Evaluating Machine Learning Interatomic Potentials for Gas Adsorption in Metal Organic Frameworks

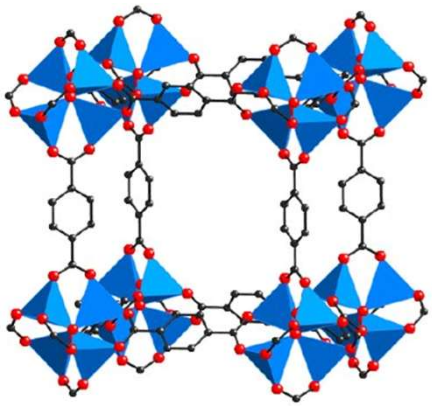
Sophie Xu, Urbana High School

Mentors: Jeffrey Kaaret, Wei Zhou

NATIONAL INSTITUTE OF STANDARDS AND TECHNOLOGY
U.S. DEPARTMENT OF COMMERCE

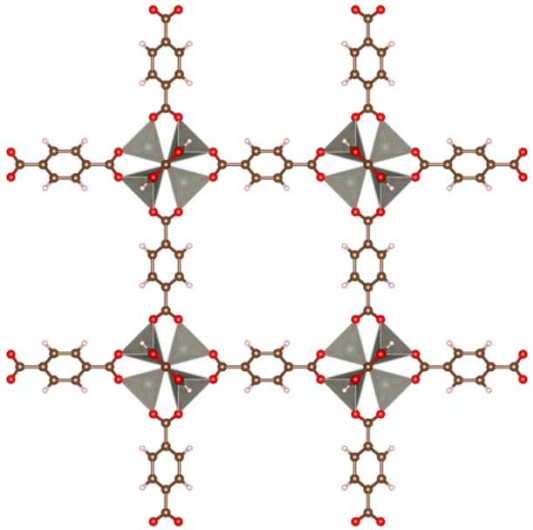
Metal-Organic Frameworks

- Metal-Organic Frameworks (MOFs) porous materials that can store gas molecules.
- Applications: Hydrogen storage (H_2) and natural gas storage (CH_4).



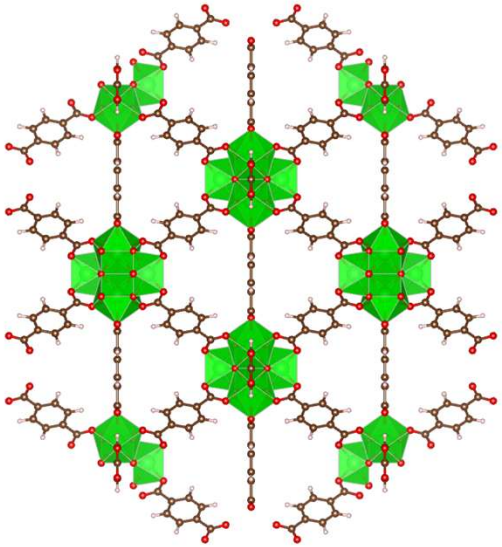
- MOF internal surface area can be as high as: 7000 square meters/ 1 gram

Metal-Organic Frameworks Candidates



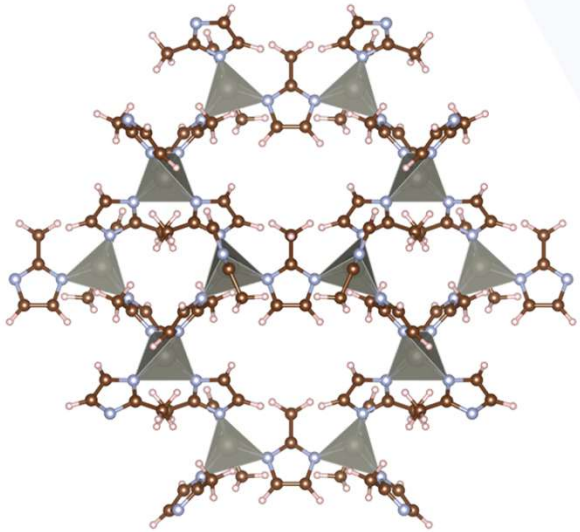
MOF-5

32Zn 104O 192C 96H



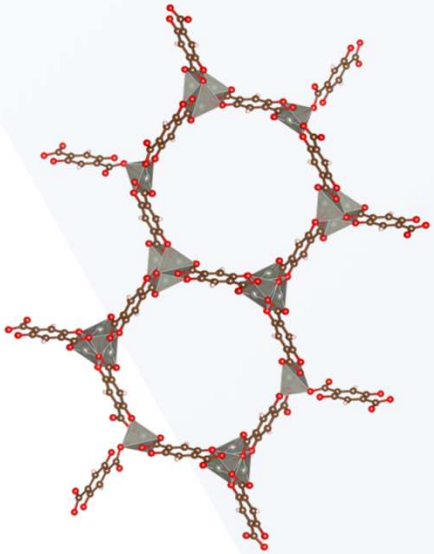
UiO-66

24Zr 128O 192C 96H



ZIF-8

12Zn 48N 96C 120H

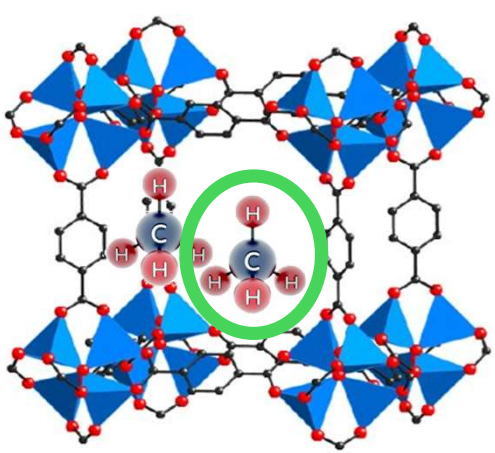


Zn-MOF-74

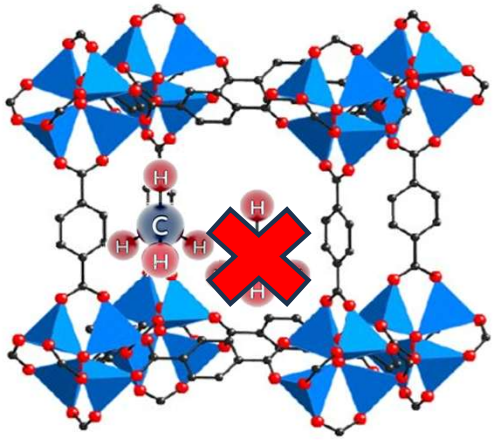
18Zn 54O 72C 18H

Simulations of Gas Adsorption

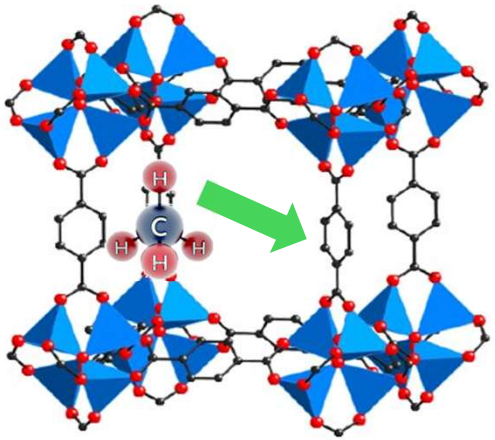
Grand Canonical Monte Carlo (GCMC) samples many structural configurations to estimate adsorption.



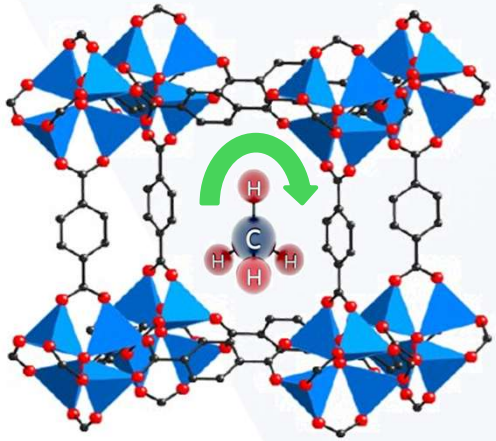
Insertion



Deletion



Translation



Rotation

Monte Carlo generates a random assortment of structures and determines their probabilities from the calculated interaction energies.

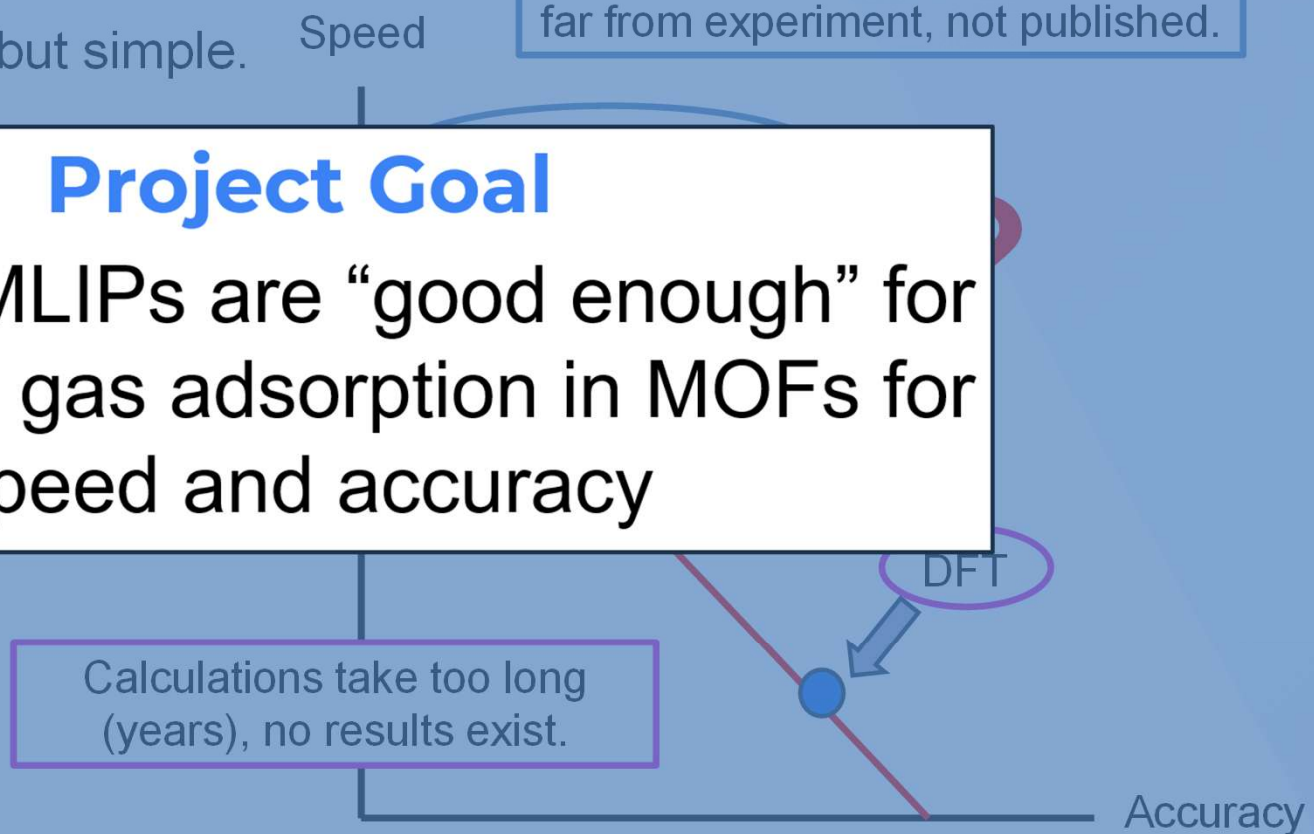
How to model interatomic interactions?

Universal Methods to calculate energy:

- Classical forcefields: fast but simple.
- Density f
  and expe
- MLIPs (M
  Potential
  ground, c

Project Goal

Assess if MLIPs are “good enough” for calculating gas adsorption in MOFs for speed and accuracy



How do the Machine learning interatomic potentials work?

Machine Learning Interatomic Potentials are neural networks trained on density functional theory results.

The networks are trained on diverse chemical datasets with the goal of calculating usable results in any material system.

These broadly trained models are referred to as ‘universal models’ since they don’t require system-specific training by users.

Universal MLIPs evaluated in this project

Fairchem-odac23

(Meta)

Orb-v3 conservative_inf_omat

(Orbital Materials)

Mace-mh-1-omat

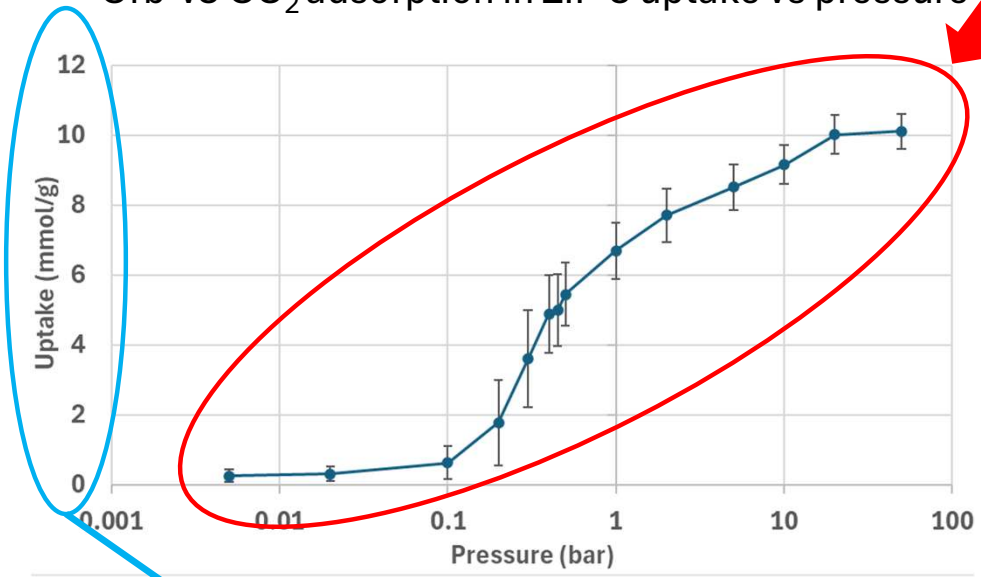
(University of Cambridge)

MACE

GCMC Code Benchmark

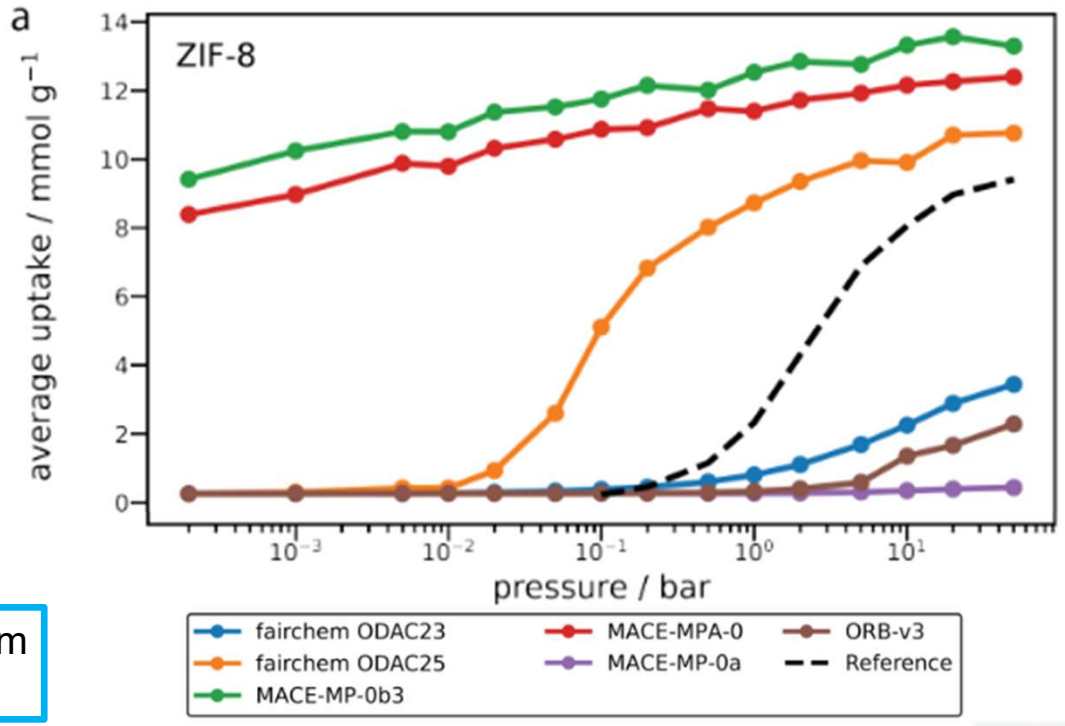
Each data point can take anywhere from 4 hours to 100 hours to calculate depending on the choice of system and pressure.

Orb-v3 CO_2 adsorption in ZIF-8 uptake vs pressure



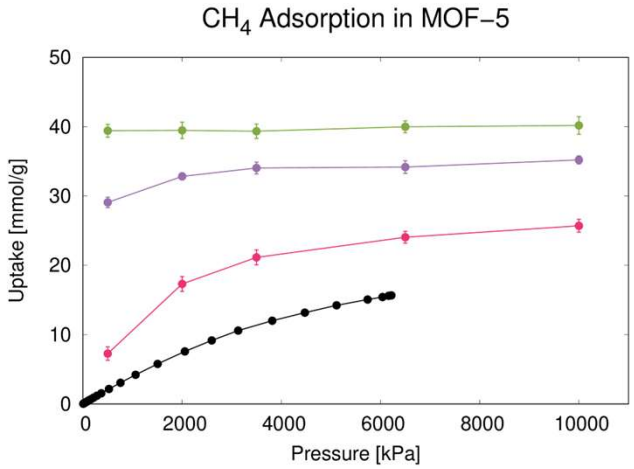
How much absorbent is absorbed per gram of the MOF as a function of pressure.

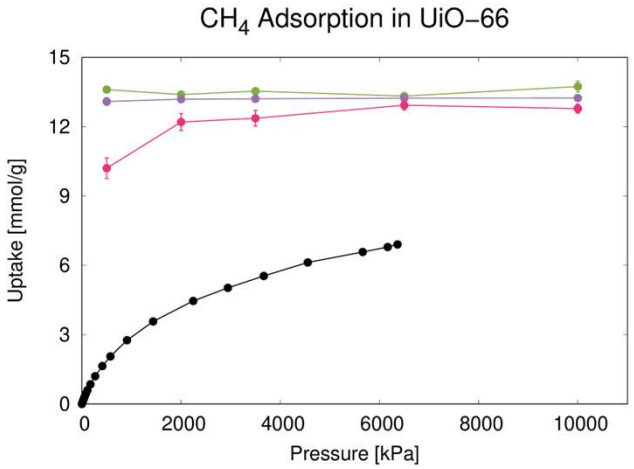
Source MLIP-MC: A Framework for Adsorption Simulations using Machine-Learned Interatomic Potentials

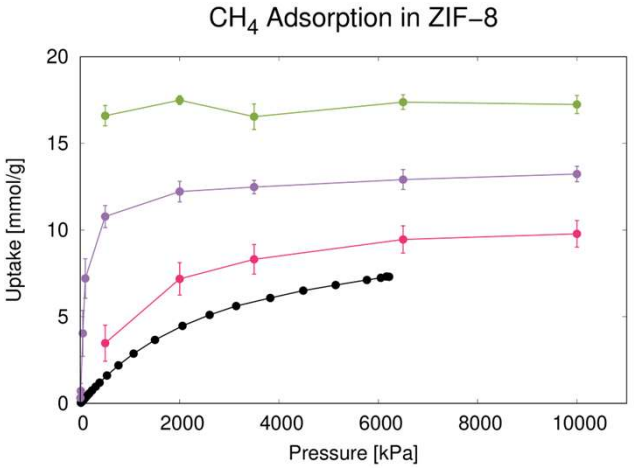


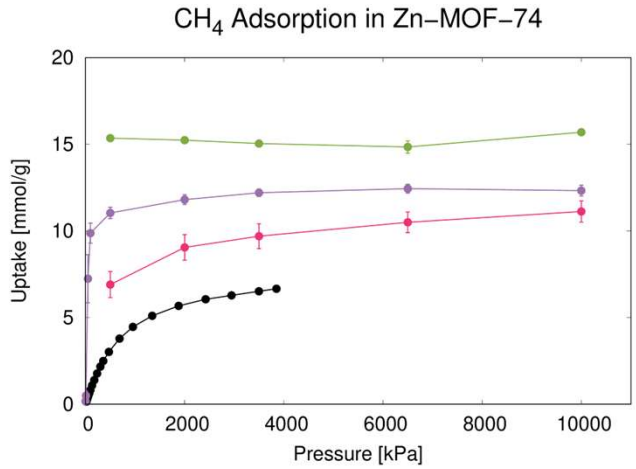
My results are comparable with theory literature values

CH_4 Adsorption Simulation Results

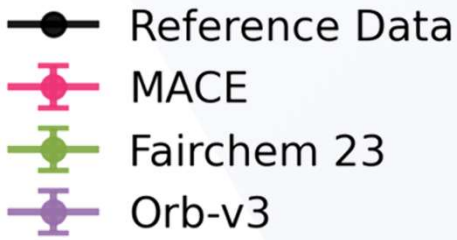








Simulations at 298 K



Models with dispersion consistently overestimated the reference data.

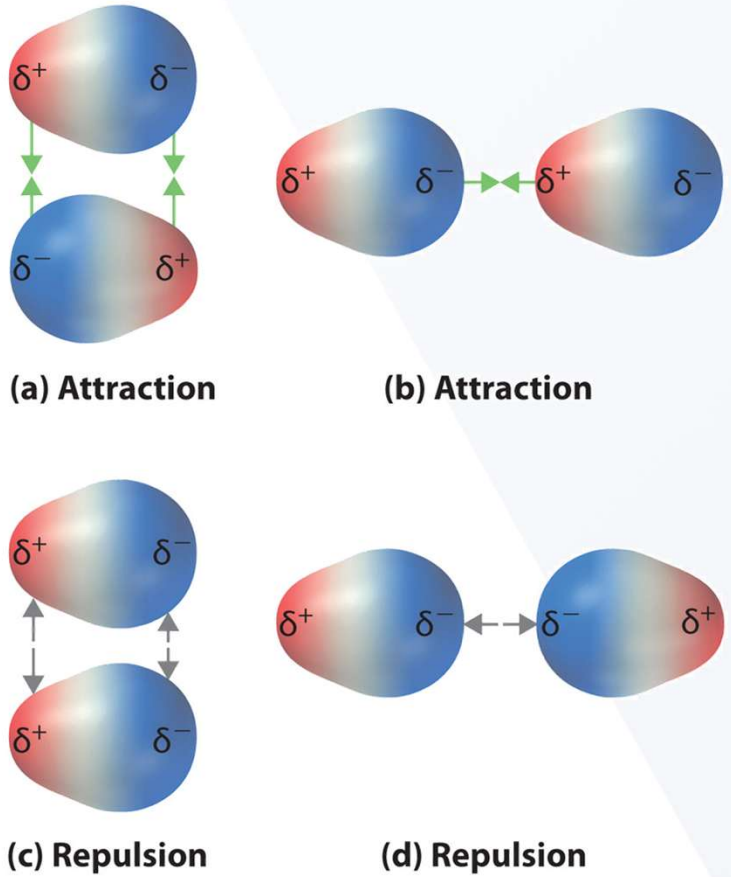
Reference data collected at NCNR

What is Dispersion?

Dispersion

Longer range forces, such as dipole-dipole interactions.

Not included in conventional DFT models as it is negligible for most materials, however it is important in MOFs.



- Fairchem-23
- Mace

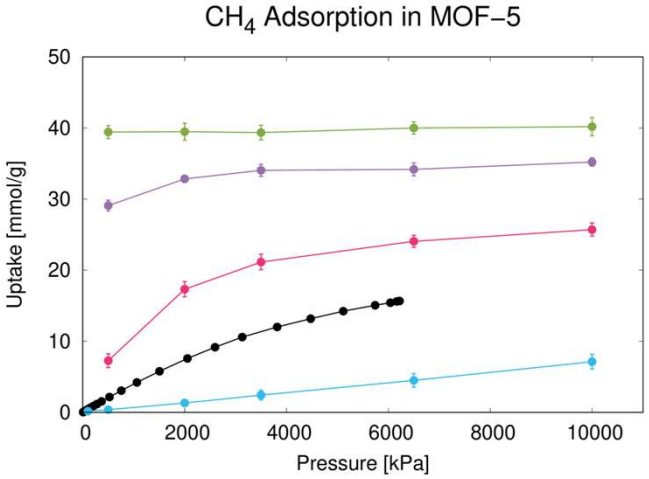
Models trained on dispersion data

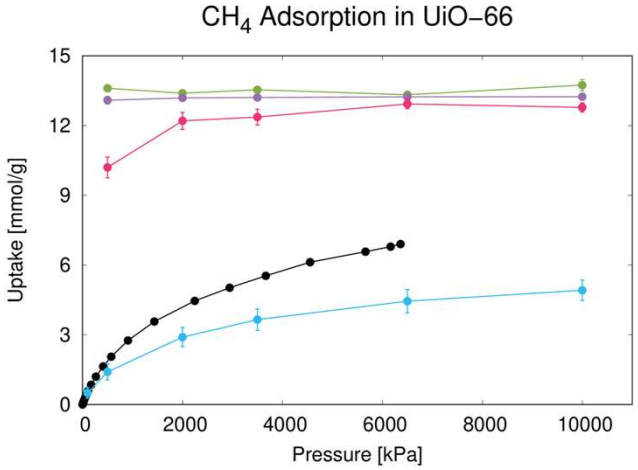
- Orb-v3

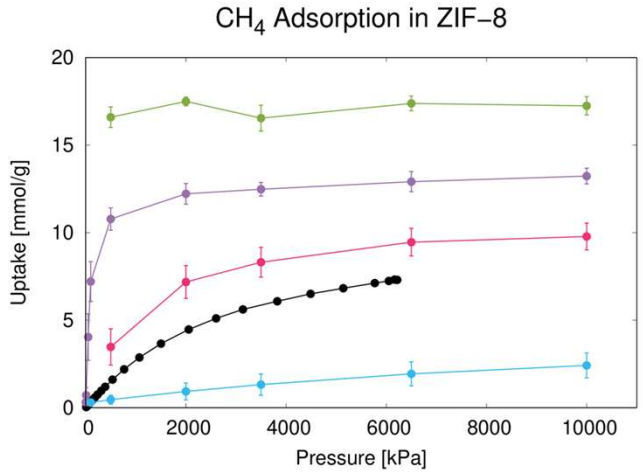
Does removing dispersion solve the overestimation?

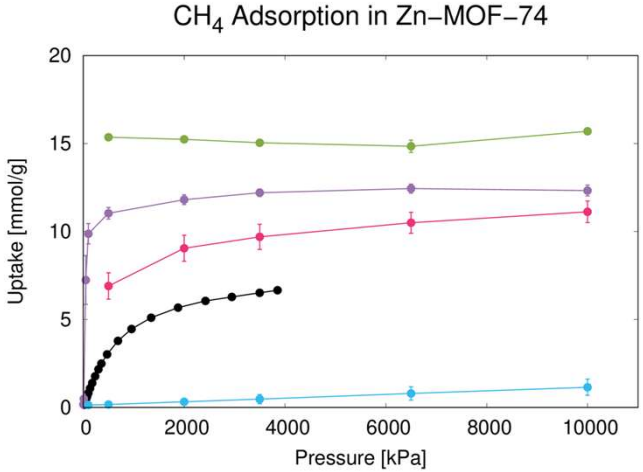
9.1: Intermolecular Forces- Dispersion, Dipole–Dipole, Hydrogen Bonding - Chemistry LibreTexts

CH_4 Adsorption Simulation Results

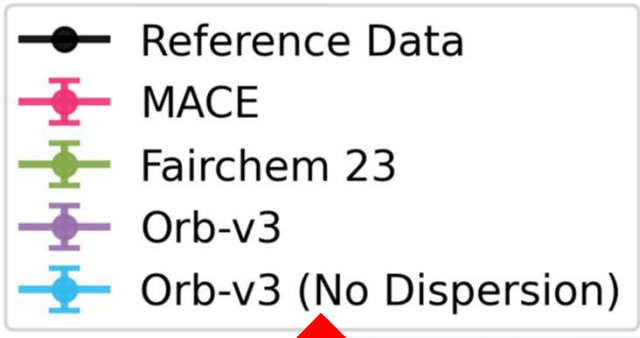








Simulations at 298 K

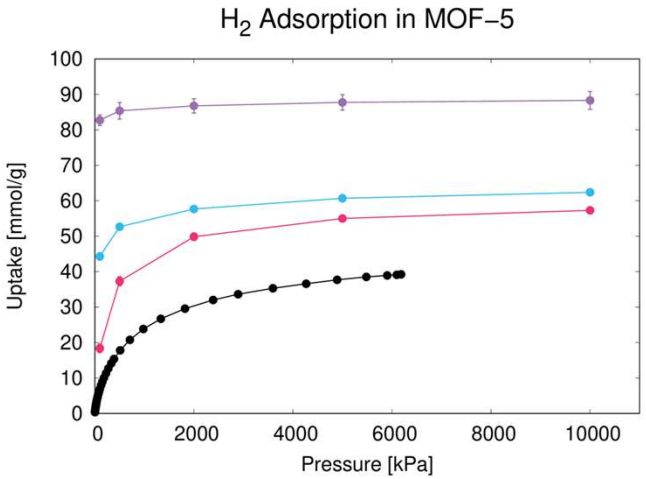


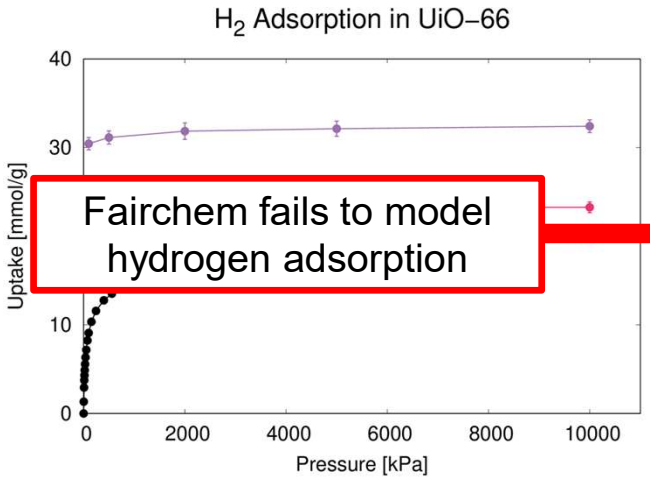
Orb-v3 without dispersion consistently underestimates CH_4 adsorption.

Reference data collected at NCNR

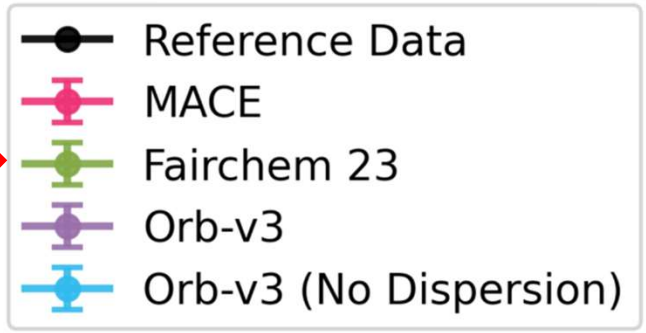
H_2 Adsorption Simulation Results

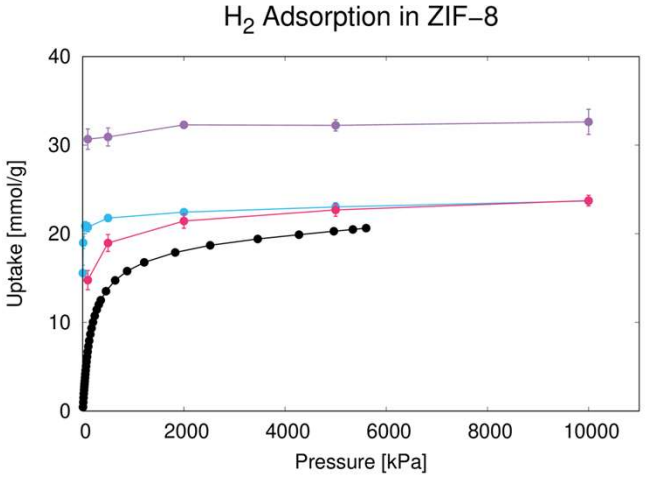
Simulations at 77 K

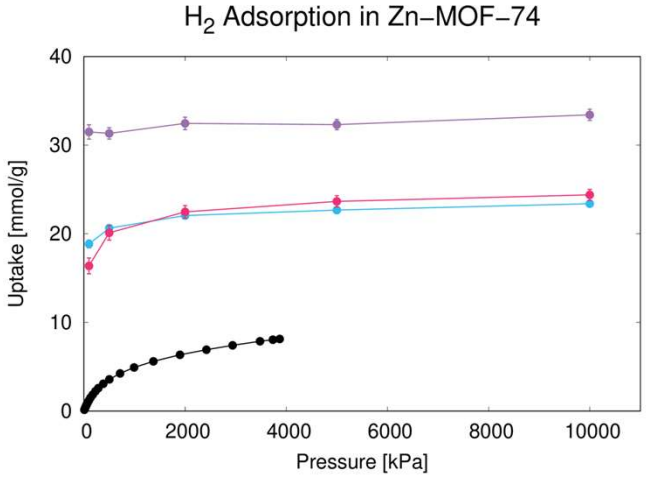




Fairchem fails to model hydrogen adsorption





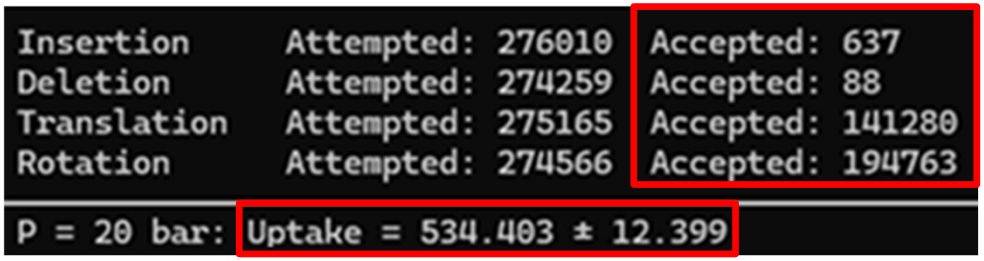


My results show Orb-v3 with dispersion, Orb-v3 without dispersion, and MACE overestimate H_2 adsorption.

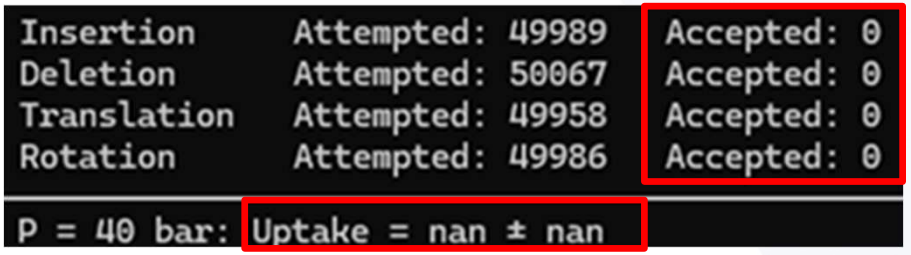
Reference data collected at NCNR

Fairchem MLIP Evaluation with H_2 Adsorption

Orb-v3



Fairchem-23



With additional testing we concluded that Fairchem modeled an unphysical energy interaction for H_2 adsorption in all the MOFs tested.

Conclusion

MLIPs evaluated in this project

MACE, Orb with dispersion, and Fairchem overestimate gas adsorption of CH_4 and H_2 in the tested MOFs, especially at lower pressures.

Orb without dispersion severely underestimates CH_4 adsorption.
Orb without dispersion overestimates H_2 adsorption to the same degree as MACE.

Of all the MLIPs evaluated, MACE appears to be the closest in reproducing results from experiments across our tested MOFs (but struggles with UiO-66). MACE could be used for refining of large-scale screening.

The least universal MLIP model, Fairchem-odac (trained on MOF data), preformed the worst.

Future

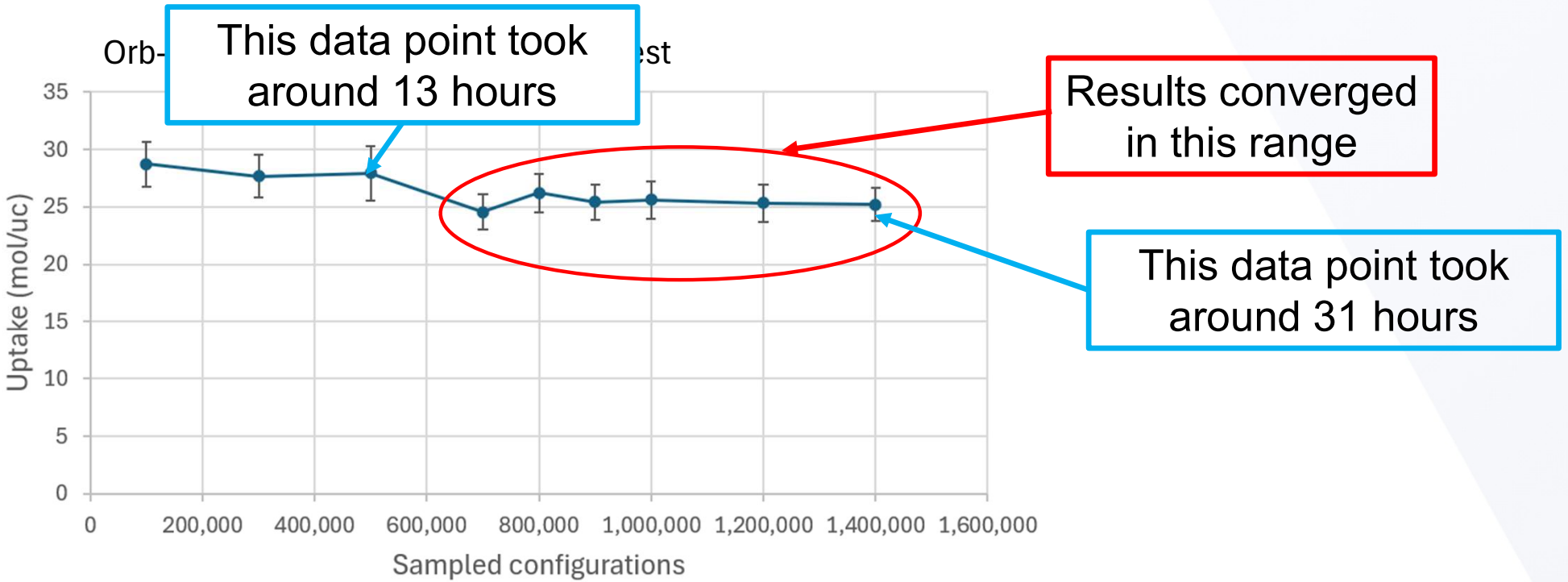
As the field is evolving very fast, we will be evaluating new and better MLIP models in the future.

Acknowledgments

Thank you so much!

- Mentors: Jeffrey Kaaret, Wei Zhou
- SHIP Directors: Cara O'Malley, Dr. Julie Borchers, Dr. Leland Harriger, Dr. Kelsi Rehmann, Dr. Susana Marujo Teixeira.
- CHRNS, NCNR staff, and NIST SHIP Program

Convergence Analysis



Determine the minimum number of computational steps for converged results.