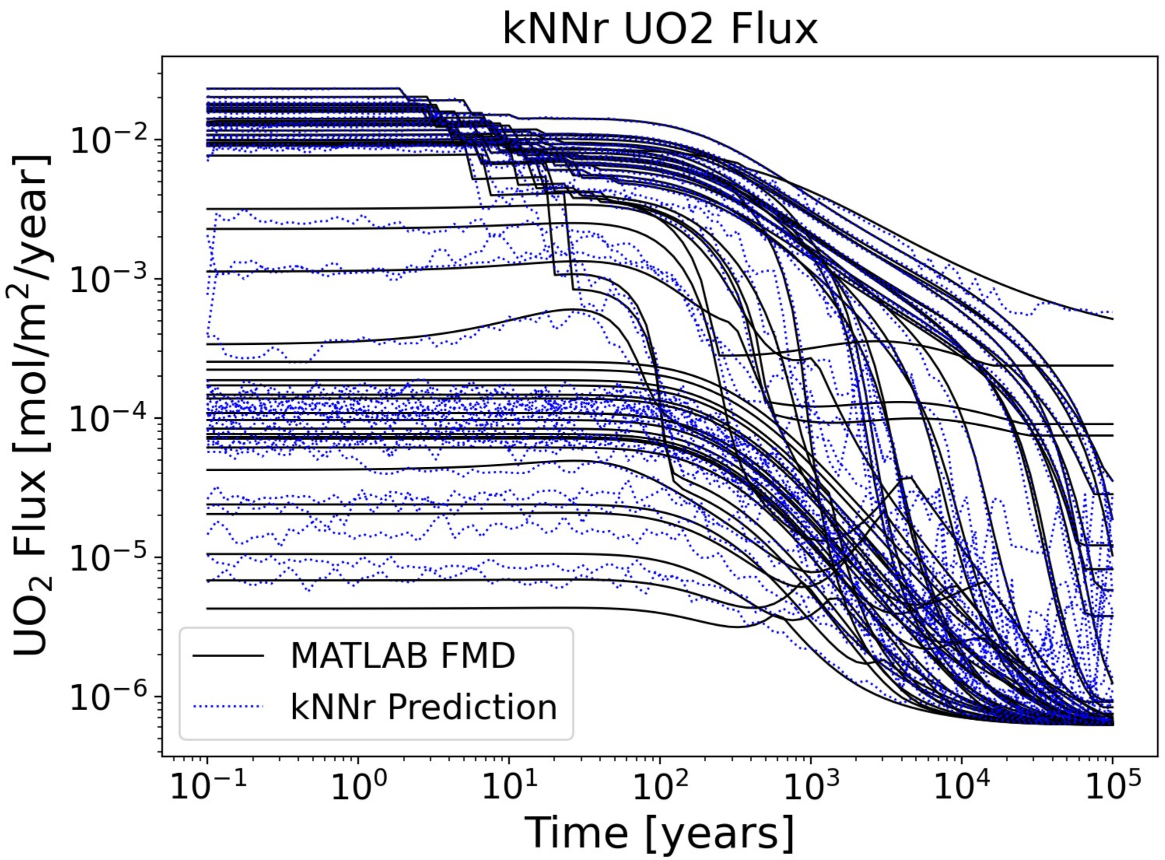


- Prediction of CLT based on test data
- No time integration used

Surrogate	nrmse	mape_f
kNNr	0.26	1.4%
ANN	0.37	2.4%

Adding the corrosion layer thickness as input gives dramatically better accuracy in fuel degradation rate prediction





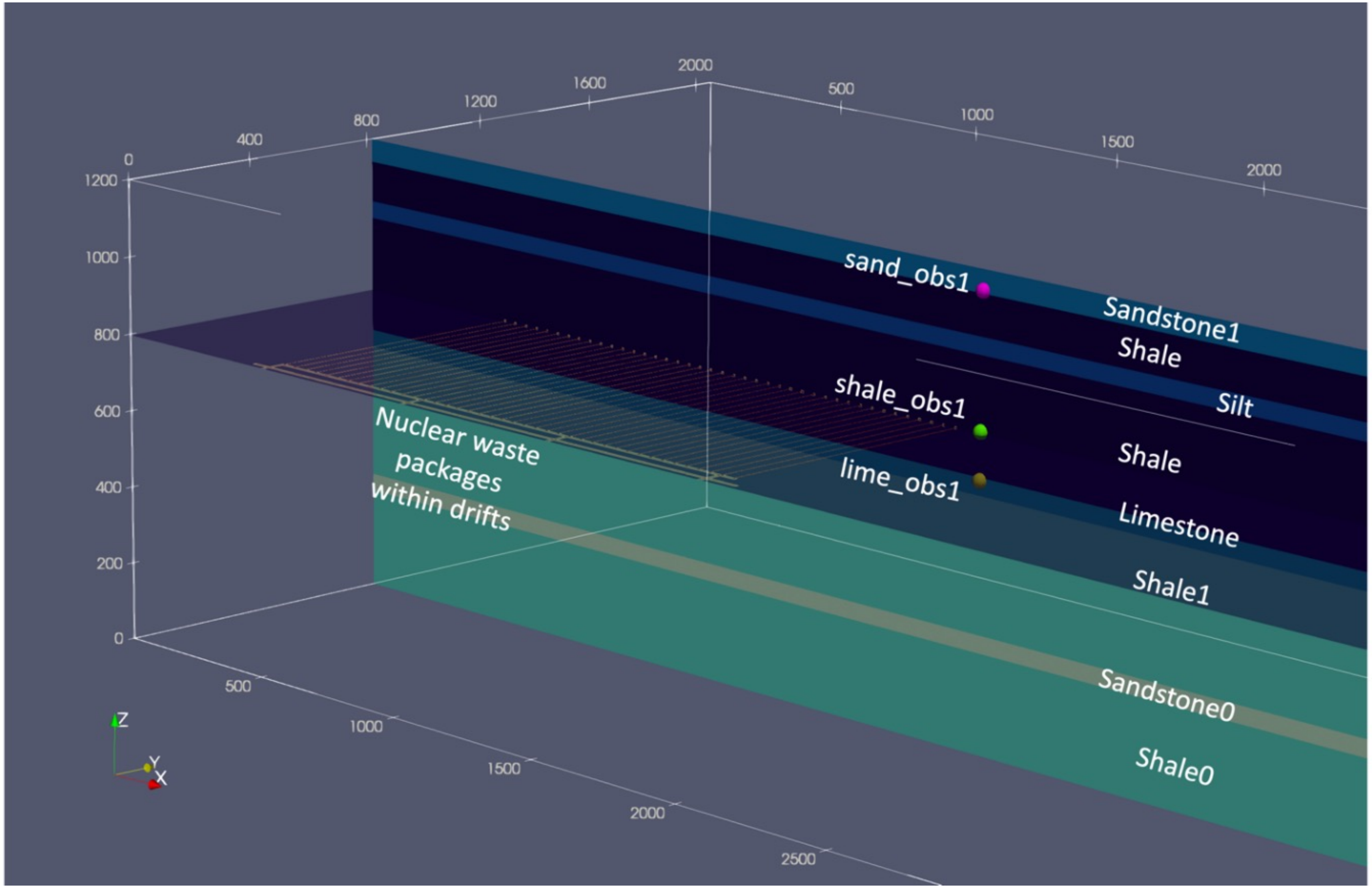
- Prediction of UO_2 flux based on test data
- No time integration used

Surrogate	nrmse	mape
kNNr	0.11	29%
ANN	0.12	14%

Application to Repository Simulation

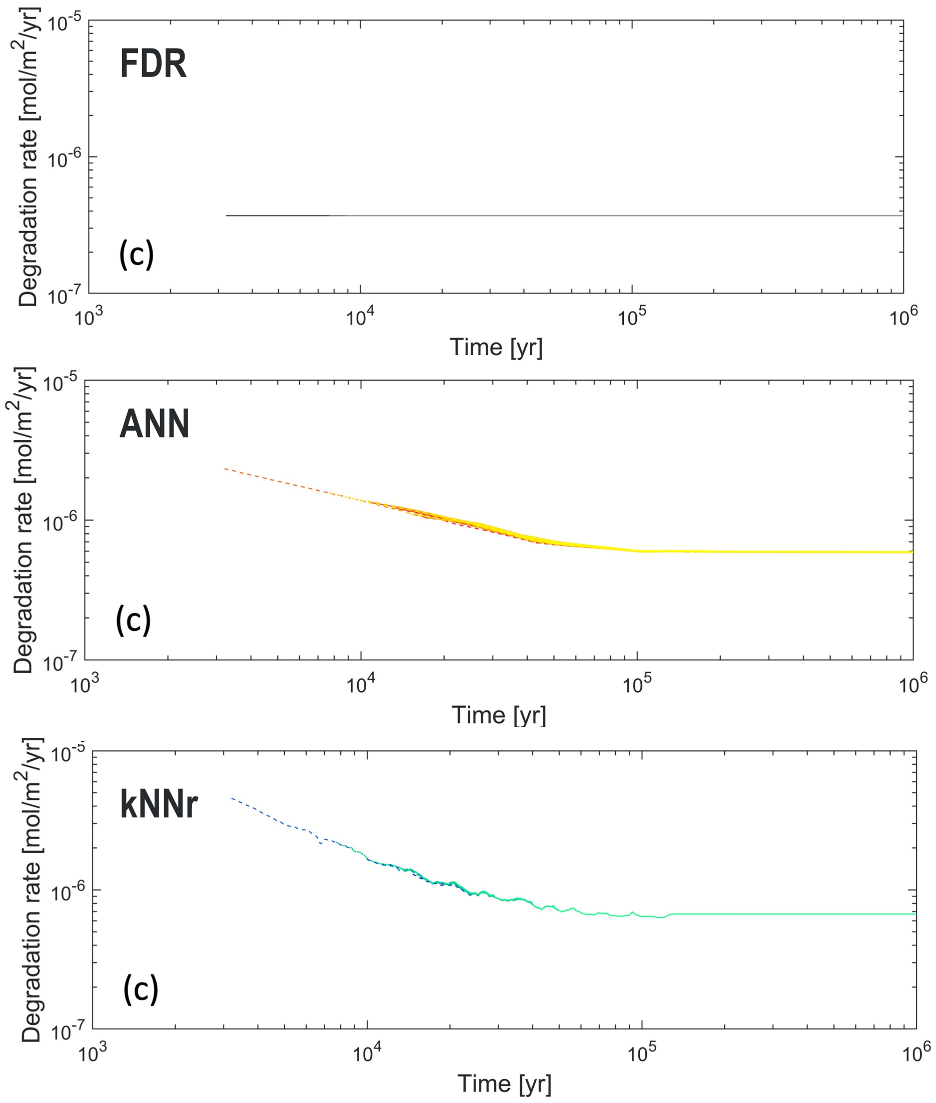
The surrogate models are demonstrated in a PFLOTRAN simulation of a generic shale repository reference case

- 2 x 41 drifts at a depth of 405m
- 10M grid cells
- 2000 4-PWR packages
  - 65 GWd/MTHM burn-up
  - 100 year Out of Reactor storage
- Sevougian et al. 2019



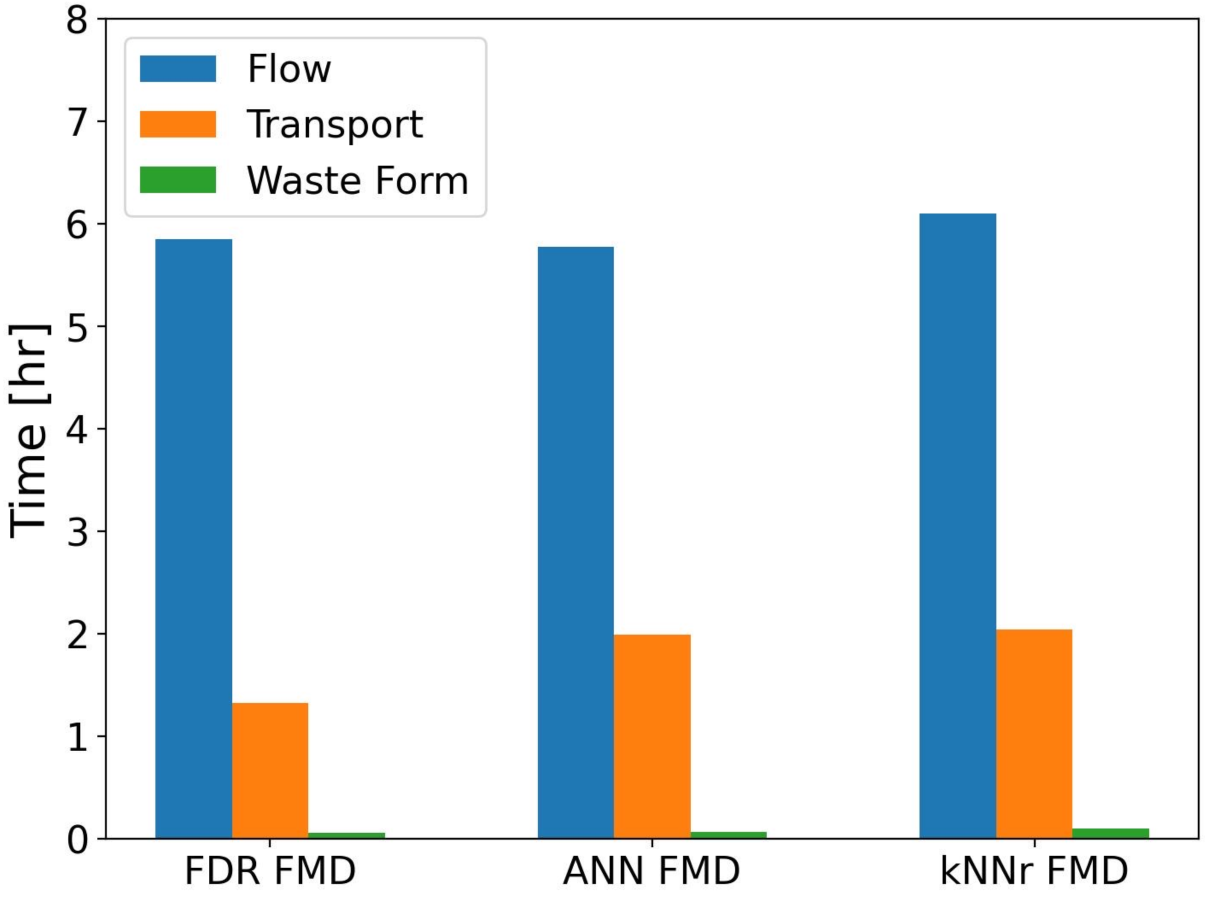
Surrogate models give more realistic prediction of fuel degradation rate than constant approximation

- Degradation starts after waste package is breached
- Fractional Degradation Rate (FDR) model assumes constant fractional rate of degradation
- ANN and kNNr surrogates provide higher fidelity by considering environmental inputs and changes in dose rate and temperature over time

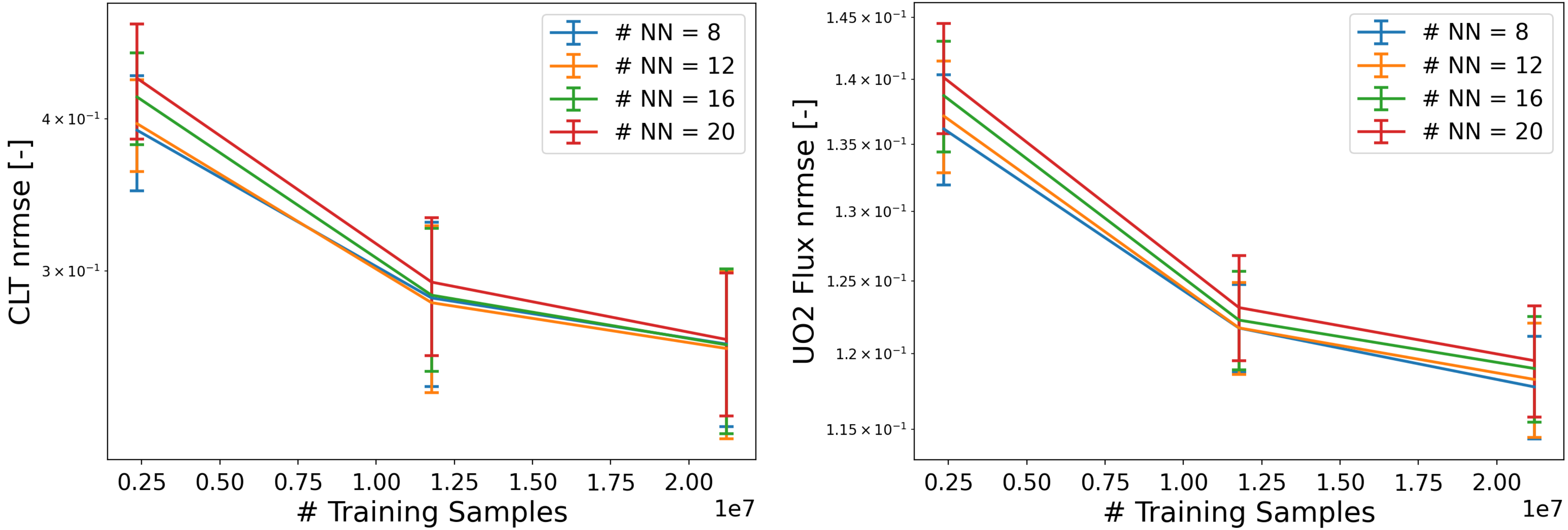


Surrogate models are comparable in computational cost to constant fractional rate approximation

- Simulations use 1024 processors
- Transport is more expensive in ANN and kNNr runs to model transport of environmental species
- Running the full FMD process model on 2000 waste packages for 1M years would not be feasible in probabilistic (UQ) setting



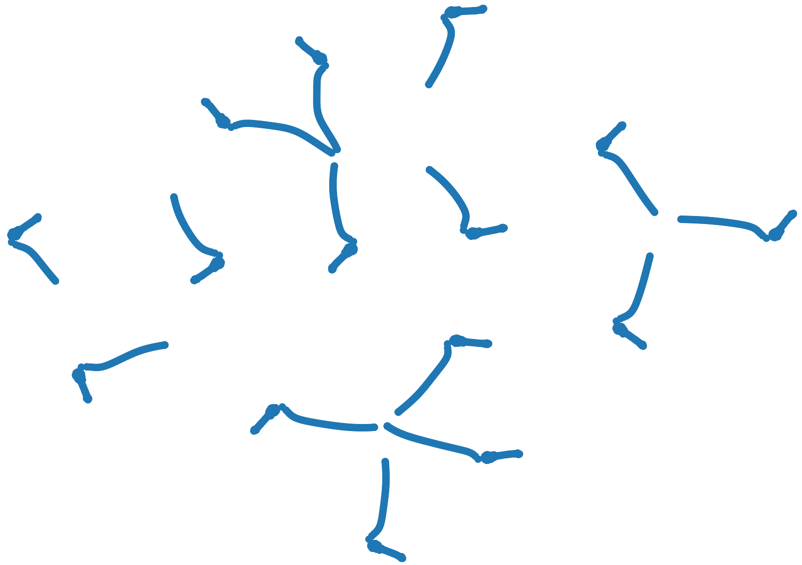
kNNr has best results with 8 – 12 nearest neighbors

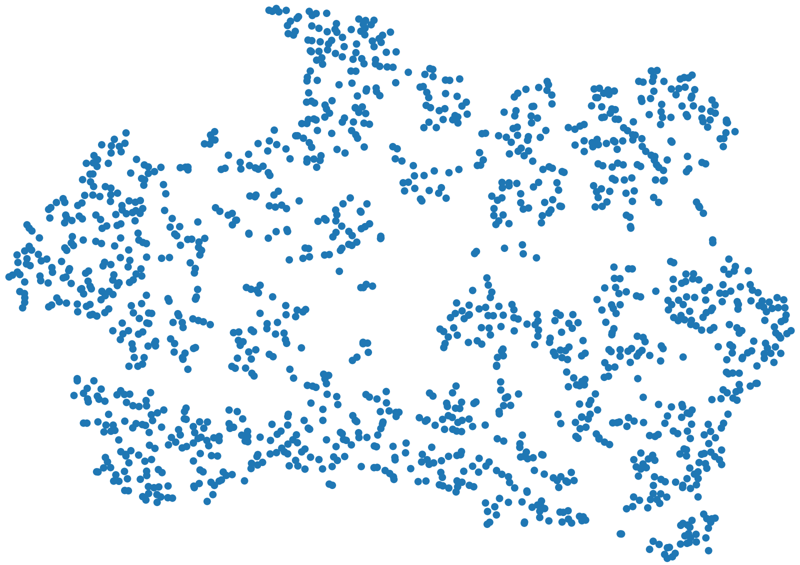


- Inputs: [CO_3^{2-}], [H_2], T, Dose Rate, CLT @ previous time, time step
- Better accuracy when using more training data
- Fewer nearest neighbors results in faster table lookup

Data conditioning improves the quality of the training data

- Remove FMD process model runs that are physically unrealistic
  - Runs that do not finish
  - Runs that stagnate at late time
  - Runs with Corrosion Layer Thicknesses that exceed physical domain size
- Log-transform data
- Subsample FMD process model runs
  - Random subset of points to reduce clustering in training data





A variety of metrics evaluate different elements of the surrogate model accuracy

- (Normalized Root) Mean Squared Error
  - Good metric for engineering purposes

$$mse = \frac{1}{N}\sum_{i=1}^{N}\left(y_{pred,i} - y_{true,i}\right)^2$$

$$nrmse = \frac{\sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(y_{pred,i} - y_{true,i}\right)^2}}{\frac{1}{N}\sum_{i=1}^{N} y_{true,i}}$$

- Mean Absolute Percentage Error
  - Highlights errors in small values

$$mape = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{y_{pred,i} - y_{true,i}}{y_{true,i}}\right| \times 100$$

- Mean Absolute Error
  - Not as sensitive to outliers

$$mae = \frac{1}{N}\sum_{i=1}^{N}\left|y_{pred,i} - y_{true,i}\right|$$