Efficient Determination of Accurate Force Fields for Porous Materials Using Ab-initio Total Energy Calculations – Supporting Information

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A. CO_2 in Mg-MOF-74 Stability Analysis

To test the robustness of our methodology, the $E_{reference,binding}$ values derived from the MP2-derived force field were varied from –4 to +4 kJ/mol in steps of 1 kJ/mol. As a result, the varying shift values to $E_{reference,binding}$ causes changes in the final K_H value. In particular, the CO_2 K_H is computed for each of the different shifts and the result is summarized in Figure SI1(a). As can be seen from Figure SI1(a), the CO_2 K_H changes from 3.30×10^{-5} mol/kg/Pa (at $E_{binding}$ = –42.22 kJ/mol) to 3.25×10^{-3} mol/kg/Pa (at $E_{binding}$ = –50.22 kJ/mol). This stability analysis illustrates the importance of obtaining accurate quantum mechanical binding energies for this system.

To extend this analysis beyond the Henry coefficient calculations, which only sheds insights on the adsorption property at infinite dilution conditions, we compute the corresponding CO_2 adsorption isotherms at $E_{binding} = E_0 - 4$, $E_0 - 2$, E_0 (reference), $E_0 + 2$, and $E_0 + 4$ kJ/mol, where E_0 = –46.22 kJ/mol, up to 1bar and compare the results with the MP2-derived force field data at T = 313 K. The results are summarized in Figure SI1(b). In all cases, the parameters for the $CO_2 – CO_2$ interactions were kept the same.

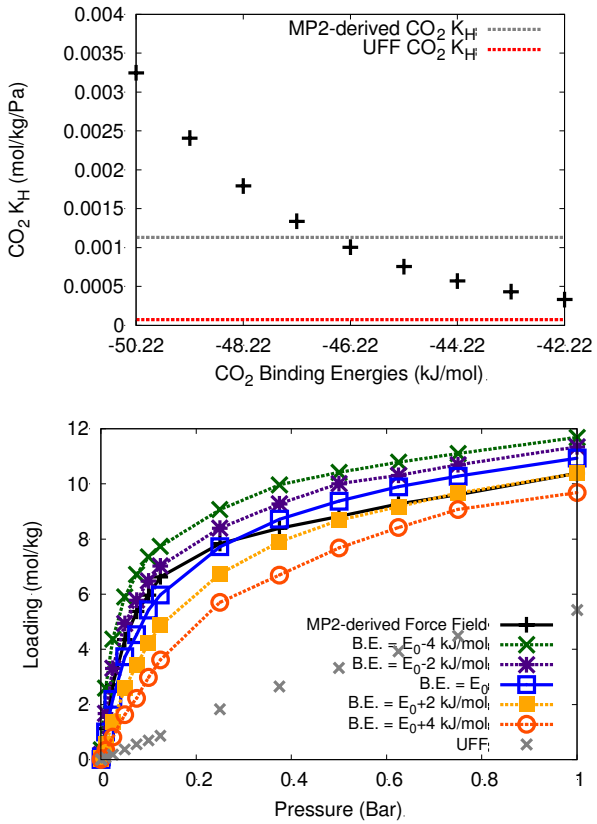
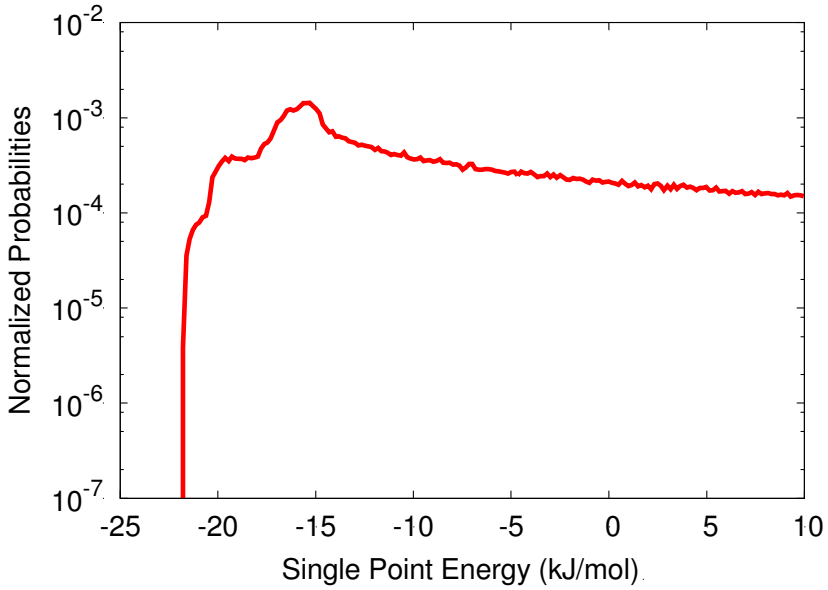


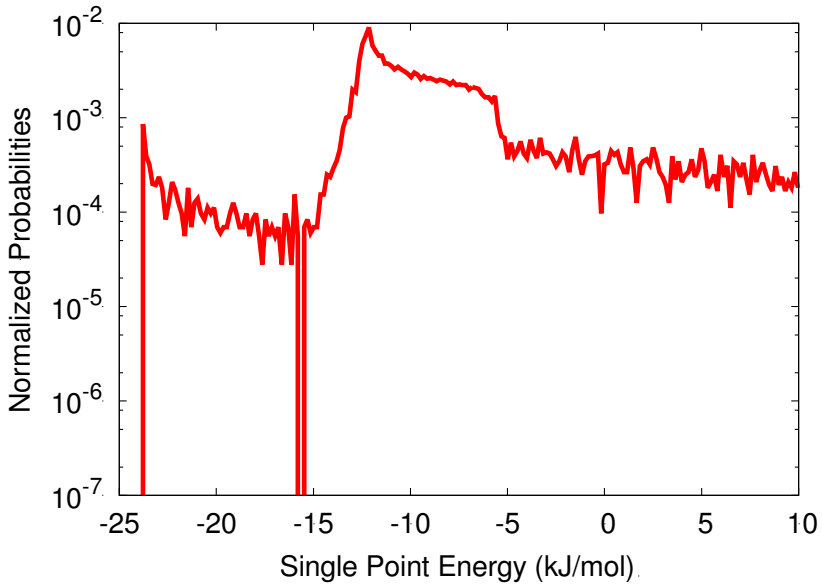
Figure S1(a) Computed CO_2 K_H as a function of the CO_2 binding energy in Mg-MOF-74 at T = 313K. The binding energy values were changed from $E_{binding} = E_0 –4$ to $E_0 + 4$ kJ/mol, with E_0 = –46.22 kJ/mol representing the accurate, original binding energy value. (b) The simulated adsorption isotherm curves for $E_{binding} = E_0 –4$ to $E_0 + 4$ kJ/mol, with E_0 = –46.22 kJ/mol.

B. CH_4 Single Point Energy Distributions for Eight Zeolites

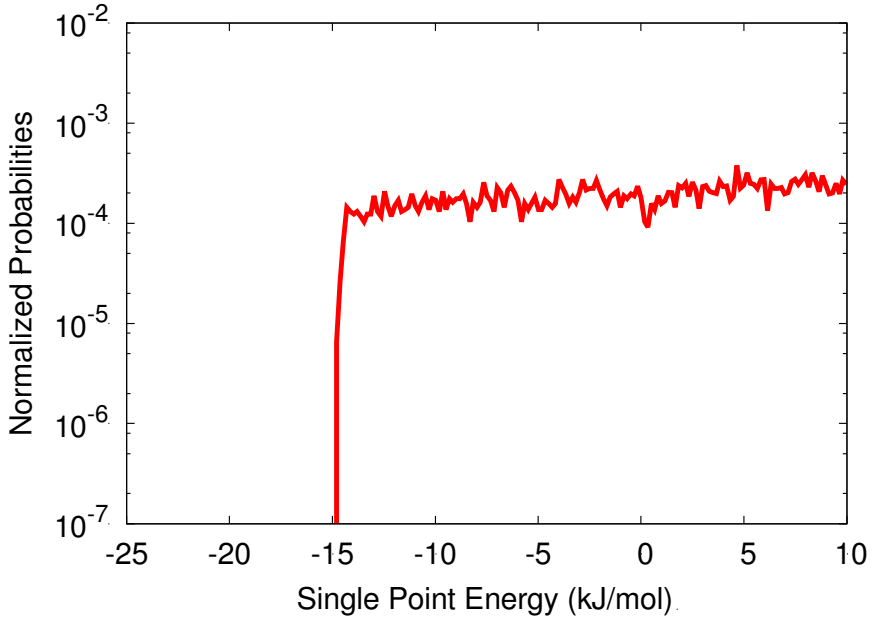
MFI



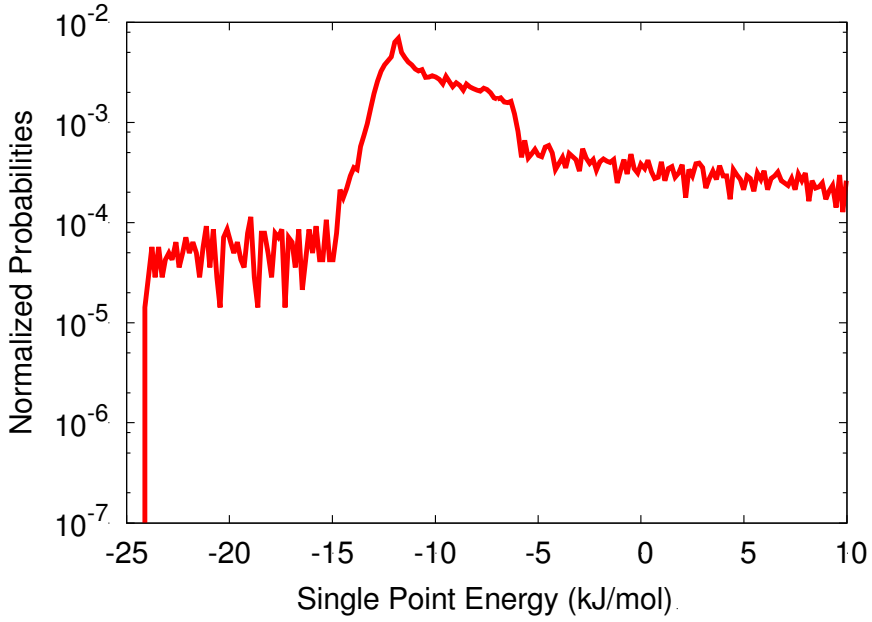
LTA



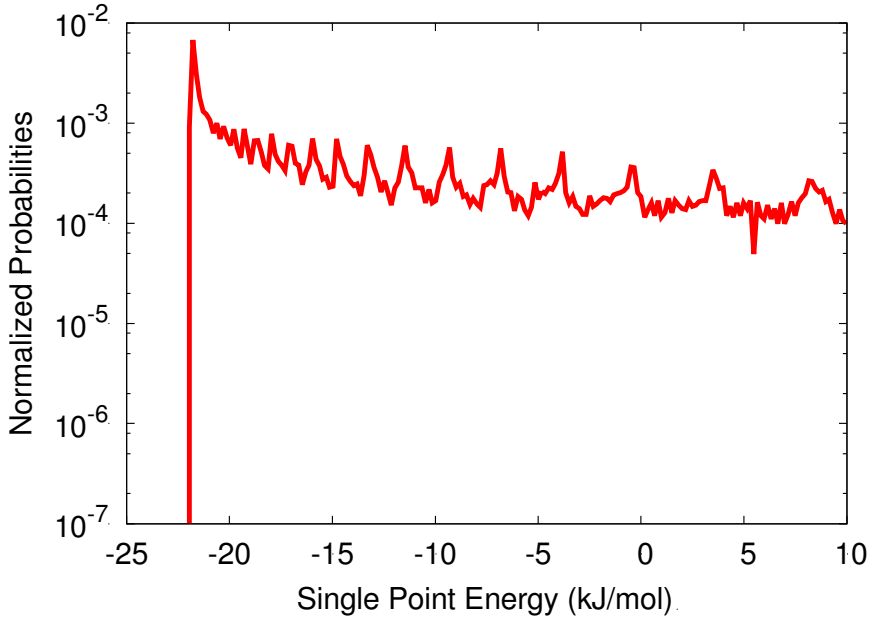
WEI


Normalized Probabilities
10-2
10-3
10-4
10-5
10-6
10-7
-25
-20
-15
-10
-5
0
5
10
Single Point Energy (kJ/mol)

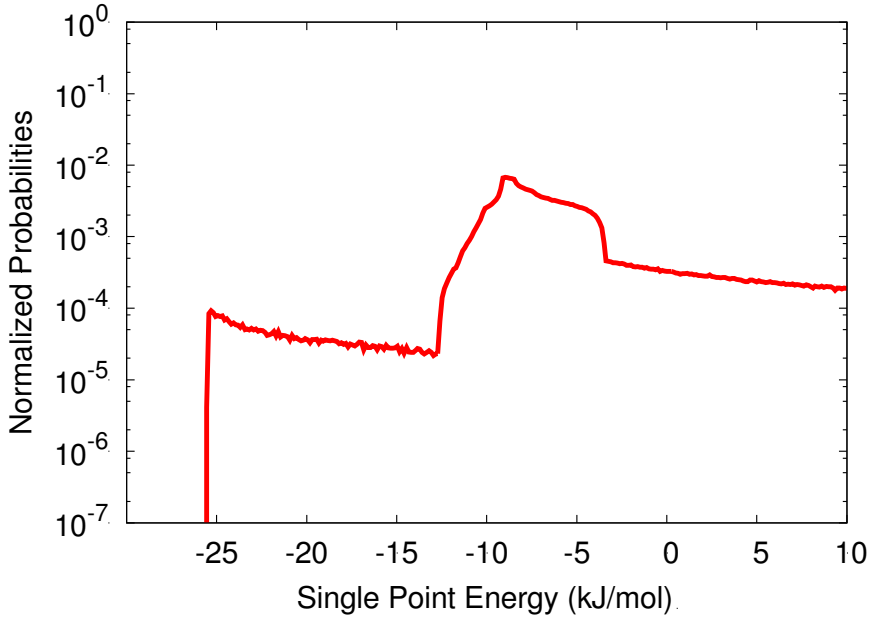
RHO


Normalized Probabilities
10-2
10-3
10-4
10-5
10-6
10-7
-25
-20
-15
-10
-5
0
5
10
Single Point Energy (kJ/mol)

SOD


Normalized Probabilities
10-2
10-3
10-4
10-5
10-6
10-7
-25
-20
-15
-10
-5
0
5
10
Single Point Energy (kJ/mol)

FAU


Normalized Probabilities
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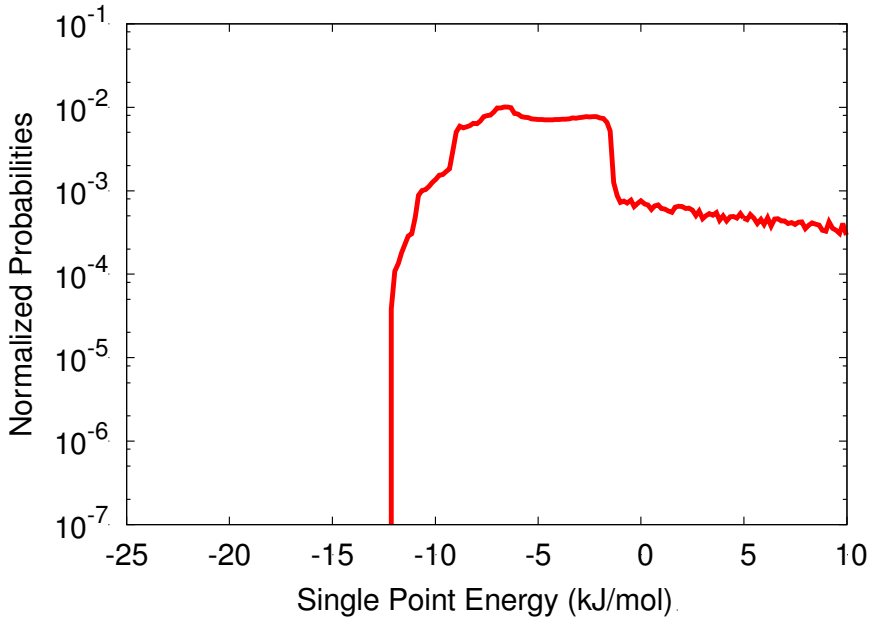
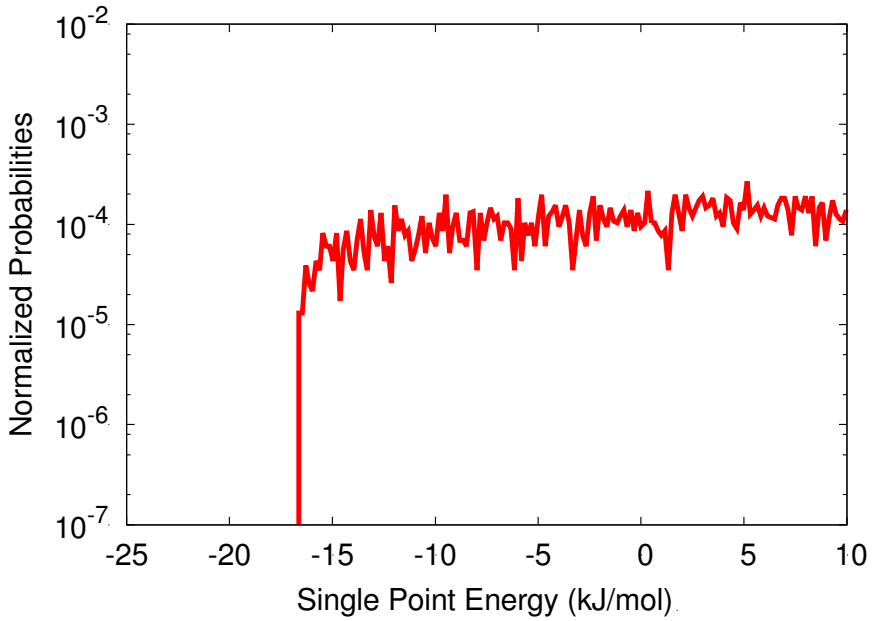
10-1
10-2
10-3
10-4
10-5
10-6
10-7
-25
-20
-15
-10
-5
0
5
10
Single Point Energy (kJ/mol)

RWY


Normalized Probabilities
10-1
10-2
10-3
10-4
10-5
10-6
10-7
-25
-20
-15
-10
-5
0
5
10
Single Point Energy (kJ/mol)

ABW


Normalized Probabilities
10-2
10-3
10-4
10-5
10-6
10-7
-25
-20
-15
-10
-5
0
5
10
Single Point Energy (kJ/mol)