Outline for MOF energy histogram work

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1 Notes

- The Guest Editors Andrew Ferguson and Johannes Hachmann for the special issue chaired my session at AIChE.
- Themed issue "Machine Learning and Data Science in Materials Design" for the journal *Molecular Systems Design & Engineering*

2 Title

- (scratch work. Let's come back to this later.)
- (from AIChE) Identifying New Descriptors for Gas Storage in Nanoporous Materials

3 Requirements and future directions

3.1 Quick tasks

- Fill in the remainder of the figures I could see for this paper (placeholders with "Figure" marking)
- Debrief with Scotty and get his input on availability and possible directions for the manuscript. He might have some good figures we should adapt for the manuscript, too.
- Do we pre-set the upper and/or lower bounds of the histograms?
- Fitting models on the 2 bar and 100 bar cases.

3.2 Potential directions

- **How general** is the method? Just hMOFs? Also ToBaCCo?
  - Could retrain on ToBaCCo and compare coefs
  - IZA database of zeolites?
  - CCDC MOFs - test model on 1000 MOFs, check agreement, then screen the rest of them?
- Identify old sources of **GCMC data for reuse** (email Diego or others?)
- **Other gases:** methane, xenon, multi-site molecules?
- **Experimental collaboration:** Screening the CCDC MOFs to identify top candidates for hydrogen storage (Would likely take a few weeks at best)
- **Isotherm prediction or Langmuir rationalization:** Repeat the predictions on multiple pressures (possibly temperatures as well). What do the coefficients look like? Is it similar to the Langmuir intuition on how much variability there will be?
- **Temperature dependence/optimization:** Could also consider changing temperature. Cryo vs. room temperature adsorption?

4 Introduction

- Hydrogen storage challenge
- MOFs and MOF databases
- Screening MOFs for hydrogen storage
  - Scotty/Jiayi paper
  - Yamil, Diego works
- Use of ML in the MOF literature
  - APRDF
  - Recent work from U Connecticut
  - See also my section in the review book
  - Relation for the Coulomb descriptor and related proxies in the literature (heat of adsorption, etc.)

5 Methods

5.1 Calculating the energy grid

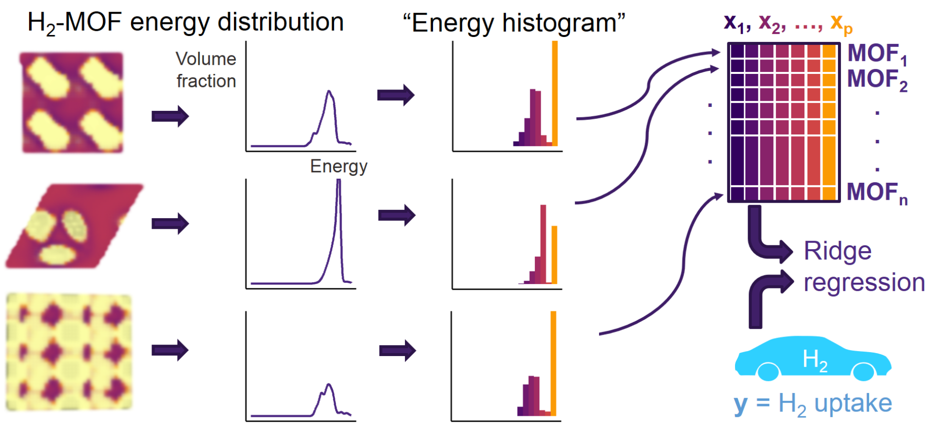


Figure 1: Calculation of the energy histogram from inputs

5.2 Molecular simulations

- RASPA
- Force field parameters for host and guests
- Number of cycles and/or source of data
- Structures
  - Wilmer's hMOFs
  - Likely ToBaCCo, so we can also look at structural diversity
  - Cleaning up the CCDC MOF subset using their solvent removal scripts, etc.

5.3 Data processing

- Ridge regression
  - Equations and loss function
  - R package `glmnet`
    - Finding the greatest lambda within 1 SE of the lambda that minimizes model error
- Data preprocessing: z-score bins and remove columns with zero variance. Also filter out unphysical uptake (¡ 0 g/L) from GCMC with giant error bars
- Define equations used for model evaluation: Q2, RMSE, MAE

6 Results and discussion

6.1 LJ metric

- Inspiration from ”binding fraction”
- Ask Scotty about this section. Figures of different distributions? Comparing MOFs that bind too strongly, etc?

6.2 Ridge regression

- Formalizing the results from LJ metric studies
- Meaning of betas and intuition

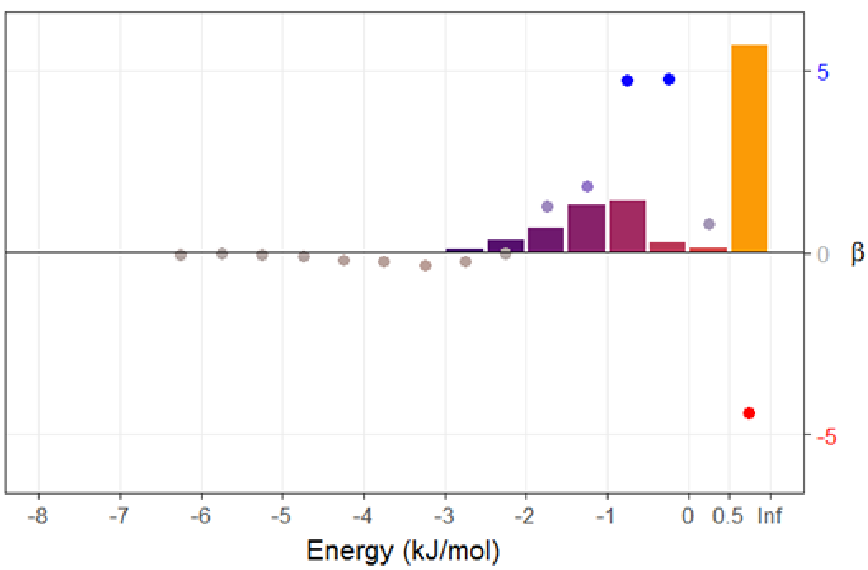


Figure 2: Regression coefficients overlaid on the histogram

- How good is the model and fit?
  - Parity plot
  - Q2 of 0.96
  - RMSE of 3 g/L, MAE/MUE of 2.4 g/L

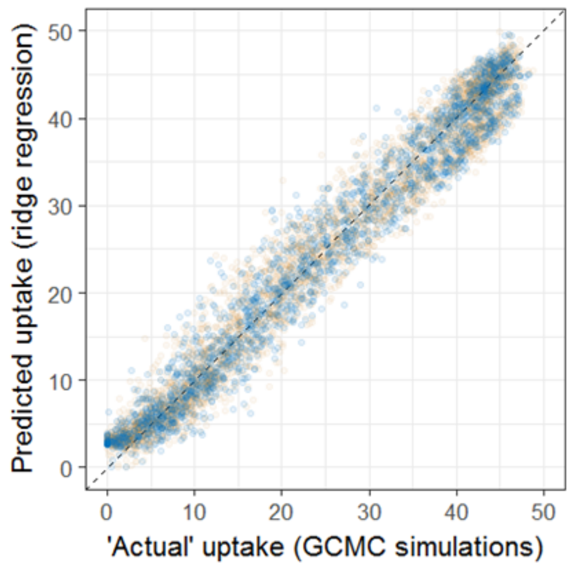


Figure 3: Parity plot for (a) training and (b) test data on the energy histograms (TODO: split into two separate figures so it's more black and white-friendly, and easier to read without an animation)

- Why ridge regression?
  - Interpretability with a simple model
  - Likely some kind of link with a multi-site Langmuir model (assuming that all sites at a given energy are identical)
  - Someone at AIChE asked why our model didn't include nonlinearity. The reason is that we don't need it to make great predictions. (my earlier work used random forest, which would avoid manual feature transformations)

6.3 Screening

- Plan of testing applicability on 1000 MOFs
- Benchmarking (maybe as a table): Full GCMC vs. energy grid calculations, feature representation, and ML

Figure 4: TODO: top candidates for experimental synthesis based on screening the CCDC MOF database

6.4 Generalizability to other gases

- See above

7 Acknowledgements

Data Science Initiative, NMGC, etc.

8 Supporting Info

8.1 Hyperparameter tuning

- Grid spacing: convergence of a few different sample MOF histograms
  - Also might be good to have a figure overlaying sampling points on top of a continuous background, to exemplify the convergence testing
- Bin width and degree of overlap (TODO: consider adding examples, and Q2 figure)
- Lambda selection

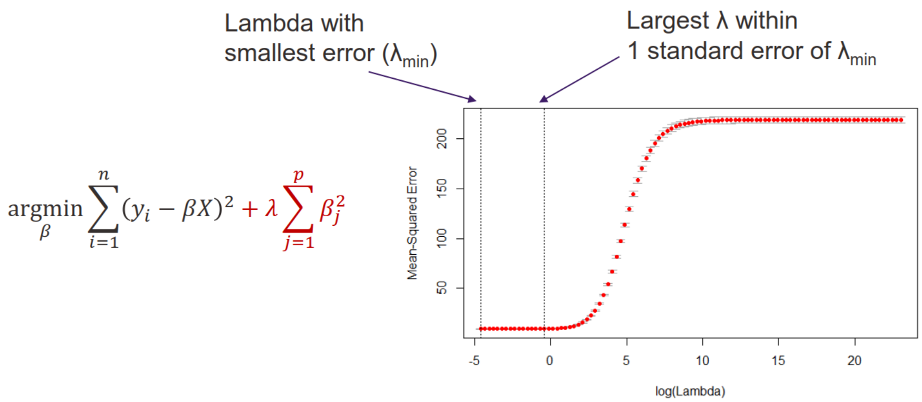


Figure 5: Determination of the regularization parameter λ for ridge regression

8.2 Model evaluation/consistency

Also consider adding a figure on "Consistency across nodes/linkers and/or other DBs" (see Zr results)

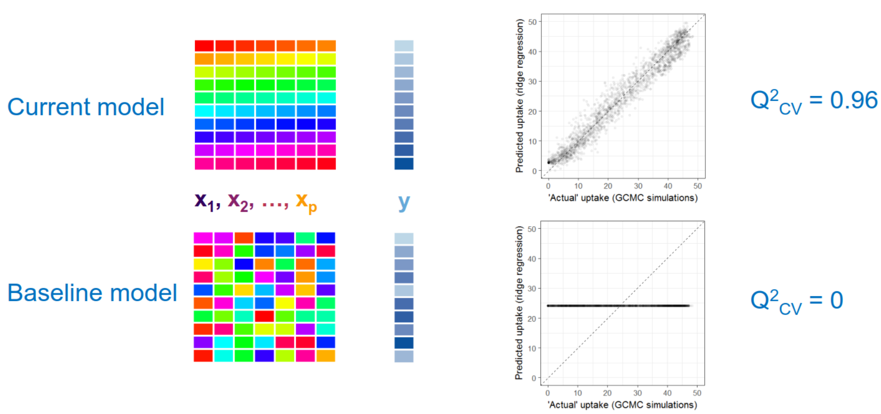


Figure 6: Comparison of Q^2 against a baseline of random data

8.3 Alternative approaches

- Benchmarking against traditional descriptors (textual properties like void fraction and density)
- LASSO figure and coefficients

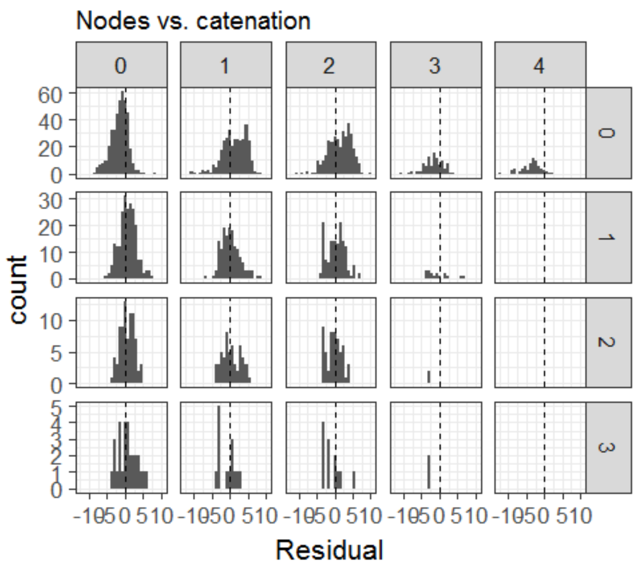


Figure 7: Consistency of model accuracy across MOF compositions. Note that Zr MOFs are less accurate (node 4), possibly due to differences in topology and undersampling relative to **pcu** MOFs.

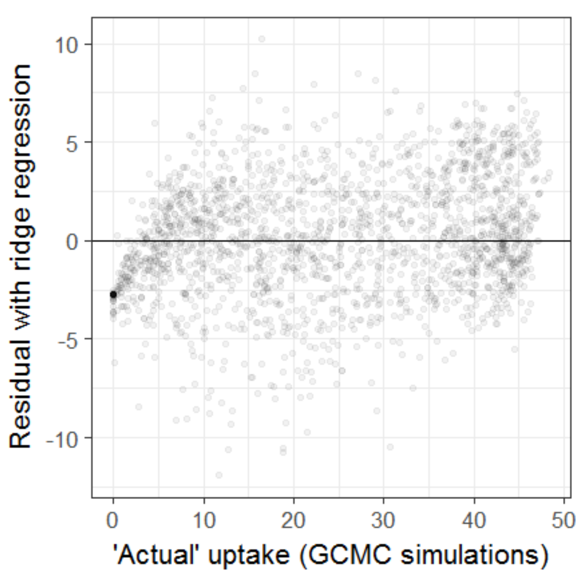
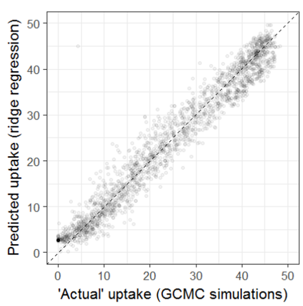
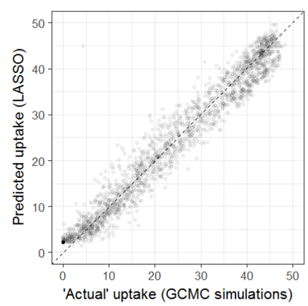


Figure 8: Residual plot for the testing data. One audience member in my talk once concerned about a long tail (?) on the top right of my parity plot, below the line of parity, possibly requiring another variable for correction. But I'm having trouble seeing it in the residual plot, so perhaps it was an optical illusion against the 45 degree line.



Beta	Ridge	LASSO
(Intercept)	24.11	24.11
4	-0.06	
5	-0.01	
6	-0.05	
7	-0.10	-0.06
8	-0.22	-0.16
9	-0.26	-0.18
10	-0.36	-0.35
11	-0.24	-0.10
12	-0.03	
13	1.26	1.25
14	1.81	1.73
15	4.71	4.86
16	4.75	4.62
17	0.79	0.69
Inf	-4.41	-4.65



Performance	Ridge	LASSO
RMSE	2.92	2.92
Rsquared	0.96	0.96
MAE	2.31	2.31

Figure 9: Ridge regression and LASSO give similar results