Secondary Structure	ϕ	φ
α	-160 to -50	-60 to +30
β	-180 to -110	+110 to +180
P^{II}	-110 to -40	+110 to +180
α^{L}	+20 to +70	-30 to +70

Table S1. The ranges used to determine residue based secondary structure populations.

ALA1	α	β	P^{II}	α^{L}	SASA
Hybrid Mod 1st shell	28.0 ± 0.5	24.5 ± 0.1	35.7 ± 0.6	3.5 ± 0.3	355.8 ± 0.0
Hybrid Mod 1st and 2nd shells	29.3 ± 1.0	23.8 ± 0.1	36.0 ± 0.1	2.8 ± 0.7	355.8 ± 0.1
ALA3	**α**	**β**	**P^{II}**	**α^{L}**	**SASA**
Hybrid Mod 1st shell	26.4 ± 0.5	22.3 ± 0.3	36.7 ± 2.1	6.9 ± 1.6	561.9 ± 0.3
Hybrid Mod 1st and 2nd shells	21.1 ± 1.6	22.6 ± 0.4	43.5 ± 2.2	4.4 ± 1.2	556.4 ± 1.1

Table S2. Populations of basins on the ϕ/ψ energy landscape corresponding to alternate secondary structures, along with average solvent accessible surface areas. These simulations employed the modified intrinsic Born radius for hydrogen bonded to oxygen, as described in the text.

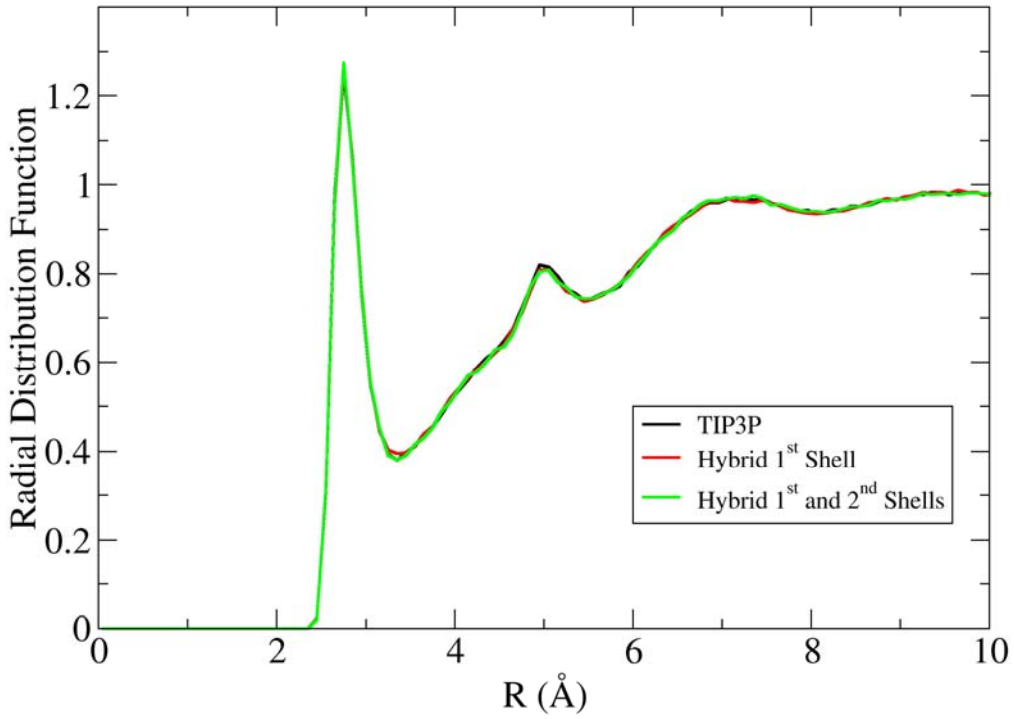

Figure S1. Radial distribution functions for water oxygen atoms around the carbonyl of Ala2 in alanine tetrapeptide, calculated using ptraj. The distributions for the hybrid models using either 1st or 1st and 2nd shells are nearly indistinguishable from those obtained using the reference standard REMD in explicit solvent.

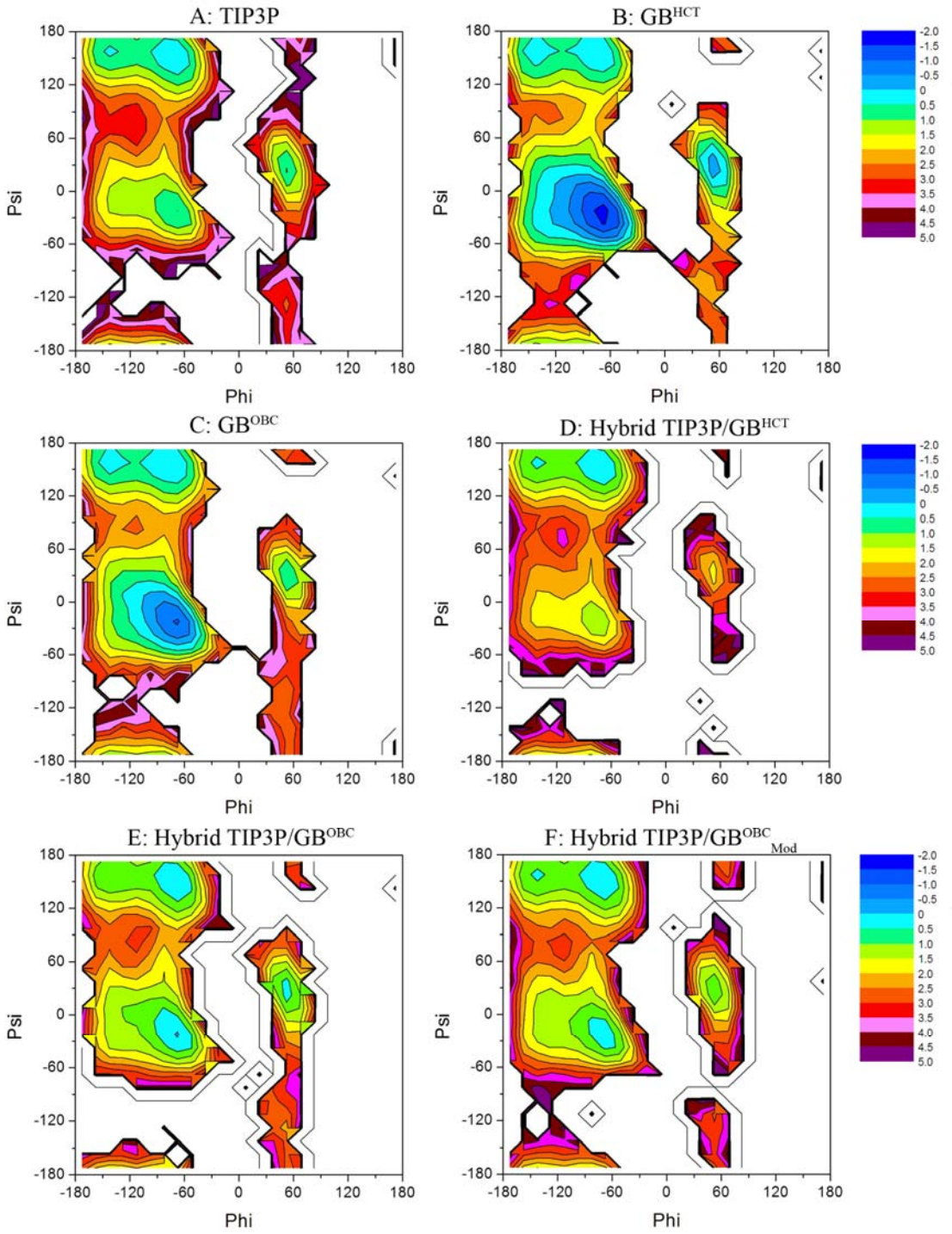

Figure S2. Free energy profiles at 300K for the central Ala5 residue from REMD in multiple solvent models. Contour levels are spaced 0.5 kcal/mol apart. Solvent models

are (A) TIP3P explicit solvent, (B) GB^{HCT}, (C) GB^{OBC}, (D) GB^{HCT}/TIP3P hybrid, (E) GB^{OBC}/TIP3P hybrid and (F) GB^{OBC}/TIP3P hybrid with intrinsic Born radius on hydrogen bonded to oxygen reduced by 0.05Å. (D), (E) and (F) correspond to fully solvated REMD simulations with the hybrid model used only for calculation of exchange probability. Basins corresponding to the major secondary structure types are all similar in free energy for models using explicit solvent; however both pure GB models show strong bias (2-3 kcal/mol) favoring α-helical conformations. Free energy landscapes were calculated using two dimensional histogram analyses of the dihedral angles of Ala5. For easier comparison between models, free energy values were normalized using the TIP3P REMD global minimum (the bin corresponding to $-75° < \varphi < -60°$, $150° < \psi < 165°$) as a free energy of zero.