

**Supporting information: Histogram-free  
reweighting with grand canonical Monte Carlo:  
Post-simulation optimization of non-bonded  
potentials for phase equilibria**

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## S1 Bonded parameters

Table S1: Equilibrium (fixed) bond lengths ( $r_{\text{eq}}$ ).  $\text{CH}_x$  and  $\text{CH}_y$  represent  $\text{CH}_3$ ,  $\text{CH}_2(\text{sp}^3)$ ,  $\text{CH}(\text{sp}^3)$ , or  $\text{C}(\text{sp}^3)$  sites.

Bond sites	$r_{\text{eq}}$ (nm)		
	TraPPE	MiPPE	NERD
$\text{CH}_x\text{-CH}_y$	0.154	0.154	0.154
$\text{C}(\text{sp})\text{-CH}_x$	–	0.146	–
$\text{CH}\equiv\text{CH}$	–	0.121	–
$\text{C}\equiv\text{CH}$	–	0.121	–

Table S2: Equilibrium bond angles ( $\theta_{\text{eq}}$ ) and force constants ( $k_\theta/k_B$ ), where  $k_B$  is the Boltzmann constant.

Bending sites	$\theta_{\text{eq}}$ (degrees)			$k_\theta/k_B$ (K/rad <sup>2</sup> )
	TraPPE	MiPPE	NERD	
$\text{CH}_x\text{-CH}_2\text{-CH}_y$	114.0	114.0	114.0	62500
$\text{CH}_x\text{-CH-CH}_y$	112.0	112.0	109.5	62500
$\text{CH}_x\text{-C-CH}_y$	109.5	109.5	109.5	62500
$\text{CH}_x\text{-CH}_2\text{-C}(\text{sp})$	–	112	–	62500
$\text{CH}_x\text{-C}(\text{sp})\equiv\text{CH}$	–	180	–	30800
$\text{CH}_x\text{-C}(\text{sp})\equiv\text{C}$	–	180	–	30800

Table S3: Fourier constants ( $c_n/k_B$ ) in units of K.

Torsion sites	$c_0/k_B$	$c_1/k_B$	$c_2/k_B$	$c_3/k_B$
$\text{CH}_x\text{-CH}_2\text{-CH}_2\text{-CH}_y$	0.0	355.03	-68.19	791.32
$\text{CH}_x\text{-CH}_2\text{-CH-CH}_y$	-251.06	428.73	-111.85	441.27
$\text{CH}_x\text{-CH}_2\text{-C-CH}_y$	0.0	0.0	0.0	461.29
$\text{CH}_x\text{-CH-CH-CH}_y$	-251.06	428.73	-111.85	441.27
$\text{CH}_x\text{-CH}_2\text{-CH}_2\text{-C}(\text{sp})$	94.88	162.00	-205.40	980.40
$\text{CH}_x\text{-CH}_2\text{-C}(\text{sp})\equiv\text{C}(\text{sp})$	0	0	0	0
$\text{CH}_x\text{-CH}_2\text{-C}(\text{sp})\equiv\text{CH}(\text{sp})$	0	0	0	0
$\text{CH}_x\text{-C}(\text{sp})\equiv\text{C}(\text{sp})\text{-CH}_y$	0	0	0	0

## S2 Fixed vs. flexible bonds

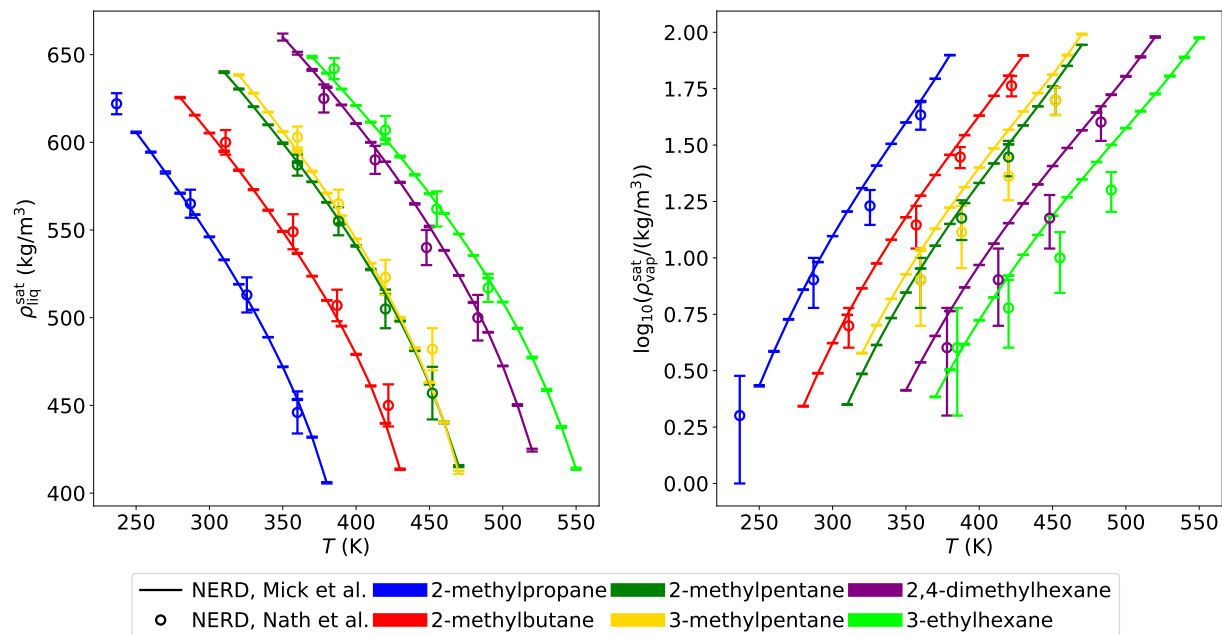


Figure S1: Comparison of saturated liquid (left panel) and vapor (right panel) densities for fixed (Mick et al.) and flexible (Nath et al.) bonds. Mick et al. used GCMC-HR while Nath et al. utilized GEMC. Note that Nath et al. did not report tabulated values for vapor pressure or enthalpy of vaporization.

### S3 CBMC acceptance rates

Table S4: Percentage acceptance of CBMC moves for cyclohexane with MiPPE force field. Averages were computed from 20 replicate simulations with  $L_{\text{box}} = 3.0$  nm.

$T$ (K)	$\mu$ (K)	Acceptance (%)
450	-4370	68.8
500	-4370	75.1
550	-4370	39.5
500	-4135	5.50
460	-4025	2.20
410	-3890	1.11
360	-3790	0.19

## S4 Compiler and hardware

With the exception of the 20 replicates performed for MiPPE cyclohexane (3.0 and 3.5 nm box length) and TAMie cyclohexane, all simulations are run on a Linux 4.4.0-112-generic x86\_64 on an Intel(R) Xeon(R) CPU E5-2699 v4 @ 2.20GHz machine. On this machine, GOMC was erroneously compiled using the sub-optimal GNU compiler collection (GCC) instead of the preferred Intel compiler. GOMC compiled with the Intel compiler typically runs approximately twice as fast as GOMC compiled with the GCC compiler.

The 20 replicate simulations for MiPPE cyclohexane (3.0 and 3.5 nm box length) and TAMie cyclohexane utilize several different machine hardware architectures, listed in Table S5. GOMC was compiled with the Intel compiler on each of these machines.

Table S5: Machine hardware for 20 replicate simulations of MiPPE cyclohexane

Intel(R) Core(TM) i7-4790K CPU @ 4.00GHz
Intel(R) Core(TM) i5-3570 CPU @ 3.40GHz
Intel(R) Core(TM) i5-2500K CPU @ 3.30GHz
Intel(R) Xeon(R) CPU X5450 @ 3.00GHz
Intel(R) Xeon(R) CPU X5355 @ 2.66GHz
Intel(R) Xeon(R) CPU E5-2640 v3 @ 2.60GHz
Intel(R) Core(TM)2 Quad CPU Q6600 @ 2.40GHz

## S5 $\epsilon$ -scaling

### S5.1 Tabulated $\psi$ values

Table S6: Optimal  $\epsilon$ -scaling parameter ( $\psi$ ) values and corresponding scoring function. Abbreviations correspond to those in Figure 2.

Molecular name	Abbreviation	Optimal $\psi$	Optimal score
Branched alkanes			
2-methylpropane	2MC <sub>3</sub>	1.0015	0.3883
2-methylbutane	2MC <sub>4</sub>	1.0025	0.4281
2-methylpentane	2MC <sub>5</sub>	1.0020	0.4770
3-methylpentane	3MC <sub>5</sub>	1.0103	0.4050
2,2-dimethylpropane	22DMC <sub>3</sub>	1.0035	0.5132
2,2-dimethylbutane	22DMC <sub>4</sub>	0.9985	0.5445
2,3-dimethylbutane	23DMC <sub>4</sub>	1.0000	0.4724
2,2,4-trimethylpentane	234TMC <sub>5</sub>	1.0005	0.4367
Alkynes			
1-ethyne	C <sub>2</sub>	1.0005	0.2931
1-propyne	C <sub>3</sub>	0.9965	0.3307
1-butyne	1C <sub>4</sub>	1.0063	1.143
2-butyne	2C <sub>4</sub>	1.0031	0.3191
1-pentyne	1C <sub>5</sub>	1.0087	1.8505
2-pentyne	2C <sub>5</sub>	1.0186	1.3801
1-hexyne	1C <sub>6</sub>	1.0063	1.908
2-hexyne	2C <sub>6</sub>	1.0228	1.0594
1-heptyne	1C <sub>7</sub>	1.0066	0.8415
1-octyne	1C <sub>8</sub>	1.0034	0.9777
1-nonyne	1C <sub>9</sub>	1.0000	0.9128

## S5.2 Tabulated phase equilibria for optimal $\psi$

### S5.2.1 Branched alkanes

Table S7: GCMC-MBAR results for 2-methylpropane with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
390	390.5 <sub>4.5</sub>	85.7 <sub>2.5</sub>	2.718 <sub>0.030</sub>	9.90 <sub>0.20</sub>	0.569 <sub>0.011</sub>
380	418.0 <sub>2.3</sub>	65.5 <sub>2.0</sub>	2.272 <sub>0.017</sub>	11.81 <sub>0.14</sub>	0.639 <sub>0.016</sub>
370	440.86 <sub>0.43</sub>	51.3 <sub>1.3</sub>	1.8856 <sub>0.0088</sub>	13.31 <sub>0.11</sub>	0.695 <sub>0.015</sub>
360	460.16 <sub>0.83</sub>	40.72 <sub>0.64</sub>	1.5515 <sub>0.0048</sub>	14.524 <sub>0.080</sub>	0.740 <sub>0.011</sub>
350	477.25 <sub>0.86</sub>	32.43 <sub>0.28</sub>	1.2638 <sub>0.0038</sub>	15.559 <sub>0.056</sub>	0.78 <sub>0.01</sub>
340	492.86 <sub>0.74</sub>	25.78 <sub>0.16</sub>	1.0177 <sub>0.0034</sub>	16.473 <sub>0.048</sub>	0.81 <sub>0.01</sub>
330	507.34 <sub>0.80</sub>	20.38 <sub>0.14</sub>	0.8090 <sub>0.0033</sub>	17.292 <sub>0.053</sub>	0.84 <sub>0.01</sub>
320	521.13 <sub>0.61</sub>	15.97 <sub>0.11</sub>	0.6336 <sub>0.0034</sub>	18.042 <sub>0.053</sub>	0.87 <sub>0.01</sub>
310	534.54 <sub>0.29</sub>	12.374 <sub>0.074</sub>	0.4881 <sub>0.0036</sub>	18.745 <sub>0.042</sub>	0.89 <sub>0.01</sub>
300	547.12 <sub>0.22</sub>	9.452 <sub>0.060</sub>	0.3690 <sub>0.0035</sub>	19.389 <sub>0.035</sub>	0.910 <sub>0.010</sub>
290	558.97 <sub>0.19</sub>	7.101 <sub>0.076</sub>	0.2732 <sub>0.0032</sub>	19.980 <sub>0.039</sub>	0.927 <sub>0.012</sub>
280	570.80 <sub>0.34</sub>	5.230 <sub>0.094</sub>	0.1975 <sub>0.0027</sub>	20.552 <sub>0.048</sub>	0.943 <sub>0.019</sub>
270	582.96 <sub>0.45</sub>	3.76 <sub>0.10</sub>	0.1389 <sub>0.0025</sub>	21.122 <sub>0.071</sub>	0.956 <sub>0.034</sub>
260	593.85 <sub>0.25</sub>	2.63 <sub>0.10</sub>	0.0946 <sub>0.0027</sub>	21.62 <sub>0.12</sub>	0.966 <sub>0.060</sub>
250	603.32 <sub>0.94</sub>	1.787 <sub>0.089</sub>	0.0623 <sub>0.0034</sub>	22.06 <sub>0.24</sub>	0.97 <sub>0.10</sub>

Table S8: GCMC-MBAR results for 2-methylbutane with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
440	403.0 <sub>2.4</sub>	84.5 <sub>1.4</sub>	2.490 <sub>0.015</sub>	11.80 <sub>0.11</sub>	0.58 <sub>0.01</sub>
430	429.5 <sub>1.4</sub>	67.3 <sub>1.1</sub>	2.1160 <sub>0.0099</sub>	13.658 <sub>0.094</sub>	0.63 <sub>0.01</sub>
420	451.56 <sub>0.83</sub>	53.86 <sub>0.75</sub>	1.7868 <sub>0.0058</sub>	15.259 <sub>0.071</sub>	0.69 <sub>0.01</sub>
410	470.40 <sub>0.84</sub>	43.55 <sub>0.44</sub>	1.4985 <sub>0.0031</sub>	16.608 <sub>0.052</sub>	0.73 <sub>0.01</sub>
400	487.2 <sub>1.1</sub>	35.38 <sub>0.25</sub>	1.2466 <sub>0.0017</sub>	17.769 <sub>0.044</sub>	0.76 <sub>0.01</sub>
390	502.5 <sub>1.3</sub>	28.74 <sub>0.17</sub>	1.0277 <sub>0.0016</sub>	18.796 <sub>0.044</sub>	0.80 <sub>0.01</sub>
380	516.7 <sub>1.4</sub>	23.25 <sub>0.13</sub>	0.8385 <sub>0.0025</sub>	19.723 <sub>0.041</sub>	0.82 <sub>0.01</sub>
370	530.1 <sub>1.2</sub>	18.699 <sub>0.091</sub>	0.6765 <sub>0.0033</sub>	20.574 <sub>0.033</sub>	0.85 <sub>0.01</sub>
360	542.77 <sub>0.83</sub>	14.917 <sub>0.061</sub>	0.5388 <sub>0.0038</sub>	21.360 <sub>0.027</sub>	0.87 <sub>0.01</sub>
350	554.94 <sub>0.49</sub>	11.782 <sub>0.058</sub>	0.4234 <sub>0.0039</sub>	22.094 <sub>0.027</sub>	0.89 <sub>0.01</sub>
340	566.86 <sub>0.43</sub>	9.196 <sub>0.079</sub>	0.3274 <sub>0.0037</sub>	22.793 <sub>0.033</sub>	0.91 <sub>0.01</sub>
330	578.69 <sub>0.60</sub>	7.08 <sub>0.10</sub>	0.2489 <sub>0.0032</sub>	23.467 <sub>0.052</sub>	0.925 <sub>0.011</sub>
320	589.78 <sub>0.55</sub>	5.36 <sub>0.12</sub>	0.1856 <sub>0.0027</sub>	24.087 <sub>0.073</sub>	0.939 <sub>0.019</sub>
310	599.68 <sub>0.39</sub>	3.99 <sub>0.12</sub>	0.1354 <sub>0.0024</sub>	24.634 <sub>0.10</sub>	0.951 <sub>0.031</sub>
300	609.37 <sub>0.41</sub>	2.90 <sub>0.11</sub>	0.0965 <sub>0.0025</sub>	25.16 <sub>0.14</sub>	0.961 <sub>0.050</sub>
290	619.67 <sub>0.47</sub>	2.065 <sub>0.092</sub>	0.0670 <sub>0.0030</sub>	25.71 <sub>0.20</sub>	0.971 <sub>0.076</sub>



Table S9: GCMC-MBAR results for 2-methylpentane with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
470	424.43 <sub>0.72</sub>	75.06 <sub>0.98</sub>	2.0415 <sub>0.0063</sub>	14.355 <sub>0.063</sub>	0.60 <sub>0.01</sub>
460	446.63 <sub>0.44</sub>	59.92 <sub>0.85</sub>	1.7383 <sub>0.0032</sub>	16.259 <sub>0.079</sub>	0.65 <sub>0.01</sub>
450	466.02 <sub>0.34</sub>	48.40 <sub>0.58</sub>	1.4722 <sub>0.0024</sub>	17.883 <sub>0.082</sub>	0.70 <sub>0.01</sub>
440	483.12 <sub>0.37</sub>	39.46 <sub>0.30</sub>	1.2388 <sub>0.0030</sub>	19.257 <sub>0.075</sub>	0.74 <sub>0.01</sub>
430	498.62 <sub>0.46</sub>	32.30 <sub>0.13</sub>	1.0347 <sub>0.0032</sub>	20.456 <sub>0.064</sub>	0.77 <sub>0.01</sub>
420	513.07 <sub>0.46</sub>	26.404 <sub>0.097</sub>	0.8567 <sub>0.0028</sub>	21.538 <sub>0.049</sub>	0.80 <sub>0.01</sub>
410	526.79 <sub>0.34</sub>	21.50 <sub>0.12</sub>	0.7026 <sub>0.0021</sub>	22.536 <sub>0.031</sub>	0.83 <sub>0.01</sub>
400	539.84 <sub>0.33</sub>	17.39 <sub>0.13</sub>	0.5701 <sub>0.0016</sub>	23.462 <sub>0.027</sub>	0.85 <sub>0.01</sub>
390	552.17 <sub>0.47</sub>	13.96 <sub>0.12</sub>	0.4573 <sub>0.0015</sub>	24.320 <sub>0.043</sub>	0.87 <sub>0.01</sub>
380	563.75 <sub>0.44</sub>	11.10 <sub>0.10</sub>	0.3622 <sub>0.0018</sub>	25.113 <sub>0.052</sub>	0.890 <sub>0.011</sub>
370	574.77 <sub>0.27</sub>	8.738 <sub>0.085</sub>	0.2829 <sub>0.0022</sub>	25.853 <sub>0.051</sub>	0.907 <sub>0.015</sub>
360	585.55 <sub>0.30</sub>	6.794 <sub>0.076</sub>	0.2177 <sub>0.0027</sub>	26.562 <sub>0.058</sub>	0.922 <sub>0.021</sub>
350	596.34 <sub>0.30</sub>	5.208 <sub>0.079</sub>	0.1647 <sub>0.0031</sub>	27.257 <sub>0.083</sub>	0.936 <sub>0.030</sub>
340	606.99 <sub>0.31</sub>	3.928 <sub>0.084</sub>	0.1222 <sub>0.0035</sub>	27.93 <sub>0.12</sub>	0.948 <sub>0.045</sub>
330	617.01 <sub>0.30</sub>	2.909 <sub>0.083</sub>	0.0888 <sub>0.0039</sub>	28.55 <sub>0.18</sub>	0.959 <sub>0.067</sub>
320	626.41 <sub>0.21</sub>	2.111 <sub>0.076</sub>	0.0631 <sub>0.0043</sub>	29.13 <sub>0.26</sub>	0.968 <sub>0.099</sub>

Table S10: GCMC-MBAR results for 3-methylpentane with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
480	428.2 <sub>1.5</sub>	75 <sub>12</sub>	2.171 <sub>0.060</sub>	14.81 <sub>0.93</sub>	0.625 <sub>0.090</sub>
470	449.9 <sub>1.6</sub>	61.1 <sub>8.5</sub>	1.863 <sub>0.021</sub>	16.56 <sub>0.84</sub>	0.672 <sub>0.089</sub>
460	468.1 <sub>1.3</sub>	50.3 <sub>4.0</sub>	1.5910 <sub>0.0071</sub>	18.01 <sub>0.53</sub>	0.712 <sub>0.060</sub>
450	484.5 <sub>1.2</sub>	41.6 <sub>1.1</sub>	1.350 <sub>0.014</sub>	19.28 <sub>0.23</sub>	0.747 <sub>0.027</sub>
440	500.3 <sub>1.1</sub>	34.45 <sub>0.21</sub>	1.137 <sub>0.015</sub>	20.464 <sub>0.074</sub>	0.78 <sub>0.01</sub>
430	515.5 <sub>1.0</sub>	28.41 <sub>0.46</sub>	0.950 <sub>0.013</sub>	21.563 <sub>0.068</sub>	0.81 <sub>0.01</sub>
420	529.38 <sub>0.62</sub>	23.34 <sub>0.40</sub>	0.786 <sub>0.010</sub>	22.556 <sub>0.054</sub>	0.83 <sub>0.01</sub>
410	541.89 <sub>0.69</sub>	19.07 <sub>0.30</sub>	0.6449 <sub>0.0079</sub>	23.443 <sub>0.043</sub>	0.85 <sub>0.01</sub>
400	553.62 <sub>0.90</sub>	15.49 <sub>0.24</sub>	0.5233 <sub>0.0062</sub>	24.264 <sub>0.049</sub>	0.88 <sub>0.01</sub>
390	565.07 <sub>0.63</sub>	12.48 <sub>0.20</sub>	0.4198 <sub>0.0048</sub>	25.051 <sub>0.039</sub>	0.89 <sub>0.01</sub>
380	575.97 <sub>0.45</sub>	9.95 <sub>0.17</sub>	0.3326 <sub>0.0039</sub>	25.789 <sub>0.043</sub>	0.912 <sub>0.014</sub>
370	586 <sub>95</sub>	7.8 <sub>1.6</sub>	0.260 <sub>0.066</sub>	26.5 <sub>4.1</sub>	0.93 <sub>0.18</sub>
360	597.06 <sub>0.27</sub>	6.10 <sub>0.13</sub>	0.2001 <sub>0.0035</sub>	27.178 <sub>0.094</sub>	0.945 <sub>0.029</sub>
350	607.29 <sub>0.23</sub>	4.67 <sub>0.12</sub>	0.1514 <sub>0.0039</sub>	27.83 <sub>0.14</sub>	0.959 <sub>0.043</sub>

Table S11: GCMC-MBAR results for 2,2-dimethylpropane with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
410	413.2 <sub>2.6</sub>	75.9 <sub>2.6</sub>	2.173 <sub>0.029</sub>	11.65 <sub>0.11</sub>	0.606 <sub>0.013</sub>
400	437.1 <sub>2.3</sub>	59.3 <sub>1.8</sub>	1.831 <sub>0.018</sub>	13.410 <sub>0.083</sub>	0.669 <sub>0.014</sub>
390	457.4 <sub>1.5</sub>	47.4 <sub>1.1</sub>	1.532 <sub>0.010</sub>	14.814 <sub>0.051</sub>	0.719 <sub>0.013</sub>
380	474.6 <sub>1.1</sub>	38.29 <sub>0.72</sub>	1.2725 <sub>0.0056</sub>	15.944 <sub>0.045</sub>	0.759 <sub>0.011</sub>
370	489.78 <sub>0.98</sub>	31.00 <sub>0.48</sub>	1.0468 <sub>0.0035</sub>	16.909 <sub>0.045</sub>	0.792 <sub>0.011</sub>
360	504.12 <sub>0.89</sub>	25.02 <sub>0.33</sub>	0.8520 <sub>0.0036</sub>	17.785 <sub>0.044</sub>	0.821 <sub>0.012</sub>
350	518.05 <sub>0.86</sub>	20.05 <sub>0.21</sub>	0.6853 <sub>0.0043</sub>	18.604 <sub>0.046</sub>	0.847 <sub>0.011</sub>
340	531.32 <sub>0.93</sub>	15.93 <sub>0.18</sub>	0.5437 <sub>0.0045</sub>	19.363 <sub>0.057</sub>	0.871 <sub>0.011</sub>
330	543.65 <sub>0.96</sub>	12.52 <sub>0.23</sub>	0.4252 <sub>0.0041</sub>	20.056 <sub>0.078</sub>	0.893 <sub>0.016</sub>
320	555.0 <sub>1.0</sub>	9.71 <sub>0.28</sub>	0.3271 <sub>0.0037</sub>	20.68 <sub>0.11</sub>	0.913 <sub>0.029</sub>
310	565.5 <sub>1.4</sub>	7.43 <sub>0.32</sub>	0.2473 <sub>0.0044</sub>	21.25 <sub>0.18</sub>	0.931 <sub>0.052</sub>
300	576.4 <sub>1.8</sub>	5.60 <sub>0.33</sub>	0.1832 <sub>0.0061</sub>	21.82 <sub>0.28</sub>	0.947 <sub>0.087</sub>
290	588.4 <sub>1.3</sub>	4.13 <sub>0.32</sub>	0.1327 <sub>0.0084</sub>	22.41 <sub>0.38</sub>	0.96 <sub>0.14</sub>
280	600.2 <sub>1.1</sub>	2.98 <sub>0.29</sub>	0.093 <sub>0.011</sub>	22.98 <sub>0.55</sub>	0.97 <sub>0.22</sub>

Table S12: GCMC-MBAR results for 2,2-dimethylbutane with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
470	410.4 <sub>5.2</sub>	86.7 <sub>8.5</sub>	2.34 <sub>0.11</sub>	12.61 <sub>0.20</sub>	0.595 <sub>0.027</sub>
460	436.5 <sub>4.9</sub>	70.3 <sub>8.7</sub>	2.014 <sub>0.071</sub>	14.48 <sub>0.34</sub>	0.645 <sub>0.049</sub>
450	457.9 <sub>3.1</sub>	57.7 <sub>7.2</sub>	1.724 <sub>0.036</sub>	16.04 <sub>0.45</sub>	0.689 <sub>0.061</sub>
440	476.4 <sub>1.5</sub>	47.7 <sub>4.6</sub>	1.467 <sub>0.011</sub>	17.37 <sub>0.41</sub>	0.725 <sub>0.057</sub>
430	493.01 <sub>0.95</sub>	39.4 <sub>2.1</sub>	1.2386 <sub>0.0045</sub>	18.53 <sub>0.25</sub>	0.757 <sub>0.039</sub>
420	508.03 <sub>0.71</sub>	32.56 <sub>0.58</sub>	1.0375 <sub>0.0094</sub>	19.573 <sub>0.098</sub>	0.786 <sub>0.020</sub>
410	521.74 <sub>0.38</sub>	26.79 <sub>0.14</sub>	0.862 <sub>0.010</sub>	20.513 <sub>0.035</sub>	0.81 <sub>0.01</sub>
400	534.75 <sub>0.29</sub>	21.94 <sub>0.16</sub>	0.7088 <sub>0.0094</sub>	21.383 <sub>0.026</sub>	0.84 <sub>0.01</sub>
390	547.49 <sub>0.34</sub>	17.86 <sub>0.11</sub>	0.5771 <sub>0.0086</sub>	22.210 <sub>0.027</sub>	0.859 <sub>0.010</sub>
380	559.93 <sub>0.37</sub>	14.416 <sub>0.067</sub>	0.4643 <sub>0.0081</sub>	22.998 <sub>0.038</sub>	0.878 <sub>0.013</sub>
370	571.83 <sub>0.42</sub>	11.528 <sub>0.058</sub>	0.3687 <sub>0.0077</sub>	23.738 <sub>0.053</sub>	0.896 <sub>0.017</sub>
360	582.92 <sub>0.45</sub>	9.115 <sub>0.074</sub>	0.2885 <sub>0.0074</sub>	24.416 <sub>0.062</sub>	0.911 <sub>0.022</sub>
350	593.10 <sub>0.52</sub>	7.115 <sub>0.099</sub>	0.2224 <sub>0.0070</sub>	25.030 <sub>0.081</sub>	0.925 <sub>0.029</sub>
340	602.70 <sub>0.55</sub>	5.47 <sub>0.12</sub>	0.1685 <sub>0.0067</sub>	25.60 <sub>0.11</sub>	0.938 <sub>0.041</sub>
330	612.20 <sub>0.41</sub>	4.14 <sub>0.14</sub>	0.1253 <sub>0.0065</sub>	26.15 <sub>0.16</sub>	0.949 <sub>0.061</sub>
320	621.65 <sub>0.33</sub>	3.08 <sub>0.13</sub>	0.0912 <sub>0.0065</sub>	26.69 <sub>0.23</sub>	0.959 <sub>0.091</sub>
310	631.20 <sub>0.30</sub>	2.24 <sub>0.12</sub>	0.0649 <sub>0.0068</sub>	27.22 <sub>0.34</sub>	0.97 <sub>0.14</sub>

Table S13: GCMC-MBAR results for 2,3-dimethylbutane with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_{\text{v}}$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
480	411.7 <sub>1.1</sub>	88.4 <sub>2.2</sub>	2.369 <sub>0.027</sub>	12.87 <sub>0.14</sub>	0.58 <sub>0.01</sub>
470	437.1 <sub>1.0</sub>	71.6 <sub>2.1</sub>	2.037 <sub>0.019</sub>	14.80 <sub>0.17</sub>	0.627 <sub>0.013</sub>
460	459.23 <sub>0.86</sub>	58.1 <sub>1.6</sub>	1.742 <sub>0.013</sub>	16.53 <sub>0.18</sub>	0.676 <sub>0.015</sub>
450	478.07 <sub>0.79</sub>	47.5 <sub>1.1</sub>	1.4813 <sub>0.0074</sub>	18.00 <sub>0.16</sub>	0.718 <sub>0.014</sub>
440	494.54 <sub>0.81</sub>	39.07 <sub>0.72</sub>	1.2513 <sub>0.0041</sub>	19.26 <sub>0.13</sub>	0.754 <sub>0.012</sub>
430	509.51 <sub>0.77</sub>	32.20 <sub>0.43</sub>	1.0493 <sub>0.0024</sub>	20.37 <sub>0.10</sub>	0.79 <sub>0.01</sub>
420	523.51 <sub>0.64</sub>	26.49 <sub>0.25</sub>	0.8726 <sub>0.0019</sub>	21.382 <sub>0.079</sub>	0.81 <sub>0.01</sub>
410	536.78 <sub>0.54</sub>	21.70 <sub>0.15</sub>	0.7189 <sub>0.0020</sub>	22.310 <sub>0.063</sub>	0.84 <sub>0.01</sub>
400	549.39 <sub>0.59</sub>	17.68 <sub>0.10</sub>	0.5863 <sub>0.0021</sub>	23.171 <sub>0.060</sub>	0.86 <sub>0.01</sub>
390	561.38 <sub>0.73</sub>	14.296 <sub>0.082</sub>	0.4727 <sub>0.0023</sub>	23.971 <sub>0.062</sub>	0.88 <sub>0.01</sub>
380	572.70 <sub>0.68</sub>	11.455 <sub>0.085</sub>	0.3764 <sub>0.0025</sub>	24.712 <sub>0.056</sub>	0.896 <sub>0.011</sub>
370	583.42 <sub>0.40</sub>	9.083 <sub>0.092</sub>	0.2957 <sub>0.0028</sub>	25.402 <sub>0.050</sub>	0.912 <sub>0.016</sub>
360	593.75 <sub>0.21</sub>	7.115 <sub>0.095</sub>	0.2289 <sub>0.0033</sub>	26.054 <sub>0.064</sub>	0.926 <sub>0.024</sub>
350	603.97 <sub>0.20</sub>	5.497 <sub>0.091</sub>	0.1742 <sub>0.0037</sub>	26.686 <sub>0.091</sub>	0.938 <sub>0.034</sub>
340	614.11 <sub>0.22</sub>	4.179 <sub>0.082</sub>	0.1302 <sub>0.0042</sub>	27.30 <sub>0.13</sub>	0.950 <sub>0.047</sub>
330	623.72 <sub>0.26</sub>	3.121 <sub>0.069</sub>	0.0953 <sub>0.0047</sub>	27.87 <sub>0.18</sub>	0.959 <sub>0.066</sub>
320	632.83 <sub>0.34</sub>	2.284 <sub>0.057</sub>	0.0682 <sub>0.0050</sub>	28.41 <sub>0.25</sub>	0.968 <sub>0.092</sub>

Table S14: GCMC-MBAR results for 2,2,4-trimethylpentane with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
530	401.6 <sub>2.0</sub>	97.3 <sub>2.8</sub>	2.113 <sub>0.013</sub>	13.65 <sub>0.15</sub>	0.563 <sub>0.014</sub>
520	428.08 <sub>0.71</sub>	79.0 <sub>1.7</sub>	1.8383 <sub>0.0085</sub>	15.95 <sub>0.17</sub>	0.615 <sub>0.012</sub>
510	449.27 <sub>0.69</sub>	64.82 <sub>0.80</sub>	1.5924 <sub>0.0067</sub>	17.85 <sub>0.12</sub>	0.66 <sub>0.01</sub>
500	467.26 <sub>0.83</sub>	53.68 <sub>0.34</sub>	1.3733 <sub>0.0059</sub>	19.449 <sub>0.068</sub>	0.70 <sub>0.01</sub>
490	484.19 <sub>0.91</sub>	44.67 <sub>0.32</sub>	1.1784 <sub>0.0050</sub>	20.898 <sub>0.058</sub>	0.74 <sub>0.01</sub>
480	500.20 <sub>0.83</sub>	37.25 <sub>0.39</sub>	1.0051 <sub>0.0038</sub>	22.224 <sub>0.066</sub>	0.77 <sub>0.01</sub>
470	514.73 <sub>0.48</sub>	31.08 <sub>0.39</sub>	0.8515 <sub>0.0027</sub>	23.414 <sub>0.060</sub>	0.80 <sub>0.01</sub>
460	527.91 <sub>0.42</sub>	25.89 <sub>0.30</sub>	0.7160 <sub>0.0022</sub>	24.486 <sub>0.050</sub>	0.83 <sub>0.01</sub>
450	540.02 <sub>0.69</sub>	21.49 <sub>0.18</sub>	0.5973 <sub>0.0026</sub>	25.458 <sub>0.051</sub>	0.85 <sub>0.01</sub>
440	551.36 <sub>0.72</sub>	17.746 <sub>0.095</sub>	0.4939 <sub>0.0030</sub>	26.355 <sub>0.050</sub>	0.87 <sub>0.01</sub>
430	562.92 <sub>0.74</sub>	14.553 <sub>0.071</sub>	0.4046 <sub>0.0031</sub>	27.239 <sub>0.050</sub>	0.89 <sub>0.01</sub>
420	574.83 <sub>0.92</sub>	11.836 <sub>0.082</sub>	0.3278 <sub>0.0033</sub>	28.116 <sub>0.049</sub>	0.906 <sub>0.013</sub>
410	585.23 <sub>0.62</sub>	9.54 <sub>0.11</sub>	0.2626 <sub>0.0037</sub>	28.885 <sub>0.042</sub>	0.922 <sub>0.022</sub>
400	594.42 <sub>0.22</sub>	7.62 <sub>0.14</sub>	0.2078 <sub>0.0042</sub>	29.567 <sub>0.098</sub>	0.937 <sub>0.036</sub>
390	604.54 <sub>0.21</sub>	6.02 <sub>0.18</sub>	0.1623 <sub>0.0051</sub>	30.29 <sub>0.17</sub>	0.949 <sub>0.058</sub>
380	615.63 <sub>0.16</sub>	4.70 <sub>0.20</sub>	0.1247 <sub>0.0061</sub>	31.04 <sub>0.27</sub>	0.959 <sub>0.089</sub>
370	625.43 <sub>0.14</sub>	3.62 <sub>0.20</sub>	0.0942 <sub>0.0072</sub>	31.71 <sub>0.40</sub>	0.97 <sub>0.13</sub>

## S5.2.2 Alkynes

Table S15: GCMC-MBAR results for ethyne with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
290	421.8 <sub>1.7</sub>	69.4 <sub>1.8</sub>	4.071 <sub>0.027</sub>	8.835 <sub>0.094</sub>	0.634 <sub>0.013</sub>
280	451.1 <sub>1.2</sub>	50.48 <sub>0.65</sub>	3.187 <sub>0.018</sub>	10.290 <sub>0.067</sub>	0.71 <sub>0.01</sub>
270	475.2 <sub>2.5</sub>	37.51 <sub>0.33</sub>	2.457 <sub>0.013</sub>	11.409 <sub>0.072</sub>	0.76 <sub>0.01</sub>
260	497.6 <sub>1.7</sub>	27.84 <sub>0.34</sub>	1.8584 <sub>0.0078</sub>	12.379 <sub>0.043</sub>	0.80 <sub>0.01</sub>
250	518.16 <sub>0.83</sub>	20.45 <sub>0.22</sub>	1.3737 <sub>0.0065</sub>	13.231 <sub>0.027</sub>	0.84 <sub>0.01</sub>
240	537.09 <sub>0.92</sub>	14.759 <sub>0.088</sub>	0.9884 <sub>0.0067</sub>	13.991 <sub>0.021</sub>	0.87 <sub>0.01</sub>
230	554.32 <sub>0.93</sub>	10.40 <sub>0.10</sub>	0.6899 <sub>0.0056</sub>	14.664 <sub>0.024</sub>	0.90 <sub>0.01</sub>
220	571.08 <sub>0.74</sub>	7.12 <sub>0.12</sub>	0.4649 <sub>0.0046</sub>	15.298 <sub>0.035</sub>	0.929 <sub>0.019</sub>

Table S16: GCMC-MBAR results for propyne with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
380	436.3 <sub>5.1</sub>	85.3 <sub>1.7</sub>	3.923 <sub>0.045</sub>	10.64 <sub>0.20</sub>	0.58 <sub>0.01</sub>
370	468.9 <sub>3.2</sub>	64.9 <sub>1.9</sub>	3.234 <sub>0.036</sub>	12.56 <sub>0.18</sub>	0.649 <sub>0.012</sub>
360	495.3 <sub>1.2</sub>	50.2 <sub>1.4</sub>	2.643 <sub>0.025</sub>	14.12 <sub>0.13</sub>	0.705 <sub>0.014</sub>
350	517.6 <sub>1.1</sub>	39.26 <sub>0.77</sub>	2.137 <sub>0.018</sub>	15.39 <sub>0.10</sub>	0.750 <sub>0.011</sub>
340	537.2 <sub>1.3</sub>	30.77 <sub>0.40</sub>	1.708 <sub>0.013</sub>	16.469 <sub>0.084</sub>	0.79 <sub>0.01</sub>
330	554.7 <sub>1.7</sub>	24.00 <sub>0.28</sub>	1.3456 <sub>0.0097</sub>	17.416 <sub>0.089</sub>	0.82 <sub>0.01</sub>
320	570.4 <sub>2.2</sub>	18.56 <sub>0.23</sub>	1.0444 <sub>0.0070</sub>	18.26 <sub>0.10</sub>	0.85 <sub>0.01</sub>
310	585.8 <sub>2.1</sub>	14.18 <sub>0.18</sub>	0.7969 <sub>0.0050</sub>	19.048 <sub>0.098</sub>	0.87 <sub>0.01</sub>
300	601.7 <sub>1.6</sub>	10.67 <sub>0.12</sub>	0.5964 <sub>0.0038</sub>	19.823 <sub>0.073</sub>	0.90 <sub>0.01</sub>
290	616.5 <sub>2.2</sub>	7.899 <sub>0.078</sub>	0.4365 <sub>0.0038</sub>	20.532 <sub>0.077</sub>	0.918 <sub>0.012</sub>

Table S17: GCMC-MBAR results for 1-butyne with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
410	452.8 <sub>4.0</sub>	70.9 <sub>5.6</sub>	2.840 <sub>0.093</sub>	13.41 <sub>0.28</sub>	0.636 <sub>0.032</sub>
400	478.29 <sub>0.92</sub>	55.9 <sub>2.7</sub>	2.366 <sub>0.066</sub>	15.10 <sub>0.20</sub>	0.689 <sub>0.016</sub>
390	500.1 <sub>1.3</sub>	44.5 <sub>1.3</sub>	1.955 <sub>0.052</sub>	16.52 <sub>0.12</sub>	0.73 <sub>0.01</sub>
380	520.1 <sub>1.4</sub>	35.6 <sub>1.1</sub>	1.599 <sub>0.043</sub>	17.771 <sub>0.095</sub>	0.77 <sub>0.01</sub>
370	538.8 <sub>1.1</sub>	28.3 <sub>1.2</sub>	1.293 <sub>0.033</sub>	18.90 <sub>0.10</sub>	0.802 <sub>0.015</sub>
360	555.7 <sub>1.3</sub>	22.5 <sub>1.2</sub>	1.032 <sub>0.021</sub>	19.92 <sub>0.14</sub>	0.831 <sub>0.031</sub>
350	571.1 <sub>1.3</sub>	17.64 <sub>0.98</sub>	0.8126 <sub>0.0098</sub>	20.83 <sub>0.15</sub>	0.856 <sub>0.041</sub>
340	585.1 <sub>1.0</sub>	13.72 <sub>0.65</sub>	0.6302 <sub>0.0033</sub>	21.64 <sub>0.16</sub>	0.879 <sub>0.044</sub>
330	598.8 <sub>2.7</sub>	10.54 <sub>0.36</sub>	0.4806 <sub>0.0057</sub>	22.41 <sub>0.19</sub>	0.899 <sub>0.042</sub>
320	613.6 <sub>4.5</sub>	7.97 <sub>0.19</sub>	0.3595 <sub>0.0080</sub>	23.21 <sub>0.25</sub>	0.917 <sub>0.039</sub>
310	627.0 <sub>2.2</sub>	5.92 <sub>0.13</sub>	0.2629 <sub>0.0086</sub>	23.93 <sub>0.17</sub>	0.932 <sub>0.040</sub>

Table S18: GCMC-MBAR results for 2-butyne with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
450	437.2 <sub>4.2</sub>	90.5 <sub>3.2</sub>	3.554 <sub>0.053</sub>	12.28 <sub>0.20</sub>	0.568 <sub>0.014</sub>
440	470.0 <sub>3.7</sub>	71.8 <sub>2.3</sub>	3.012 <sub>0.037</sub>	14.30 <sub>0.20</sub>	0.620 <sub>0.017</sub>
430	494.6 <sub>1.6</sub>	57.4 <sub>1.2</sub>	2.535 <sub>0.029</sub>	15.96 <sub>0.15</sub>	0.668 <sub>0.014</sub>
420	514.58 <sub>0.50</sub>	46.27 <sub>0.57</sub>	2.118 <sub>0.025</sub>	17.336 <sub>0.085</sub>	0.71 <sub>0.01</sub>
410	532.82 <sub>0.48</sub>	37.39 <sub>0.49</sub>	1.757 <sub>0.022</sub>	18.554 <sub>0.040</sub>	0.75 <sub>0.01</sub>
400	550.44 <sub>0.50</sub>	30.16 <sub>0.62</sub>	1.444 <sub>0.017</sub>	19.684 <sub>0.065</sub>	0.78 <sub>0.01</sub>
390	567.00 <sub>0.68</sub>	24.23 <sub>0.63</sub>	1.175 <sub>0.012</sub>	20.72 <sub>0.11</sub>	0.809 <sub>0.013</sub>
380	582.34 <sub>0.91</sub>	19.36 <sub>0.53</sub>	0.9452 <sub>0.0067</sub>	21.66 <sub>0.13</sub>	0.836 <sub>0.017</sub>
370	596.57 <sub>0.71</sub>	15.35 <sub>0.39</sub>	0.7513 <sub>0.0033</sub>	22.51 <sub>0.12</sub>	0.860 <sub>0.019</sub>
360	610.15 <sub>0.68</sub>	12.07 <sub>0.25</sub>	0.5893 <sub>0.0027</sub>	23.32 <sub>0.12</sub>	0.882 <sub>0.019</sub>
350	623.0 <sub>1.1</sub>	9.39 <sub>0.13</sub>	0.4552 <sub>0.0037</sub>	24.06 <sub>0.12</sub>	0.901 <sub>0.018</sub>
340	635.13 <sub>0.92</sub>	7.213 <sub>0.060</sub>	0.3458 <sub>0.0041</sub>	24.75 <sub>0.10</sub>	0.917 <sub>0.016</sub>
330	647.43 <sub>0.75</sub>	5.454 <sub>0.041</sub>	0.2577 <sub>0.0039</sub>	25.410 <sub>0.086</sub>	0.931 <sub>0.015</sub>

Table S19: GCMC-MBAR results for 1-pentyne with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
450	445.1 <sub>1.3</sub>	77.7 <sub>1.0</sub>	2.582 <sub>0.012</sub>	14.14 <sub>0.12</sub>	0.61 <sub>0.01</sub>
440	471.10 <sub>0.57</sub>	61.18 <sub>0.83</sub>	2.1759 <sub>0.0091</sub>	16.143 <sub>0.079</sub>	0.66 <sub>0.01</sub>
430	493.03 <sub>0.73</sub>	48.93 <sub>0.57</sub>	1.8221 <sub>0.0082</sub>	17.791 <sub>0.054</sub>	0.71 <sub>0.01</sub>
420	512.1 <sub>1.0</sub>	39.44 <sub>0.30</sub>	1.5143 <sub>0.0083</sub>	19.182 <sub>0.025</sub>	0.75 <sub>0.01</sub>
410	529.2 <sub>1.4</sub>	31.85 <sub>0.22</sub>	1.2476 <sub>0.0079</sub>	20.400 <sub>0.045</sub>	0.78 <sub>0.01</sub>
400	545.2 <sub>1.3</sub>	25.65 <sub>0.31</sub>	1.0180 <sub>0.0064</sub>	21.503 <sub>0.073</sub>	0.81 <sub>0.01</sub>
390	560.39 <sub>0.90</sub>	20.55 <sub>0.33</sub>	0.8216 <sub>0.0042</sub>	22.528 <sub>0.075</sub>	0.84 <sub>0.01</sub>
380	574.87 <sub>0.92</sub>	16.34 <sub>0.27</sub>	0.6551 <sub>0.0022</sub>	23.483 <sub>0.088</sub>	0.865 <sub>0.012</sub>
370	588.2 <sub>1.7</sub>	12.87 <sub>0.20</sub>	0.5155 <sub>0.0014</sub>	24.35 <sub>0.13</sub>	0.887 <sub>0.014</sub>
360	600.8 <sub>2.1</sub>	10.03 <sub>0.16</sub>	0.4000 <sub>0.0020</sub>	25.16 <sub>0.15</sub>	0.907 <sub>0.018</sub>
350	613.6 <sub>1.5</sub>	7.72 <sub>0.15</sub>	0.3052 <sub>0.0029</sub>	25.96 <sub>0.14</sub>	0.925 <sub>0.026</sub>
340	626.2 <sub>1.2</sub>	5.86 <sub>0.16</sub>	0.2288 <sub>0.0041</sub>	26.72 <sub>0.17</sub>	0.941 <sub>0.042</sub>

Table S20: GCMC-MBAR results for 2-pentyne with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
470	470.2 <sub>2.0</sub>	67.8 <sub>1.5</sub>	2.443 <sub>0.022</sub>	16.285 <sub>0.075</sub>	0.628 <sub>0.016</sub>
460	493.37 <sub>0.97</sub>	54.31 <sub>0.93</sub>	2.066 <sub>0.024</sub>	18.127 <sub>0.084</sub>	0.677 <sub>0.016</sub>
450	513.26 <sub>0.50</sub>	43.98 <sub>0.45</sub>	1.735 <sub>0.025</sub>	19.667 <sub>0.065</sub>	0.718 <sub>0.013</sub>
440	531.38 <sub>0.38</sub>	35.74 <sub>0.41</sub>	1.447 <sub>0.023</sub>	21.022 <sub>0.031</sub>	0.75 <sub>0.01</sub>
430	548.11 <sub>0.47</sub>	29.01 <sub>0.58</sub>	1.197 <sub>0.020</sub>	22.242 <sub>0.067</sub>	0.79 <sub>0.01</sub>
420	563.44 <sub>0.72</sub>	23.48 <sub>0.64</sub>	0.981 <sub>0.015</sub>	23.34 <sub>0.12</sub>	0.81 <sub>0.01</sub>
410	577.72 <sub>0.97</sub>	18.92 <sub>0.59</sub>	0.796 <sub>0.011</sub>	24.35 <sub>0.16</sub>	0.840 <sub>0.015</sub>
400	591.5 <sub>1.4</sub>	15.15 <sub>0.47</sub>	0.6386 <sub>0.0070</sub>	25.30 <sub>0.20</sub>	0.863 <sub>0.018</sub>
390	604.8 <sub>1.9</sub>	12.03 <sub>0.32</sub>	0.5062 <sub>0.0042</sub>	26.19 <sub>0.22</sub>	0.884 <sub>0.018</sub>
380	617.3 <sub>1.8</sub>	9.46 <sub>0.18</sub>	0.3957 <sub>0.0027</sub>	27.01 <sub>0.19</sub>	0.902 <sub>0.014</sub>
370	629.1 <sub>1.5</sub>	7.353 <sub>0.077</sub>	0.3048 <sub>0.0022</sub>	27.78 <sub>0.14</sub>	0.92 <sub>0.01</sub>
360	640.7 <sub>1.8</sub>	5.637 <sub>0.054</sub>	0.2308 <sub>0.0024</sub>	28.52 <sub>0.12</sub>	0.932 <sub>0.016</sub>
350	652.0 <sub>1.7</sub>	4.255 <sub>0.079</sub>	0.1717 <sub>0.0030</sub>	29.229 <sub>0.082</sub>	0.944 <sub>0.032</sub>



Table S21: GCMC-MBAR results for 1-hexyne with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
490	433.3 <sub>7.6</sub>	81.5 <sub>3.5</sub>	2.435 <sub>0.037</sub>	15.09 <sub>0.17</sub>	0.602 <sub>0.017</sub>
480	461.1 <sub>2.9</sub>	65.5 <sub>2.5</sub>	2.080 <sub>0.022</sub>	17.26 <sub>0.14</sub>	0.654 <sub>0.019</sub>
470	483.65 <sub>0.76</sub>	53.2 <sub>1.3</sub>	1.766 <sub>0.014</sub>	19.07 <sub>0.11</sub>	0.698 <sub>0.013</sub>
460	502.56 <sub>0.55</sub>	43.45 <sub>0.54</sub>	1.489 <sub>0.010</sub>	20.586 <sub>0.041</sub>	0.74 <sub>0.01</sub>
450	519.59 <sub>0.69</sub>	35.56 <sub>0.36</sub>	1.2468 <sub>0.0083</sub>	21.936 <sub>0.038</sub>	0.77 <sub>0.01</sub>
440	536.0 <sub>1.0</sub>	29.06 <sub>0.41</sub>	1.0351 <sub>0.0065</sub>	23.201 <sub>0.073</sub>	0.80 <sub>0.01</sub>
430	551.44 <sub>0.96</sub>	23.66 <sub>0.47</sub>	0.8514 <sub>0.0043</sub>	24.36 <sub>0.10</sub>	0.827 <sub>0.014</sub>
420	565.34 <sub>0.57</sub>	19.17 <sub>0.42</sub>	0.6934 <sub>0.0031</sub>	25.40 <sub>0.11</sub>	0.851 <sub>0.019</sub>
410	578.56 <sub>0.49</sub>	15.43 <sub>0.30</sub>	0.5583 <sub>0.0037</sub>	26.378 <sub>0.089</sub>	0.872 <sub>0.021</sub>
400	591.16 <sub>0.88</sub>	12.31 <sub>0.18</sub>	0.4443 <sub>0.0049</sub>	27.296 <sub>0.062</sub>	0.892 <sub>0.022</sub>
390	602.46 <sub>0.56</sub>	9.72 <sub>0.10</sub>	0.3488 <sub>0.0056</sub>	28.119 <sub>0.069</sub>	0.909 <sub>0.022</sub>
380	612.80 <sub>0.61</sub>	7.587 <sub>0.080</sub>	0.2702 <sub>0.0059</sub>	28.86 <sub>0.10</sub>	0.926 <sub>0.027</sub>
370	624.2 <sub>1.5</sub>	5.851 <sub>0.083</sub>	0.2061 <sub>0.0060</sub>	29.65 <sub>0.15</sub>	0.940 <sub>0.037</sub>
360	637.2 <sub>1.1</sub>	4.445 <sub>0.086</sub>	0.1543 <sub>0.0064</sub>	30.50 <sub>0.19</sub>	0.953 <sub>0.053</sub>

Table S22: GCMC-MBAR results for 2-hexyne with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
500	467.54 <sub>0.99</sub>	66.01 <sub>0.94</sub>	2.140 <sub>0.019</sub>	17.945 <sub>0.093</sub>	0.64 <sub>0.01</sub>
490	488.44 <sub>0.86</sub>	53.67 <sub>0.62</sub>	1.825 <sub>0.016</sub>	19.766 <sub>0.070</sub>	0.69 <sub>0.01</sub>
480	506.42 <sub>0.87</sub>	44.07 <sub>0.54</sub>	1.547 <sub>0.012</sub>	21.286 <sub>0.048</sub>	0.72 <sub>0.01</sub>
470	523.2 <sub>1.3</sub>	36.28 <sub>0.58</sub>	1.3025 <sub>0.0090</sub>	22.660 <sub>0.039</sub>	0.75 <sub>0.01</sub>
460	539.7 <sub>1.9</sub>	29.82 <sub>0.56</sub>	1.0881 <sub>0.0054</sub>	23.957 <sub>0.044</sub>	0.784 <sub>0.012</sub>
450	555.2 <sub>1.7</sub>	24.42 <sub>0.46</sub>	0.9014 <sub>0.0029</sub>	25.157 <sub>0.042</sub>	0.810 <sub>0.015</sub>
440	569.3 <sub>1.2</sub>	19.91 <sub>0.33</sub>	0.7397 <sub>0.0032</sub>	26.240 <sub>0.053</sub>	0.834 <sub>0.016</sub>
430	582.7 <sub>1.3</sub>	16.13 <sub>0.20</sub>	0.6010 <sub>0.0043</sub>	27.253 <sub>0.058</sub>	0.856 <sub>0.016</sub>
420	596.1 <sub>1.2</sub>	12.96 <sub>0.11</sub>	0.4831 <sub>0.0051</sub>	28.236 <sub>0.062</sub>	0.876 <sub>0.015</sub>
410	608.84 <sub>0.86</sub>	10.324 <sub>0.065</sub>	0.3835 <sub>0.0054</sub>	29.161 <sub>0.058</sub>	0.895 <sub>0.014</sub>
400	620.4 <sub>1.4</sub>	8.136 <sub>0.057</sub>	0.3005 <sub>0.0053</sub>	30.003 <sub>0.073</sub>	0.912 <sub>0.015</sub>
390	631.7 <sub>1.6</sub>	6.340 <sub>0.060</sub>	0.2321 <sub>0.0051</sub>	30.807 <sub>0.083</sub>	0.928 <sub>0.018</sub>
380	642.43 <sub>0.53</sub>	4.878 <sub>0.064</sub>	0.1766 <sub>0.0049</sub>	31.568 <sub>0.066</sub>	0.941 <sub>0.026</sub>
370	652.13 <sub>0.63</sub>	3.702 <sub>0.067</sub>	0.1322 <sub>0.0047</sub>	32.24 <sub>0.12</sub>	0.953 <sub>0.040</sub>

Table S23: GCMC-MBAR results for 1-heptyne with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
520	437.2 <sub>2.6</sub>	77.0 <sub>2.2</sub>	2.122 <sub>0.017</sub>	16.99 <sub>0.12</sub>	0.613 <sub>0.013</sub>
510	462.1 <sub>1.5</sub>	62.4 <sub>1.4</sub>	1.822 <sub>0.011</sub>	19.188 <sub>0.099</sub>	0.662 <sub>0.013</sub>
500	483.1 <sub>1.1</sub>	51.09 <sub>0.74</sub>	1.5560 <sub>0.0076</sub>	21.050 <sub>0.060</sub>	0.70 <sub>0.01</sub>
490	501.4 <sub>1.0</sub>	42.07 <sub>0.40</sub>	1.3207 <sub>0.0063</sub>	22.662 <sub>0.042</sub>	0.74 <sub>0.01</sub>
480	518.0 <sub>1.1</sub>	34.69 <sub>0.32</sub>	1.1135 <sub>0.0053</sub>	24.100 <sub>0.057</sub>	0.77 <sub>0.01</sub>
470	533.2 <sub>1.2</sub>	28.57 <sub>0.30</sub>	0.9318 <sub>0.0044</sub>	25.399 <sub>0.069</sub>	0.80 <sub>0.01</sub>
460	547.3 <sub>1.1</sub>	23.46 <sub>0.24</sub>	0.7736 <sub>0.0042</sub>	26.588 <sub>0.061</sub>	0.83 <sub>0.01</sub>
450	560.81 <sub>1.0</sub>	19.18 <sub>0.16</sub>	0.6364 <sub>0.0046</sub>	27.703 <sub>0.047</sub>	0.853 <sub>0.011</sub>
440	573.98 <sub>0.87</sub>	15.59 <sub>0.12</sub>	0.5185 <sub>0.0052</sub>	28.768 <sub>0.035</sub>	0.874 <sub>0.015</sub>
430	586.56 <sub>0.51</sub>	12.58 <sub>0.10</sub>	0.4179 <sub>0.0057</sub>	29.770 <sub>0.052</sub>	0.894 <sub>0.019</sub>
420	598.39 <sub>0.36</sub>	10.06 <sub>0.10</sub>	0.3329 <sub>0.0061</sub>	30.703 <sub>0.082</sub>	0.911 <sub>0.024</sub>
410	609.60 <sub>0.41</sub>	7.97 <sub>0.12</sub>	0.2618 <sub>0.0064</sub>	31.576 <sub>0.098</sub>	0.927 <sub>0.030</sub>
400	620.43 <sub>0.28</sub>	6.24 <sub>0.16</sub>	0.2031 <sub>0.0065</sub>	32.41 <sub>0.13</sub>	0.941 <sub>0.042</sub>
390	631.53 <sub>0.29</sub>	4.82 <sub>0.19</sub>	0.1551 <sub>0.0067</sub>	33.24 <sub>0.20</sub>	0.953 <sub>0.062</sub>

Table S24: GCMC-MBAR results for 1-octyne with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
550	424.4 <sub>3.1</sub>	85.7 <sub>2.4</sub>	2.025 <sub>0.020</sub>	16.856 <sub>0.096</sub>	0.570 <sub>0.012</sub>
540	449.6 <sub>2.4</sub>	68.1 <sub>1.6</sub>	1.745 <sub>0.012</sub>	19.509 <sub>0.082</sub>	0.629 <sub>0.011</sub>
530	471.3 <sub>1.4</sub>	55.11 <sub>0.94</sub>	1.4972 <sub>0.0080</sub>	21.746 <sub>0.072</sub>	0.68 <sub>0.01</sub>
520	490.29 <sub>0.85</sub>	45.27 <sub>0.56</sub>	1.2786 <sub>0.0057</sub>	23.612 <sub>0.065</sub>	0.72 <sub>0.01</sub>
510	507.10 <sub>0.64</sub>	37.43 <sub>0.38</sub>	1.0854 <sub>0.0043</sub>	25.219 <sub>0.062</sub>	0.75 <sub>0.01</sub>
500	522.35 <sub>0.60</sub>	30.99 <sub>0.30</sub>	0.9152 <sub>0.0033</sub>	26.653 <sub>0.065</sub>	0.78 <sub>0.01</sub>
490	536.64 <sub>0.61</sub>	25.61 <sub>0.25</sub>	0.7660 <sub>0.0026</sub>	27.971 <sub>0.072</sub>	0.81 <sub>0.01</sub>
480	550.42 <sub>0.57</sub>	21.08 <sub>0.21</sub>	0.6360 <sub>0.0023</sub>	29.216 <sub>0.075</sub>	0.83 <sub>0.01</sub>
470	563.64 <sub>0.48</sub>	17.26 <sub>0.16</sub>	0.5234 <sub>0.0024</sub>	30.392 <sub>0.075</sub>	0.85 <sub>0.01</sub>
460	576.05 <sub>0.40</sub>	14.05 <sub>0.13</sub>	0.4265 <sub>0.0026</sub>	31.485 <sub>0.074</sub>	0.875 <sub>0.012</sub>
450	587.80 <sub>0.34</sub>	11.35 <sub>0.11</sub>	0.3440 <sub>0.0029</sub>	32.510 <sub>0.077</sub>	0.893 <sub>0.014</sub>
440	599.21 <sub>0.42</sub>	9.09 <sub>0.11</sub>	0.2743 <sub>0.0031</sub>	33.488 <sub>0.090</sub>	0.909 <sub>0.018</sub>
430	610.23 <sub>0.53</sub>	7.210 <sub>0.099</sub>	0.2160 <sub>0.0034</sub>	34.42 <sub>0.11</sub>	0.923 <sub>0.023</sub>
420	620.82 <sub>0.50</sub>	5.656 <sub>0.089</sub>	0.1678 <sub>0.0037</sub>	35.31 <sub>0.12</sub>	0.936 <sub>0.031</sub>
410	631.54 <sub>0.36</sub>	4.383 <sub>0.076</sub>	0.1285 <sub>0.0039</sub>	36.19 <sub>0.15</sub>	0.948 <sub>0.041</sub>

Table S25: GCMC-MBAR results for 1-nonyne with the iMiPPE force field (optimal  $\psi$  value from Table S6). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
570	427.1 <sub>1.4</sub>	80.51 <sub>0.73</sub>	1.776 <sub>0.010</sub>	18.58 <sub>0.12</sub>	0.58 <sub>0.01</sub>
560	450.90 <sub>0.83</sub>	64.43 <sub>0.54</sub>	1.5311 <sub>0.0083</sub>	21.279 <sub>0.086</sub>	0.63 <sub>0.01</sub>
550	471.72 <sub>0.65</sub>	52.31 <sub>0.42</sub>	1.3149 <sub>0.0067</sub>	23.583 <sub>0.093</sub>	0.68 <sub>0.01</sub>
540	489.89 <sub>0.61</sub>	43.02 <sub>0.39</sub>	1.1239 <sub>0.0056</sub>	25.52 <sub>0.11</sub>	0.72 <sub>0.01</sub>
530	506.26 <sub>0.52</sub>	35.61 <sub>0.32</sub>	0.9552 <sub>0.0047</sub>	27.21 <sub>0.11</sub>	0.76 <sub>0.01</sub>
520	521.47 <sub>0.54</sub>	29.51 <sub>0.25</sub>	0.8067 <sub>0.0040</sub>	28.739 <sub>0.079</sub>	0.79 <sub>0.01</sub>
510	535.75 <sub>0.66</sub>	24.42 <sub>0.23</sub>	0.6762 <sub>0.0031</sub>	30.149 <sub>0.058</sub>	0.81 <sub>0.01</sub>
500	549.10 <sub>0.71</sub>	20.14 <sub>0.24</sub>	0.5626 <sub>0.0022</sub>	31.452 <sub>0.059</sub>	0.83 <sub>0.01</sub>
490	561.65 <sub>0.67</sub>	16.54 <sub>0.23</sub>	0.4641 <sub>0.0013</sub>	32.666 <sub>0.073</sub>	0.856 <sub>0.010</sub>
480	573.68 <sub>0.55</sub>	13.50 <sub>0.21</sub>	0.37923 <sub>0.00095</sub>	33.811 <sub>0.085</sub>	0.875 <sub>0.014</sub>
470	585.26 <sub>0.38</sub>	10.94 <sub>0.18</sub>	0.3068 <sub>0.0015</sub>	34.896 <sub>0.092</sub>	0.892 <sub>0.018</sub>
460	596.35 <sub>0.30</sub>	8.79 <sub>0.16</sub>	0.2455 <sub>0.0022</sub>	35.92 <sub>0.10</sub>	0.907 <sub>0.023</sub>
450	607.27 <sub>0.32</sub>	7.00 <sub>0.14</sub>	0.1942 <sub>0.0030</sub>	36.92 <sub>0.13</sub>	0.921 <sub>0.031</sub>
440	618.21 <sub>0.22</sub>	5.52 <sub>0.12</sub>	0.1516 <sub>0.0036</sub>	37.91 <sub>0.16</sub>	0.933 <sub>0.041</sub>
430	628.74 <sub>0.19</sub>	4.29 <sub>0.11</sub>	0.1166 <sub>0.0042</sub>	38.85 <sub>0.22</sub>	0.944 <sub>0.055</sub>
420	638.50 <sub>0.35</sub>	3.296 <sub>0.094</sub>	0.0885 <sub>0.0047</sub>	39.72 <sub>0.29</sub>	0.955 <sub>0.074</sub>

## S6 Case study: Cyclohexane optimization

### S6.1 Phase equilibria plots

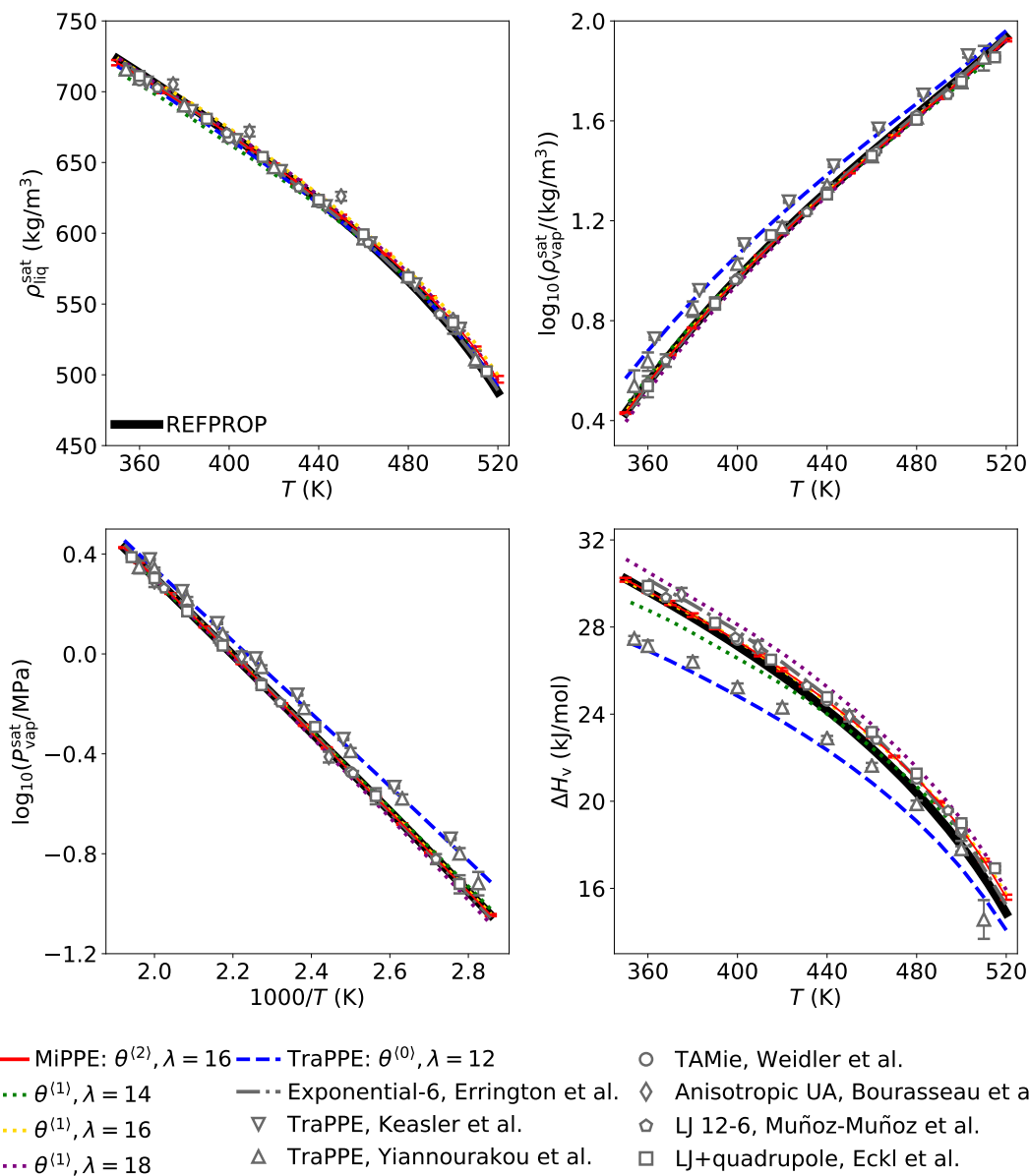


Figure S2: Phase equilibria for MiPPE ( $\theta^{(2)}, \lambda_{\text{CH}_2} = 16$ ), zeroth iteration (TraPPE:  $\theta^{(0)}$ ), first iterations ( $\theta^{(1)}, \lambda_{\text{CH}_2} = 14, 16, 18, 20$ ), and several literature force fields. See caption of Figure 9 in manuscript for details.

## S6.2 Twenty replicates

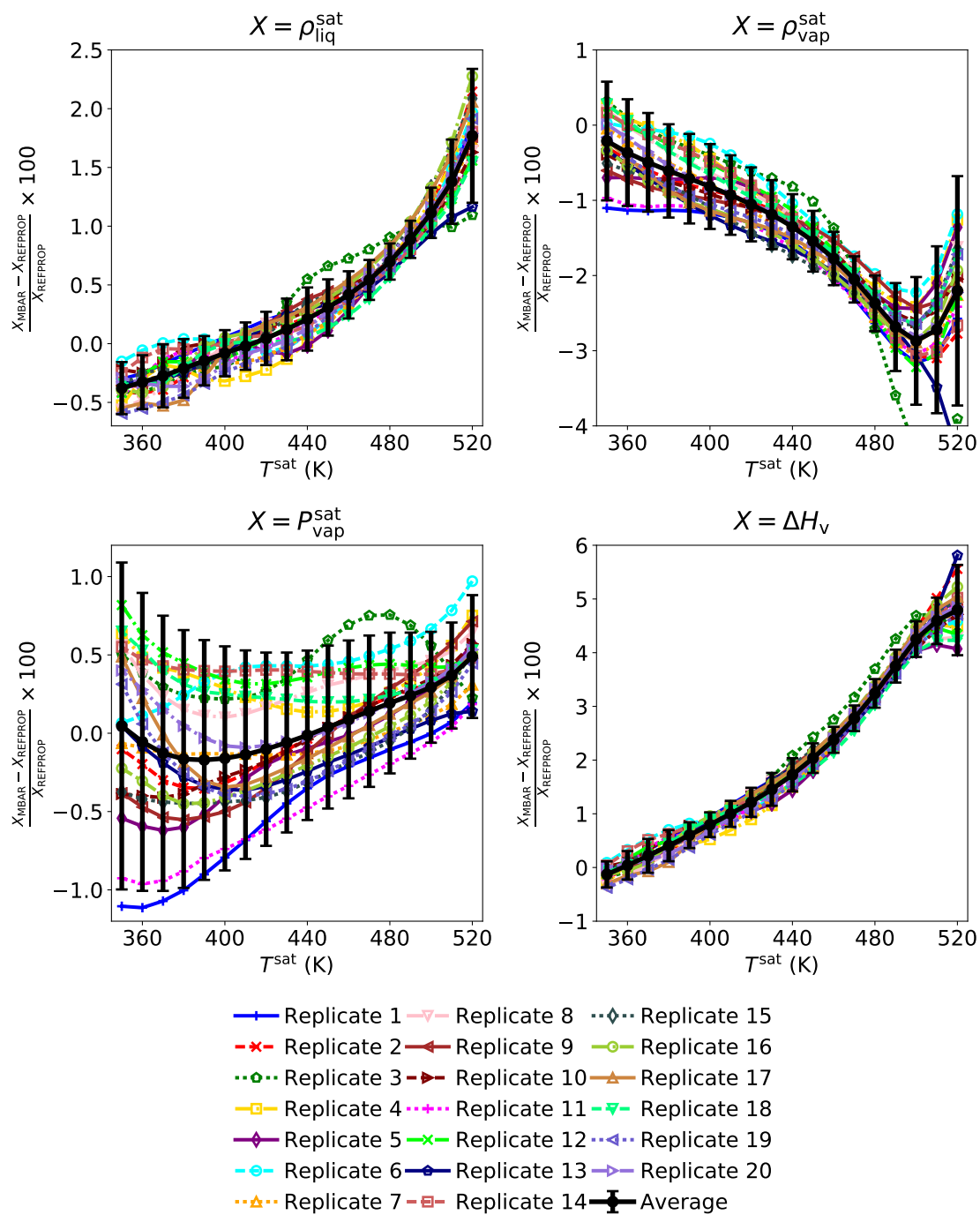


Figure S3: Percent deviations of MiPPE force field relative to REFPROP cyclohexane values for twenty replicates and average.

### S6.3 TAMie comparison

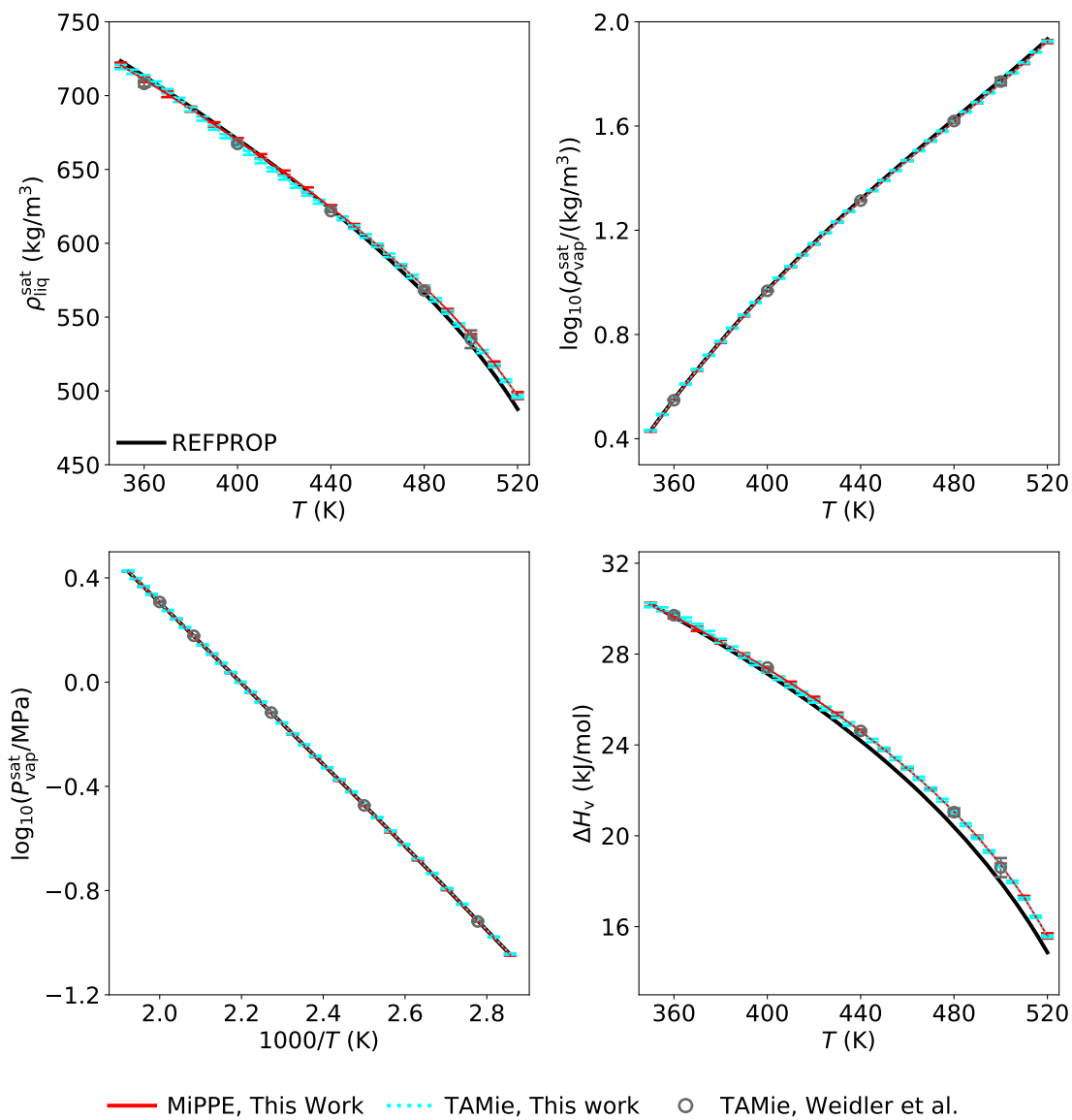


Figure S4: Comparison of MiPPE and TAMie (from this work and Weidler et al.) cyclohexane phase equilibria.

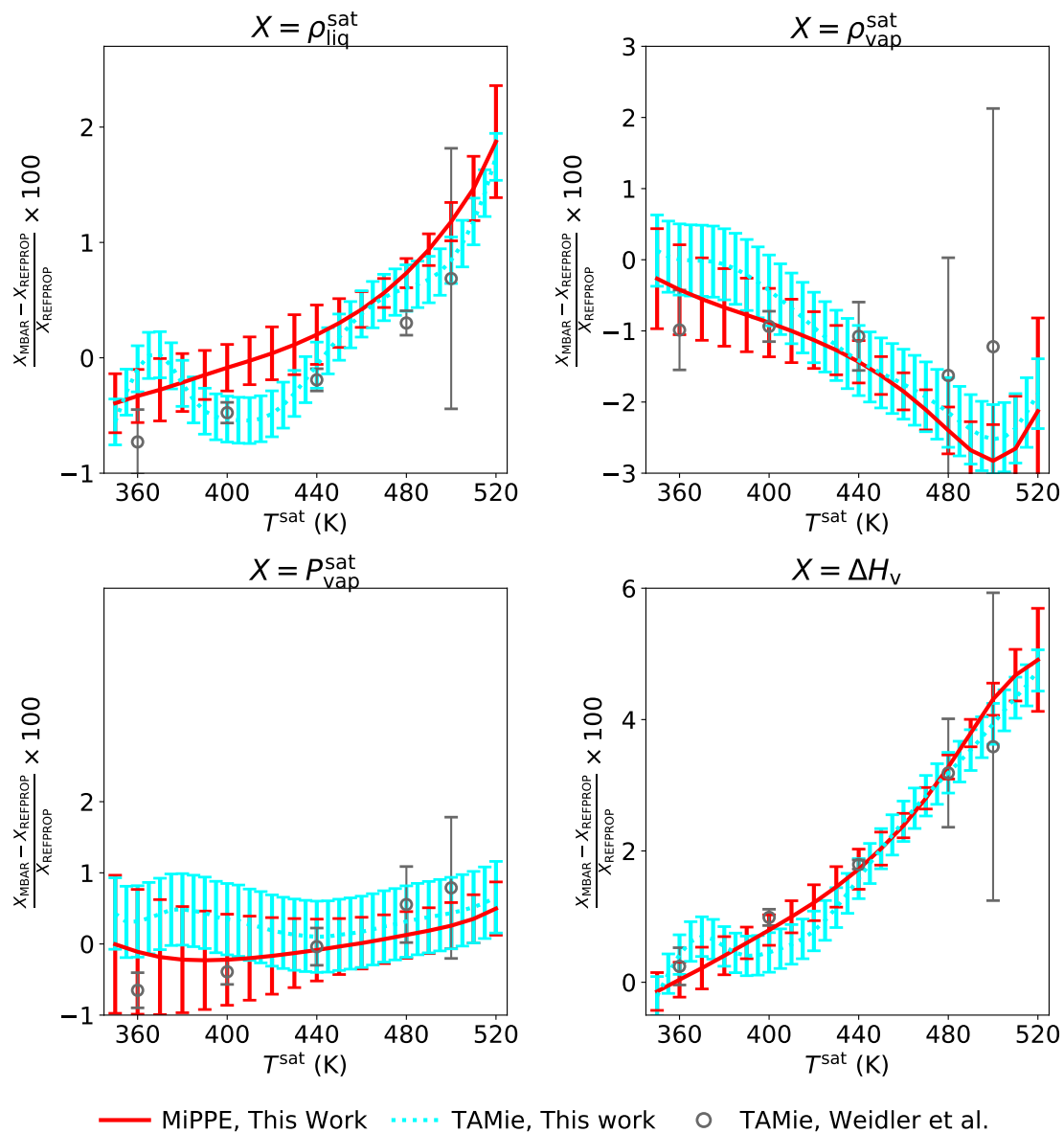


Figure S5: Comparison of MiPPE and TAMie (from this work and Weidler et al.) deviations relative to REFPROP cyclohexane values.



## S6.4 Minimum number of effective snapshots

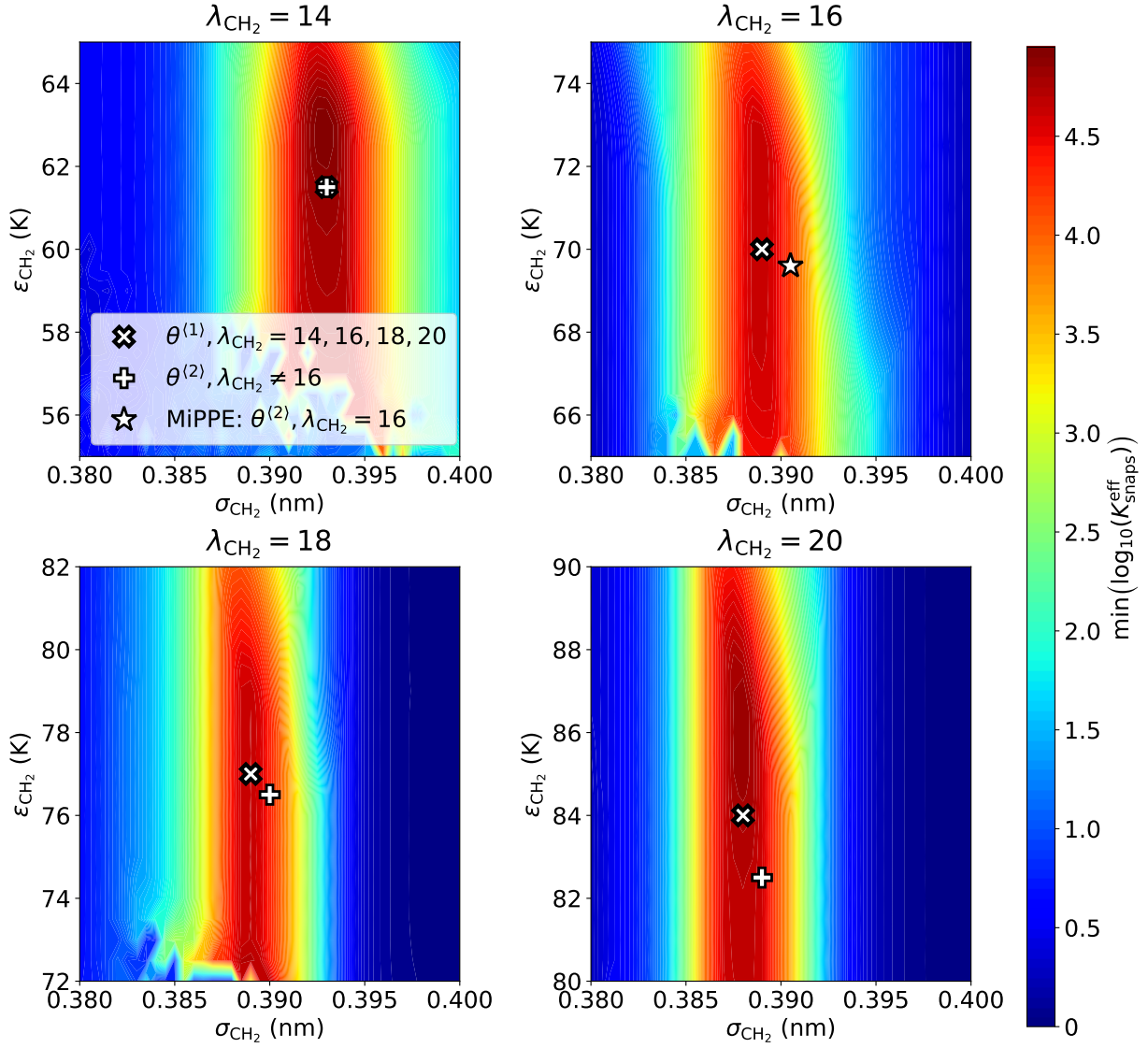


Figure S6: Second iteration minimum number of effective snapshots ( $\min(K_{\text{snaps}}^{\text{eff}})$ ) with respect to  $\epsilon_{\text{CH}_2}$  and  $\sigma_{\text{CH}_2}$  for cyclohexane. Optimization has converged as  $\min(K_{\text{snaps}}^{\text{eff}}) \gg 50$  for the optimal  $\epsilon_{\text{CH}_2}$ ,  $\sigma_{\text{CH}_2}$ ,  $\lambda_{\text{CH}_2}$  parameter set. Top-left, top-right, bottom-left, and bottom-right panels correspond  $\lambda_{\text{CH}_2} = 14$ ,  $\lambda_{\text{CH}_2} = 16$ ,  $\lambda_{\text{CH}_2} = 18$ , and  $\lambda_{\text{CH}_2} = 12$ , respectively. White star represents the optimal parameter set, i.e., the lowest value of  $S$ , for a given  $\lambda_{\text{CH}_2}$ .

## S6.5 Finite-size effects

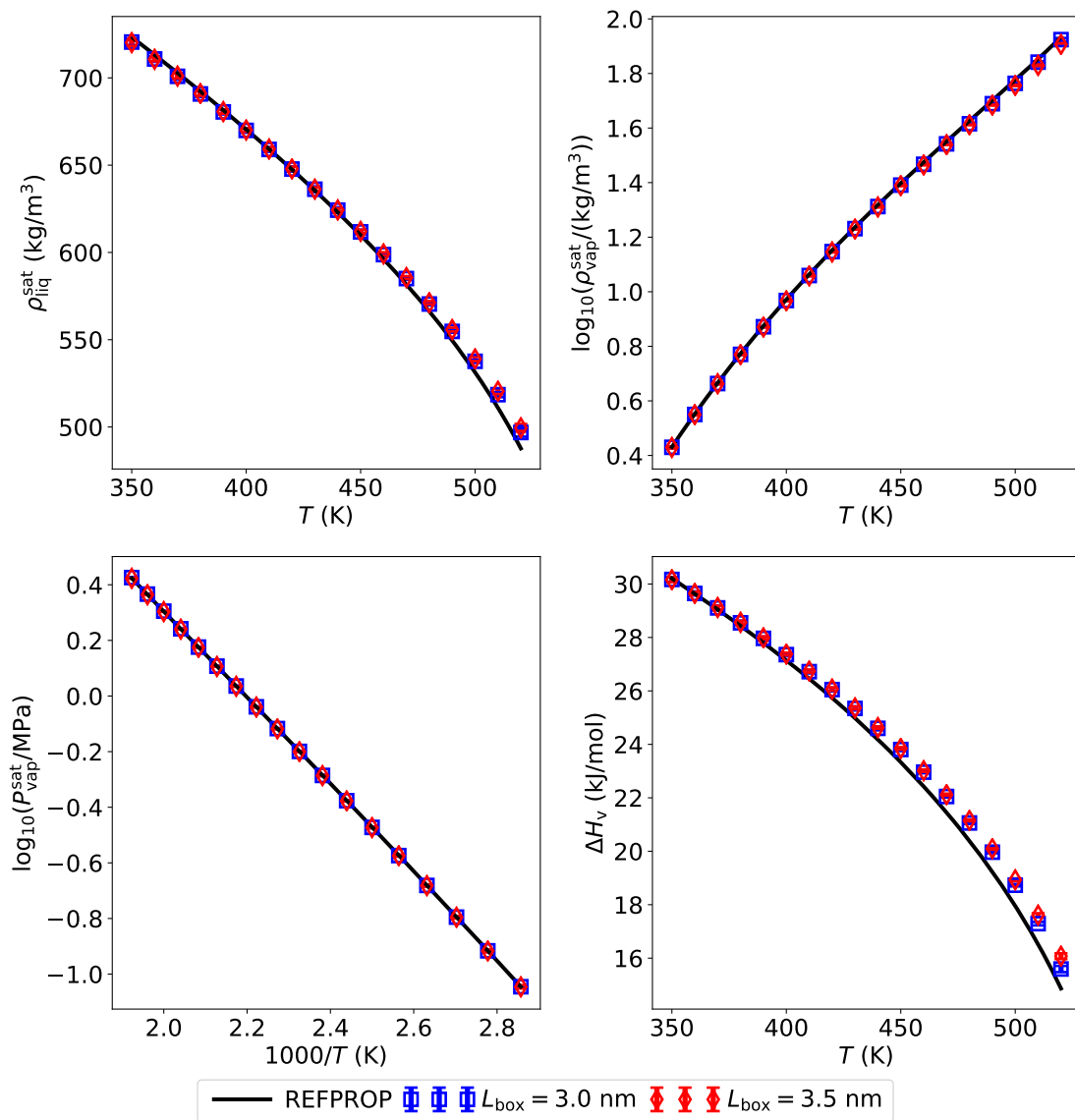


Figure S7: Comparison of phase equilibria using simulations with 3.0 nm and 3.5 nm box lengths for cyclohexane with MiPPE force field. Estimated values and uncertainties are obtained from 20 independent replicate simulations. See Figure S8 for quantitative assessment of finite-size effects.

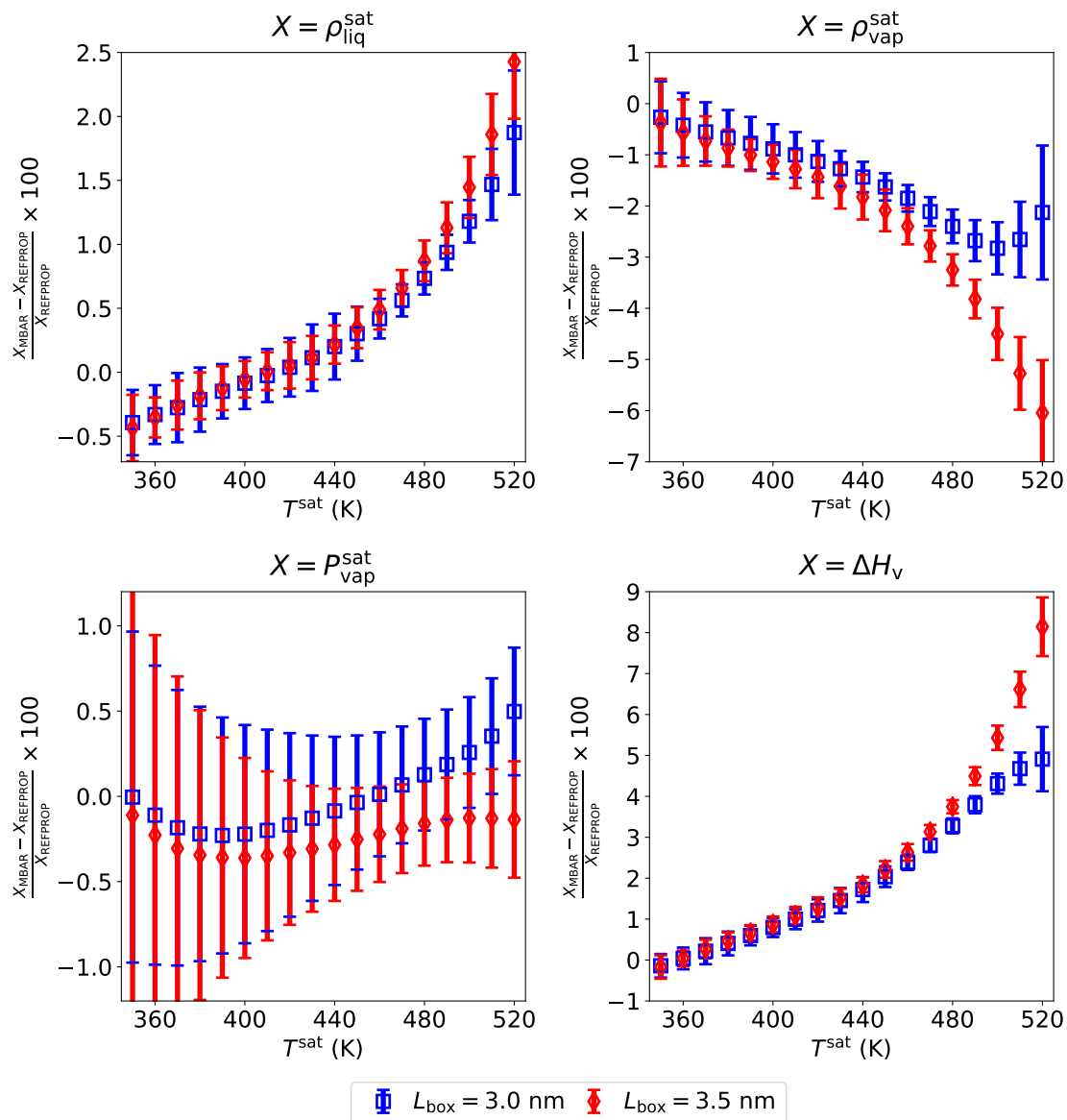


Figure S8: Comparison of percent deviations using simulations with 3.0 nm and 3.5 nm box lengths for cyclohexane with MiPPE force field. Agreement is typically within the combined uncertainties for low to moderate temperatures. Finite-size effects are most prevalent in  $\rho_{\text{vap}}^{\text{sat}}$  and  $\Delta H_v$  near the critical temperature ( $T^{\text{sat}} > 480$  K). Estimated values and uncertainties are obtained from 20 independent replicate simulations.

Table S26: GCMC-MBAR results for the MiPPE force field with 3.5 nm box length to test finite-size effects. Subscripts correspond to the 95% confidence interval computed with twenty independent replicate GCMC simulations at each state point.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
360	710.7 <sub>1.2</sub>	3.545 <sub>0.025</sub>	0.1211 <sub>0.0015</sub>	29.648 <sub>0.061</sub>	0.961 <sub>0.014</sub>
370	701.0 <sub>1.4</sub>	4.600 <sub>0.024</sub>	0.1601 <sub>0.0017</sub>	29.122 <sub>0.071</sub>	0.952 <sub>0.011</sub>
380	691.0 <sub>1.4</sub>	5.882 <sub>0.023</sub>	0.2081 <sub>0.0019</sub>	28.567 <sub>0.062</sub>	0.9424 <sub>0.0093</sub>
390	680.7 <sub>1.2</sub>	7.424 <sub>0.025</sub>	0.2665 <sub>0.0020</sub>	27.984 <sub>0.057</sub>	0.9315 <sub>0.0077</sub>
400	670.1 <sub>1.0</sub>	9.261 <sub>0.033</sub>	0.3365 <sub>0.0021</sub>	27.379 <sub>0.056</sub>	0.9195 <sub>0.0067</sub>
410	659.2 <sub>1.0</sub>	11.434 <sub>0.046</sub>	0.4196 <sub>0.0022</sub>	26.743 <sub>0.060</sub>	0.9061 <sub>0.0061</sub>
420	647.9 <sub>1.3</sub>	13.987 <sub>0.063</sub>	0.5173 <sub>0.0023</sub>	26.070 <sub>0.066</sub>	0.8913 <sub>0.0057</sub>
430	636.2 <sub>1.2</sub>	16.970 <sub>0.081</sub>	0.6309 <sub>0.0025</sub>	25.364 <sub>0.057</sub>	0.8752 <sub>0.0054</sub>
440	624.3 <sub>1.0</sub>	20.444 <sub>0.098</sub>	0.7621 <sub>0.0027</sub>	24.627 <sub>0.047</sub>	0.8576 <sub>0.0051</sub>
450	612.1 <sub>1.0</sub>	24.48 <sub>0.11</sub>	0.9125 <sub>0.0029</sub>	23.849 <sub>0.050</sub>	0.8385 <sub>0.0046</sub>
460	599.17 <sub>0.99</sub>	29.16 <sub>0.11</sub>	1.0837 <sub>0.0032</sub>	23.015 <sub>0.049</sub>	0.8178 <sub>0.0040</sub>
470	585.58 <sub>0.89</sub>	34.59 <sub>0.12</sub>	1.2774 <sub>0.0036</sub>	22.120 <sub>0.039</sub>	0.7953 <sub>0.0035</sub>
480	571.22 <sub>0.96</sub>	40.91 <sub>0.14</sub>	1.4953 <sub>0.0040</sub>	21.154 <sub>0.035</sub>	0.7707 <sub>0.0033</sub>
490	555.8 <sub>1.2</sub>	48.31 <sub>0.20</sub>	1.7394 <sub>0.0046</sub>	20.099 <sub>0.045</sub>	0.7438 <sub>0.0037</sub>
500	539.0 <sub>1.4</sub>	57.04 <sub>0.33</sub>	2.0115 <sub>0.0056</sub>	18.930 <sub>0.057</sub>	0.7139 <sub>0.0045</sub>
510	520.5 <sub>1.7</sub>	67.54 <sub>0.54</sub>	2.3139 <sub>0.0072</sub>	17.610 <sub>0.076</sub>	0.6799 <sub>0.0058</sub>
520	499.5 <sub>2.3</sub>	80.59 <sub>0.95</sub>	2.6492 <sub>0.0097</sub>	16.07 <sub>0.11</sub>	0.6399 <sub>0.0079</sub>

## S6.6 Tabulated phase equilibria for iterations

Table S27: GCMC-MBAR results for the MiPPE force field (second iteration,  $\theta^{(2)} \lambda_{\text{CH}_2} = 16$ ). Subscripts correspond to the 95% confidence interval computed with twenty independent replicate GCMC simulations at each state point.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
360	710.9 <sub>1.7</sub>	3.552 <sub>0.027</sub>	0.1213 <sub>0.0012</sub>	29.648 <sub>0.084</sub>	0.960 <sub>0.012</sub>
370	700.9 <sub>2.0</sub>	4.611 <sub>0.032</sub>	0.1604 <sub>0.0015</sub>	29.106 <sub>0.098</sub>	0.952 <sub>0.011</sub>
380	690.8 <sub>1.8</sub>	5.898 <sub>0.039</sub>	0.2085 <sub>0.0018</sub>	28.548 <sub>0.088</sub>	0.942 <sub>0.010</sub>
390	680.5 <sub>1.5</sub>	7.446 <sub>0.048</sub>	0.2670 <sub>0.0022</sub>	27.967 <sub>0.071</sub>	0.9306 <sub>0.0097</sub>
400	669.9 <sub>1.4</sub>	9.291 <sub>0.056</sub>	0.3372 <sub>0.0026</sub>	27.359 <sub>0.067</sub>	0.9183 <sub>0.0090</sub>
410	659.0 <sub>1.4</sub>	11.474 <sub>0.065</sub>	0.4205 <sub>0.0030</sub>	26.720 <sub>0.069</sub>	0.9048 <sub>0.0083</sub>
420	647.8 <sub>1.6</sub>	14.040 <sub>0.075</sub>	0.5185 <sub>0.0034</sub>	26.050 <sub>0.074</sub>	0.8899 <sub>0.0075</sub>
430	636.3 <sub>1.8</sub>	17.043 <sub>0.085</sub>	0.6325 <sub>0.0039</sub>	25.346 <sub>0.082</sub>	0.8736 <sub>0.0069</sub>
440	624.3 <sub>1.8</sub>	20.543 <sub>0.096</sub>	0.7642 <sub>0.0044</sub>	24.600 <sub>0.080</sub>	0.8558 <sub>0.0064</sub>
450	611.8 <sub>1.6</sub>	24.61 <sub>0.11</sub>	0.9152 <sub>0.0051</sub>	23.807 <sub>0.069</sub>	0.8363 <sub>0.0059</sub>
460	598.7 <sub>1.3</sub>	29.35 <sub>0.11</sub>	1.0871 <sub>0.0058</sub>	22.958 <sub>0.058</sub>	0.8151 <sub>0.0054</sub>
470	584.9 <sub>1.1</sub>	34.85 <sub>0.12</sub>	1.2816 <sub>0.0066</sub>	22.044 <sub>0.055</sub>	0.7920 <sub>0.0049</sub>
480	570.25 <sub>0.93</sub>	41.29 <sub>0.17</sub>	1.5006 <sub>0.0072</sub>	21.051 <sub>0.058</sub>	0.7665 <sub>0.0048</sub>
490	554.49 <sub>0.93</sub>	48.88 <sub>0.31</sub>	1.7460 <sub>0.0075</sub>	19.956 <sub>0.063</sub>	0.7379 <sub>0.0057</sub>
500	537.3 <sub>1.2</sub>	58.02 <sub>0.54</sub>	2.0200 <sub>0.0076</sub>	18.718 <sub>0.064</sub>	0.7049 <sub>0.0071</sub>
510	518.0 <sub>2.0</sub>	69.36 <sub>0.85</sub>	2.3254 <sub>0.0084</sub>	17.276 <sub>0.076</sub>	0.6654 <sub>0.0085</sub>
520	496.3 <sub>3.0</sub>	83.9 <sub>1.4</sub>	2.666 <sub>0.011</sub>	15.58 <sub>0.13</sub>	0.619 <sub>0.011</sub>

Table S28: GCMC-MBAR results for the TraPPE force field (zeroth iteration,  $\theta^{(0)}$ ). Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
360	709.02 <sub>0.33</sub>	4.775 <sub>0.062</sub>	0.161 <sub>0.016</sub>	26.916 <sub>0.050</sub>	0.946 <sub>0.096</sub>
370	698.93 <sub>0.39</sub>	6.067 <sub>0.072</sub>	0.208 <sub>0.012</sub>	26.440 <sub>0.042</sub>	0.937 <sub>0.055</sub>
380	688.03 <sub>0.44</sub>	7.610 <sub>0.088</sub>	0.26 <sub>0.01</sub>	25.919 <sub>0.036</sub>	0.927 <sub>0.026</sub>
390	677.14 <sub>0.49</sub>	9.43 <sub>0.11</sub>	0.33 <sub>0.01</sub>	25.382 <sub>0.033</sub>	0.916 <sub>0.015</sub>
400	666.43 <sub>0.55</sub>	11.57 <sub>0.12</sub>	0.41 <sub>0.01</sub>	24.835 <sub>0.030</sub>	0.904 <sub>0.023</sub>
410	655.56 <sub>0.39</sub>	14.07 <sub>0.13</sub>	0.507 <sub>0.017</sub>	24.266 <sub>0.023</sub>	0.890 <sub>0.032</sub>
420	644.33 <sub>0.48</sub>	16.97 <sub>0.13</sub>	0.616 <sub>0.025</sub>	23.665 <sub>0.027</sub>	0.875 <sub>0.036</sub>
430	632.76 <sub>0.54</sub>	20.32 <sub>0.12</sub>	0.741 <sub>0.031</sub>	23.030 <sub>0.030</sub>	0.858 <sub>0.037</sub>
440	620.79 <sub>0.67</sub>	24.18 <sub>0.26</sub>	0.883 <sub>0.039</sub>	22.356 <sub>0.051</sub>	0.840 <sub>0.038</sub>
450	608.2 <sub>1.2</sub>	28.64 <sub>0.96</sub>	1.045 <sub>0.052</sub>	21.63 <sub>0.10</sub>	0.821 <sub>0.049</sub>
460	595.0 <sub>2.9</sub>	33.8 <sub>2.4</sub>	1.227 <sub>0.098</sub>	20.85 <sub>0.20</sub>	0.799 <sub>0.085</sub>
470	581.0 <sub>4.6</sub>	39.7 <sub>3.8</sub>	1.43 <sub>0.17</sub>	20.01 <sub>0.30</sub>	0.78 <sub>0.12</sub>
480	566.2 <sub>4.8</sub>	46.7 <sub>4.6</sub>	1.66 <sub>0.24</sub>	19.08 <sub>0.33</sub>	0.75 <sub>0.13</sub>
490	550.4 <sub>4.5</sub>	54.8 <sub>4.7</sub>	1.91 <sub>0.29</sub>	18.06 <sub>0.38</sub>	0.72 <sub>0.13</sub>
500	533.2 <sub>5.4</sub>	64.6 <sub>3.8</sub>	2.19 <sub>0.38</sub>	16.913 <sub>0.088</sub>	0.69 <sub>0.13</sub>
510	513.8 <sub>5.3</sub>	76.6 <sub>5.4</sub>	2.50 <sub>0.37</sub>	15.59 <sub>0.33</sub>	0.65 <sub>0.11</sub>
520	491.6 <sub>3.8</sub>	91.4 <sub>7.6</sub>	2.85 <sub>0.28</sub>	14.08 <sub>0.34</sub>	0.607 <sub>0.078</sub>

Table S29: GCMC-MBAR results for the first iteration ( $\theta^{(1)}$ )  $\lambda_{\text{CH}_2} = 14$  force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
360	704.93 <sub>0.26</sub>	3.743 <sub>0.073</sub>	0.127 <sub>0.044</sub>	28.78 <sub>0.10</sub>	0.96 <sub>0.33</sub>
370	695.36 <sub>0.63</sub>	4.822 <sub>0.057</sub>	0.167 <sub>0.048</sub>	28.28 <sub>0.11</sub>	0.95 <sub>0.27</sub>
380	684.68 <sub>0.79</sub>	6.125 <sub>0.057</sub>	0.216 <sub>0.049</sub>	27.73 <sub>0.11</sub>	0.94 <sub>0.21</sub>
390	673.71 <sub>0.68</sub>	7.680 <sub>0.084</sub>	0.274 <sub>0.048</sub>	27.144 <sub>0.099</sub>	0.93 <sub>0.16</sub>
400	663.17 <sub>0.45</sub>	9.52 <sub>0.13</sub>	0.345 <sub>0.044</sub>	26.568 <sub>0.086</sub>	0.92 <sub>0.12</sub>
410	652.73 <sub>0.24</sub>	11.68 <sub>0.17</sub>	0.427 <sub>0.039</sub>	25.980 <sub>0.062</sub>	0.903 <sub>0.084</sub>
420	641.98 <sub>0.82</sub>	14.21 <sub>0.23</sub>	0.524 <sub>0.033</sub>	25.360 <sub>0.039</sub>	0.889 <sub>0.058</sub>
430	630.8 <sub>1.2</sub>	17.15 <sub>0.30</sub>	0.636 <sub>0.032</sub>	24.703 <sub>0.036</sub>	0.873 <sub>0.046</sub>
440	619.3 <sub>1.1</sub>	20.57 <sub>0.39</sub>	0.765 <sub>0.041</sub>	24.005 <sub>0.044</sub>	0.856 <sub>0.049</sub>
450	607.24 <sub>0.80</sub>	24.53 <sub>0.54</sub>	0.912 <sub>0.062</sub>	23.261 <sub>0.063</sub>	0.837 <sub>0.060</sub>
460	594.62 <sub>0.73</sub>	29.11 <sub>0.77</sub>	1.080 <sub>0.093</sub>	22.463 <sub>0.085</sub>	0.816 <sub>0.074</sub>
470	581.33 <sub>0.87</sub>	34.4 <sub>1.1</sub>	1.27 <sub>0.14</sub>	21.60 <sub>0.11</sub>	0.793 <sub>0.091</sub>
480	567.3 <sub>1.1</sub>	40.6 <sub>1.6</sub>	1.48 <sub>0.21</sub>	20.68 <sub>0.13</sub>	0.77 <sub>0.11</sub>
490	552.4 <sub>1.5</sub>	47.9 <sub>2.1</sub>	1.72 <sub>0.30</sub>	19.66 <sub>0.14</sub>	0.74 <sub>0.13</sub>
500	536.3 <sub>2.5</sub>	56.5 <sub>2.6</sub>	1.98 <sub>0.41</sub>	18.52 <sub>0.14</sub>	0.71 <sub>0.15</sub>
510	518.2 <sub>4.3</sub>	67.2 <sub>2.8</sub>	2.27 <sub>0.54</sub>	17.20 <sub>0.16</sub>	0.67 <sub>0.16</sub>

Table S30: GCMC-MBAR results for the first iteration ( $\theta^{(1)}$ )  $\lambda_{\text{CH}_2} = 16$  force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
360	713.64 <sub>0.38</sub>	3.583 <sub>0.049</sub>	0.122 <sub>0.039</sub>	29.59 <sub>0.13</sub>	0.96 <sub>0.31</sub>
370	704.1 <sub>1.2</sub>	4.647 <sub>0.057</sub>	0.162 <sub>0.036</sub>	29.065 <sub>0.088</sub>	0.95 <sub>0.21</sub>
380	695.0 <sub>2.4</sub>	5.943 <sub>0.071</sub>	0.210 <sub>0.032</sub>	28.551 <sub>0.098</sub>	0.94 <sub>0.14</sub>
390	685.2 <sub>2.3</sub>	7.505 <sub>0.090</sub>	0.269 <sub>0.027</sub>	27.993 <sub>0.084</sub>	0.931 <sub>0.094</sub>
400	674.3 <sub>1.1</sub>	9.37 <sub>0.11</sub>	0.340 <sub>0.024</sub>	27.376 <sub>0.037</sub>	0.918 <sub>0.066</sub>
410	662.91 <sub>0.76</sub>	11.57 <sub>0.12</sub>	0.424 <sub>0.026</sub>	26.717 <sub>0.040</sub>	0.905 <sub>0.056</sub>
420	651.2 <sub>1.0</sub>	14.16 <sub>0.14</sub>	0.523 <sub>0.032</sub>	26.027 <sub>0.065</sub>	0.890 <sub>0.055</sub>
430	639.4 <sub>1.1</sub>	17.19 <sub>0.18</sub>	0.638 <sub>0.040</sub>	25.310 <sub>0.078</sub>	0.873 <sub>0.056</sub>
440	627.31 <sub>0.89</sub>	20.71 <sub>0.22</sub>	0.770 <sub>0.050</sub>	24.565 <sub>0.072</sub>	0.856 <sub>0.056</sub>
450	614.82 <sub>0.58</sub>	24.79 <sub>0.24</sub>	0.922 <sub>0.059</sub>	23.777 <sub>0.051</sub>	0.837 <sub>0.055</sub>
460	601.71 <sub>0.56</sub>	29.53 <sub>0.25</sub>	1.095 <sub>0.067</sub>	22.935 <sub>0.053</sub>	0.816 <sub>0.050</sub>
470	587.98 <sub>0.65</sub>	35.03 <sub>0.48</sub>	1.291 <sub>0.067</sub>	22.033 <sub>0.094</sub>	0.794 <sub>0.042</sub>
480	573.59 <sub>0.74</sub>	41.45 <sub>0.99</sub>	1.511 <sub>0.063</sub>	21.06 <sub>0.15</sub>	0.768 <sub>0.037</sub>
490	558.19 <sub>0.80</sub>	49.0 <sub>1.7</sub>	1.757 <sub>0.086</sub>	19.99 <sub>0.20</sub>	0.740 <sub>0.045</sub>
500	541.27 <sub>0.93</sub>	58.2 <sub>2.6</sub>	2.03 <sub>0.16</sub>	18.78 <sub>0.24</sub>	0.707 <sub>0.064</sub>
510	522.1 <sub>1.7</sub>	69.5 <sub>3.4</sub>	2.34 <sub>0.28</sub>	17.36 <sub>0.25</sub>	0.668 <sub>0.087</sub>
520	499.8 <sub>2.6</sub>	83.7 <sub>3.9</sub>	2.68 <sub>0.43</sub>	15.71 <sub>0.22</sub>	0.62 <sub>0.10</sub>



Table S31: GCMC-MBAR results for the first iteration ( $\theta^{(1)}$ )  $\lambda_{\text{CH}_2} = 18$  force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
360	713.60 <sub>0.89</sub>	3.294 <sub>0.099</sub>	0.113 <sub>0.020</sub>	30.527 <sub>0.066</sub>	0.96 <sub>0.17</sub>
370	702.86 <sub>0.81</sub>	4.306 <sub>0.092</sub>	0.150 <sub>0.015</sub>	29.921 <sub>0.044</sub>	0.95 <sub>0.10</sub>
380	692.51 <sub>0.62</sub>	5.545 <sub>0.081</sub>	0.196 <sub>0.013</sub>	29.327 <sub>0.039</sub>	0.943 <sub>0.063</sub>
390	682.23 <sub>0.53</sub>	7.043 <sub>0.088</sub>	0.253 <sub>0.011</sub>	28.725 <sub>0.043</sub>	0.932 <sub>0.043</sub>
400	671.99 <sub>0.38</sub>	8.84 <sub>0.13</sub>	0.32 <sub>0.01</sub>	28.110 <sub>0.043</sub>	0.920 <sub>0.030</sub>
410	661.74 <sub>0.33</sub>	10.98 <sub>0.17</sub>	0.403 <sub>0.013</sub>	27.478 <sub>0.038</sub>	0.906 <sub>0.033</sub>
420	650.92 <sub>0.46</sub>	13.51 <sub>0.21</sub>	0.499 <sub>0.024</sub>	26.801 <sub>0.037</sub>	0.891 <sub>0.046</sub>
430	639.09 <sub>0.44</sub>	16.48 <sub>0.24</sub>	0.613 <sub>0.039</sub>	26.056 <sub>0.039</sub>	0.875 <sub>0.057</sub>
440	626.49 <sub>0.39</sub>	19.96 <sub>0.27</sub>	0.744 <sub>0.055</sub>	25.253 <sub>0.042</sub>	0.857 <sub>0.065</sub>
450	613.53 <sub>0.38</sub>	24.02 <sub>0.35</sub>	0.894 <sub>0.073</sub>	24.409 <sub>0.048</sub>	0.837 <sub>0.069</sub>
460	600.27 <sub>0.57</sub>	28.75 <sub>0.70</sub>	1.066 <sub>0.096</sub>	23.525 <sub>0.086</sub>	0.816 <sub>0.076</sub>
470	586.50 <sub>0.90</sub>	34.3 <sub>1.6</sub>	1.26 <sub>0.14</sub>	22.58 <sub>0.16</sub>	0.793 <sub>0.095</sub>
480	571.9 <sub>1.5</sub>	40.7 <sub>2.6</sub>	1.48 <sub>0.23</sub>	21.56 <sub>0.21</sub>	0.77 <sub>0.13</sub>
490	556.3 <sub>2.5</sub>	48.4 <sub>3.2</sub>	1.73 <sub>0.35</sub>	20.44 <sub>0.25</sub>	0.74 <sub>0.16</sub>
500	538.9 <sub>3.0</sub>	57.7 <sub>3.1</sub>	2.01 <sub>0.46</sub>	19.16 <sub>0.25</sub>	0.71 <sub>0.16</sub>
510	519.1 <sub>2.0</sub>	69.1 <sub>2.7</sub>	2.32 <sub>0.54</sub>	17.67 <sub>0.18</sub>	0.67 <sub>0.16</sub>
520	495.9 <sub>1.6</sub>	83.5 <sub>2.4</sub>	2.67 <sub>0.62</sub>	15.93 <sub>0.12</sub>	0.62 <sub>0.15</sub>

Table S32: GCMC-MBAR results for the first iteration ( $\theta^{(1)}$ )  $\lambda_{\text{CH}_2} = 20$  force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
360	721.21 <sub>0.17</sub>	2.885 <sub>0.047</sub>	0.099 <sub>0.059</sub>	31.72 <sub>0.18</sub>	0.96 <sub>0.57</sub>
370	711.38 <sub>0.48</sub>	3.812 <sub>0.043</sub>	0.133 <sub>0.056</sub>	31.13 <sub>0.11</sub>	0.96 <sub>0.40</sub>
380	701.58 <sub>0.81</sub>	4.955 <sub>0.041</sub>	0.176 <sub>0.054</sub>	30.545 <sub>0.066</sub>	0.95 <sub>0.29</sub>
390	691.47 <sub>0.70</sub>	6.351 <sub>0.048</sub>	0.229 <sub>0.054</sub>	29.931 <sub>0.055</sub>	0.94 <sub>0.22</sub>
400	680.14 <sub>0.36</sub>	8.033 <sub>0.065</sub>	0.293 <sub>0.055</sub>	29.243 <sub>0.046</sub>	0.92 <sub>0.17</sub>
410	668.26 <sub>0.63</sub>	10.039 <sub>0.086</sub>	0.370 <sub>0.057</sub>	28.510 <sub>0.057</sub>	0.91 <sub>0.14</sub>
420	656.5 <sub>1.6</sub>	12.41 <sub>0.11</sub>	0.462 <sub>0.061</sub>	27.77 <sub>0.10</sub>	0.90 <sub>0.12</sub>
430	644.5 <sub>2.3</sub>	15.21 <sub>0.13</sub>	0.570 <sub>0.065</sub>	27.00 <sub>0.14</sub>	0.88 <sub>0.10</sub>
440	631.8 <sub>1.8</sub>	18.50 <sub>0.16</sub>	0.695 <sub>0.068</sub>	26.17 <sub>0.12</sub>	0.864 <sub>0.085</sub>
450	618.8 <sub>1.1</sub>	22.34 <sub>0.20</sub>	0.840 <sub>0.072</sub>	25.308 <sub>0.082</sub>	0.845 <sub>0.072</sub>
460	605.44 <sub>0.73</sub>	26.84 <sub>0.33</sub>	1.005 <sub>0.072</sub>	24.399 <sub>0.086</sub>	0.824 <sub>0.060</sub>
470	591.81 <sub>0.70</sub>	32.11 <sub>0.65</sub>	1.194 <sub>0.069</sub>	23.44 <sub>0.13</sub>	0.801 <sub>0.049</sub>
480	577.60 <sub>0.68</sub>	38.3 <sub>1.2</sub>	1.408 <sub>0.074</sub>	22.41 <sub>0.19</sub>	0.775 <sub>0.048</sub>
490	562.27 <sub>0.91</sub>	45.7 <sub>2.1</sub>	1.65 <sub>0.12</sub>	21.28 <sub>0.23</sub>	0.746 <sub>0.064</sub>
500	545.2 <sub>2.6</sub>	54.6 <sub>2.9</sub>	1.92 <sub>0.23</sub>	20.00 <sub>0.19</sub>	0.713 <sub>0.092</sub>
510	526.1 <sub>5.6</sub>	65.5 <sub>3.4</sub>	2.23 <sub>0.38</sub>	18.526 <sub>0.093</sub>	0.68 <sub>0.12</sub>
520	504.1 <sub>9.3</sub>	79.2 <sub>3.3</sub>	2.57 <sub>0.57</sub>	16.81 <sub>0.22</sub>	0.63 <sub>0.14</sub>

## S7 Compressibility factor

### S7.1 Validation of GCMC-MBAR

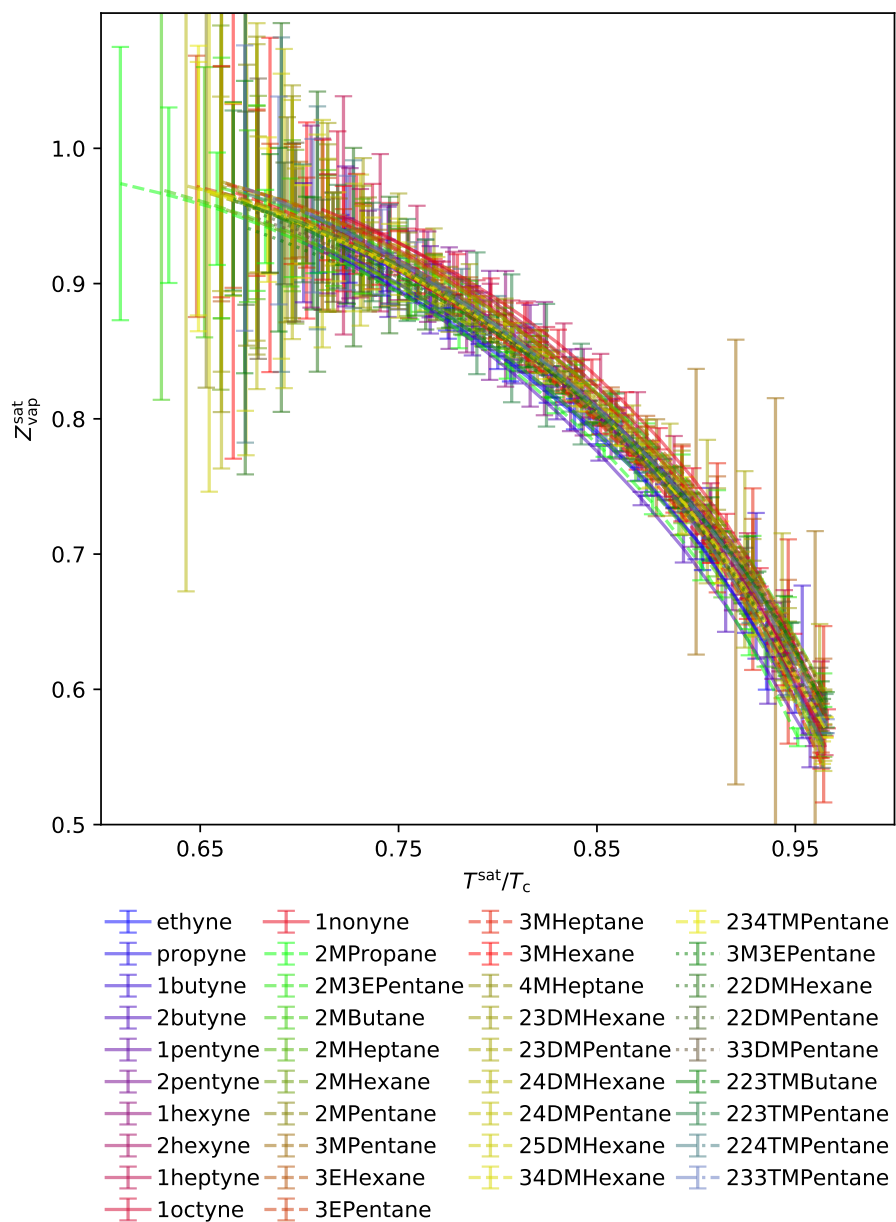


Figure S9: Compressibility factor in saturated vapor phase ( $Z_{\text{vap}}^{\text{sat}}$ ) for all compounds simulated in Mick et al. and Soroush Barhaghi et al. Note that symmetric (normal) 95% confidence intervals are ill-suited when  $Z_{\text{vap}}^{\text{sat}} \approx 1$ , as this assumption can result in  $Z_{\text{vap}}^{\text{sat}} > 1$ .

## S7.2 Case study: Cyclohexane optimization

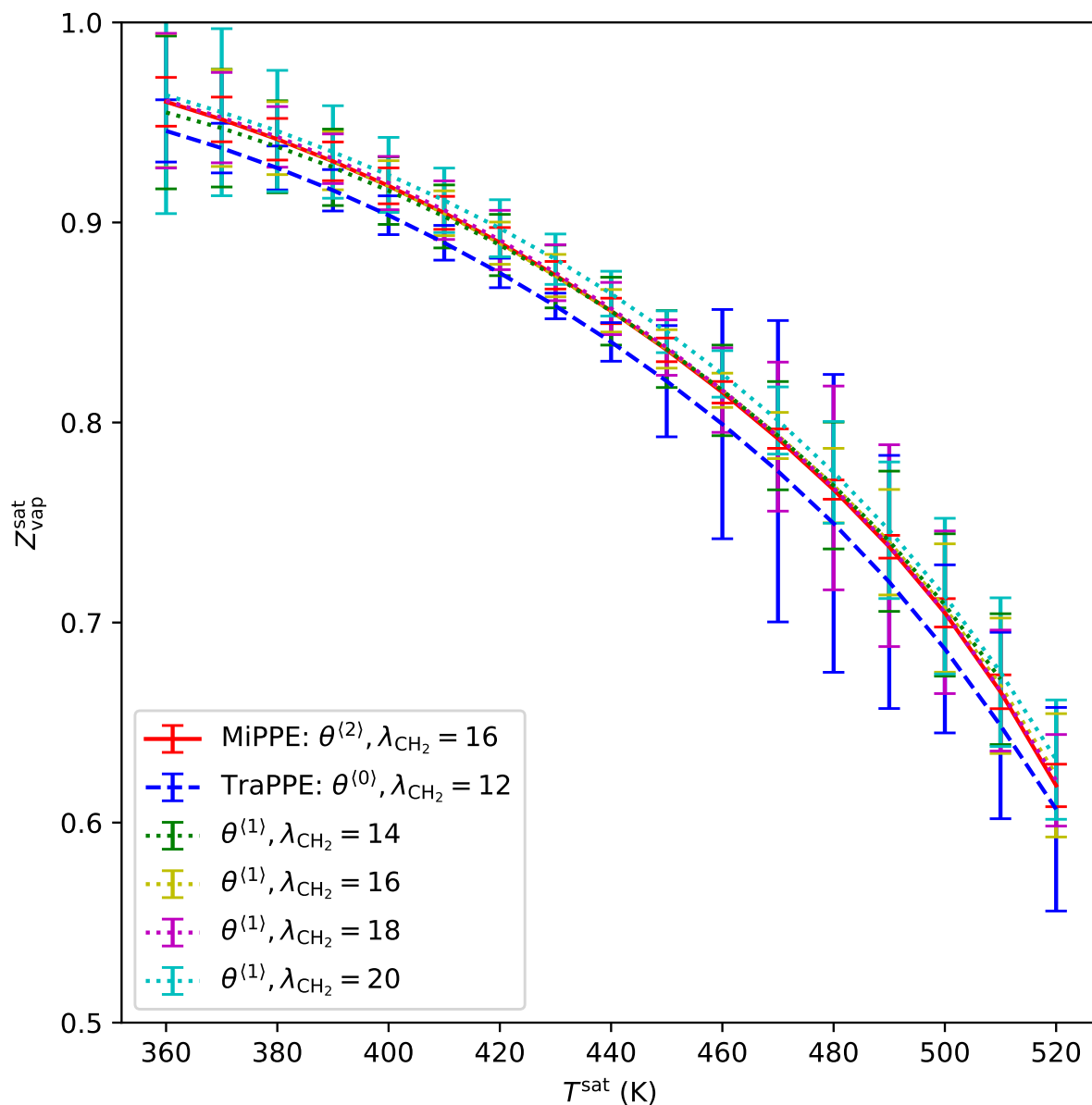


Figure S10: Compressibility factor in saturated vapor phase ( $Z_{\text{vap}}^{\text{sat}}$ ) for iterations of cyclohexane optimization. Note that symmetric (normal) 95% confidence intervals are ill-suited when  $Z_{\text{vap}}^{\text{sat}} \approx 1$ , as this assumption can result in  $Z_{\text{vap}}^{\text{sat}} > 1$ .

## S8 Tabulated phase equilibria for validation of GCMC-MBAR

### S8.1 Branched alkanes

Table S33: GCMC-MBAR results for 2-methylpropane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
390	388.8 <sub>8.4</sub>	87.1 <sub>1.6</sub>	2.742 <sub>0.026</sub>	9.76 <sub>0.34</sub>	0.56 <sub>0.01</sub>
380	416.6 <sub>4.6</sub>	66.4 <sub>1.2</sub>	2.292 <sub>0.026</sub>	11.69 <sub>0.17</sub>	0.63 <sub>0.01</sub>
370	439.7 <sub>1.3</sub>	51.95 <sub>0.77</sub>	1.903 <sub>0.025</sub>	13.219 <sub>0.077</sub>	0.692 <sub>0.011</sub>
360	459.18 <sub>0.47</sub>	41.22 <sub>0.53</sub>	1.566 <sub>0.023</sub>	14.442 <sub>0.060</sub>	0.74 <sub>0.01</sub>
350	476.39 <sub>0.48</sub>	32.82 <sub>0.53</sub>	1.276 <sub>0.019</sub>	15.485 <sub>0.049</sub>	0.78 <sub>0.01</sub>
340	492.09 <sub>0.59</sub>	26.09 <sub>0.54</sub>	1.028 <sub>0.015</sub>	16.404 <sub>0.052</sub>	0.81 <sub>0.01</sub>
330	506.65 <sub>0.67</sub>	20.62 <sub>0.48</sub>	0.817 <sub>0.011</sub>	17.227 <sub>0.055</sub>	0.84 <sub>0.01</sub>
320	520.48 <sub>0.53</sub>	16.16 <sub>0.39</sub>	0.6403 <sub>0.0069</sub>	17.980 <sub>0.058</sub>	0.865 <sub>0.013</sub>
310	533.93 <sub>0.54</sub>	12.53 <sub>0.27</sub>	0.4935 <sub>0.0043</sub>	18.686 <sub>0.062</sub>	0.889 <sub>0.015</sub>
300	546.58 <sub>0.63</sub>	9.57 <sub>0.17</sub>	0.3733 <sub>0.0032</sub>	19.332 <sub>0.062</sub>	0.909 <sub>0.016</sub>
290	558.46 <sub>0.48</sub>	7.19 <sub>0.12</sub>	0.2765 <sub>0.0030</sub>	19.925 <sub>0.061</sub>	0.927 <sub>0.019</sub>
280	570.30 <sub>0.67</sub>	5.300 <sub>0.094</sub>	0.2000 <sub>0.0032</sub>	20.498 <sub>0.077</sub>	0.942 <sub>0.027</sub>
270	582.5 <sub>1.0</sub>	3.815 <sub>0.083</sub>	0.1408 <sub>0.0037</sub>	21.07 <sub>0.11</sub>	0.955 <sub>0.042</sub>
260	593.47 <sub>0.60</sub>	2.672 <sub>0.071</sub>	0.0959 <sub>0.0043</sub>	21.57 <sub>0.14</sub>	0.965 <sub>0.065</sub>
250	603.0 <sub>1.0</sub>	1.815 <sub>0.057</sub>	0.0632 <sub>0.0048</sub>	22.01 <sub>0.22</sub>	0.97 <sub>0.10</sub>

Table S34: GCMC-MBAR results for 2-methylbutane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_{\text{v}}$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
440	399.8 <sub>3.5</sub>	86.6 <sub>2.1</sub>	2.528 <sub>0.020</sub>	11.558 <sub>0.090</sub>	0.58 <sub>0.01</sub>
430	426.9 <sub>2.6</sub>	68.9 <sub>1.7</sub>	2.149 <sub>0.011</sub>	13.44 <sub>0.11</sub>	0.629 <sub>0.012</sub>
420	449.4 <sub>1.4</sub>	55.1 <sub>1.2</sub>	1.8149 <sub>0.0059</sub>	15.07 <sub>0.11</sub>	0.680 <sub>0.012</sub>
410	468.58 <sub>0.70</sub>	44.49 <sub>0.66</sub>	1.5225 <sub>0.0039</sub>	16.438 <sub>0.094</sub>	0.724 <sub>0.010</sub>
400	485.59 <sub>0.52</sub>	36.12 <sub>0.31</sub>	1.2671 <sub>0.0037</sub>	17.615 <sub>0.066</sub>	0.76 <sub>0.01</sub>
390	501.08 <sub>0.68</sub>	29.33 <sub>0.15</sub>	1.0450 <sub>0.0036</sub>	18.654 <sub>0.048</sub>	0.79 <sub>0.01</sub>
380	515.41 <sub>0.89</sub>	23.73 <sub>0.12</sub>	0.8532 <sub>0.0034</sub>	19.590 <sub>0.055</sub>	0.82 <sub>0.01</sub>
370	528.88 <sub>0.98</sub>	19.09 <sub>0.11</sub>	0.6887 <sub>0.0031</sub>	20.447 <sub>0.064</sub>	0.85 <sub>0.01</sub>
360	541.7 <sub>1.0</sub>	15.230 <sub>0.098</sub>	0.5489 <sub>0.0029</sub>	21.238 <sub>0.067</sub>	0.87 <sub>0.01</sub>
350	553.9 <sub>1.1</sub>	12.03 <sub>0.10</sub>	0.4315 <sub>0.0028</sub>	21.976 <sub>0.071</sub>	0.889 <sub>0.010</sub>
340	565.8 <sub>1.3</sub>	9.40 <sub>0.11</sub>	0.3341 <sub>0.0029</sub>	22.678 <sub>0.085</sub>	0.907 <sub>0.016</sub>
330	577.7 <sub>1.7</sub>	7.24 <sub>0.12</sub>	0.2541 <sub>0.0033</sub>	23.35 <sub>0.12</sub>	0.923 <sub>0.025</sub>
320	588.9 <sub>1.7</sub>	5.48 <sub>0.14</sub>	0.1896 <sub>0.0039</sub>	23.98 <sub>0.16</sub>	0.938 <sub>0.040</sub>
310	598.9 <sub>1.2</sub>	4.08 <sub>0.14</sub>	0.1385 <sub>0.0047</sub>	24.53 <sub>0.20</sub>	0.950 <sub>0.064</sub>
300	608.6 <sub>1.2</sub>	2.98 <sub>0.14</sub>	0.0988 <sub>0.0055</sub>	25.06 <sub>0.28</sub>	0.960 <sub>0.10</sub>
290	618.9 <sub>1.3</sub>	2.12 <sub>0.13</sub>	0.0686 <sub>0.0064</sub>	25.61 <sub>0.41</sub>	0.97 <sub>0.16</sub>

Table S35: GCMC-MBAR results for 2-methylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
470	422.20 <sub>0.56</sub>	76.7 <sub>1.5</sub>	2.068 <sub>0.017</sub>	14.14 <sub>0.11</sub>	0.59 <sub>0.01</sub>
460	444.71 <sub>0.79</sub>	61.2 <sub>1.2</sub>	1.761 <sub>0.013</sub>	16.06 <sub>0.12</sub>	0.65 <sub>0.01</sub>
450	464.38 <sub>0.87</sub>	49.31 <sub>0.88</sub>	1.4918 <sub>0.0096</sub>	17.71 <sub>0.11</sub>	0.70 <sub>0.01</sub>
440	481.69 <sub>0.77</sub>	40.17 <sub>0.64</sub>	1.2557 <sub>0.0068</sub>	19.106 <sub>0.090</sub>	0.74 <sub>0.01</sub>
430	497.34 <sub>0.70</sub>	32.86 <sub>0.47</sub>	1.0491 <sub>0.0045</sub>	20.318 <sub>0.074</sub>	0.77 <sub>0.01</sub>
420	511.89 <sub>0.78</sub>	26.86 <sub>0.36</sub>	0.8690 <sub>0.0027</sub>	21.408 <sub>0.068</sub>	0.80 <sub>0.01</sub>
410	525.69 <sub>0.84</sub>	21.87 <sub>0.29</sub>	0.7130 <sub>0.0017</sub>	22.412 <sub>0.069</sub>	0.824 <sub>0.011</sub>
400	538.82 <sub>0.88</sub>	17.70 <sub>0.25</sub>	0.5788 <sub>0.0020</sub>	23.344 <sub>0.069</sub>	0.848 <sub>0.014</sub>
390	551.23 <sub>0.87</sub>	14.21 <sub>0.22</sub>	0.4645 <sub>0.0031</sub>	24.207 <sub>0.075</sub>	0.869 <sub>0.018</sub>
380	562.89 <sub>0.80</sub>	11.30 <sub>0.19</sub>	0.3681 <sub>0.0042</sub>	25.004 <sub>0.096</sub>	0.888 <sub>0.024</sub>
370	573.96 <sub>0.88</sub>	8.90 <sub>0.16</sub>	0.2878 <sub>0.0051</sub>	25.75 <sub>0.14</sub>	0.906 <sub>0.032</sub>
360	584.8 <sub>1.1</sub>	6.92 <sub>0.14</sub>	0.2215 <sub>0.0059</sub>	26.46 <sub>0.18</sub>	0.922 <sub>0.042</sub>
350	595.6 <sub>1.2</sub>	5.31 <sub>0.12</sub>	0.1677 <sub>0.0065</sub>	27.15 <sub>0.22</sub>	0.935 <sub>0.056</sub>
340	606.3 <sub>1.2</sub>	4.01 <sub>0.10</sub>	0.1245 <sub>0.0070</sub>	27.83 <sub>0.27</sub>	0.947 <sub>0.076</sub>
330	616.4 <sub>1.0</sub>	2.969 <sub>0.085</sub>	0.0906 <sub>0.0074</sub>	28.45 <sub>0.32</sub>	0.96 <sub>0.10</sub>
320	625.82 <sub>0.57</sub>	2.156 <sub>0.068</sub>	0.0644 <sub>0.0078</sub>	29.04 <sub>0.40</sub>	0.97 <sub>0.14</sub>

Table S36: GCMC-MBAR results for 2-methylhexane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
510	406.8 <sub>3.4</sub>	88.4 <sub>2.0</sub>	2.092 <sub>0.011</sub>	14.14 <sub>0.12</sub>	0.56 <sub>0.01</sub>
500	431.2 <sub>2.2</sub>	70.0 <sub>1.6</sub>	1.7989 <sub>0.0052</sub>	16.506 <sub>0.085</sub>	0.620 <sub>0.013</sub>
490	452.1 <sub>1.2</sub>	56.5 <sub>1.0</sub>	1.5403 <sub>0.0041</sub>	18.472 <sub>0.088</sub>	0.671 <sub>0.013</sub>
480	470.38 <sub>0.73</sub>	46.30 <sub>0.50</sub>	1.3120 <sub>0.0056</sub>	20.092 <sub>0.065</sub>	0.712 <sub>0.010</sub>
470	486.88 <sub>0.50</sub>	38.21 <sub>0.17</sub>	1.1104 <sub>0.0062</sub>	21.497 <sub>0.034</sub>	0.75 <sub>0.01</sub>
460	502.20 <sub>0.37</sub>	31.56 <sub>0.16</sub>	0.9327 <sub>0.0058</sub>	22.762 <sub>0.022</sub>	0.77 <sub>0.01</sub>
450	516.52 <sub>0.39</sub>	26.01 <sub>0.17</sub>	0.7772 <sub>0.0050</sub>	23.921 <sub>0.024</sub>	0.80 <sub>0.01</sub>
440	530.01 <sub>0.34</sub>	21.34 <sub>0.15</sub>	0.6417 <sub>0.0043</sub>	24.994 <sub>0.024</sub>	0.82 <sub>0.01</sub>
430	542.86 <sub>0.37</sub>	17.41 <sub>0.14</sub>	0.5245 <sub>0.0039</sub>	25.998 <sub>0.031</sub>	0.84 <sub>0.01</sub>
420	554.93 <sub>0.46</sub>	14.10 <sub>0.13</sub>	0.4241 <sub>0.0037</sub>	26.928 <sub>0.041</sub>	0.86 <sub>0.01</sub>
410	566.22 <sub>0.43</sub>	11.32 <sub>0.14</sub>	0.3390 <sub>0.0036</sub>	27.789 <sub>0.046</sub>	0.881 <sub>0.014</sub>
400	577.21 <sub>0.43</sub>	8.99 <sub>0.14</sub>	0.2674 <sub>0.0037</sub>	28.610 <sub>0.060</sub>	0.896 <sub>0.020</sub>
390	588.02 <sub>0.38</sub>	7.06 <sub>0.15</sub>	0.2080 <sub>0.0039</sub>	29.403 <sub>0.089</sub>	0.910 <sub>0.030</sub>
380	598.35 <sub>0.22</sub>	5.47 <sub>0.16</sub>	0.1593 <sub>0.0044</sub>	30.15 <sub>0.14</sub>	0.923 <sub>0.044</sub>
370	608.04 <sub>0.19</sub>	4.18 <sub>0.16</sub>	0.1200 <sub>0.0050</sub>	30.85 <sub>0.20</sub>	0.935 <sub>0.064</sub>
360	617.49 <sub>0.27</sub>	3.14 <sub>0.16</sub>	0.0888 <sub>0.0057</sub>	31.52 <sub>0.28</sub>	0.945 <sub>0.093</sub>
350	627.22 <sub>0.26</sub>	2.32 <sub>0.15</sub>	0.0644 <sub>0.0065</sub>	32.20 <sub>0.39</sub>	0.96 <sub>0.13</sub>



Table S37: GCMC-MBAR results for 2-methylheptane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
540	401.3 <sub>3.7</sub>	88.7 <sub>2.6</sub>	1.947 <sub>0.032</sub>	15.17 <sub>0.13</sub>	0.56 <sub>0.01</sub>
530	426.3 <sub>2.7</sub>	71.5 <sub>2.6</sub>	1.683 <sub>0.023</sub>	17.60 <sub>0.12</sub>	0.610 <sub>0.015</sub>
520	448.1 <sub>1.4</sub>	58.0 <sub>2.1</sub>	1.447 <sub>0.015</sub>	19.75 <sub>0.16</sub>	0.660 <sub>0.020</sub>
510	467.12 <sub>0.68</sub>	47.5 <sub>1.5</sub>	1.2389 <sub>0.0097</sub>	21.59 <sub>0.16</sub>	0.702 <sub>0.019</sub>
500	483.89 <sub>0.51</sub>	39.23 <sub>0.90</sub>	1.0545 <sub>0.0068</sub>	23.17 <sub>0.13</sub>	0.738 <sub>0.016</sub>
490	499.04 <sub>0.51</sub>	32.49 <sub>0.54</sub>	0.8917 <sub>0.0055</sub>	24.571 <sub>0.096</sub>	0.770 <sub>0.012</sub>
480	513.07 <sub>0.62</sub>	26.88 <sub>0.35</sub>	0.7488 <sub>0.0048</sub>	25.842 <sub>0.079</sub>	0.80 <sub>0.01</sub>
470	526.33 <sub>0.63</sub>	22.18 <sub>0.26</sub>	0.6238 <sub>0.0042</sub>	27.019 <sub>0.072</sub>	0.82 <sub>0.01</sub>
460	538.86 <sub>0.70</sub>	18.22 <sub>0.22</sub>	0.5151 <sub>0.0038</sub>	28.112 <sub>0.079</sub>	0.844 <sub>0.010</sub>
450	550.62 <sub>0.83</sub>	14.88 <sub>0.19</sub>	0.4215 <sub>0.0036</sub>	29.125 <sub>0.091</sub>	0.865 <sub>0.013</sub>
440	561.64 <sub>0.82</sub>	12.07 <sub>0.16</sub>	0.3414 <sub>0.0037</sub>	30.06 <sub>0.10</sub>	0.883 <sub>0.017</sub>
430	572.07 <sub>0.80</sub>	9.71 <sub>0.14</sub>	0.2735 <sub>0.0040</sub>	30.94 <sub>0.11</sub>	0.900 <sub>0.022</sub>
420	582.24 <sub>0.77</sub>	7.74 <sub>0.11</sub>	0.2165 <sub>0.0043</sub>	31.78 <sub>0.13</sub>	0.915 <sub>0.028</sub>
410	592.62 <sub>0.67</sub>	6.108 <sub>0.092</sub>	0.1691 <sub>0.0045</sub>	32.62 <sub>0.14</sub>	0.927 <sub>0.035</sub>
400	603.09 <sub>0.65</sub>	4.758 <sub>0.074</sub>	0.1302 <sub>0.0047</sub>	33.44 <sub>0.16</sub>	0.940 <sub>0.044</sub>
390	612.67 <sub>0.70</sub>	3.655 <sub>0.061</sub>	0.0986 <sub>0.0049</sub>	34.20 <sub>0.19</sub>	0.950 <sub>0.056</sub>
380	621.39 <sub>0.62</sub>	2.766 <sub>0.052</sub>	0.0734 <sub>0.0049</sub>	34.89 <sub>0.24</sub>	0.960 <sub>0.071</sub>
370	630.41 <sub>0.64</sub>	2.057 <sub>0.045</sub>	0.0536 <sub>0.0049</sub>	35.58 <sub>0.31</sub>	0.967 <sub>0.093</sub>

Table S38: GCMC-MBAR results for 3-methylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
480	415.8 <sub>1.5</sub>	84 <sub>24</sub>	2.31 <sub>0.24</sub>	13.7 <sub>1.3</sub>	0.60 <sub>0.12</sub>
470	440.0 <sub>2.3</sub>	67 <sub>24</sub>	1.99 <sub>0.14</sub>	15.6 <sub>1.7</sub>	0.65 <sub>0.17</sub>
460	459.8 <sub>2.5</sub>	55 <sub>18</sub>	1.698 <sub>0.059</sub>	17.2 <sub>1.6</sub>	0.69 <sub>0.16</sub>
450	477.1 <sub>1.9</sub>	45.4 <sub>7.6</sub>	1.443 <sub>0.012</sub>	18.52 <sub>0.94</sub>	0.73 <sub>0.11</sub>
440	493.2 <sub>1.2</sub>	37.5 <sub>1.5</sub>	1.217 <sub>0.014</sub>	19.74 <sub>0.27</sub>	0.764 <sub>0.036</sub>
430	508.9 <sub>1.4</sub>	30.95 <sub>0.38</sub>	1.019 <sub>0.015</sub>	20.87 <sub>0.11</sub>	0.794 <sub>0.013</sub>
420	523.6 <sub>1.1</sub>	25.42 <sub>0.36</sub>	0.845 <sub>0.014</sub>	21.917 <sub>0.094</sub>	0.821 <sub>0.016</sub>
410	536.8 <sub>1.4</sub>	20.78 <sub>0.32</sub>	0.695 <sub>0.014</sub>	22.85 <sub>0.12</sub>	0.845 <sub>0.020</sub>
400	548.9 <sub>1.7</sub>	16.89 <sub>0.24</sub>	0.565 <sub>0.013</sub>	23.69 <sub>0.15</sub>	0.867 <sub>0.021</sub>
390	560.5 <sub>1.5</sub>	13.63 <sub>0.21</sub>	0.455 <sub>0.012</sub>	24.49 <sub>0.13</sub>	0.887 <sub>0.018</sub>
380	571.8 <sub>1.2</sub>	10.88 <sub>0.20</sub>	0.361 <sub>0.010</sub>	25.250 <sub>0.099</sub>	0.905 <sub>0.016</sub>
370	582.3 <sub>1.1</sub>	8.59 <sub>0.18</sub>	0.2829 <sub>0.0089</sub>	25.950 <sub>0.093</sub>	0.922 <sub>0.018</sub>
360	593.0 <sub>1.1</sub>	6.70 <sub>0.16</sub>	0.2185 <sub>0.0077</sub>	26.64 <sub>0.12</sub>	0.939 <sub>0.024</sub>
350	603.8 <sub>1.0</sub>	5.15 <sub>0.14</sub>	0.1661 <sub>0.0067</sub>	27.33 <sub>0.14</sub>	0.955 <sub>0.035</sub>

Table S39: GCMC-MBAR results for 3-methylhexane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
520	398.4 <sub>4.6</sub>	98.1 <sub>1.2</sub>	2.307 <sub>0.018</sub>	13.25 <sub>0.29</sub>	0.55 <sub>0.01</sub>
510	426.2 <sub>3.4</sub>	77.3 <sub>1.3</sub>	1.996 <sub>0.015</sub>	15.86 <sub>0.21</sub>	0.61 <sub>0.01</sub>
500	449.0 <sub>1.8</sub>	62.6 <sub>1.1</sub>	1.720 <sub>0.012</sub>	17.92 <sub>0.14</sub>	0.66 <sub>0.01</sub>
490	467.85 <sub>0.87</sub>	51.64 <sub>0.76</sub>	1.474 <sub>0.011</sub>	19.56 <sub>0.10</sub>	0.70 <sub>0.01</sub>
480	484.12 <sub>0.71</sub>	42.82 <sub>0.51</sub>	1.2562 <sub>0.0093</sub>	20.963 <sub>0.074</sub>	0.74 <sub>0.01</sub>
470	498.93 <sub>0.76</sub>	35.54 <sub>0.44</sub>	1.0638 <sub>0.0075</sub>	22.215 <sub>0.069</sub>	0.77 <sub>0.01</sub>
460	512.9 <sub>1.0</sub>	29.46 <sub>0.42</sub>	0.8944 <sub>0.0056</sub>	23.366 <sub>0.088</sub>	0.80 <sub>0.01</sub>
450	526.2 <sub>1.3</sub>	24.34 <sub>0.36</sub>	0.7463 <sub>0.0040</sub>	24.442 <sub>0.093</sub>	0.82 <sub>0.01</sub>
440	539.09 <sub>0.79</sub>	20.03 <sub>0.26</sub>	0.6171 <sub>0.0029</sub>	25.454 <sub>0.048</sub>	0.84 <sub>0.01</sub>
430	551.25 <sub>0.90</sub>	16.38 <sub>0.15</sub>	0.5055 <sub>0.0025</sub>	26.397 <sub>0.089</sub>	0.86 <sub>0.01</sub>
420	563.0 <sub>1.6</sub>	13.302 <sub>0.087</sub>	0.4097 <sub>0.0025</sub>	27.29 <sub>0.16</sub>	0.88 <sub>0.01</sub>
410	574.5 <sub>1.4</sub>	10.70 <sub>0.12</sub>	0.3283 <sub>0.0024</sub>	28.15 <sub>0.18</sub>	0.90 <sub>0.01</sub>
400	585.3 <sub>1.3</sub>	8.53 <sub>0.18</sub>	0.2598 <sub>0.0021</sub>	28.96 <sub>0.19</sub>	0.918 <sub>0.019</sub>
390	595.7 <sub>2.4</sub>	6.72 <sub>0.26</sub>	0.2028 <sub>0.0021</sub>	29.72 <sub>0.30</sub>	0.933 <sub>0.039</sub>
380	605.9 <sub>2.8</sub>	5.23 <sub>0.33</sub>	0.1560 <sub>0.0032</sub>	30.45 <sub>0.41</sub>	0.947 <sub>0.073</sub>
370	615.4 <sub>1.3</sub>	4.01 <sub>0.38</sub>	0.1181 <sub>0.0053</sub>	31.13 <sub>0.47</sub>	0.96 <sub>0.12</sub>
360	624.92 <sub>0.55</sub>	3.04 <sub>0.40</sub>	0.0878 <sub>0.0081</sub>	31.78 <sub>0.63</sub>	0.97 <sub>0.20</sub>

Table S40: GCMC-MBAR results for 3-methylheptane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
540	417.2 <sub>1.2</sub>	83 <sub>12</sub>	1.890 <sub>0.063</sub>	16.23 <sub>0.82</sub>	0.582 <sub>0.065</sub>
530	440.3 <sub>1.2</sub>	66.7 <sub>9.0</sub>	1.634 <sub>0.033</sub>	18.58 <sub>0.89</sub>	0.635 <sub>0.076</sub>
520	460.5 <sub>1.2</sub>	54.5 <sub>5.6</sub>	1.407 <sub>0.012</sub>	20.58 <sub>0.75</sub>	0.681 <sub>0.067</sub>
510	478.1 <sub>1.2</sub>	45.1 <sub>2.8</sub>	1.2045 <sub>0.0059</sub>	22.26 <sub>0.50</sub>	0.719 <sub>0.048</sub>
500	494.01 <sub>0.92</sub>	37.5 <sub>1.1</sub>	1.0252 <sub>0.0098</sub>	23.73 <sub>0.26</sub>	0.752 <sub>0.028</sub>
490	508.73 <sub>0.65</sub>	31.13 <sub>0.27</sub>	0.867 <sub>0.011</sub>	25.068 <sub>0.10</sub>	0.781 <sub>0.013</sub>
480	522.52 <sub>0.51</sub>	25.80 <sub>0.29</sub>	0.728 <sub>0.010</sub>	26.295 <sub>0.066</sub>	0.81 <sub>0.01</sub>
470	535.46 <sub>0.54</sub>	21.31 <sub>0.32</sub>	0.6061 <sub>0.0088</sub>	27.431 <sub>0.087</sub>	0.83 <sub>0.01</sub>
460	547.59 <sub>0.69</sub>	17.52 <sub>0.28</sub>	0.5005 <sub>0.0075</sub>	28.484 <sub>0.098</sub>	0.85 <sub>0.01</sub>
450	559.13 <sub>0.85</sub>	14.31 <sub>0.24</sub>	0.4094 <sub>0.0063</sub>	29.47 <sub>0.11</sub>	0.87 <sub>0.01</sub>
440	570.31 <sub>0.88</sub>	11.61 <sub>0.22</sub>	0.3315 <sub>0.0051</sub>	30.41 <sub>0.11</sub>	0.89 <sub>0.01</sub>
430	581.03 <sub>0.75</sub>	9.34 <sub>0.20</sub>	0.2656 <sub>0.0041</sub>	31.30 <sub>0.11</sub>	0.91 <sub>0.01</sub>
420	591.11 <sub>0.65</sub>	7.44 <sub>0.18</sub>	0.2102 <sub>0.0031</sub>	32.12 <sub>0.12</sub>	0.924 <sub>0.015</sub>
410	600.65 <sub>0.78</sub>	5.87 <sub>0.16</sub>	0.1642 <sub>0.0025</sub>	32.89 <sub>0.15</sub>	0.937 <sub>0.022</sub>
400	610.1 <sub>1.1</sub>	4.58 <sub>0.14</sub>	0.1265 <sub>0.0021</sub>	33.65 <sub>0.20</sub>	0.949 <sub>0.032</sub>
390	620.1 <sub>1.3</sub>	3.52 <sub>0.12</sub>	0.0959 <sub>0.0022</sub>	34.43 <sub>0.24</sub>	0.959 <sub>0.044</sub>
380	630.35 <sub>0.84</sub>	2.67 <sub>0.10</sub>	0.0714 <sub>0.0025</sub>	35.23 <sub>0.26</sub>	0.967 <sub>0.062</sub>
370	640.62 <sub>0.56</sub>	1.986 <sub>0.080</sub>	0.0522 <sub>0.0029</sub>	36.01 <sub>0.30</sub>	0.975 <sub>0.085</sub>

Table S41: GCMC-MBAR results for 3-ethylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_{\text{v}}$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
520	405.9 <sub>2.5</sub>	92.5 <sub>3.7</sub>	2.279 <sub>0.020</sub>	13.93 <sub>0.27</sub>	0.571 <sub>0.018</sub>
510	433.2 <sub>1.4</sub>	75.3 <sub>2.9</sub>	1.975 <sub>0.011</sub>	16.14 <sub>0.27</sub>	0.620 <sub>0.021</sub>
500	456.01 <sub>0.97</sub>	61.6 <sub>1.8</sub>	1.7031 <sub>0.0070</sub>	18.08 <sub>0.24</sub>	0.667 <sub>0.019</sub>
490	474.95 <sub>0.93</sub>	50.83 <sub>0.86</sub>	1.4607 <sub>0.0069</sub>	19.70 <sub>0.17</sub>	0.707 <sub>0.013</sub>
480	491.46 <sub>0.83</sub>	42.20 <sub>0.30</sub>	1.2454 <sub>0.0072</sub>	21.10 <sub>0.11</sub>	0.74 <sub>0.01</sub>
470	506.63 <sub>0.68</sub>	35.08 <sub>0.18</sub>	1.0550 <sub>0.0067</sub>	22.354 <sub>0.060</sub>	0.77 <sub>0.01</sub>
460	521.02 <sub>0.50</sub>	29.12 <sub>0.22</sub>	0.8870 <sub>0.0058</sub>	23.514 <sub>0.046</sub>	0.80 <sub>0.01</sub>
450	534.61 <sub>0.44</sub>	24.08 <sub>0.23</sub>	0.7398 <sub>0.0048</sub>	24.589 <sub>0.057</sub>	0.82 <sub>0.01</sub>
440	547.23 <sub>0.42</sub>	19.82 <sub>0.21</sub>	0.6115 <sub>0.0037</sub>	25.575 <sub>0.058</sub>	0.85 <sub>0.01</sub>
430	559.10 <sub>0.45</sub>	16.21 <sub>0.17</sub>	0.5007 <sub>0.0028</sub>	26.491 <sub>0.043</sub>	0.87 <sub>0.01</sub>
420	570.66 <sub>0.61</sub>	13.16 <sub>0.11</sub>	0.4058 <sub>0.0023</sub>	27.365 <sub>0.031</sub>	0.88 <sub>0.01</sub>
410	582.02 <sub>0.53</sub>	10.587 <sub>0.082</sub>	0.3251 <sub>0.0022</sub>	28.205 <sub>0.034</sub>	0.90 <sub>0.01</sub>
400	592.79 <sub>0.41</sub>	8.438 <sub>0.086</sub>	0.2572 <sub>0.0023</sub>	28.992 <sub>0.054</sub>	0.918 <sub>0.015</sub>
390	602.92 <sub>0.51</sub>	6.653 <sub>0.098</sub>	0.2007 <sub>0.0026</sub>	29.724 <sub>0.089</sub>	0.932 <sub>0.023</sub>
380	612.81 <sub>0.53</sub>	5.18 <sub>0.10</sub>	0.1544 <sub>0.0030</sub>	30.43 <sub>0.12</sub>	0.945 <sub>0.033</sub>
370	622.47 <sub>0.37</sub>	3.982 <sub>0.097</sub>	0.1168 <sub>0.0035</sub>	31.10 <sub>0.15</sub>	0.956 <sub>0.048</sub>
360	631.88 <sub>0.27</sub>	3.012 <sub>0.090</sub>	0.0868 <sub>0.0040</sub>	31.75 <sub>0.21</sub>	0.965 <sub>0.068</sub>
350	641.27 <sub>0.30</sub>	2.238 <sub>0.081</sub>	0.0632 <sub>0.0044</sub>	32.39 <sub>0.29</sub>	0.972 <sub>0.097</sub>

Table S42: GCMC-MBAR results for 3-ethylhexane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
540	425.4 <sub>1.8</sub>	79.4 <sub>4.9</sub>	1.866 <sub>0.043</sub>	16.85 <sub>0.40</sub>	0.598 <sub>0.025</sub>
530	449.1 <sub>1.4</sub>	64.7 <sub>4.4</sub>	1.614 <sub>0.029</sub>	19.05 <sub>0.45</sub>	0.647 <sub>0.034</sub>
520	468.9 <sub>1.0</sub>	53.1 <sub>3.2</sub>	1.390 <sub>0.017</sub>	20.94 <sub>0.42</sub>	0.691 <sub>0.035</sub>
510	486.13 <sub>0.88</sub>	44.0 <sub>2.0</sub>	1.1907 <sub>0.0092</sub>	22.56 <sub>0.34</sub>	0.728 <sub>0.029</sub>
500	501.71 <sub>0.68</sub>	36.6 <sub>1.1</sub>	1.0141 <sub>0.0047</sub>	23.99 <sub>0.23</sub>	0.760 <sub>0.021</sub>
490	516.13 <sub>0.49</sub>	30.49 <sub>0.54</sub>	0.8581 <sub>0.0029</sub>	25.28 <sub>0.14</sub>	0.789 <sub>0.013</sub>
480	529.64 <sub>0.54</sub>	25.32 <sub>0.27</sub>	0.7207 <sub>0.0024</sub>	26.475 <sub>0.089</sub>	0.81 <sub>0.01</sub>
470	542.43 <sub>0.64</sub>	20.95 <sub>0.17</sub>	0.6006 <sub>0.0021</sub>	27.583 <sub>0.069</sub>	0.84 <sub>0.01</sub>
460	554.62 <sub>0.65</sub>	17.25 <sub>0.13</sub>	0.4962 <sub>0.0019</sub>	28.624 <sub>0.065</sub>	0.86 <sub>0.01</sub>
450	566.22 <sub>0.66</sub>	14.11 <sub>0.11</sub>	0.4063 <sub>0.0017</sub>	29.600 <sub>0.066</sub>	0.88 <sub>0.01</sub>
440	577.33 <sub>0.67</sub>	11.466 <sub>0.088</sub>	0.3292 <sub>0.0016</sub>	30.521 <sub>0.065</sub>	0.90 <sub>0.01</sub>
430	588.23 <sub>0.68</sub>	9.237 <sub>0.067</sub>	0.2638 <sub>0.0016</sub>	31.409 <sub>0.064</sub>	0.91 <sub>0.01</sub>
420	598.94 <sub>0.67</sub>	7.369 <sub>0.055</sub>	0.2088 <sub>0.0018</sub>	32.268 <sub>0.063</sub>	0.927 <sub>0.013</sub>
410	609.11 <sub>0.53</sub>	5.815 <sub>0.055</sub>	0.1630 <sub>0.0019</sub>	33.074 <sub>0.060</sub>	0.939 <sub>0.019</sub>
400	618.75 <sub>0.46</sub>	4.534 <sub>0.059</sub>	0.1255 <sub>0.0022</sub>	33.831 <sub>0.070</sub>	0.950 <sub>0.028</sub>
390	628.18 <sub>0.46</sub>	3.487 <sub>0.060</sub>	0.0950 <sub>0.0025</sub>	34.56 <sub>0.10</sub>	0.960 <sub>0.041</sub>
380	637.85 <sub>0.36</sub>	2.641 <sub>0.057</sub>	0.0707 <sub>0.0028</sub>	35.31 <sub>0.15</sub>	0.968 <sub>0.060</sub>
370	647.74 <sub>0.30</sub>	1.965 <sub>0.051</sub>	0.0515 <sub>0.0031</sub>	36.05 <sub>0.23</sub>	0.974 <sub>0.087</sub>

Table S43: GCMC-MBAR results for 4-methylheptane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
540	418.9 <sub>1.9</sub>	80.1 <sub>1.6</sub>	1.858 <sub>0.017</sub>	16.62 <sub>0.17</sub>	0.59 <sub>0.01</sub>
530	442.2 <sub>1.4</sub>	65.0 <sub>1.3</sub>	1.606 <sub>0.015</sub>	18.87 <sub>0.15</sub>	0.640 <sub>0.010</sub>
520	462.3 <sub>1.1</sub>	53.3 <sub>1.1</sub>	1.382 <sub>0.013</sub>	20.81 <sub>0.15</sub>	0.685 <sub>0.011</sub>
510	479.9 <sub>1.1</sub>	44.10 <sub>0.88</sub>	1.183 <sub>0.010</sub>	22.48 <sub>0.16</sub>	0.722 <sub>0.010</sub>
500	495.8 <sub>1.0</sub>	36.63 <sub>0.73</sub>	1.0060 <sub>0.0083</sub>	23.95 <sub>0.16</sub>	0.755 <sub>0.011</sub>
490	510.3 <sub>1.0</sub>	30.43 <sub>0.61</sub>	0.8501 <sub>0.0062</sub>	25.28 <sub>0.16</sub>	0.783 <sub>0.011</sub>
480	523.91 <sub>0.96</sub>	25.22 <sub>0.47</sub>	0.7132 <sub>0.0044</sub>	26.49 <sub>0.15</sub>	0.809 <sub>0.011</sub>
470	536.77 <sub>0.78</sub>	20.83 <sub>0.32</sub>	0.5936 <sub>0.0029</sub>	27.62 <sub>0.11</sub>	0.83 <sub>0.01</sub>
460	548.98 <sub>0.49</sub>	17.12 <sub>0.19</sub>	0.4897 <sub>0.0020</sub>	28.679 <sub>0.071</sub>	0.85 <sub>0.01</sub>
450	560.48 <sub>0.39</sub>	13.982 <sub>0.094</sub>	0.4002 <sub>0.0014</sub>	29.660 <sub>0.044</sub>	0.87 <sub>0.01</sub>
440	571.22 <sub>0.45</sub>	11.338 <sub>0.062</sub>	0.3238 <sub>0.0012</sub>	30.567 <sub>0.048</sub>	0.89 <sub>0.01</sub>
430	581.42 <sub>0.48</sub>	9.118 <sub>0.092</sub>	0.2591 <sub>0.0013</sub>	31.419 <sub>0.060</sub>	0.908 <sub>0.012</sub>
420	591.67 <sub>0.57</sub>	7.26 <sub>0.12</sub>	0.2049 <sub>0.0018</sub>	32.257 <sub>0.078</sub>	0.923 <sub>0.023</sub>
410	602.13 <sub>0.53</sub>	5.72 <sub>0.14</sub>	0.1598 <sub>0.0025</sub>	33.10 <sub>0.12</sub>	0.936 <sub>0.037</sub>
400	612.18 <sub>0.47</sub>	4.45 <sub>0.14</sub>	0.1228 <sub>0.0033</sub>	33.90 <sub>0.18</sub>	0.947 <sub>0.056</sub>
390	621.73 <sub>0.54</sub>	3.42 <sub>0.14</sub>	0.0928 <sub>0.0042</sub>	34.65 <sub>0.25</sub>	0.957 <sub>0.082</sub>
380	631.63 <sub>0.45</sub>	2.58 <sub>0.12</sub>	0.0690 <sub>0.0050</sub>	35.41 <sub>0.37</sub>	0.97 <sub>0.12</sub>
370	641.49 <sub>0.32</sub>	1.92 <sub>0.11</sub>	0.0502 <sub>0.0057</sub>	36.15 <sub>0.52</sub>	0.97 <sub>0.17</sub>

Table S44: GCMC-MBAR results for 2,3-dimethylbutane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
480	411.7 <sub>1.1</sub>	88.4 <sub>2.2</sub>	2.369 <sub>0.027</sub>	12.87 <sub>0.14</sub>	0.58 <sub>0.01</sub>
470	437.1 <sub>1.0</sub>	71.6 <sub>2.1</sub>	2.037 <sub>0.019</sub>	14.80 <sub>0.17</sub>	0.627 <sub>0.013</sub>
460	459.23 <sub>0.86</sub>	58.1 <sub>1.6</sub>	1.742 <sub>0.013</sub>	16.53 <sub>0.18</sub>	0.676 <sub>0.015</sub>
450	478.07 <sub>0.79</sub>	47.5 <sub>1.1</sub>	1.4813 <sub>0.0074</sub>	18.00 <sub>0.16</sub>	0.718 <sub>0.014</sub>
440	494.54 <sub>0.81</sub>	39.07 <sub>0.72</sub>	1.2513 <sub>0.0041</sub>	19.26 <sub>0.13</sub>	0.754 <sub>0.012</sub>
430	509.51 <sub>0.77</sub>	32.20 <sub>0.43</sub>	1.0493 <sub>0.0024</sub>	20.37 <sub>0.10</sub>	0.79 <sub>0.01</sub>
420	523.51 <sub>0.64</sub>	26.49 <sub>0.25</sub>	0.8726 <sub>0.0019</sub>	21.382 <sub>0.079</sub>	0.81 <sub>0.01</sub>
410	536.78 <sub>0.54</sub>	21.70 <sub>0.15</sub>	0.7189 <sub>0.0020</sub>	22.310 <sub>0.063</sub>	0.84 <sub>0.01</sub>
400	549.39 <sub>0.59</sub>	17.68 <sub>0.10</sub>	0.5863 <sub>0.0021</sub>	23.171 <sub>0.060</sub>	0.86 <sub>0.01</sub>
390	561.38 <sub>0.73</sub>	14.296 <sub>0.082</sub>	0.4727 <sub>0.0023</sub>	23.971 <sub>0.062</sub>	0.88 <sub>0.01</sub>
380	572.70 <sub>0.68</sub>	11.455 <sub>0.085</sub>	0.3764 <sub>0.0025</sub>	24.712 <sub>0.056</sub>	0.896 <sub>0.011</sub>
370	583.42 <sub>0.40</sub>	9.083 <sub>0.092</sub>	0.2957 <sub>0.0028</sub>	25.402 <sub>0.050</sub>	0.912 <sub>0.016</sub>
360	593.75 <sub>0.21</sub>	7.115 <sub>0.095</sub>	0.2289 <sub>0.0033</sub>	26.054 <sub>0.064</sub>	0.926 <sub>0.024</sub>
350	603.97 <sub>0.20</sub>	5.497 <sub>0.091</sub>	0.1742 <sub>0.0037</sub>	26.686 <sub>0.091</sub>	0.938 <sub>0.034</sub>
340	614.11 <sub>0.22</sub>	4.179 <sub>0.082</sub>	0.1302 <sub>0.0042</sub>	27.30 <sub>0.13</sub>	0.950 <sub>0.047</sub>
330	623.72 <sub>0.26</sub>	3.121 <sub>0.069</sub>	0.0953 <sub>0.0047</sub>	27.87 <sub>0.18</sub>	0.959 <sub>0.066</sub>
320	632.83 <sub>0.34</sub>	2.284 <sub>0.057</sub>	0.0682 <sub>0.0050</sub>	28.41 <sub>0.25</sub>	0.968 <sub>0.092</sub>



Table S45: GCMC-MBAR results for 2,3-dimethylhexane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
540	422.2 <sub>1.1</sub>	84.5 <sub>3.7</sub>	1.948 <sub>0.048</sub>	15.98 <sub>0.12</sub>	0.586 <sub>0.012</sub>
530	445.87 <sub>0.82</sub>	68.6 <sub>3.6</sub>	1.689 <sub>0.036</sub>	18.23 <sub>0.22</sub>	0.638 <sub>0.022</sub>
520	466.30 <sub>0.66</sub>	56.3 <sub>3.0</sub>	1.458 <sub>0.024</sub>	20.17 <sub>0.26</sub>	0.684 <sub>0.027</sub>
510	484.13 <sub>0.59</sub>	46.7 <sub>2.1</sub>	1.253 <sub>0.015</sub>	21.81 <sub>0.23</sub>	0.722 <sub>0.026</sub>
500	499.99 <sub>0.52</sub>	39.0 <sub>1.4</sub>	1.0703 <sub>0.0092</sub>	23.24 <sub>0.18</sub>	0.755 <sub>0.022</sub>
490	514.34 <sub>0.48</sub>	32.51 <sub>0.80</sub>	0.9084 <sub>0.0054</sub>	24.51 <sub>0.13</sub>	0.783 <sub>0.016</sub>
480	527.67 <sub>0.54</sub>	27.08 <sub>0.47</sub>	0.7657 <sub>0.0032</sub>	25.677 <sub>0.096</sub>	0.809 <sub>0.012</sub>
470	540.41 <sub>0.64</sub>	22.49 <sub>0.30</sub>	0.6406 <sub>0.0019</sub>	26.765 <sub>0.082</sub>	0.833 <sub>0.010</sub>
460	552.77 <sub>0.76</sub>	18.58 <sub>0.23</sub>	0.5313 <sub>0.0015</sub>	27.797 <sub>0.085</sub>	0.854 <sub>0.011</sub>
450	564.84 <sub>0.87</sub>	15.26 <sub>0.22</sub>	0.4366 <sub>0.0018</sub>	28.78 <sub>0.11</sub>	0.874 <sub>0.015</sub>
440	576.56 <sub>0.95</sub>	12.43 <sub>0.23</sub>	0.3553 <sub>0.0025</sub>	29.73 <sub>0.14</sub>	0.892 <sub>0.022</sub>
430	587.55 <sub>0.97</sub>	10.05 <sub>0.23</sub>	0.2860 <sub>0.0034</sub>	30.60 <sub>0.18</sub>	0.909 <sub>0.032</sub>
420	597.72 <sub>0.83</sub>	8.05 <sub>0.21</sub>	0.2274 <sub>0.0044</sub>	31.40 <sub>0.21</sub>	0.924 <sub>0.043</sub>
410	607.46 <sub>0.53</sub>	6.39 <sub>0.18</sub>	0.1787 <sub>0.0053</sub>	32.16 <sub>0.22</sub>	0.937 <sub>0.055</sub>
400	617.12 <sub>0.48</sub>	5.01 <sub>0.14</sub>	0.1384 <sub>0.0061</sub>	32.90 <sub>0.24</sub>	0.949 <sub>0.070</sub>
390	626.72 <sub>0.62</sub>	3.88 <sub>0.10</sub>	0.1056 <sub>0.0067</sub>	33.64 <sub>0.28</sub>	0.959 <sub>0.087</sub>
380	636.43 <sub>0.54</sub>	2.959 <sub>0.070</sub>	0.0792 <sub>0.0071</sub>	34.36 <sub>0.35</sub>	0.97 <sub>0.11</sub>
370	646.47 <sub>0.59</sub>	2.219 <sub>0.049</sub>	0.0583 <sub>0.0072</sub>	35.10 <sub>0.46</sub>	0.98 <sub>0.14</sub>

Table S46: GCMC-MBAR results for 2,3-dimethylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
510	427.9 <sub>1.6</sub>	81 <sub>10</sub>	2.082 <sub>0.091</sub>	15.20 <sub>0.53</sub>	0.606 <sub>0.042</sub>
500	451.7 <sub>1.0</sub>	66.3 <sub>9.6</sub>	1.800 <sub>0.057</sub>	17.17 <sub>0.70</sub>	0.654 <sub>0.061</sub>
490	471.41 <sub>0.86</sub>	54.7 <sub>7.1</sub>	1.549 <sub>0.028</sub>	18.83 <sub>0.70</sub>	0.696 <sub>0.065</sub>
480	488.48 <sub>0.89</sub>	45.5 <sub>4.0</sub>	1.3245 <sub>0.0092</sub>	20.25 <sub>0.54</sub>	0.731 <sub>0.053</sub>
470	503.90 <sub>0.73</sub>	37.9 <sub>1.7</sub>	1.1257 <sub>0.0050</sub>	21.50 <sub>0.32</sub>	0.762 <sub>0.034</sub>
460	518.28 <sub>0.52</sub>	31.50 <sub>0.62</sub>	0.9501 <sub>0.0072</sub>	22.64 <sub>0.14</sub>	0.790 <sub>0.020</sub>
450	532.03 <sub>0.63</sub>	26.12 <sub>0.25</sub>	0.7956 <sub>0.0080</sub>	23.709 <sub>0.054</sub>	0.816 <sub>0.013</sub>
440	545.26 <sub>0.62</sub>	21.57 <sub>0.18</sub>	0.6606 <sub>0.0080</sub>	24.711 <sub>0.037</sub>	0.839 <sub>0.011</sub>
430	557.77 <sub>0.54</sub>	17.70 <sub>0.16</sub>	0.5435 <sub>0.0075</sub>	25.646 <sub>0.038</sub>	0.860 <sub>0.010</sub>
420	569.33 <sub>0.53</sub>	14.43 <sub>0.15</sub>	0.4426 <sub>0.0070</sub>	26.504 <sub>0.037</sub>	0.880 <sub>0.011</sub>
410	580.03 <sub>0.59</sub>	11.68 <sub>0.14</sub>	0.3565 <sub>0.0063</sub>	27.290 <sub>0.040</sub>	0.897 <sub>0.012</sub>
400	590.33 <sub>0.66</sub>	9.36 <sub>0.13</sub>	0.2837 <sub>0.0057</sub>	28.032 <sub>0.047</sub>	0.913 <sub>0.014</sub>
390	600.50 <sub>0.61</sub>	7.43 <sub>0.11</sub>	0.2229 <sub>0.0052</sub>	28.747 <sub>0.057</sub>	0.927 <sub>0.019</sub>
380	610.33 <sub>0.59</sub>	5.831 <sub>0.094</sub>	0.1726 <sub>0.0048</sub>	29.424 <sub>0.079</sub>	0.939 <sub>0.025</sub>
370	620.01 <sub>0.64</sub>	4.512 <sub>0.077</sub>	0.1315 <sub>0.0046</sub>	30.08 <sub>0.11</sub>	0.950 <sub>0.036</sub>
360	630.25 <sub>0.55</sub>	3.438 <sub>0.066</sub>	0.0984 <sub>0.0045</sub>	30.75 <sub>0.14</sub>	0.959 <sub>0.050</sub>
350	640.58 <sub>0.49</sub>	2.572 <sub>0.058</sub>	0.0722 <sub>0.0046</sub>	31.42 <sub>0.20</sub>	0.966 <sub>0.072</sub>

Table S47: GCMC-MBAR results for 2,4-dimethylhexane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
540	406.0 <sub>3.8</sub>	97.8 <sub>2.1</sub>	2.090 <sub>0.036</sub>	14.21 <sub>0.22</sub>	0.54 <sub>0.01</sub>
530	430.3 <sub>3.2</sub>	78.1 <sub>2.3</sub>	1.810 <sub>0.028</sub>	16.71 <sub>0.16</sub>	0.60 <sub>0.01</sub>
520	452.1 <sub>1.9</sub>	63.1 <sub>2.2</sub>	1.562 <sub>0.019</sub>	18.90 <sub>0.15</sub>	0.654 <sub>0.016</sub>
510	470.9 <sub>1.1</sub>	51.8 <sub>1.9</sub>	1.342 <sub>0.013</sub>	20.72 <sub>0.19</sub>	0.698 <sub>0.020</sub>
500	487.5 <sub>1.1</sub>	42.9 <sub>1.4</sub>	1.1472 <sub>0.0080</sub>	22.26 <sub>0.19</sub>	0.734 <sub>0.020</sub>
490	502.5 <sub>1.2</sub>	35.72 <sub>0.85</sub>	0.9745 <sub>0.0054</sub>	23.60 <sub>0.16</sub>	0.765 <sub>0.017</sub>
480	516.3 <sub>1.4</sub>	29.69 <sub>0.47</sub>	0.8222 <sub>0.0043</sub>	24.83 <sub>0.13</sub>	0.793 <sub>0.012</sub>
470	529.5 <sub>1.6</sub>	24.62 <sub>0.27</sub>	0.6886 <sub>0.0037</sub>	25.96 <sub>0.11</sub>	0.82 <sub>0.01</sub>
460	542.1 <sub>1.7</sub>	20.34 <sub>0.18</sub>	0.5720 <sub>0.0031</sub>	27.026 <sub>0.098</sub>	0.84 <sub>0.01</sub>
450	554.3 <sub>1.8</sub>	16.71 <sub>0.13</sub>	0.4708 <sub>0.0026</sub>	28.031 <sub>0.098</sub>	0.86 <sub>0.01</sub>
440	566.1 <sub>1.8</sub>	13.63 <sub>0.10</sub>	0.3836 <sub>0.0024</sub>	28.98 <sub>0.10</sub>	0.88 <sub>0.01</sub>
430	577.4 <sub>1.6</sub>	11.03 <sub>0.11</sub>	0.3092 <sub>0.0023</sub>	29.89 <sub>0.11</sub>	0.895 <sub>0.010</sub>
420	588.1 <sub>1.4</sub>	8.85 <sub>0.15</sub>	0.2463 <sub>0.0022</sub>	30.73 <sub>0.13</sub>	0.910 <sub>0.019</sub>
410	598.1 <sub>1.1</sub>	7.02 <sub>0.19</sub>	0.1937 <sub>0.0024</sub>	31.51 <sub>0.17</sub>	0.924 <sub>0.033</sub>
400	607.97 <sub>0.72</sub>	5.51 <sub>0.22</sub>	0.1503 <sub>0.0031</sub>	32.27 <sub>0.22</sub>	0.937 <sub>0.056</sub>
390	617.69 <sub>0.31</sub>	4.27 <sub>0.23</sub>	0.1148 <sub>0.0042</sub>	33.01 <sub>0.29</sub>	0.947 <sub>0.088</sub>
380	627.04 <sub>0.22</sub>	3.26 <sub>0.22</sub>	0.0862 <sub>0.0055</sub>	33.71 <sub>0.42</sub>	0.96 <sub>0.14</sub>
370	635.81 <sub>0.19</sub>	2.45 <sub>0.20</sub>	0.0636 <sub>0.0068</sub>	34.36 <sub>0.62</sub>	0.97 <sub>0.20</sub>
360	644.04 <sub>0.32</sub>	1.80 <sub>0.18</sub>	0.0460 <sub>0.0080</sub>	34.97 <sub>0.91</sub>	0.97 <sub>0.30</sub>

Table S48: GCMC-MBAR results for 2,4-dimethylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
500	420.0 <sub>1.7</sub>	81.9 <sub>2.0</sub>	2.003 <sub>0.018</sub>	14.77 <sub>0.12</sub>	0.590 <sub>0.011</sub>
490	443.0 <sub>1.0</sub>	65.6 <sub>1.4</sub>	1.722 <sub>0.014</sub>	16.87 <sub>0.12</sub>	0.646 <sub>0.011</sub>
480	462.83 <sub>0.68</sub>	53.40 <sub>0.95</sub>	1.473 <sub>0.011</sub>	18.632 <sub>0.099</sub>	0.69 <sub>0.01</sub>
470	480.50 <sub>0.59</sub>	43.96 <sub>0.74</sub>	1.2527 <sub>0.0088</sub>	20.125 <sub>0.093</sub>	0.73 <sub>0.01</sub>
460	496.63 <sub>0.64</sub>	36.33 <sub>0.64</sub>	1.0583 <sub>0.0067</sub>	21.44 <sub>0.10</sub>	0.763 <sub>0.010</sub>
450	511.50 <sub>0.67</sub>	30.02 <sub>0.54</sub>	0.8873 <sub>0.0049</sub>	22.62 <sub>0.10</sub>	0.792 <sub>0.012</sub>
440	525.28 <sub>0.65</sub>	24.73 <sub>0.40</sub>	0.7377 <sub>0.0038</sub>	23.700 <sub>0.095</sub>	0.817 <sub>0.012</sub>
430	538.33 <sub>0.62</sub>	20.28 <sub>0.26</sub>	0.6076 <sub>0.0032</sub>	24.704 <sub>0.085</sub>	0.840 <sub>0.011</sub>
420	550.86 <sub>0.66</sub>	16.52 <sub>0.19</sub>	0.4955 <sub>0.0029</sub>	25.650 <sub>0.080</sub>	0.86 <sub>0.01</sub>
410	562.59 <sub>0.67</sub>	13.35 <sub>0.21</sub>	0.3996 <sub>0.0024</sub>	26.522 <sub>0.078</sub>	0.880 <sub>0.014</sub>
400	573.32 <sub>0.65</sub>	10.70 <sub>0.23</sub>	0.3184 <sub>0.0024</sub>	27.312 <sub>0.092</sub>	0.897 <sub>0.023</sub>
390	583.50 <sub>0.57</sub>	8.49 <sub>0.25</sub>	0.2504 <sub>0.0031</sub>	28.05 <sub>0.13</sub>	0.911 <sub>0.037</sub>
380	593.48 <sub>0.38</sub>	6.66 <sub>0.25</sub>	0.1942 <sub>0.0044</sub>	28.75 <sub>0.18</sub>	0.925 <sub>0.056</sub>
370	603.16 <sub>0.41</sub>	5.15 <sub>0.26</sub>	0.1483 <sub>0.0059</sub>	29.42 <sub>0.26</sub>	0.937 <sub>0.084</sub>
360	613.02 <sub>0.44</sub>	3.93 <sub>0.28</sub>	0.1113 <sub>0.0076</sub>	30.09 <sub>0.36</sub>	0.95 <sub>0.13</sub>
350	623.28 <sub>0.28</sub>	2.94 <sub>0.28</sub>	0.0819 <sub>0.0093</sub>	30.77 <sub>0.52</sub>	0.96 <sub>0.18</sub>

Table S49: GCMC-MBAR results for 2,5-dimethylhexane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
530	405.4 <sub>2.2</sub>	92.0 <sub>1.7</sub>	1.963 <sub>0.011</sub>	14.59 <sub>0.15</sub>	0.55 <sub>0.01</sub>
520	429.1 <sub>1.3</sub>	73.2 <sub>1.3</sub>	1.6929 <sub>0.0078</sub>	17.04 <sub>0.13</sub>	0.61 <sub>0.01</sub>
510	450.43 <sub>0.78</sub>	59.06 <sub>0.93</sub>	1.4547 <sub>0.0055</sub>	19.17 <sub>0.12</sub>	0.66 <sub>0.01</sub>
500	469.12 <sub>0.68</sub>	48.33 <sub>0.60</sub>	1.2442 <sub>0.0042</sub>	20.967 <sub>0.088</sub>	0.71 <sub>0.01</sub>
490	485.77 <sub>0.64</sub>	39.90 <sub>0.39</sub>	1.0581 <sub>0.0034</sub>	22.495 <sub>0.063</sub>	0.74 <sub>0.01</sub>
480	500.89 <sub>0.55</sub>	33.04 <sub>0.27</sub>	0.8939 <sub>0.0028</sub>	23.843 <sub>0.049</sub>	0.77 <sub>0.01</sub>
470	514.91 <sub>0.60</sub>	27.35 <sub>0.18</sub>	0.7498 <sub>0.0025</sub>	25.066 <sub>0.046</sub>	0.80 <sub>0.01</sub>
460	528.20 <sub>0.87</sub>	22.57 <sub>0.11</sub>	0.6237 <sub>0.0025</sub>	26.198 <sub>0.050</sub>	0.83 <sub>0.01</sub>
450	540.9 <sub>1.0</sub>	18.541 <sub>0.071</sub>	0.5141 <sub>0.0025</sub>	27.259 <sub>0.054</sub>	0.85 <sub>0.01</sub>
440	553.03 <sub>0.89</sub>	15.132 <sub>0.065</sub>	0.4196 <sub>0.0026</sub>	28.254 <sub>0.049</sub>	0.87 <sub>0.01</sub>
430	564.47 <sub>0.80</sub>	12.255 <sub>0.087</sub>	0.3388 <sub>0.0028</sub>	29.185 <sub>0.043</sub>	0.883 <sub>0.012</sub>
420	575.38 <sub>0.95</sub>	9.83 <sub>0.12</sub>	0.2703 <sub>0.0031</sub>	30.060 <sub>0.042</sub>	0.899 <sub>0.019</sub>
410	586.0 <sub>1.0</sub>	7.81 <sub>0.15</sub>	0.2130 <sub>0.0036</sub>	30.900 <sub>0.064</sub>	0.914 <sub>0.030</sub>
400	596.47 <sub>0.83</sub>	6.13 <sub>0.17</sub>	0.1654 <sub>0.0044</sub>	31.71 <sub>0.12</sub>	0.927 <sub>0.047</sub>
390	606.50 <sub>0.59</sub>	4.75 <sub>0.18</sub>	0.1266 <sub>0.0053</sub>	32.48 <sub>0.20</sub>	0.939 <sub>0.071</sub>
380	615.90 <sub>0.53</sub>	3.63 <sub>0.18</sub>	0.0953 <sub>0.0064</sub>	33.20 <sub>0.30</sub>	0.95 <sub>0.11</sub>
370	624.82 <sub>0.36</sub>	2.73 <sub>0.18</sub>	0.0704 <sub>0.0075</sub>	33.87 <sub>0.44</sub>	0.96 <sub>0.15</sub>
360	633.58 <sub>0.39</sub>	2.01 <sub>0.16</sub>	0.0510 <sub>0.0085</sub>	34.52 <sub>0.65</sub>	0.97 <sub>0.22</sub>

Table S50: GCMC-MBAR results for 3,4-dimethylhexane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_{\text{v}}$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
550	416.97 <sub>0.84</sub>	91.7 <sub>2.1</sub>	2.100 <sub>0.024</sub>	15.29 <sub>0.11</sub>	0.57 <sub>0.01</sub>
540	441.75 <sub>0.84</sub>	75.2 <sub>2.0</sub>	1.828 <sub>0.018</sub>	17.53 <sub>0.13</sub>	0.619 <sub>0.011</sub>
530	462.97 <sub>0.65</sub>	61.9 <sub>1.6</sub>	1.584 <sub>0.012</sub>	19.50 <sub>0.14</sub>	0.664 <sub>0.014</sub>
520	481.27 <sub>0.52</sub>	51.4 <sub>1.2</sub>	1.3670 <sub>0.0076</sub>	21.20 <sub>0.13</sub>	0.703 <sub>0.014</sub>
510	497.59 <sub>0.51</sub>	42.88 <sub>0.80</sub>	1.1732 <sub>0.0044</sub>	22.68 <sub>0.12</sub>	0.737 <sub>0.012</sub>
500	512.45 <sub>0.50</sub>	35.87 <sub>0.49</sub>	1.0010 <sub>0.0025</sub>	23.994 <sub>0.090</sub>	0.77 <sub>0.01</sub>
490	526.21 <sub>0.55</sub>	30.00 <sub>0.31</sub>	0.8485 <sub>0.0015</sub>	25.192 <sub>0.073</sub>	0.79 <sub>0.01</sub>
480	539.20 <sub>0.79</sub>	25.02 <sub>0.22</sub>	0.7142 <sub>0.0014</sub>	26.302 <sub>0.072</sub>	0.82 <sub>0.01</sub>
470	551.6 <sub>1.1</sub>	20.79 <sub>0.18</sub>	0.5963 <sub>0.0017</sub>	27.341 <sub>0.083</sub>	0.84 <sub>0.01</sub>
460	563.5 <sub>1.3</sub>	17.18 <sub>0.15</sub>	0.4936 <sub>0.0023</sub>	28.319 <sub>0.097</sub>	0.858 <sub>0.011</sub>
450	574.7 <sub>1.3</sub>	14.10 <sub>0.14</sub>	0.4050 <sub>0.0028</sub>	29.24 <sub>0.11</sub>	0.877 <sub>0.014</sub>
440	585.4 <sub>1.2</sub>	11.49 <sub>0.12</sub>	0.3287 <sub>0.0033</sub>	30.09 <sub>0.11</sub>	0.893 <sub>0.018</sub>
430	595.6 <sub>1.1</sub>	9.29 <sub>0.11</sub>	0.2642 <sub>0.0037</sub>	30.90 <sub>0.11</sub>	0.909 <sub>0.022</sub>
420	605.36 <sub>0.91</sub>	7.437 <sub>0.097</sub>	0.2097 <sub>0.0040</sub>	31.66 <sub>0.11</sub>	0.922 <sub>0.027</sub>
410	614.74 <sub>0.65</sub>	5.893 <sub>0.085</sub>	0.1644 <sub>0.0042</sub>	32.38 <sub>0.12</sub>	0.935 <sub>0.033</sub>
400	624.02 <sub>0.41</sub>	4.615 <sub>0.075</sub>	0.1271 <sub>0.0044</sub>	33.08 <sub>0.14</sub>	0.946 <sub>0.041</sub>
390	633.41 <sub>0.31</sub>	3.566 <sub>0.067</sub>	0.0967 <sub>0.0045</sub>	33.78 <sub>0.17</sub>	0.955 <sub>0.053</sub>
380	642.95 <sub>0.22</sub>	2.715 <sub>0.059</sub>	0.0724 <sub>0.0045</sub>	34.48 <sub>0.22</sub>	0.964 <sub>0.070</sub>
370	652.67 <sub>0.19</sub>	2.032 <sub>0.051</sub>	0.0531 <sub>0.0046</sub>	35.18 <sub>0.29</sub>	0.970 <sub>0.094</sub>

Table S51: GCMC-MBAR results for 2,3,4-trimethylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
550	415.7 <sub>1.4</sub>	96.1 <sub>3.2</sub>	2.199 <sub>0.049</sub>	14.71 <sub>0.16</sub>	0.57 <sub>0.01</sub>
540	442.1 <sub>1.2</sub>	79.3 <sub>3.4</sub>	1.920 <sub>0.039</sub>	16.92 <sub>0.21</sub>	0.616 <sub>0.015</sub>
530	464.77 <sub>0.78</sub>	65.5 <sub>3.0</sub>	1.669 <sub>0.029</sub>	18.91 <sub>0.25</sub>	0.661 <sub>0.021</sub>
520	483.71 <sub>0.55</sub>	54.4 <sub>2.4</sub>	1.444 <sub>0.021</sub>	20.61 <sub>0.24</sub>	0.701 <sub>0.023</sub>
510	499.94 <sub>0.54</sub>	45.5 <sub>1.7</sub>	1.243 <sub>0.015</sub>	22.07 <sub>0.20</sub>	0.736 <sub>0.021</sub>
500	514.37 <sub>0.58</sub>	38.2 <sub>1.1</sub>	1.064 <sub>0.011</sub>	23.34 <sub>0.15</sub>	0.766 <sub>0.017</sub>
490	527.58 <sub>0.61</sub>	32.03 <sub>0.69</sub>	0.9054 <sub>0.0087</sub>	24.49 <sub>0.10</sub>	0.793 <sub>0.013</sub>
480	540.01 <sub>0.71</sub>	26.83 <sub>0.52</sub>	0.7652 <sub>0.0066</sub>	25.541 <sub>0.081</sub>	0.816 <sub>0.011</sub>
470	552.10 <sub>0.82</sub>	22.39 <sub>0.43</sub>	0.6419 <sub>0.0048</sub>	26.539 <sub>0.074</sub>	0.838 <sub>0.011</sub>
460	564.06 <sub>0.86</sub>	18.59 <sub>0.35</sub>	0.5341 <sub>0.0032</sub>	27.497 <sub>0.070</sub>	0.858 <sub>0.013</sub>
450	575.70 <sub>0.84</sub>	15.35 <sub>0.26</sub>	0.4403 <sub>0.0020</sub>	28.409 <sub>0.064</sub>	0.876 <sub>0.014</sub>
440	586.68 <sub>0.81</sub>	12.58 <sub>0.18</sub>	0.3594 <sub>0.0017</sub>	29.258 <sub>0.061</sub>	0.892 <sub>0.013</sub>
430	596.87 <sub>0.75</sub>	10.22 <sub>0.11</sub>	0.2903 <sub>0.0018</sub>	30.041 <sub>0.060</sub>	0.907 <sub>0.013</sub>
420	606.38 <sub>0.61</sub>	8.235 <sub>0.070</sub>	0.2318 <sub>0.0019</sub>	30.766 <sub>0.064</sub>	0.921 <sub>0.013</sub>
410	615.73 <sub>0.46</sub>	6.567 <sub>0.082</sub>	0.1828 <sub>0.0020</sub>	31.466 <sub>0.074</sub>	0.933 <sub>0.018</sub>
400	625.40 <sub>0.41</sub>	5.18 <sub>0.11</sub>	0.1422 <sub>0.0022</sub>	32.176 <sub>0.095</sub>	0.944 <sub>0.029</sub>
390	634.86 <sub>0.32</sub>	4.03 <sub>0.13</sub>	0.1090 <sub>0.0025</sub>	32.86 <sub>0.14</sub>	0.953 <sub>0.046</sub>
380	643.49 <sub>0.23</sub>	3.09 <sub>0.13</sub>	0.0822 <sub>0.0031</sub>	33.48 <sub>0.20</sub>	0.962 <sub>0.071</sub>
370	652.26 <sub>0.34</sub>	2.33 <sub>0.13</sub>	0.0609 <sub>0.0038</sub>	34.10 <sub>0.29</sub>	0.97 <sub>0.11</sub>

Table S52: GCMC-MBAR results for 2-methyl-3-ethylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
540	433.4 <sub>2.7</sub>	82.6 <sub>3.0</sub>	1.946 <sub>0.032</sub>	16.49 <sub>0.24</sub>	0.599 <sub>0.012</sub>
530	455.6 <sub>2.0</sub>	67.4 <sub>2.8</sub>	1.688 <sub>0.024</sub>	18.61 <sub>0.30</sub>	0.649 <sub>0.019</sub>
520	474.7 <sub>1.3</sub>	55.6 <sub>2.2</sub>	1.458 <sub>0.016</sub>	20.42 <sub>0.29</sub>	0.692 <sub>0.021</sub>
510	491.79 <sub>0.85</sub>	46.4 <sub>1.5</sub>	1.253 <sub>0.011</sub>	21.98 <sub>0.23</sub>	0.728 <sub>0.019</sub>
500	507.34 <sub>0.64</sub>	38.79 <sub>0.93</sub>	1.0711 <sub>0.0081</sub>	23.35 <sub>0.17</sub>	0.759 <sub>0.015</sub>
490	521.73 <sub>0.53</sub>	32.44 <sub>0.55</sub>	0.9094 <sub>0.0062</sub>	24.59 <sub>0.11</sub>	0.786 <sub>0.011</sub>
480	535.29 <sub>0.40</sub>	27.07 <sub>0.36</sub>	0.7666 <sub>0.0049</sub>	25.746 <sub>0.076</sub>	0.81 <sub>0.01</sub>
470	548.25 <sub>0.30</sub>	22.50 <sub>0.28</sub>	0.6411 <sub>0.0038</sub>	26.827 <sub>0.056</sub>	0.83 <sub>0.01</sub>
460	560.62 <sub>0.34</sub>	18.60 <sub>0.23</sub>	0.5316 <sub>0.0028</sub>	27.843 <sub>0.047</sub>	0.85 <sub>0.01</sub>
450	572.31 <sub>0.35</sub>	15.28 <sub>0.20</sub>	0.4368 <sub>0.0020</sub>	28.791 <sub>0.049</sub>	0.87 <sub>0.01</sub>
440	583.21 <sub>0.31</sub>	12.46 <sub>0.17</sub>	0.3554 <sub>0.0014</sub>	29.667 <sub>0.055</sub>	0.891 <sub>0.012</sub>
430	593.38 <sub>0.30</sub>	10.08 <sub>0.14</sub>	0.2861 <sub>0.0014</sub>	30.476 <sub>0.064</sub>	0.907 <sub>0.015</sub>
420	602.98 <sub>0.33</sub>	8.09 <sub>0.13</sub>	0.2277 <sub>0.0018</sub>	31.230 <sub>0.085</sub>	0.921 <sub>0.020</sub>
410	612.40 <sub>0.54</sub>	6.42 <sub>0.11</sub>	0.1789 <sub>0.0022</sub>	31.96 <sub>0.12</sub>	0.933 <sub>0.026</sub>
400	622.18 <sub>0.63</sub>	5.04 <sub>0.10</sub>	0.1387 <sub>0.0027</sub>	32.69 <sub>0.15</sub>	0.944 <sub>0.036</sub>
390	632.26 <sub>0.45</sub>	3.907 <sub>0.092</sub>	0.1058 <sub>0.0031</sub>	33.42 <sub>0.18</sub>	0.954 <sub>0.049</sub>
380	641.81 <sub>0.31</sub>	2.981 <sub>0.084</sub>	0.0794 <sub>0.0036</sub>	34.12 <sub>0.22</sub>	0.963 <sub>0.069</sub>
370	650.71 <sub>0.52</sub>	2.238 <sub>0.074</sub>	0.0585 <sub>0.0039</sub>	34.76 <sub>0.31</sub>	0.971 <sub>0.096</sub>



Table S53: GCMC-MBAR results for 3-methyl-3-ethylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_{\text{v}}$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
560	429.6 <sub>1.9</sub>	93.3 <sub>3.0</sub>	2.224 <sub>0.025</sub>	15.51 <sub>0.15</sub>	0.585 <sub>0.013</sub>
550	453.4 <sub>1.3</sub>	76.7 <sub>2.3</sub>	1.949 <sub>0.017</sub>	17.72 <sub>0.13</sub>	0.635 <sub>0.015</sub>
540	473.9 <sub>1.5</sub>	64.1 <sub>1.4</sub>	1.702 <sub>0.012</sub>	19.55 <sub>0.10</sub>	0.676 <sub>0.012</sub>
530	491.5 <sub>1.3</sub>	54.07 <sub>0.82</sub>	1.4787 <sub>0.0089</sub>	21.089 <sub>0.076</sub>	0.71 <sub>0.01</sub>
520	506.85 <sub>0.72</sub>	45.77 <sub>0.59</sub>	1.2780 <sub>0.0068</sub>	22.420 <sub>0.057</sub>	0.74 <sub>0.01</sub>
510	520.59 <sub>0.37</sub>	38.74 <sub>0.54</sub>	1.0984 <sub>0.0049</sub>	23.608 <sub>0.064</sub>	0.76 <sub>0.01</sub>
500	533.49 <sub>0.41</sub>	32.75 <sub>0.51</sub>	0.9383 <sub>0.0034</sub>	24.705 <sub>0.078</sub>	0.787 <sub>0.011</sub>
490	545.92 <sub>0.68</sub>	27.61 <sub>0.41</sub>	0.7960 <sub>0.0027</sub>	25.736 <sub>0.080</sub>	0.808 <sub>0.012</sub>
480	557.91 <sub>0.83</sub>	23.19 <sub>0.27</sub>	0.6704 <sub>0.0029</sub>	26.707 <sub>0.072</sub>	0.827 <sub>0.012</sub>
470	569.50 <sub>0.65</sub>	19.38 <sub>0.14</sub>	0.5599 <sub>0.0033</sub>	27.623 <sub>0.058</sub>	0.84 <sub>0.01</sub>
460	580.74 <sub>0.49</sub>	16.096 <sub>0.094</sub>	0.4634 <sub>0.0033</sub>	28.496 <sub>0.048</sub>	0.86 <sub>0.01</sub>
450	591.73 <sub>0.55</sub>	13.27 <sub>0.13</sub>	0.3799 <sub>0.0029</sub>	29.336 <sub>0.044</sub>	0.87 <sub>0.01</sub>
440	602.39 <sub>0.47</sub>	10.84 <sub>0.16</sub>	0.3082 <sub>0.0025</sub>	30.140 <sub>0.046</sub>	0.888 <sub>0.011</sub>
430	612.22 <sub>0.39</sub>	8.78 <sub>0.18</sub>	0.2472 <sub>0.0021</sub>	30.879 <sub>0.073</sub>	0.900 <sub>0.017</sub>
420	621.24 <sub>0.63</sub>	7.03 <sub>0.20</sub>	0.1959 <sub>0.0020</sub>	31.56 <sub>0.12</sub>	0.911 <sub>0.026</sub>
410	630.11 <sub>0.71</sub>	5.57 <sub>0.23</sub>	0.1533 <sub>0.0023</sub>	32.21 <sub>0.17</sub>	0.922 <sub>0.041</sub>
400	639.21 <sub>0.40</sub>	4.36 <sub>0.28</sub>	0.1182 <sub>0.0032</sub>	32.87 <sub>0.24</sub>	0.932 <sub>0.068</sub>
390	648.92 <sub>0.26</sub>	3.36 <sub>0.32</sub>	0.0898 <sub>0.0047</sub>	33.56 <sub>0.35</sub>	0.94 <sub>0.11</sub>

Table S54: GCMC-MBAR results for 2,2-dimethylbutane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
470	412.4 <sub>5.2</sub>	85.4 <sub>8.5</sub>	2.32 <sub>0.11</sub>	12.77 <sub>0.20</sub>	0.598 <sub>0.027</sub>
460	438.1 <sub>4.9</sub>	69.3 <sub>8.7</sub>	1.996 <sub>0.071</sub>	14.62 <sub>0.34</sub>	0.649 <sub>0.049</sub>
450	459.2 <sub>3.1</sub>	56.9 <sub>7.2</sub>	1.709 <sub>0.036</sub>	16.16 <sub>0.45</sub>	0.691 <sub>0.061</sub>
440	477.5 <sub>1.5</sub>	47.1 <sub>4.6</sub>	1.453 <sub>0.011</sub>	17.47 <sub>0.41</sub>	0.727 <sub>0.057</sub>
430	494.03 <sub>0.95</sub>	39.0 <sub>2.1</sub>	1.2267 <sub>0.0045</sub>	18.63 <sub>0.25</sub>	0.759 <sub>0.039</sub>
420	508.92 <sub>0.71</sub>	32.17 <sub>0.58</sub>	1.0273 <sub>0.0094</sub>	19.664 <sub>0.098</sub>	0.788 <sub>0.020</sub>
410	522.56 <sub>0.38</sub>	26.47 <sub>0.14</sub>	0.853 <sub>0.010</sub>	20.599 <sub>0.035</sub>	0.81 <sub>0.01</sub>
400	535.52 <sub>0.29</sub>	21.68 <sub>0.16</sub>	0.7014 <sub>0.0094</sub>	21.466 <sub>0.026</sub>	0.84 <sub>0.01</sub>
390	548.22 <sub>0.34</sub>	17.64 <sub>0.11</sub>	0.5708 <sub>0.0086</sub>	22.291 <sub>0.027</sub>	0.860 <sub>0.010</sub>
380	560.63 <sub>0.37</sub>	14.238 <sub>0.067</sub>	0.4591 <sub>0.0081</sub>	23.076 <sub>0.038</sub>	0.879 <sub>0.013</sub>
370	572.47 <sub>0.42</sub>	11.382 <sub>0.058</sub>	0.3644 <sub>0.0077</sub>	23.813 <sub>0.053</sub>	0.897 <sub>0.017</sub>
360	583.49 <sub>0.45</sub>	8.997 <sub>0.074</sub>	0.2850 <sub>0.0074</sub>	24.488 <sub>0.062</sub>	0.912 <sub>0.022</sub>
350	593.61 <sub>0.52</sub>	7.021 <sub>0.099</sub>	0.2196 <sub>0.0070</sub>	25.098 <sub>0.081</sub>	0.926 <sub>0.029</sub>
340	603.19 <sub>0.55</sub>	5.40 <sub>0.12</sub>	0.1662 <sub>0.0067</sub>	25.66 <sub>0.11</sub>	0.939 <sub>0.041</sub>
330	612.67 <sub>0.41</sub>	4.09 <sub>0.14</sub>	0.1236 <sub>0.0065</sub>	26.22 <sub>0.16</sub>	0.950 <sub>0.061</sub>
320	622.10 <sub>0.33</sub>	3.03 <sub>0.13</sub>	0.0899 <sub>0.0065</sub>	26.75 <sub>0.23</sub>	0.959 <sub>0.091</sub>
310	631.64 <sub>0.30</sub>	2.21 <sub>0.12</sub>	0.0640 <sub>0.0068</sub>	27.29 <sub>0.34</sub>	0.97 <sub>0.14</sub>

Table S55: GCMC-MBAR results for 2,2-dimethylhexane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
530	405.4 <sub>1.1</sub>	88.4 <sub>2.4</sub>	1.979 <sub>0.012</sub>	14.84 <sub>0.19</sub>	0.580 <sub>0.013</sub>
520	431.48 <sub>0.93</sub>	72.7 <sub>1.8</sub>	1.7154 <sub>0.0075</sub>	16.99 <sub>0.19</sub>	0.623 <sub>0.013</sub>
510	453.8 <sub>1.0</sub>	59.8 <sub>1.2</sub>	1.4785 <sub>0.0047</sub>	18.93 <sub>0.16</sub>	0.666 <sub>0.012</sub>
500	472.8 <sub>1.0</sub>	49.35 <sub>0.64</sub>	1.2675 <sub>0.0038</sub>	20.62 <sub>0.13</sub>	0.71 <sub>0.01</sub>
490	489.55 <sub>0.82</sub>	40.92 <sub>0.33</sub>	1.0801 <sub>0.0034</sub>	22.090 <sub>0.076</sub>	0.74 <sub>0.01</sub>
480	504.38 <sub>0.54</sub>	33.96 <sub>0.19</sub>	0.9145 <sub>0.0030</sub>	23.386 <sub>0.037</sub>	0.77 <sub>0.01</sub>
470	517.67 <sub>0.43</sub>	28.16 <sub>0.15</sub>	0.7689 <sub>0.0025</sub>	24.539 <sub>0.032</sub>	0.80 <sub>0.01</sub>
460	530.06 <sub>0.45</sub>	23.29 <sub>0.13</sub>	0.6417 <sub>0.0022</sub>	25.596 <sub>0.035</sub>	0.82 <sub>0.01</sub>
450	542.23 <sub>0.55</sub>	19.19 <sub>0.13</sub>	0.5309 <sub>0.0021</sub>	26.601 <sub>0.035</sub>	0.84 <sub>0.01</sub>
440	554.29 <sub>0.63</sub>	15.71 <sub>0.16</sub>	0.4353 <sub>0.0023</sub>	27.566 <sub>0.050</sub>	0.865 <sub>0.012</sub>
430	565.90 <sub>0.55</sub>	12.78 <sub>0.20</sub>	0.3531 <sub>0.0029</sub>	28.479 <sub>0.074</sub>	0.883 <sub>0.020</sub>
420	577.12 <sub>0.44</sub>	10.30 <sub>0.25</sub>	0.2833 <sub>0.0040</sub>	29.35 <sub>0.10</sub>	0.899 <sub>0.033</sub>
410	588.18 <sub>0.45</sub>	8.23 <sub>0.27</sub>	0.2243 <sub>0.0053</sub>	30.19 <sub>0.16</sub>	0.914 <sub>0.050</sub>
400	598.75 <sub>0.43</sub>	6.49 <sub>0.27</sub>	0.1753 <sub>0.0068</sub>	30.99 <sub>0.23</sub>	0.927 <sub>0.073</sub>
390	608.66 <sub>0.49</sub>	5.06 <sub>0.26</sub>	0.1349 <sub>0.0082</sub>	31.74 <sub>0.32</sub>	0.94 <sub>0.10</sub>
380	618.76 <sub>0.62</sub>	3.89 <sub>0.23</sub>	0.1021 <sub>0.0096</sub>	32.48 <sub>0.43</sub>	0.95 <sub>0.14</sub>
370	629.14 <sub>0.47</sub>	2.94 <sub>0.20</sub>	0.076 <sub>0.011</sub>	33.23 <sub>0.60</sub>	0.96 <sub>0.20</sub>

Table S56: GCMC-MBAR results for 2,2-dimethylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
500	411.2 <sub>4.8</sub>	88.4 <sub>3.0</sub>	2.127 <sub>0.025</sub>	13.748 <sub>0.081</sub>	0.580 <sub>0.013</sub>
490	436.8 <sub>4.1</sub>	71.5 <sub>2.2</sub>	1.835 <sub>0.014</sub>	15.833 <sub>0.068</sub>	0.631 <sub>0.015</sub>
480	457.9 <sub>2.5</sub>	58.3 <sub>1.4</sub>	1.5750 <sub>0.0068</sub>	17.601 <sub>0.049</sub>	0.678 <sub>0.014</sub>
470	476.0 <sub>1.4</sub>	48.10 <sub>0.81</sub>	1.3443 <sub>0.0036</sub>	19.097 <sub>0.040</sub>	0.717 <sub>0.012</sub>
460	492.22 <sub>0.92</sub>	39.84 <sub>0.47</sub>	1.1402 <sub>0.0035</sub>	20.399 <sub>0.036</sub>	0.75 <sub>0.01</sub>
450	506.85 <sub>0.87</sub>	33.01 <sub>0.29</sub>	0.9600 <sub>0.0041</sub>	21.553 <sub>0.034</sub>	0.78 <sub>0.01</sub>
440	520.4 <sub>1.0</sub>	27.29 <sub>0.18</sub>	0.8020 <sub>0.0047</sub>	22.600 <sub>0.037</sub>	0.80 <sub>0.01</sub>
430	533.27 <sub>0.96</sub>	22.47 <sub>0.10</sub>	0.6642 <sub>0.0050</sub>	23.572 <sub>0.034</sub>	0.83 <sub>0.01</sub>
420	545.67 <sub>0.39</sub>	18.398 <sub>0.066</sub>	0.5448 <sub>0.0050</sub>	24.481 <sub>0.029</sub>	0.85 <sub>0.01</sub>
410	557.49 <sub>0.37</sub>	14.958 <sub>0.060</sub>	0.4422 <sub>0.0048</sub>	25.331 <sub>0.052</sub>	0.87 <sub>0.01</sub>
400	568.93 <sub>0.50</sub>	12.058 <sub>0.076</sub>	0.3547 <sub>0.0047</sub>	26.137 <sub>0.064</sub>	0.886 <sub>0.013</sub>
390	580.08 <sub>0.47</sub>	9.622 <sub>0.094</sub>	0.2810 <sub>0.0046</sub>	26.908 <sub>0.069</sub>	0.902 <sub>0.018</sub>
380	590.67 <sub>0.42</sub>	7.59 <sub>0.11</sub>	0.2195 <sub>0.0045</sub>	27.631 <sub>0.075</sub>	0.917 <sub>0.023</sub>
370	601.07 <sub>0.39</sub>	5.91 <sub>0.14</sub>	0.1689 <sub>0.0043</sub>	28.333 <sub>0.091</sub>	0.931 <sub>0.031</sub>
360	611.44 <sub>0.41</sub>	4.53 <sub>0.18</sub>	0.1279 <sub>0.0041</sub>	29.02 <sub>0.13</sub>	0.945 <sub>0.045</sub>
350	620.82 <sub>0.40</sub>	3.42 <sub>0.21</sub>	0.0950 <sub>0.0040</sub>	29.64 <sub>0.19</sub>	0.956 <sub>0.071</sub>

Table S57: GCMC-MBAR results for 2,2-dimethylpropane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_{\text{v}}$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
410	409.5 <sub>2.6</sub>	78.8 <sub>2.6</sub>	2.218 <sub>0.029</sub>	11.33 <sub>0.11</sub>	0.596 <sub>0.013</sub>
400	433.9 <sub>2.3</sub>	61.3 <sub>1.8</sub>	1.869 <sub>0.018</sub>	13.140 <sub>0.083</sub>	0.661 <sub>0.014</sub>
390	454.8 <sub>1.5</sub>	48.8 <sub>1.1</sub>	1.565 <sub>0.010</sub>	14.591 <sub>0.051</sub>	0.713 <sub>0.013</sub>
380	472.4 <sub>1.1</sub>	39.38 <sub>0.72</sub>	1.3004 <sub>0.0056</sub>	15.750 <sub>0.045</sub>	0.754 <sub>0.011</sub>
370	487.88 <sub>0.98</sub>	31.87 <sub>0.48</sub>	1.0704 <sub>0.0035</sub>	16.732 <sub>0.045</sub>	0.788 <sub>0.011</sub>
360	502.34 <sub>0.89</sub>	25.71 <sub>0.33</sub>	0.8720 <sub>0.0036</sub>	17.617 <sub>0.044</sub>	0.817 <sub>0.012</sub>
350	516.37 <sub>0.86</sub>	20.61 <sub>0.21</sub>	0.7019 <sub>0.0043</sub>	18.443 <sub>0.046</sub>	0.844 <sub>0.011</sub>
340	529.79 <sub>0.93</sub>	16.38 <sub>0.18</sub>	0.5574 <sub>0.0045</sub>	19.210 <sub>0.057</sub>	0.868 <sub>0.011</sub>
330	542.27 <sub>0.96</sub>	12.88 <sub>0.23</sub>	0.4363 <sub>0.0041</sub>	19.910 <sub>0.078</sub>	0.891 <sub>0.016</sub>
320	553.7 <sub>1.0</sub>	10.00 <sub>0.28</sub>	0.3360 <sub>0.0037</sub>	20.54 <sub>0.11</sub>	0.911 <sub>0.029</sub>
310	564.4 <sub>1.4</sub>	7.66 <sub>0.32</sub>	0.2543 <sub>0.0044</sub>	21.12 <sub>0.18</sub>	0.930 <sub>0.052</sub>
300	575.3 <sub>1.8</sub>	5.77 <sub>0.33</sub>	0.1887 <sub>0.0061</sub>	21.68 <sub>0.28</sub>	0.946 <sub>0.087</sub>
290	587.1 <sub>1.3</sub>	4.27 <sub>0.32</sub>	0.1368 <sub>0.0084</sub>	22.27 <sub>0.38</sub>	0.96 <sub>0.14</sub>
280	599.1 <sub>1.1</sub>	3.08 <sub>0.29</sub>	0.096 <sub>0.011</sub>	22.85 <sub>0.55</sub>	0.97 <sub>0.22</sub>

Table S58: GCMC-MBAR results for 3,3-dimethylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
510	433.8 <sub>1.4</sub>	83.3 <sub>2.1</sub>	2.128 <sub>0.013</sub>	14.92 <sub>0.15</sub>	0.604 <sub>0.012</sub>
500	456.2 <sub>1.8</sub>	68.2 <sub>1.6</sub>	1.8431 <sub>0.0076</sub>	16.79 <sub>0.16</sub>	0.651 <sub>0.013</sub>
490	475.3 <sub>1.5</sub>	56.40 <sub>0.99</sub>	1.5893 <sub>0.0041</sub>	18.38 <sub>0.12</sub>	0.693 <sub>0.012</sub>
480	492.10 <sub>0.90</sub>	47.00 <sub>0.51</sub>	1.3630 <sub>0.0031</sub>	19.752 <sub>0.070</sub>	0.73 <sub>0.01</sub>
470	507.53 <sub>0.52</sub>	39.28 <sub>0.29</sub>	1.1617 <sub>0.0029</sub>	20.968 <sub>0.041</sub>	0.76 <sub>0.01</sub>
460	521.91 <sub>0.42</sub>	32.81 <sub>0.22</sub>	0.9831 <sub>0.0027</sub>	22.072 <sub>0.042</sub>	0.78 <sub>0.01</sub>
450	535.23 <sub>0.39</sub>	27.34 <sub>0.16</sub>	0.8258 <sub>0.0025</sub>	23.077 <sub>0.038</sub>	0.81 <sub>0.01</sub>
440	547.46 <sub>0.40</sub>	22.676 <sub>0.096</sub>	0.6879 <sub>0.0023</sub>	23.990 <sub>0.031</sub>	0.83 <sub>0.01</sub>
430	559.01 <sub>0.43</sub>	18.706 <sub>0.065</sub>	0.5678 <sub>0.0022</sub>	24.839 <sub>0.027</sub>	0.85 <sub>0.01</sub>
420	570.36 <sub>0.53</sub>	15.325 <sub>0.054</sub>	0.4642 <sub>0.0020</sub>	25.651 <sub>0.032</sub>	0.87 <sub>0.01</sub>
410	581.43 <sub>0.85</sub>	12.456 <sub>0.067</sub>	0.3754 <sub>0.0020</sub>	26.425 <sub>0.058</sub>	0.89 <sub>0.01</sub>
400	591.91 <sub>0.96</sub>	10.03 <sub>0.10</sub>	0.3000 <sub>0.0021</sub>	27.146 <sub>0.083</sub>	0.901 <sub>0.013</sub>
390	601.87 <sub>0.71</sub>	8.00 <sub>0.14</sub>	0.2368 <sub>0.0024</sub>	27.82 <sub>0.10</sub>	0.915 <sub>0.024</sub>
380	611.41 <sub>0.54</sub>	6.30 <sub>0.17</sub>	0.1842 <sub>0.0031</sub>	28.46 <sub>0.14</sub>	0.927 <sub>0.040</sub>
370	620.56 <sub>0.38</sub>	4.90 <sub>0.19</sub>	0.1411 <sub>0.0040</sub>	29.06 <sub>0.21</sub>	0.938 <sub>0.066</sub>
360	629.62 <sub>0.19</sub>	3.76 <sub>0.19</sub>	0.1063 <sub>0.0052</sub>	29.64 <sub>0.30</sub>	0.95 <sub>0.10</sub>

Table S59: GCMC-MBAR results for 2,2,3-trimethylbutane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
520	427.6 <sub>2.7</sub>	90.4 <sub>3.4</sub>	2.310 <sub>0.041</sub>	14.20 <sub>0.20</sub>	0.592 <sub>0.014</sub>
510	451.1 <sub>1.5</sub>	73.9 <sub>3.6</sub>	2.014 <sub>0.030</sub>	16.22 <sub>0.27</sub>	0.644 <sub>0.025</sub>
500	471.00 <sub>0.92</sub>	61.5 <sub>2.9</sub>	1.748 <sub>0.020</sub>	17.84 <sub>0.29</sub>	0.684 <sub>0.029</sub>
490	488.37 <sub>0.82</sub>	51.6 <sub>1.9</sub>	1.509 <sub>0.014</sub>	19.22 <sub>0.23</sub>	0.719 <sub>0.023</sub>
480	503.84 <sub>0.76</sub>	43.39 <sub>0.99</sub>	1.295 <sub>0.011</sub>	20.44 <sub>0.15</sub>	0.749 <sub>0.015</sub>
470	518.11 <sub>0.61</sub>	36.42 <sub>0.54</sub>	1.1046 <sub>0.0087</sub>	21.544 <sub>0.083</sub>	0.78 <sub>0.01</sub>
460	531.60 <sub>0.50</sub>	30.51 <sub>0.45</sub>	0.9358 <sub>0.0068</sub>	22.566 <sub>0.064</sub>	0.80 <sub>0.01</sub>
450	544.35 <sub>0.55</sub>	25.49 <sub>0.41</sub>	0.7870 <sub>0.0049</sub>	23.512 <sub>0.070</sub>	0.83 <sub>0.01</sub>
440	556.68 <sub>0.54</sub>	21.21 <sub>0.32</sub>	0.6564 <sub>0.0032</sub>	24.402 <sub>0.069</sub>	0.85 <sub>0.01</sub>
430	568.85 <sub>0.50</sub>	17.55 <sub>0.23</sub>	0.5424 <sub>0.0020</sub>	25.255 <sub>0.062</sub>	0.87 <sub>0.01</sub>
420	580.37 <sub>0.55</sub>	14.42 <sub>0.15</sub>	0.4436 <sub>0.0014</sub>	26.050 <sub>0.058</sub>	0.88 <sub>0.01</sub>
410	590.79 <sub>0.86</sub>	11.748 <sub>0.088</sub>	0.3590 <sub>0.0014</sub>	26.769 <sub>0.064</sub>	0.90 <sub>0.01</sub>
400	600.6 <sub>1.0</sub>	9.476 <sub>0.069</sub>	0.2870 <sub>0.0014</sub>	27.440 <sub>0.077</sub>	0.91 <sub>0.01</sub>
390	610.62 <sub>0.83</sub>	7.560 <sub>0.096</sub>	0.2267 <sub>0.0013</sub>	28.107 <sub>0.086</sub>	0.927 <sub>0.013</sub>
380	621.8 <sub>1.5</sub>	5.96 <sub>0.14</sub>	0.1765 <sub>0.0014</sub>	28.82 <sub>0.16</sub>	0.940 <sub>0.025</sub>
370	633.5 <sub>2.1</sub>	4.63 <sub>0.17</sub>	0.1353 <sub>0.0019</sub>	29.56 <sub>0.26</sub>	0.952 <sub>0.044</sub>
360	643.07 <sub>0.71</sub>	3.54 <sub>0.20</sub>	0.1019 <sub>0.0029</sub>	30.16 <sub>0.26</sub>	0.963 <sub>0.071</sub>

Table S60: GCMC-MBAR results for 2,2,3-trimethylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
550	424.5 <sub>1.7</sub>	94.6 <sub>5.8</sub>	2.204 <sub>0.085</sub>	14.90 <sub>0.26</sub>	0.582 <sub>0.014</sub>
540	446.63 <sub>0.90</sub>	77.5 <sub>4.4</sub>	1.931 <sub>0.070</sub>	17.04 <sub>0.27</sub>	0.634 <sub>0.015</sub>
530	466.95 <sub>0.61</sub>	64.6 <sub>2.9</sub>	1.685 <sub>0.058</sub>	18.86 <sub>0.22</sub>	0.676 <sub>0.010</sub>
520	485.81 <sub>0.76</sub>	54.4 <sub>1.9</sub>	1.463 <sub>0.050</sub>	20.45 <sub>0.13</sub>	0.71 <sub>0.01</sub>
510	502.62 <sub>0.63</sub>	45.9 <sub>1.6</sub>	1.263 <sub>0.044</sub>	21.840 <sub>0.082</sub>	0.74 <sub>0.01</sub>
500	517.03 <sub>0.59</sub>	38.7 <sub>1.7</sub>	1.084 <sub>0.037</sub>	23.05 <sub>0.11</sub>	0.77 <sub>0.01</sub>
490	529.98 <sub>0.76</sub>	32.6 <sub>1.9</sub>	0.925 <sub>0.029</sub>	24.14 <sub>0.18</sub>	0.795 <sub>0.024</sub>
480	542.7 <sub>1.0</sub>	27.4 <sub>1.8</sub>	0.784 <sub>0.021</sub>	25.18 <sub>0.25</sub>	0.818 <sub>0.037</sub>
470	555.2 <sub>1.2</sub>	23.0 <sub>1.6</sub>	0.659 <sub>0.013</sub>	26.17 <sub>0.29</sub>	0.840 <sub>0.045</sub>
460	566.7 <sub>1.3</sub>	19.1 <sub>1.2</sub>	0.5500 <sub>0.0068</sub>	27.09 <sub>0.30</sub>	0.860 <sub>0.047</sub>
450	577.6 <sub>1.2</sub>	15.82 <sub>0.81</sub>	0.4550 <sub>0.0027</sub>	27.94 <sub>0.29</sub>	0.878 <sub>0.045</sub>
440	588.5 <sub>3.8</sub>	13.0 <sub>1.5</sub>	0.4 <sub>1.3</sub>	28.8 <sub>7.3</sub>	0.9 <sub>2.0</sub>
430	599.6 <sub>2.4</sub>	10.61 <sub>0.31</sub>	0.3023 <sub>0.0035</sub>	29.58 <sub>0.30</sub>	0.910 <sub>0.038</sub>
420	610.1 <sub>2.1</sub>	8.58 <sub>0.19</sub>	0.2423 <sub>0.0044</sub>	30.34 <sub>0.26</sub>	0.923 <sub>0.036</sub>
410	619.5 <sub>1.4</sub>	6.88 <sub>0.12</sub>	0.1918 <sub>0.0049</sub>	31.02 <sub>0.22</sub>	0.935 <sub>0.038</sub>
400	628.4 <sub>1.5</sub>	5.447 <sub>0.098</sub>	0.1498 <sub>0.0051</sub>	31.66 <sub>0.24</sub>	0.945 <sub>0.044</sub>
380	645.33 <sub>0.68</sub>	3.285 <sub>0.097</sub>	0.0874 <sub>0.0051</sub>	32.86 <sub>0.25</sub>	0.961 <sub>0.067</sub>



Table S61: GCMC-MBAR results for 2,2,4-trimethylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
530	400.9 <sub>2.2</sub>	97.9 <sub>4.8</sub>	2.120 <sub>0.038</sub>	13.58 <sub>0.32</sub>	0.561 <sub>0.019</sub>
520	427.5 <sub>1.3</sub>	79.4 <sub>3.2</sub>	1.844 <sub>0.028</sub>	15.89 <sub>0.27</sub>	0.614 <sub>0.018</sub>
510	448.78 <sub>0.77</sub>	65.1 <sub>1.8</sub>	1.597 <sub>0.021</sub>	17.79 <sub>0.18</sub>	0.661 <sub>0.012</sub>
500	466.83 <sub>0.75</sub>	53.9 <sub>1.1</sub>	1.378 <sub>0.018</sub>	19.40 <sub>0.13</sub>	0.70 <sub>0.01</sub>
490	483.78 <sub>0.81</sub>	44.87 <sub>0.89</sub>	1.182 <sub>0.015</sub>	20.85 <sub>0.12</sub>	0.74 <sub>0.01</sub>
480	499.84 <sub>0.70</sub>	37.41 <sub>0.89</sub>	1.009 <sub>0.012</sub>	22.18 <sub>0.12</sub>	0.772 <sub>0.012</sub>
470	514.41 <sub>0.57</sub>	31.21 <sub>0.86</sub>	0.8545 <sub>0.0083</sub>	23.38 <sub>0.13</sub>	0.800 <sub>0.016</sub>
460	527.62 <sub>0.53</sub>	26.00 <sub>0.75</sub>	0.7187 <sub>0.0053</sub>	24.45 <sub>0.13</sub>	0.826 <sub>0.020</sub>
450	539.75 <sub>0.68</sub>	21.58 <sub>0.59</sub>	0.5995 <sub>0.0030</sub>	25.42 <sub>0.12</sub>	0.848 <sub>0.022</sub>
440	551.12 <sub>0.66</sub>	17.82 <sub>0.43</sub>	0.4958 <sub>0.0022</sub>	26.32 <sub>0.12</sub>	0.869 <sub>0.022</sub>
430	562.67 <sub>0.50</sub>	14.62 <sub>0.31</sub>	0.4062 <sub>0.0027</sub>	27.21 <sub>0.13</sub>	0.888 <sub>0.023</sub>
420	574.59 <sub>0.49</sub>	11.89 <sub>0.25</sub>	0.3292 <sub>0.0035</sub>	28.09 <sub>0.13</sub>	0.906 <sub>0.027</sub>
410	585.04 <sub>0.41</sub>	9.58 <sub>0.25</sub>	0.2637 <sub>0.0042</sub>	28.86 <sub>0.15</sub>	0.922 <sub>0.037</sub>
400	594.23 <sub>0.30</sub>	7.66 <sub>0.27</sub>	0.2087 <sub>0.0051</sub>	29.54 <sub>0.20</sub>	0.936 <sub>0.055</sub>
390	604.33 <sub>0.26</sub>	6.05 <sub>0.29</sub>	0.1630 <sub>0.0063</sub>	30.26 <sub>0.28</sub>	0.949 <sub>0.082</sub>
380	615.43 <sub>0.23</sub>	4.72 <sub>0.30</sub>	0.1252 <sub>0.0077</sub>	31.01 <sub>0.41</sub>	0.96 <sub>0.12</sub>
370	625.27 <sub>0.23</sub>	3.63 <sub>0.29</sub>	0.0946 <sub>0.0092</sub>	31.68 <sub>0.58</sub>	0.97 <sub>0.18</sub>

Table S62: GCMC-MBAR results for 2,3,3-trimethylpentane with the MiPPE-SL force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
560	426.5 <sub>9.5</sub>	98.7 <sub>1.6</sub>	2.304 <sub>0.024</sub>	14.76 <sub>0.51</sub>	0.57 <sub>0.01</sub>
550	452.4 <sub>5.9</sub>	80.7 <sub>1.4</sub>	2.023 <sub>0.020</sub>	17.10 <sub>0.29</sub>	0.63 <sub>0.01</sub>
540	473.3 <sub>2.0</sub>	67.2 <sub>1.1</sub>	1.769 <sub>0.018</sub>	18.98 <sub>0.12</sub>	0.67 <sub>0.01</sub>
530	490.60 <sub>0.59</sub>	56.53 <sub>0.74</sub>	1.540 <sub>0.016</sub>	20.523 <sub>0.065</sub>	0.71 <sub>0.01</sub>
520	505.89 <sub>0.92</sub>	47.80 <sub>0.63</sub>	1.335 <sub>0.014</sub>	21.863 <sub>0.043</sub>	0.74 <sub>0.01</sub>
510	520.2 <sub>1.1</sub>	40.46 <sub>0.65</sub>	1.151 <sub>0.012</sub>	23.086 <sub>0.038</sub>	0.77 <sub>0.01</sub>
500	534.1 <sub>1.0</sub>	34.21 <sub>0.66</sub>	0.9866 <sub>0.0088</sub>	24.232 <sub>0.057</sub>	0.79 <sub>0.01</sub>
490	546.97 <sub>0.93</sub>	28.87 <sub>0.60</sub>	0.8402 <sub>0.0063</sub>	25.283 <sub>0.071</sub>	0.816 <sub>0.012</sub>
480	558.82 <sub>0.80</sub>	24.29 <sub>0.47</sub>	0.7108 <sub>0.0044</sub>	26.241 <sub>0.071</sub>	0.837 <sub>0.013</sub>
470	570.25 <sub>0.87</sub>	20.36 <sub>0.30</sub>	0.5967 <sub>0.0036</sub>	27.143 <sub>0.070</sub>	0.857 <sub>0.013</sub>
460	581.5 <sub>1.1</sub>	16.96 <sub>0.17</sub>	0.4969 <sub>0.0037</sub>	28.007 <sub>0.066</sub>	0.875 <sub>0.013</sub>
450	592.0 <sub>1.0</sub>	14.04 <sub>0.13</sub>	0.4101 <sub>0.0041</sub>	28.816 <sub>0.071</sub>	0.892 <sub>0.016</sub>
440	602.0 <sub>1.1</sub>	11.53 <sub>0.15</sub>	0.3353 <sub>0.0046</sub>	29.578 <sub>0.085</sub>	0.908 <sub>0.022</sub>
430	612.3 <sub>1.6</sub>	9.39 <sub>0.16</sub>	0.2714 <sub>0.0052</sub>	30.34 <sub>0.10</sub>	0.924 <sub>0.031</sub>
420	622.8 <sub>1.6</sub>	7.58 <sub>0.16</sub>	0.2171 <sub>0.0058</sub>	31.10 <sub>0.13</sub>	0.937 <sub>0.042</sub>
410	632.87 <sub>0.75</sub>	6.05 <sub>0.16</sub>	0.1716 <sub>0.0064</sub>	31.82 <sub>0.20</sub>	0.950 <sub>0.057</sub>
400	642.39 <sub>0.29</sub>	4.78 <sub>0.17</sub>	0.1338 <sub>0.0070</sub>	32.50 <sub>0.28</sub>	0.961 <sub>0.077</sub>
390	651.59 <sub>0.31</sub>	3.73 <sub>0.18</sub>	0.1028 <sub>0.0075</sub>	33.14 <sub>0.37</sub>	0.97 <sub>0.11</sub>

## S8.2 Alkynes

Table S63: GCMC-MBAR results for ethyne with the MiPPE force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
290	421.3 <sub>3.5</sub>	69.7 <sub>3.3</sub>	4.083 <sub>0.020</sub>	8.80 <sub>0.21</sub>	0.632 <sub>0.033</sub>
280	450.7 <sub>1.9</sub>	50.69 <sub>0.91</sub>	3.196 <sub>0.026</sub>	10.27 <sub>0.12</sub>	0.705 <sub>0.017</sub>
270	474.9 <sub>3.0</sub>	37.66 <sub>0.39</sub>	2.465 <sub>0.024</sub>	11.390 <sub>0.086</sub>	0.76 <sub>0.01</sub>
260	497.3 <sub>1.1</sub>	27.95 <sub>0.53</sub>	1.865 <sub>0.017</sub>	12.361 <sub>0.034</sub>	0.804 <sub>0.011</sub>
250	517.91 <sub>0.59</sub>	20.53 <sub>0.34</sub>	1.378 <sub>0.012</sub>	13.214 <sub>0.042</sub>	0.841 <sub>0.013</sub>
240	536.87 <sub>0.67</sub>	14.82 <sub>0.12</sub>	0.992 <sub>0.010</sub>	13.975 <sub>0.028</sub>	0.87 <sub>0.01</sub>
230	554.12 <sub>0.54</sub>	10.447 <sub>0.095</sub>	0.6925 <sub>0.0089</sub>	14.649 <sub>0.037</sub>	0.903 <sub>0.012</sub>
220	570.90 <sub>0.53</sub>	7.155 <sub>0.097</sub>	0.4667 <sub>0.0082</sub>	15.283 <sub>0.049</sub>	0.929 <sub>0.021</sub>

Table S64: GCMC-MBAR results for propyne with the MiPPE force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
380	441.0 <sub>7.9</sub>	82.2 <sub>3.1</sub>	3.839 <sub>0.090</sub>	10.96 <sub>0.34</sub>	0.59 <sub>0.01</sub>
370	472.7 <sub>5.0</sub>	62.7 <sub>2.7</sub>	3.164 <sub>0.073</sub>	12.83 <sub>0.31</sub>	0.657 <sub>0.015</sub>
360	498.3 <sub>2.8</sub>	48.6 <sub>1.9</sub>	2.584 <sub>0.057</sub>	14.34 <sub>0.24</sub>	0.711 <sub>0.015</sub>
350	520.1 <sub>2.4</sub>	38.1 <sub>1.3</sub>	2.088 <sub>0.045</sub>	15.58 <sub>0.19</sub>	0.754 <sub>0.011</sub>
340	539.4 <sub>2.1</sub>	29.88 <sub>0.90</sub>	1.667 <sub>0.035</sub>	16.65 <sub>0.15</sub>	0.79 <sub>0.01</sub>
330	556.6 <sub>1.4</sub>	23.31 <sub>0.72</sub>	1.313 <sub>0.027</sub>	17.58 <sub>0.11</sub>	0.822 <sub>0.010</sub>
320	572.1 <sub>1.3</sub>	18.02 <sub>0.60</sub>	1.018 <sub>0.019</sub>	18.41 <sub>0.11</sub>	0.850 <sub>0.014</sub>
310	587.6 <sub>1.3</sub>	13.76 <sub>0.47</sub>	0.776 <sub>0.012</sub>	19.20 <sub>0.12</sub>	0.876 <sub>0.017</sub>
300	603.3 <sub>1.5</sub>	10.35 <sub>0.32</sub>	0.5799 <sub>0.0068</sub>	19.97 <sub>0.13</sub>	0.900 <sub>0.019</sub>
290	617.9 <sub>2.2</sub>	7.65 <sub>0.20</sub>	0.4240 <sub>0.0040</sub>	20.67 <sub>0.15</sub>	0.921 <sub>0.020</sub>

Table S65: GCMC-MBAR results for 1-butyne with the MiPPE force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
410	445.1 <sub>5.3</sub>	76 <sub>12</sub>	2.95 <sub>0.10</sub>	12.82 <sub>0.72</sub>	0.620 <sub>0.056</sub>
400	472.3 <sub>3.4</sub>	59.2 <sub>6.8</sub>	2.464 <sub>0.050</sub>	14.61 <sub>0.60</sub>	0.677 <sub>0.054</sub>
390	495.0 <sub>2.6</sub>	47.1 <sub>2.3</sub>	2.038 <sub>0.030</sub>	16.08 <sub>0.32</sub>	0.722 <sub>0.027</sub>
380	515.5 <sub>2.0</sub>	37.54 <sub>0.86</sub>	1.669 <sub>0.023</sub>	17.37 <sub>0.16</sub>	0.76 <sub>0.01</sub>
370	534.5 <sub>1.3</sub>	29.90 <sub>0.70</sub>	1.351 <sub>0.018</sub>	18.53 <sub>0.13</sub>	0.79 <sub>0.01</sub>
360	552.0 <sub>1.6</sub>	23.69 <sub>0.62</sub>	1.081 <sub>0.013</sub>	19.57 <sub>0.16</sub>	0.824 <sub>0.013</sub>
350	567.8 <sub>1.9</sub>	18.62 <sub>0.51</sub>	0.8524 <sub>0.0086</sub>	20.50 <sub>0.18</sub>	0.851 <sub>0.017</sub>
340	582.2 <sub>1.1</sub>	14.49 <sub>0.38</sub>	0.6623 <sub>0.0060</sub>	21.34 <sub>0.13</sub>	0.874 <sub>0.019</sub>
330	595.8 <sub>1.1</sub>	11.15 <sub>0.24</sub>	0.5061 <sub>0.0049</sub>	22.112 <sub>0.082</sub>	0.895 <sub>0.019</sub>
320	610.6 <sub>2.8</sub>	8.45 <sub>0.14</sub>	0.3795 <sub>0.0049</sub>	22.905 <sub>0.095</sub>	0.913 <sub>0.019</sub>
310	624.6 <sub>1.8</sub>	6.284 <sub>0.087</sub>	0.2782 <sub>0.0051</sub>	23.650 <sub>0.059</sub>	0.929 <sub>0.022</sub>

Table S66: GCMC-MBAR results for 2-butyne with the MiPPE force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
450	431.8 <sub>7.4</sub>	93.5 <sub>4.3</sub>	3.624 <sub>0.050</sub>	11.93 <sub>0.26</sub>	0.561 <sub>0.018</sub>
440	466.0 <sub>5.5</sub>	74.1 <sub>3.5</sub>	3.073 <sub>0.028</sub>	14.00 <sub>0.24</sub>	0.613 <sub>0.024</sub>
430	491.6 <sub>2.0</sub>	59.1 <sub>1.9</sub>	2.587 <sub>0.016</sub>	15.71 <sub>0.17</sub>	0.662 <sub>0.020</sub>
420	512.10 <sub>0.77</sub>	47.58 <sub>0.69</sub>	2.163 <sub>0.014</sub>	17.114 <sub>0.094</sub>	0.704 <sub>0.010</sub>
410	530.5 <sub>1.1</sub>	38.42 <sub>0.30</sub>	1.795 <sub>0.013</sub>	18.346 <sub>0.054</sub>	0.74 <sub>0.01</sub>
400	548.28 <sub>0.94</sub>	30.99 <sub>0.31</sub>	1.476 <sub>0.011</sub>	19.487 <sub>0.051</sub>	0.77 <sub>0.01</sub>
390	565.05 <sub>0.94</sub>	24.89 <sub>0.27</sub>	1.2013 <sub>0.0089</sub>	20.533 <sub>0.061</sub>	0.81 <sub>0.01</sub>
380	580.6 <sub>1.1</sub>	19.89 <sub>0.25</sub>	0.9674 <sub>0.0068</sub>	21.483 <sub>0.073</sub>	0.83 <sub>0.01</sub>
370	594.97 <sub>0.83</sub>	15.78 <sub>0.25</sub>	0.7697 <sub>0.0050</sub>	22.349 <sub>0.074</sub>	0.858 <sub>0.011</sub>
360	608.65 <sub>0.59</sub>	12.41 <sub>0.22</sub>	0.6041 <sub>0.0038</sub>	23.157 <sub>0.070</sub>	0.880 <sub>0.017</sub>
350	621.69 <sub>0.66</sub>	9.66 <sub>0.19</sub>	0.4672 <sub>0.0041</sub>	23.912 <sub>0.091</sub>	0.899 <sub>0.024</sub>
340	633.85 <sub>0.59</sub>	7.42 <sub>0.16</sub>	0.3552 <sub>0.0052</sub>	24.60 <sub>0.12</sub>	0.916 <sub>0.033</sub>
330	646.13 <sub>0.73</sub>	5.62 <sub>0.13</sub>	0.2651 <sub>0.0064</sub>	25.26 <sub>0.16</sub>	0.930 <sub>0.044</sub>

Table S67: GCMC-MBAR results for 1-pentyne with the MiPPE force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
450	433.7 <sub>2.3</sub>	85.6 <sub>2.5</sub>	2.732 <sub>0.022</sub>	13.15 <sub>0.18</sub>	0.581 <sub>0.014</sub>
440	461.7 <sub>1.7</sub>	66.9 <sub>2.0</sub>	2.304 <sub>0.015</sub>	15.29 <sub>0.18</sub>	0.641 <sub>0.018</sub>
430	485.2 <sub>1.5</sub>	53.1 <sub>1.2</sub>	1.932 <sub>0.012</sub>	17.06 <sub>0.16</sub>	0.693 <sub>0.016</sub>
420	505.4 <sub>1.6</sub>	42.65 <sub>0.51</sub>	1.608 <sub>0.012</sub>	18.54 <sub>0.11</sub>	0.735 <sub>0.011</sub>
410	523.3 <sub>1.4</sub>	34.37 <sub>0.30</sub>	1.327 <sub>0.010</sub>	19.810 <sub>0.078</sub>	0.77 <sub>0.01</sub>
400	539.7 <sub>1.2</sub>	27.67 <sub>0.31</sub>	1.0844 <sub>0.0077</sub>	20.949 <sub>0.055</sub>	0.80 <sub>0.01</sub>
390	555.3 <sub>1.1</sub>	22.17 <sub>0.30</sub>	0.8770 <sub>0.0052</sub>	21.998 <sub>0.041</sub>	0.83 <sub>0.01</sub>
380	570.2 <sub>1.0</sub>	17.64 <sub>0.27</sub>	0.7009 <sub>0.0033</sub>	22.977 <sub>0.033</sub>	0.857 <sub>0.012</sub>
370	584.03 <sub>0.90</sub>	13.91 <sub>0.25</sub>	0.5528 <sub>0.0028</sub>	23.874 <sub>0.050</sub>	0.880 <sub>0.018</sub>
360	596.86 <sub>0.83</sub>	10.86 <sub>0.22</sub>	0.4300 <sub>0.0040</sub>	24.697 <sub>0.083</sub>	0.901 <sub>0.026</sub>
350	609.71 <sub>0.69</sub>	8.37 <sub>0.19</sub>	0.3291 <sub>0.0055</sub>	25.49 <sub>0.11</sub>	0.920 <sub>0.037</sub>
340	622.58 <sub>0.99</sub>	6.37 <sub>0.15</sub>	0.2474 <sub>0.0070</sub>	26.27 <sub>0.13</sub>	0.936 <sub>0.050</sub>

Table S68: GCMC-MBAR results for 2-pentyne with the MiPPE force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
470	445.8 <sub>3.1</sub>	83.4 <sub>6.2</sub>	2.77 <sub>0.12</sub>	14.09 <sub>0.22</sub>	0.578 <sub>0.020</sub>
460	473.8 <sub>1.9</sub>	65.6 <sub>5.4</sub>	2.343 <sub>0.088</sub>	16.27 <sub>0.30</sub>	0.636 <sub>0.028</sub>
450	496.8 <sub>1.0</sub>	52.5 <sub>3.7</sub>	1.973 <sub>0.064</sub>	18.06 <sub>0.28</sub>	0.685 <sub>0.026</sub>
440	516.64 <sub>0.73</sub>	42.4 <sub>2.3</sub>	1.650 <sub>0.048</sub>	19.56 <sub>0.22</sub>	0.725 <sub>0.020</sub>
430	534.83 <sub>0.74</sub>	34.3 <sub>1.7</sub>	1.369 <sub>0.036</sub>	20.89 <sub>0.19</sub>	0.761 <sub>0.017</sub>
420	551.57 <sub>0.81</sub>	27.7 <sub>1.4</sub>	1.126 <sub>0.025</sub>	22.08 <sub>0.19</sub>	0.792 <sub>0.022</sub>
410	566.89 <sub>0.86</sub>	22.3 <sub>1.2</sub>	0.917 <sub>0.016</sub>	23.16 <sub>0.22</sub>	0.821 <sub>0.030</sub>
400	581.3 <sub>1.1</sub>	17.9 <sub>1.0</sub>	0.7394 <sub>0.0080</sub>	24.15 <sub>0.26</sub>	0.847 <sub>0.038</sub>
390	595.3 <sub>1.6</sub>	14.24 <sub>0.71</sub>	0.5890 <sub>0.0039</sub>	25.09 <sub>0.28</sub>	0.869 <sub>0.040</sub>
380	608.6 <sub>1.7</sub>	11.23 <sub>0.40</sub>	0.4631 <sub>0.0052</sub>	25.95 <sub>0.25</sub>	0.889 <sub>0.038</sub>
370	621.0 <sub>1.3</sub>	8.76 <sub>0.19</sub>	0.3587 <sub>0.0066</sub>	26.75 <sub>0.19</sub>	0.907 <sub>0.032</sub>
360	632.99 <sub>0.96</sub>	6.75 <sub>0.16</sub>	0.2735 <sub>0.0069</sub>	27.52 <sub>0.12</sub>	0.922 <sub>0.031</sub>
350	644.7 <sub>1.0</sub>	5.12 <sub>0.20</sub>	0.2049 <sub>0.0065</sub>	28.249 <sub>0.10</sub>	0.937 <sub>0.044</sub>

Table S69: GCMC-MBAR results for 1-hexyne with the MiPPE force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
490	423.3 <sub>3.5</sub>	87.3 <sub>2.1</sub>	2.538 <sub>0.068</sub>	14.25 <sub>0.14</sub>	0.586 <sub>0.010</sub>
480	453.3 <sub>2.0</sub>	69.9 <sub>1.5</sub>	2.170 <sub>0.060</sub>	16.54 <sub>0.12</sub>	0.639 <sub>0.015</sub>
470	477.4 <sub>1.3</sub>	56.51 <sub>0.96</sub>	1.844 <sub>0.056</sub>	18.448 <sub>0.089</sub>	0.686 <sub>0.016</sub>
460	497.3 <sub>1.4</sub>	46.06 <sub>0.95</sub>	1.556 <sub>0.051</sub>	20.041 <sub>0.074</sub>	0.726 <sub>0.011</sub>
450	514.8 <sub>1.3</sub>	37.6 <sub>1.3</sub>	1.304 <sub>0.044</sub>	21.428 <sub>0.10</sub>	0.76 <sub>0.01</sub>
440	531.5 <sub>1.2</sub>	30.7 <sub>1.7</sub>	1.084 <sub>0.035</sub>	22.71 <sub>0.16</sub>	0.792 <sub>0.017</sub>
430	547.4 <sub>1.2</sub>	25.0 <sub>1.7</sub>	0.893 <sub>0.024</sub>	23.91 <sub>0.22</sub>	0.820 <sub>0.032</sub>
420	561.77 <sub>0.87</sub>	20.3 <sub>1.5</sub>	0.728 <sub>0.013</sub>	24.98 <sub>0.23</sub>	0.845 <sub>0.042</sub>
410	575.17 <sub>0.62</sub>	16.34 <sub>0.99</sub>	0.5875 <sub>0.0050</sub>	25.97 <sub>0.20</sub>	0.867 <sub>0.043</sub>
400	588.08 <sub>0.75</sub>	13.04 <sub>0.55</sub>	0.4682 <sub>0.0027</sub>	26.90 <sub>0.15</sub>	0.887 <sub>0.038</sub>
390	599.80 <sub>0.63</sub>	10.31 <sub>0.28</sub>	0.3684 <sub>0.0047</sub>	27.75 <sub>0.11</sub>	0.905 <sub>0.033</sub>
380	610.32 <sub>0.79</sub>	8.06 <sub>0.17</sub>	0.2858 <sub>0.0061</sub>	28.507 <sub>0.080</sub>	0.922 <sub>0.036</sub>
370	621.3 <sub>2.9</sub>	6.23 <sub>0.13</sub>	0.2185 <sub>0.0072</sub>	29.267 <sub>0.086</sub>	0.937 <sub>0.048</sub>
360	634.3 <sub>3.1</sub>	4.74 <sub>0.11</sub>	0.1640 <sub>0.0083</sub>	30.118 <sub>0.078</sub>	0.950 <sub>0.067</sub>

Table S70: GCMC-MBAR results for 2-hexyne with the MiPPE force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
500	438.2 <sub>2.9</sub>	85.9 <sub>4.0</sub>	2.495 <sub>0.026</sub>	14.93 <sub>0.36</sub>	0.574 <sub>0.024</sub>
490	464.9 <sub>1.8</sub>	67.7 <sub>2.5</sub>	2.131 <sub>0.016</sub>	17.31 <sub>0.29</sub>	0.635 <sub>0.021</sub>
480	486.7 <sub>1.5</sub>	54.7 <sub>1.3</sub>	1.811 <sub>0.011</sub>	19.18 <sub>0.19</sub>	0.682 <sub>0.016</sub>
470	505.2 <sub>1.7</sub>	44.67 <sub>0.74</sub>	1.5302 <sub>0.0087</sub>	20.71 <sub>0.14</sub>	0.720 <sub>0.012</sub>
460	522.5 <sub>2.3</sub>	36.61 <sub>0.52</sub>	1.2839 <sub>0.0074</sub>	22.09 <sub>0.14</sub>	0.753 <sub>0.011</sub>
450	539.3 <sub>2.5</sub>	29.96 <sub>0.40</sub>	1.0686 <sub>0.0062</sub>	23.39 <sub>0.15</sub>	0.783 <sub>0.010</sub>
440	555.2 <sub>1.8</sub>	24.43 <sub>0.31</sub>	0.8815 <sub>0.0050</sub>	24.60 <sub>0.11</sub>	0.81 <sub>0.01</sub>
430	569.62 <sub>0.68</sub>	19.82 <sub>0.28</sub>	0.7201 <sub>0.0040</sub>	25.678 <sub>0.066</sub>	0.835 <sub>0.010</sub>
420	583.3 <sub>1.4</sub>	15.98 <sub>0.26</sub>	0.5821 <sub>0.0034</sub>	26.69 <sub>0.11</sub>	0.857 <sub>0.014</sub>
410	597.0 <sub>1.4</sub>	12.77 <sub>0.22</sub>	0.4651 <sub>0.0035</sub>	27.67 <sub>0.12</sub>	0.877 <sub>0.017</sub>
400	609.9 <sub>1.1</sub>	10.11 <sub>0.16</sub>	0.3669 <sub>0.0039</sub>	28.59 <sub>0.11</sub>	0.896 <sub>0.020</sub>
390	621.7 <sub>1.4</sub>	7.92 <sub>0.14</sub>	0.2857 <sub>0.0045</sub>	29.42 <sub>0.14</sub>	0.914 <sub>0.026</sub>
380	633.2 <sub>1.2</sub>	6.12 <sub>0.16</sub>	0.2190 <sub>0.0050</sub>	30.23 <sub>0.16</sub>	0.930 <sub>0.040</sub>
370	644.01 <sub>0.40</sub>	4.67 <sub>0.18</sub>	0.1653 <sub>0.0059</sub>	30.98 <sub>0.19</sub>	0.944 <sub>0.063</sub>

Table S71: GCMC-MBAR results for 1-heptyne with the MiPPE force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
520	427.5 <sub>6.0</sub>	82.9 <sub>6.4</sub>	2.218 <sub>0.071</sub>	16.05 <sub>0.64</sub>	0.595 <sub>0.026</sub>
510	454.3 <sub>2.5</sub>	66.9 <sub>4.8</sub>	1.906 <sub>0.055</sub>	18.37 <sub>0.49</sub>	0.646 <sub>0.027</sub>
500	476.5 <sub>1.4</sub>	54.5 <sub>3.3</sub>	1.629 <sub>0.041</sub>	20.34 <sub>0.35</sub>	0.691 <sub>0.024</sub>
490	495.7 <sub>1.4</sub>	44.8 <sub>2.4</sub>	1.384 <sub>0.030</sub>	22.02 <sub>0.25</sub>	0.730 <sub>0.023</sub>
480	512.9 <sub>1.6</sub>	36.9 <sub>1.8</sub>	1.168 <sub>0.020</sub>	23.51 <sub>0.18</sub>	0.764 <sub>0.024</sub>
470	528.7 <sub>2.0</sub>	30.3 <sub>1.4</sub>	0.979 <sub>0.012</sub>	24.85 <sub>0.13</sub>	0.794 <sub>0.026</sub>
460	543.2 <sub>1.9</sub>	24.91 <sub>0.95</sub>	0.8138 <sub>0.0072</sub>	26.066 <sub>0.092</sub>	0.821 <sub>0.027</sub>
450	556.9 <sub>1.5</sub>	20.37 <sub>0.57</sub>	0.6705 <sub>0.0058</sub>	27.199 <sub>0.070</sub>	0.846 <sub>0.025</sub>
440	570.2 <sub>1.0</sub>	16.57 <sub>0.25</sub>	0.5472 <sub>0.0063</sub>	28.279 <sub>0.062</sub>	0.868 <sub>0.019</sub>
430	583.08 <sub>0.58</sub>	13.38 <sub>0.11</sub>	0.4419 <sub>0.0062</sub>	29.301 <sub>0.062</sub>	0.888 <sub>0.012</sub>
420	595.19 <sub>0.32</sub>	10.71 <sub>0.23</sub>	0.3526 <sub>0.0055</sub>	30.253 <sub>0.097</sub>	0.907 <sub>0.017</sub>
410	606.63 <sub>0.36</sub>	8.50 <sub>0.30</sub>	0.2779 <sub>0.0046</sub>	31.14 <sub>0.16</sub>	0.923 <sub>0.035</sub>
400	617.58 <sub>0.39</sub>	6.66 <sub>0.33</sub>	0.2160 <sub>0.0048</sub>	31.98 <sub>0.23</sub>	0.938 <sub>0.058</sub>
390	628.63 <sub>0.36</sub>	5.16 <sub>0.31</sub>	0.1654 <sub>0.0061</sub>	32.81 <sub>0.31</sub>	0.950 <sub>0.088</sub>

Table S72: GCMC-MBAR results for 1-octyne with the MiPPE force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
550	419.4 <sub>3.6</sub>	89.5 <sub>1.2</sub>	2.075 <sub>0.015</sub>	16.27 <sub>0.24</sub>	0.56 <sub>0.01</sub>
540	445.2 <sub>3.0</sub>	70.95 <sub>0.88</sub>	1.788 <sub>0.015</sub>	18.98 <sub>0.20</sub>	0.62 <sub>0.01</sub>
530	467.6 <sub>2.6</sub>	57.19 <sub>0.55</sub>	1.535 <sub>0.015</sub>	21.30 <sub>0.17</sub>	0.67 <sub>0.01</sub>
520	487.1 <sub>2.4</sub>	46.85 <sub>0.42</sub>	1.311 <sub>0.014</sub>	23.22 <sub>0.15</sub>	0.71 <sub>0.01</sub>
510	504.3 <sub>2.0</sub>	38.69 <sub>0.48</sub>	1.114 <sub>0.012</sub>	24.87 <sub>0.13</sub>	0.75 <sub>0.01</sub>
500	519.8 <sub>1.5</sub>	32.01 <sub>0.56</sub>	0.9395 <sub>0.0099</sub>	26.33 <sub>0.12</sub>	0.78 <sub>0.01</sub>
490	534.3 <sub>1.2</sub>	26.45 <sub>0.57</sub>	0.7869 <sub>0.0075</sub>	27.66 <sub>0.13</sub>	0.805 <sub>0.010</sub>
480	548.2 <sub>1.2</sub>	21.77 <sub>0.52</sub>	0.6539 <sub>0.0053</sub>	28.92 <sub>0.15</sub>	0.829 <sub>0.014</sub>
470	561.6 <sub>1.2</sub>	17.83 <sub>0.40</sub>	0.5385 <sub>0.0038</sub>	30.10 <sub>0.15</sub>	0.852 <sub>0.016</sub>
460	574.15 <sub>0.95</sub>	14.52 <sub>0.26</sub>	0.4392 <sub>0.0033</sub>	31.21 <sub>0.13</sub>	0.872 <sub>0.016</sub>
450	586.02 <sub>0.57</sub>	11.73 <sub>0.12</sub>	0.3546 <sub>0.0033</sub>	32.245 <sub>0.084</sub>	0.890 <sub>0.012</sub>
440	597.51 <sub>0.35</sub>	9.404 <sub>0.077</sub>	0.2830 <sub>0.0032</sub>	33.229 <sub>0.046</sub>	0.91 <sub>0.01</sub>
430	608.64 <sub>0.40</sub>	7.46 <sub>0.15</sub>	0.2231 <sub>0.0029</sub>	34.171 <sub>0.089</sub>	0.921 <sub>0.019</sub>
420	619.31 <sub>0.42</sub>	5.86 <sub>0.20</sub>	0.1736 <sub>0.0027</sub>	35.06 <sub>0.16</sub>	0.935 <sub>0.039</sub>
410	630.00 <sub>0.38</sub>	4.55 <sub>0.22</sub>	0.1330 <sub>0.0032</sub>	35.94 <sub>0.24</sub>	0.946 <sub>0.067</sub>

Table S73: GCMC-MBAR results for 1-nonyne with the MiPPE force field. Subscripts correspond to the 95% confidence interval computed with bootstrap re-sampling.

$T^{\text{sat}}$ (K)	$\rho_{\text{liq}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$\rho_{\text{vap}}^{\text{sat}}$ (kg/m <sup>3</sup> )	$P_{\text{vap}}^{\text{sat}}$ (MPa)	$\Delta H_v$ (kJ/mol)	$Z_{\text{vap}}^{\text{sat}}$
570	427.1 <sub>1.2</sub>	80.5 <sub>1.0</sub>	1.776 <sub>0.015</sub>	18.58 <sub>0.17</sub>	0.58 <sub>0.01</sub>
560	450.9 <sub>1.3</sub>	64.43 <sub>0.86</sub>	1.531 <sub>0.013</sub>	21.28 <sub>0.20</sub>	0.63 <sub>0.01</sub>
550	471.7 <sub>1.6</sub>	52.31 <sub>0.73</sub>	1.315 <sub>0.010</sub>	23.58 <sub>0.20</sub>	0.68 <sub>0.01</sub>
540	489.9 <sub>1.5</sub>	43.02 <sub>0.62</sub>	1.1239 <sub>0.0080</sub>	25.52 <sub>0.17</sub>	0.72 <sub>0.01</sub>
530	506.3 <sub>1.1</sub>	35.61 <sub>0.52</sub>	0.9552 <sub>0.0059</sub>	27.21 <sub>0.13</sub>	0.76 <sub>0.01</sub>
520	521.47 <sub>0.66</sub>	29.51 <sub>0.42</sub>	0.8067 <sub>0.0041</sub>	28.739 <sub>0.093</sub>	0.79 <sub>0.01</sub>
510	535.75 <sub>0.67</sub>	24.42 <sub>0.33</sub>	0.6762 <sub>0.0026</sub>	30.149 <sub>0.083</sub>	0.81 <sub>0.01</sub>
500	549.10 <sub>0.85</sub>	20.14 <sub>0.26</sub>	0.5626 <sub>0.0015</sub>	31.452 <sub>0.094</sub>	0.83 <sub>0.01</sub>
490	561.65 <sub>0.81</sub>	16.54 <sub>0.20</sub>	0.46410 <sub>0.00097</sub>	32.666 <sub>0.094</sub>	0.856 <sub>0.010</sub>
480	573.68 <sub>0.62</sub>	13.50 <sub>0.15</sub>	0.3792 <sub>0.0010</sub>	33.811 <sub>0.080</sub>	0.875 <sub>0.011</sub>
470	585.26 <sub>0.49</sub>	10.94 <sub>0.11</sub>	0.3068 <sub>0.0014</sub>	34.896 <sub>0.067</sub>	0.892 <sub>0.012</sub>
460	596.35 <sub>0.58</sub>	8.794 <sub>0.096</sub>	0.2455 <sub>0.0017</sub>	35.924 <sub>0.069</sub>	0.907 <sub>0.015</sub>
450	607.27 <sub>0.71</sub>	7.003 <sub>0.091</sub>	0.1942 <sub>0.0020</sub>	36.924 <sub>0.081</sub>	0.921 <sub>0.019</sub>
440	618.21 <sub>0.61</sub>	5.516 <sub>0.091</sub>	0.1516 <sub>0.0023</sub>	37.91 <sub>0.10</sub>	0.933 <sub>0.027</sub>
430	628.74 <sub>0.51</sub>	4.292 <sub>0.090</sub>	0.1166 <sub>0.0026</sub>	38.85 <sub>0.14</sub>	0.944 <sub>0.037</sub>
420	638.50 <sub>0.50</sub>	3.296 <sub>0.086</sub>	0.0885 <sub>0.0030</sub>	39.72 <sub>0.18</sub>	0.955 <sub>0.052</sub>



## S9 Simulation state points

### S9.1 Cyclohexane

Table S74: State points simulated to test finite-size effects for cyclohexane with the MiPPE force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
450	-4370	3.5
510	-4370	3.5
550	-4370	3.5
510	-4177	3.5
470	-4048	3.5
430	-3936	3.5
400	-3865	3.5
370	-3806	3.5
350	-3774	3.5

Table S75: State points simulated for cyclohexane with the MiPPE force field (second iteration,  $\theta^{(2)} \lambda_{\text{CH}_2} = 16$ ).

$T$ (K)	$\mu$ (K)	$L$ (nm)
450	-4370	3.0
500	-4370	3.0
550	-4370	3.0
500	-4135	3.0
460	-4025	3.0
410	-3890	3.0
360	-3790	3.0

Table S76: State points simulated for cyclohexane with the TraPPE force field (zeroth iteration,  $\theta^{(0)}$ ).

$T$ (K)	$\mu$ (K)	$L$ (nm)
450	-4350	3.0
500	-4350	3.0
550	-4350	3.0
500	-4120	3.0
460	-3977	3.0
410	-3790	3.0
350	-3562	3.0

Table S77: State points simulated for cyclohexane with the first iteration ( $\theta^{(1)}$ )  $\lambda_{\text{CH}_2} = 14$  force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
450	-4389	3.0
500	-4389	3.0
550	-4389	3.0
500	-4164	3.0
460	-4033	3.0
410	-3891	3.0
360	-3780	3.0

Table S78: State points simulated for cyclohexane with the first iteration ( $\theta^{(1)}$ )  $\lambda_{\text{CH}_2} = 16$  force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
450	-4367	3.0
500	-4367	3.0
550	-4367	3.0
500	-4149	3.0
460	-4024	3.0
410	-3893	3.0
360	-3792	3.0

Table S79: State points simulated for cyclohexane with the first iteration ( $\theta^{(1)}$ )  $\lambda_{\text{CH}_2} = 18$  force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
450	-4370	3.0
500	-4370	3.0
550	-4370	3.0
500	-4158	3.0
460	-4037	3.0
410	-3912	3.0
360	-3825	3.0

Table S80: State points simulated for cyclohexane with the first iteration ( $\theta^{(1)}$ )  $\lambda_{\text{CH}_2} = 20$  force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
450	-4386	3.0
500	-4386	3.0
550	-4386	3.0
500	-4178	3.0
460	-4062	3.0
410	-3946	3.0
360	-3866	3.0

## S9.2 Branched alkanes

Table S81: State points simulated for 2-methylpropane with the TraPPE force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
350	-3120	3.0
380	-3120	3.0
405	-3117	3.0
380	-2980	3.0
350	-2880	3.0
320	-2790	3.0
290	-2705	3.0
260	-2645	3.0
230	-2600	3.0
200	-2570	3.0

Table S82: State points simulated for 2,2-dimethylpropane with the TraPPE force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
380	-3405	3.0
410	-3405	3.0
440	-3405	3.0
410	-3250	3.0
380	-3140	3.0
350	-3037	3.0
330	-2970	3.0
300	-2900	3.0
270	-2820	3.0

Table S83: State points simulated for 2,2-dimethylbutane with the TraPPE force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
420	-3860	3.5
450	-3860	3.5
480	-3860	3.5
450	-3719	3.5
420	-3600	3.5
400	-3524	3.5
380	-3450	3.5
360	-3368	3.5
340	-3288	3.5
310	-3280	3.5

Table S84: State points simulated for 2,3-dimethylbutane with the TraPPE force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
440	-4015	3.0
470	-4015	3.0
500	-4011	3.0
470	-3845	3.0
440	-3735	3.0
410	-3635	3.0
380	-3555	3.0
350	-3480	3.0
320	-3415	3.0

Table S85: State points simulated for 3,3-dimethylhexane with the TraPPE force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
500	-4670	3.5
530	-4670	3.5
560	-4670	3.5
520	-4476	3.5
490	-4370	3.5
460	-4268	3.5
430	-4164	3.5
400	-4039	3.5
370	-3925	3.5

Table S86: State points simulated for 3-methyl-3-ethylpentane with the TraPPE force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
500	-4785	4.0
550	-4785	4.0
580	-4785	4.0
550	-4636	4.0
520	-4520	4.0
490	-4400	4.0
460	-4280	4.0
430	-4160	4.0
410	-4080	4.0
390	-3990	4.0

Table S87: State points simulated for 2,3,4-trimethylpentane with the TraPPE force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
480	-4740	3.5
520	-4740	3.5
565	-4735	3.5
530	-4549	3.5
500	-4436	3.5
470	-4337	3.5
440	-4241	3.5
410	-4182	3.5
380	-4090	3.5
350	-4020	3.5

Table S88: State points simulated for 2,2,4-trimethylpentane with the TraPPE force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
480	-4600	4.0
530	-4600	4.0
560	-4600	4.0
530	-4450	4.0
500	-4330	4.0
470	-4210	4.0
440	-4090	4.0
410	-3960	4.0
380	-3840	4.0

Table S89: State points simulated for 2-methylpropane with the MiPPE-gen force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
350	-3150	3.0
380	-3150	3.0
410	-3145	3.0
380	-3010	3.0
350	-2910	3.0
320	-2830	3.0
290	-2760	3.0
260	-2700	3.0
230	-2670	3.0
200	-2640	3.0

Table S90: State points simulated for 2,2-dimethylpropane with the MiPPE-gen force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
368	-3344	3.0
398	-3344	3.0
430	-3400	3.0
398	-3216	3.0
372	-3124	3.0
346	-3032	3.0
326	-2961	3.0
299	-2865	3.0
270	-2759	3.0



Table S91: State points simulated for 2,2-dimethylbutane with the MiPPE-gen force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
415	-3873	3.5
445	-3873	3.5
480	-3895	3.5
450	-3756	3.5
420	-3654	3.5
400	-3588	3.5
380	-3521	3.5
360	-3454	3.5
340	-3384	3.5
310	-3380	3.5

Table S92: State points simulated for 2,3-dimethylbutane with the MiPPE-gen force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
440	-4010	3.0
470	-4010	3.0
500	-4009	3.0
470	-3860	3.0
440	-3760	3.0
410	-3670	3.0
380	-3600	3.0
350	-3530	3.0
320	-3480	3.0

Table S93: State points simulated for 2,3,4-trimethylpentane with the MiPPE-gen force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
480	-4720	3.5
520	-4720	3.5
565	-4713	3.5
530	-4540	3.5
500	-4360	3.5
470	-4355	3.5
440	-4275	3.5
410	-4205	3.5
380	-4165	3.5
350	-4115	3.5

Table S94: State points simulated for 2,2,4-trimethylpentane with the MiPPE-gen force field.

$T$ (K)	$\mu$ (K)	$L$ (nm)
470	-4570	4.0
520	-4570	4.0
550	-4570	4.0
520	-4420	4.0
490	-4300	4.0
460	-4170	4.0
430	-4050	4.0
400	-3920	4.0
370	-3790	4.0