Uncertainty Quantification of Machine Learning Interatomic Potential Models

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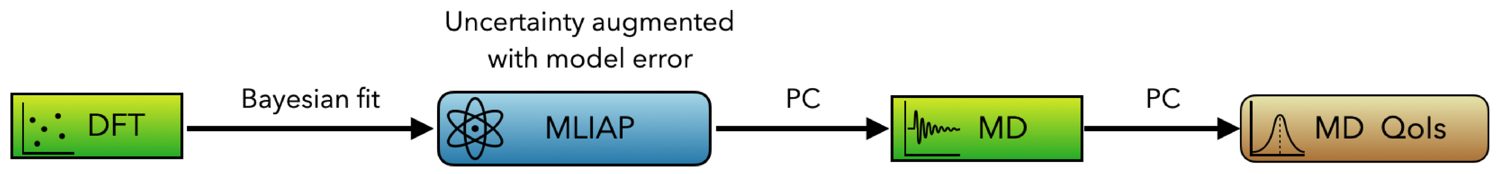
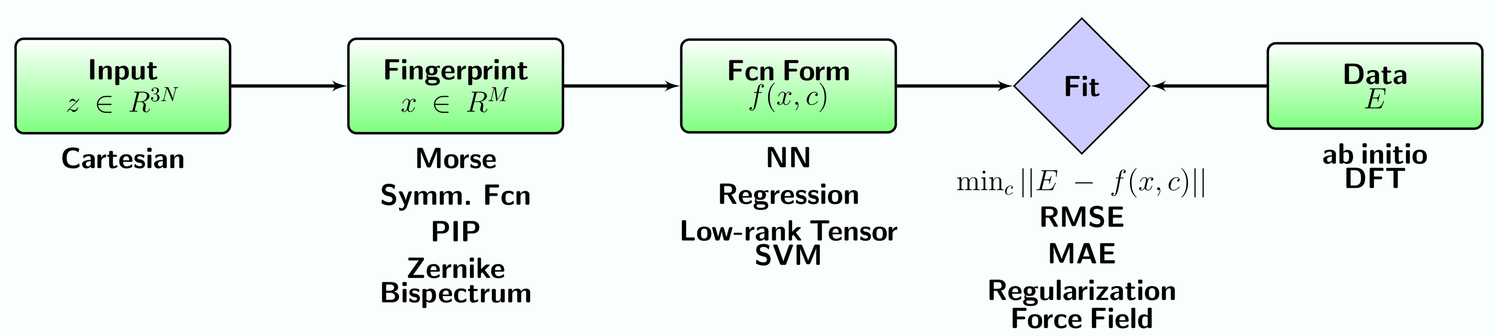
Big Picture

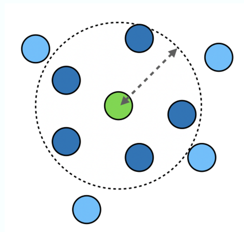
Bayesian Inference
Model Error
Polynomial Chaos Propagation



ML Interatomic Potentials (MLIAPs)



- Partition the interatomic interaction energy into individual contributions of the atoms
  $$E_{\text{total}} = \sum_{i=1}^{N} E_i$$
- Assume flexible functional forms with respect to positions of the neighboring atoms
  $$E \approx f(x, c)$$



Bayesian Inference of MLIAPs

Parameter inference

- Given a model $f(x,c)$ and data $y_i = y(x_i)$, calibrate parameters c such that $y_i \approx f(x_i, c)$
  - Linear model $y \approx Ac$ with coefficients c, or
  - NN model $y \approx NN_c(x)$ with weights/biases c.
- Weighted least-squares fit:
  $c^* = \text{argmin}_c \sum_{i=1}^{N} w_i^2 \left(f(x_i, c) - y_i\right)^2$

Likelihood function or data model

Posterior PDF, Prior PDF, Model, Data

$$\underbrace{p(c|y)}_{\text{Posterior PDF}} \propto \underbrace{p(y|c)}_{\text{Likelihood}}\underbrace{p(c)}_{\text{Prior PDF}} \propto \prod_{i=1}^{N} \exp\left(-\frac{(f(x_i,c) - y_i)^2}{2\sigma_i^2}\right)$$

Likelihood

✦ Prior contains previous knowledge or regularization

✦ Likelihood contains data noise modeling assumptions,

e.g. $y_i = f(x_i, c) + \sigma_i \epsilon_i$, where $\epsilon_i \sim \mathcal{N}(0,1)$

Data Model

Model Error

Elephant in the room: model error

$$\underbrace{y_i}_{\text{Truth}} = \underbrace{f(x_i, c)}_{\text{Model}} + \underbrace{\sigma_i \epsilon_i}_{\text{Data err.}}$$

Model ≠ Truth

Ignoring model error hurts in a few ways:

- Biased estimates of parameters c (crucial if the model is physical, and/or c is propagated through other models)
- More data leads to overconfident predictions (we become more and more certain about the wrong values of the data)

Capturing model error in data model

External correction (Kennedy-O'Hagan):

$$y_i = f(x_i, c) + \delta(x_i) + \sigma_i \epsilon_i$$

- Kennedy, O'Hagan, "Bayesian Calibration of Computer Models". *J Royal Stat Soc: Series B (Stat Meth)*, 63: 425-464, 2001.

Internal correction (embedded model error):

$$y_i = f(x_i, c + \delta(x_i)) + \sigma_i \epsilon_i$$

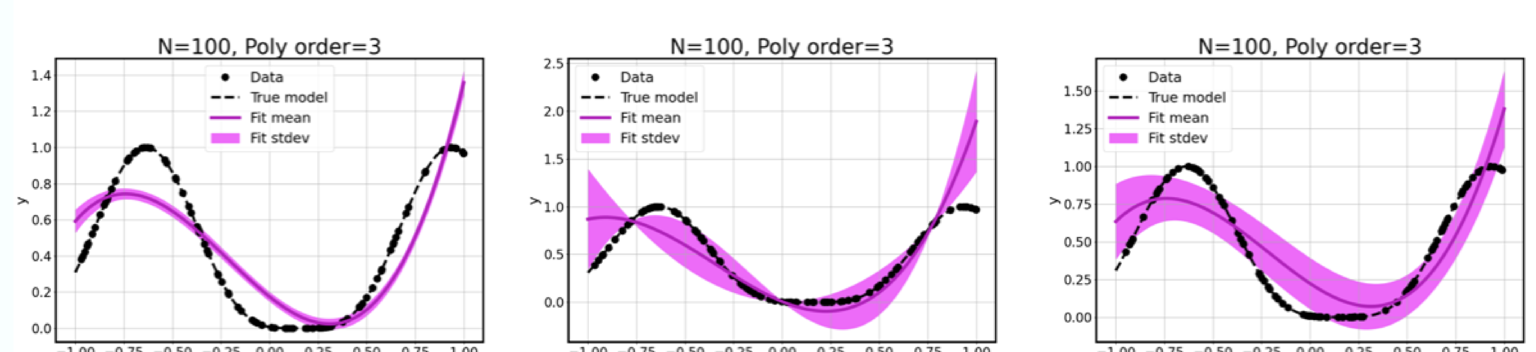
- Allows meaningful usage of calibrated model
- 'Leftover' noise term even with no data error
- Respects physics (not too relevant in our context)

- Sargsyan, Najm, Ghanem, "On the Statistical Calibration of Physical Models". *Int. J. Chem. Kinet.*, 47: 246-276, 2015.
- Sargsyan, Huan, Najm, "Embedded Model Error Representation for Bayesian Model Calibration". *Int. J. Uncert. Quantif.*, 9(4): 365-394, 2019.

Embedded model error method

- Statistical correction inside the model
  $$y_i \approx f\left(x_i, c + d\mathcal{N}(0,1)\right)$$
- Can be done non-intrusively, with a surrogate
- Jointly infer parameters of model and model error
  $$p(c, d|y) \propto p(y|c, d)p(c, d)$$
- Degenerate likelihood: needs approximations
  - Independent output approximation (IID):
    $$p(c, d|y) \propto \prod_{i=1}^{N} \exp\left(-\frac{(\mu_f(x_i,c) - y_i)^2}{2\sigma_f^2(x_i,c)}\right)$$
  - Approximate Bayesian computation (ABC):
    $$p(c, d|y) \propto \prod_{i=1}^{N} \exp\left(-\frac{(\mu_f(x_i,c) - y_i)^2 + (\sigma_f(x_i,c) - \alpha|\mu_f(x_i,c) - y_i|)^2}{2\epsilon^2}\right)$$

Classical case — Model error, IID likelihood — Model error, ABC likelihood

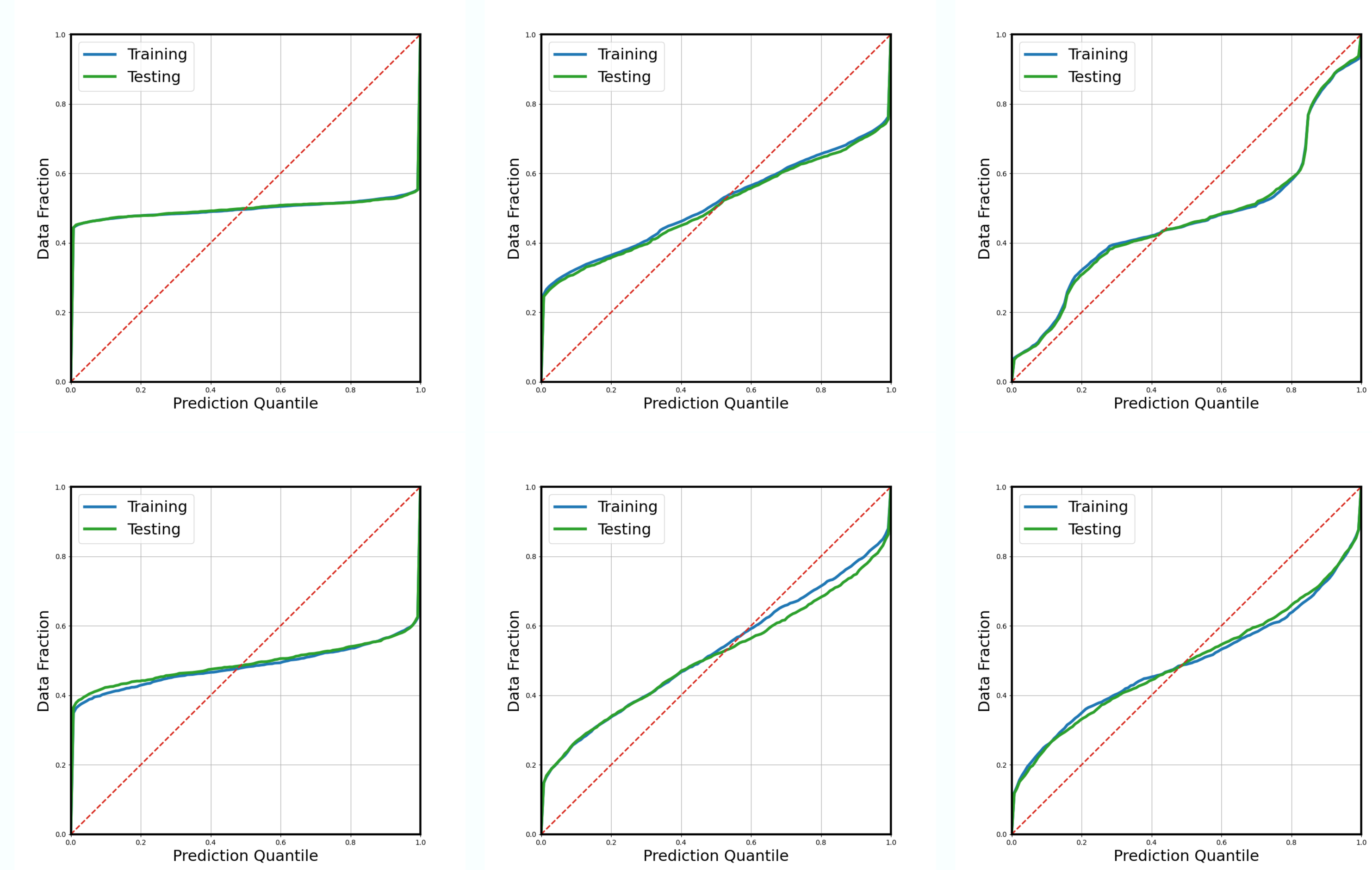


Uncertainty Validation

- We employ Spectral Neighbor Analysis Potential (SNAP) and FitSNAP (`https://github.com/FitSNAP/FitSNAP`)
- Embedded approach leads to better calibrated uncertainties enabling efficient active learning and uncertainty propagation.

Two examples with SNAP

No model error — Embedded, IID — Embedded, ABC



Uncertainty Propagation

Forward UQ via Polynomial Chaos (PC)

- Based on Bayesian MLIAP fit, construct input PC for MLIAP parameters $c = \sum_{k=0}^{K} a_k \Psi_k(\xi)$
- Sample input parameters and IAPs, $E(x) = f(x, c)$
- Obtain molecular dynamics QoIs $h = MD(E(x))$
- Build PC expansion for MD QoIs: $h = \sum_{k=0}^{K} b_k \Psi_k(\xi)$ via regression
- Evaluate QoIs statistics, compare to DFT benchmarks
- Variance-based decomposition (global sensitivity analysis) of the output PCs

Summary

- Bayesian fit of ML interatomic potentials: supervised ML
- Embedded model error with baked-in uncertainty
  - Model-error uncertainty capturing the true residual
- Polynomial chaos based uncertainty propagation through molecular dynamics

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