Supporting Information: Enzymatic Kinetic Isotope Effects from First Principles Path Sampling Calculations

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ABSTRACT

This provides supporting information to our manuscript, with information on the CMD calculated forces relative to that of a 16 bead system.

Molecular dynamics simulations were run for 1000 timesteps (500 fs) using normal mode centroid molecular dynamics (CMD) for the propagation of the transferring particle. Two non-reactive trajectories were obtained using 8 bead- and 16 bead-CMD. Each trajectory was obtained from the equilibrated structure of yeast alcohol dehydrogenase, and velocities were chosen from a Gaussian distribution with different seeds. The magnitude of the CMD force vector at each timeslice was plotted along the trajectory. Histograms of these force vector magnitudes were obtained for each simulation (Figure 1). Values of force magnitude higher than 100 kcal/(mol*Å) were removed

from the histograms so that the x-axes have the same scale (0 for 8 bead- and 15 for 16 bead-CMD). Each distribution exhibits similar peaks and shape, peaking at 22 kcal/(mol*Å) and 25 kcal/(mol*Å) for 8 bead- and 16 bead-CMD, respectively, indicating that CMD force is similar in each of the CMD simulations. In addition, while 16 bead-CMD exhibits a slightly sharper distribution, this sharpness comes with increased computation time required for the trajectory to be obtained, up to a 2-fold increase in CPU hours required.

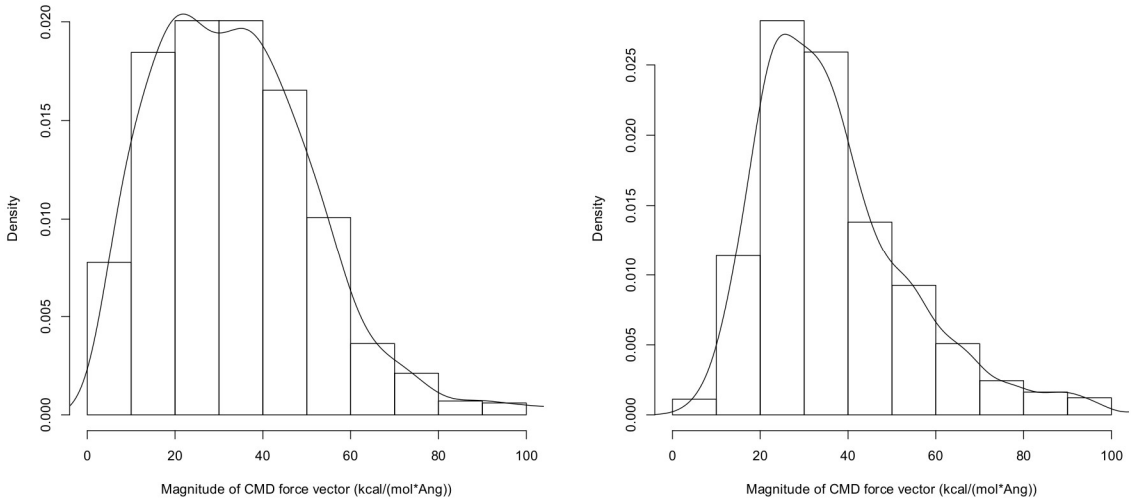

Figure 1: Histograms of the distribution of the magnitude of the CMD force vectors for a trajectory run with 8 bead-CMD (left) and 16 bead-CMD (right), with density fits.