CO$_2$ capture is required to mitigate climate change

Inorganic solids have potential to capture CO$_2$, but they are not yet economically viable. 😞
Role of Pore Chemistry and Topology in the CO$_2$ Capture Capabilities of MOFs: From Molecular Simulation to Machine Learning

- MOF = metal organic framework
- Machine learning: input = property, output = descriptors
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\[
S = \frac{\frac{N_{\text{ads}}}{y_{\text{CO}_2}}}{\frac{N_{i,\text{ads}}}{y_i}}
\]

\[
WC = N_{\text{ads}}^{\text{CO}_2} - N_{\text{des}}^{\text{CO}_2}
\]
Test many MOFs, many linkers
DFT binding energies and geometries

- DFT can estimate exchange-correlation energy
- A self consistent field method is still used to find the energies and wavefunctions.

\[ E_{\text{binding}} = E_{\text{complex}} - E_{\text{CO}_2} - E_{\text{benzene}} \]
The calculation is pretty good

- CO\textsubscript{2} loading = mole of CO\textsubscript{2} per mole of MOF
- GCMC = Grand canonical Monte Carlo, a computational method to statistically investigate a reaction (such as CO\textsubscript{2} adsorption)
The DFT functional for systems with dispersion forces!

- Literature: M06-L with Def2SVP
  - I suggested: B3LYP and 6-31+G(d,p) because I got errors when I tried to run
    
    \#P M06L/def2SVP opt iop(1/7=30) integral=ultrafine
  
- However, the benzene and CO$_2$ molecules did not “bind” with B3LYP and 6-31+G(d,p)!
  - Eureka! M06-L is better than B3LYP for dispersion.
  - With testing, I found the error was Def2VSP (not M06-L).

- Please run M06L and 6-311+G(d,p) or 6-31+G(d,p).
  - I think the larger basis set will be more accurate, but it takes twice the amount of time (I tested on benzene geo opt).
  - New input files and analysis script is on iLearn.
Locations of CO$_2$ binding in MOF

- $(H)_4$
- $(CH_3)_2$
- $(CN)_4$
Finding the best MOFs and linkers

“parent” MOF LPD increases

$\Delta CO_2$ [mol/kg]

15:85 CO$_2$:N$_2$ mixture@1bar

$\Delta S$

“parent” MOF topology

$\Delta CO_2$ [mol/kg]

20:80 CO$_2$:H$_2$ mixture@20bar

$\Delta S$

“parent” MOF topology