Supplementary Table 1. Total ^{2}H isotope shifts for ^{15}N, $^{1}H^{N}$, and $^{13}C'$ measured for amino acid pairs with multiple occurrences in aS.

KT pairs

Res #	N	H
22	-127	2
33	-121	-3
44	-115	-2
59	-131	-5
81	-159	-2
Rmsd	**17.1**	**2.7**

Res #	CO
21	40
32	35
43	36
58	10
80	40
	12.6

AA pairs

Res #	N	H
18	-278	-9
19	-256	-10
30	-250	-11
90	-262	-6
91	-260	-9
Rmsd	**10.6**	**2.0**

Res #	CO
17	41
18	24
29	19
89	43
90	34
	10.5

GV pairs

Res #	N	H
15		
26	-66	-3
37	-54	2
48	-60	1
52	-51	-2
74	-55	-1
Rmsd	**5.7**	**2.0**

Res #	CO
14	3
25	3
36	4
47	6
51	3
73	8
	2.2

Table S2. Total ^{2}H isotope shifts measured from a mixture of protonated and perdeuterated aS. Values are given in parts per billion (ppb).

Residue	$^{13}C^{\alpha}$	$^{13}C^{\beta}$	$^{13}C'$	^{15}N	$^{1}H^{N}$
D2	-382	-665	-12	—	—
V3	-520	-960	39	-154	-2
F4	-434	-839	12	-263	-7
M5	-466	-903	11	-189	-3
K6	-493	-1022	42	-179	-9
G7	-483	—	0	-216	-15
L8	-440	-1104	43	-112	0
S9	-455	-709	2	-233	-4
K10	-453	-1028	43	—	—
A11	-484	-876	22	-252	-3
K12	-460	-1027	—	-180	-8
E13	-504	-870	30	-209	-7
G14	-475	—	3	-197	-11
V15	-523	-964	39	—	—
V16	-541	-964	61	-282	-11
A17	-475	-880	41	-380	-12
A18	-485	-868	24	-278	-9
A19	-490	-870	18	-256	-10
E20	-498	-884	16	-169	-9
K21	-480	-1026	40	-168	-1
T22	-455	-572	19	-127	2
K23	-469	-1026	39	-204	-5
Q24	-518	-892	29	—	—
G25	-475	—	3	-199	-11
V26	-525	-962	43	-66	-3
A27	-496	-873	23	-350	-11
E28	-505	-880	24	-200	-11
A29	-472	-874	19	-241	-1
A30	-477	-873	39	-225	-6
G31	-474	—	6	-188	-11
K32	-443	-1046	35	-81	-3
T33	-413	-584	6	-121	-3
K34	—	-1014	38	—	—
E35	—	-872	—	-201	-5
G36	-490	—	4	-181	-9
V37	-501	-963	44	-54	2
L38	-447	-1116	40	-273	-9

Table S2 (continued)

Residue	$^{13}C^{\alpha}$	$^{13}C^{\beta}$	$^{13}C'$	^{15}N	$^{1}H^{N}$
Y39	-433	-867	17	-212	-4
V40	-500	-979	49	-139	-5
G41	-477	—	9	-327	-29
S42	-447	-709	-1	-129	-5
K43	-448	-1033	36	-178	-8
T44	-423	-579	10	-115	-2
K45	-471	-1027	—	-201	-9
E46	—	—	—	-216	-7
G47	-470	—	6	-184	-9
V48	-503	-971	38	-60	1
V49	-509	-969	43	-276	-8
H50	-453	-674	14	-346	-11
G51	—	—	3	-197	-11
V52	-470	-938	48	-51	-2
A53	-471	-881	29	-372	-12
T54	-428	-570	37	-148	-2
V55	-508	-962	54	-197	-11
A56	-472	-871	30	-366	-11
E57	-490	-881	16	-208	-9
K58	-456	-1033	10	-180	-5
T59	-444	-572	14	-131	1
K60	-475	-1042	—	—	—
E61	—	—	19	-215	-9
Q62	-480	-856	15	—	—
V63	-509	-964	40	-144	-2
T64	-418	-576	20	-203	-2
N65	-377	-613	-15	-247	-4
V66	-519	-952	45	-114	1
G67	-475	—	14	-299	-16
G68	-469	—	13	-108	-7
A69	-466	-879	23	-169	-3
V70	-521	-967	38	-182	-9
V71	-487	-952	46	-288	-13
T72	-430	-573	33	-257	-5
G73	-467	—	8	-235	-14
V74	-500	-972	38	-55	-1
T75	-438	-572	31	-226	-5

Table S2 (continued)

Residue	$^{13}C^{\alpha}$	$^{13}C^{\beta}$	$^{13}C'$	^{15}N	$^{1}H^{N}$
A76	-465	-878	26	-294	-7
V77	-506	-964	53	-160	-5
A78	-480	-885	31	-365	-10
Q79	-488	-866	20	—	—
K80	-447	-1040	40	-188	-6
T81	-429	-576	16	-159	-2
V82	-504	-961	47	-202	-6
E83	-498	-883	34	-308	-12
G84	-471	—	15	-215	-10
A85	-475	-875	29	-174	-3
G86	-470	—	8	-190	-13
S87	-434	-705	1	-122	-2
I88	-464	-1019	62	-132	-4
A89	-474	-881	43	-347	-7
A90	-466	-881	34	-262	-6
A91	-454	-895	26	-260	-9
T92	-428	-561	31	-114	0
G93	-467	—	9	-227	-12
F94	-425	-854	17	-112	-2
V95	-492	-983	42	-186	-7
K96	-467	-1051	32	-300	-13
K97	-450	-1040	41	—	—
D98	-381	-650	-12	-236	-7
Q99	-483	-842	11	-163	0
L100	-447	-1094	48	—	—
G101	-478	—	1	-232	-10
K102	-446	-1042	29	-97	-4
N103	-392	-610	-30	-243	-10
E104	-494	-875	11	-172	-8
E105	-491	-875	28	-172	-7
G106	-475	—	17	-200	-6
A107	-449	-866	—	-172	-1
P108	-450	-931	40	—	—
Q109	-498	-850	23	-240	-8
E110	-480	-882	31	-195	-6
G111	-472	—	3	-195	-8
I112	-477	-1019	35	-33	-3

Table S2 (continued)

Residue	$^{13}C^{\alpha}$	$^{13}C^{\beta}$	$^{13}C'$	^{15}N	$^{1}H^{N}$
I112	-477	-1019	35	-33	-3
L113	-458	-1102	28	-283	-9
E114	-485	-877	28	-227	-9
D115	-387	-657	-5	-214	-1
M116	-430	-877	—	-163	4
P117	-453	-913	49	—	—
V118	-492	-972	51	-195	-8
D119	-395	-680	—	-312	-9
P120	-433	-899	40	—	—
D121	-391	-651	-3	-241	-4
N122	-392	-628	-19	-206	2
E123	-484	-880	17	-208	-4
A124	-468	-869	22	-231	-5
Y125	-435	-854	14	-157	1
E126	-483	-889	25	-179	-3
M127	-434	-875	—	-199	-6
P128	-448	-909	47	—	—
S129	-450	-702	-1	-250	-10
E130	-473	-873	14	-203	-10
E131	-503	-884	25	—	—
G132	-484	—	0	-200	-9
Y133	-427	-852	2	-88	1
Q134	-499	-844	9	-212	-4
D135	-384	-656	0	-232	-6
Y136	-444	-854	4	-179	2
E137	-471	-893	—	-202	-2
P138	-452	-909	42	—	—
E139	-476	-884	31	-240	-13
A140	-489	-914	—	-273	-4