

Supporting Tables:

Table S1. $^1J_{\text{NH}}$ couplings and $^1D_{\text{NH}}$ RDCs measured for the inhibitor-free PR and the DMP323-bound PR at 293 K. The average experimental error is 0.05 Hz in the measured $^1J_{\text{NH}}$ and 0.15 Hz in the measured $^1D_{\text{NH}}$.

Residue	Inhibitor-free PR		DMP323-bound PR	
	$^1J_{\text{NH}}$ (Hz)	$^1D_{\text{NH}}$ (Hz)	$^1J_{\text{NH}}$ (Hz)	$^1D_{\text{NH}}$ (Hz)
Q2	-91.9	-3.2	-91.8	-3.4
V3	-93.4	-2.5	-93.1	-2.5
T4	-92.7	-2.6	-93.0	-2.8
W6	-92.5	-1.2	-92.4	-2.5
K7	-91.5	0.6	-91.7	-0.7
R8	-93.6	-2.6	-93.4	-2.5
L10	-92.1	0.7	-92.5	1.4
V11	-93.2	-2.0	-93.0	-2.4
T12	-92.4	-1.5	-92.4	-2.2
I13	-93.1	-2.5	-92.9	-2.6
K14	-92.0	-3.0	-92.4	-3.4
I15	-93.9	-2.3	-93.7	-2.9
G16	-94.8	-2.0	-94.5	-2.7
G17	-93.9	-2.1	-93.2	-2.5
Q18	-92.7	-3.2	-92.1	-3.1
L19	-92.3	-2.5	-92.9	-3.1
K20	-93.4	-1.7	-93.2	-2.2
E21	-92.1	-2.1	-92.5	-2.5
A22	-93.5	-1.6	-93.4	-2.2
L23	-91.7	-1.4	-91.9	-2.5
L24	-	-	-94.5	-0.8
N25	-94.6	4.0	-92.8	4.0
T26	-93.4	-0.6	-92.0	1.4
G27	-92.1	-2.7	-93.9	-3.2
A28	-95.0	-2.0	-93.7	-2.4
D29	-93.1	-1.1	-92.9	-0.9
T31	-92.7	1.6	-92.8	2.8
V32	-91.0	1.4	-91.9	1.9
I33	-92.5	0.7	-93.1	1.1
E34	-92.9	-0.2	-93.2	-0.8
E35	-92.3	2.4	-92.6	3.2
M36	-92.3	-2.7	-92.4	-2.5
S37	-93.0	-2.8	-93.1	-3.6
L38	-92.7	-2.0	-93.1	-2.9
G40	-93.8	-0.1	-93.9	0.5
R41	-92.2	-1.5	-92.0	-0.6
W42	-92.7	0.4	-92.2	-0.7
K43	-92.3	1.3	-92.7	0.4
K45	-93.3	-3.5	-93.4	-3.5
M46	-92.8	-3.1	-92.8	-3.1
I47	-93.2	-2.9	-93.1	-3.4
G48	-94.1	-2.7	-94.0	-2.6
G49	-	-	-94.1	-2.8
I50	-92.4	-2.0	-92.5	-2.0
G51	-93.2	0.6	-93.1	0.6
G52	-93.9	-1.3	-94.4	-1.8
I54	-92.1	-2.1	-92.2	-3.6
K55	-92.3	-2.3	-92.4	-2.2
V56	-93.7	-2.7	-93.8	-3.3
R57	-91.6	-1.5	-91.6	-2.2
Q58	-92.5	1.1	-92.4	-1.0
Y59	-93.4	2.8	-93.0	3.8
D60	-92.8	2.9	-93.2	2.7

Q61	-92.4	1.1	-92.2	0.5
I62	-92.8	2.4	-92.9	2.3
I63	-92.6	2.0	-92.6	1.9
I64	-93.5	-2.5	-93.7	-2.8
E65	-92.3	-1.7	-91.9	-2.7
I66	-92.7	-2.2	-92.8	-1.7
A67	-93.7	-2.6	-93.8	-2.4
G68	-93.3	-1.5	-93.3	-2.0
H69	-92.1	1.6	-92.4	2.3
K70	-92.9	-1.5	-92.7	-1.7
A71	-93.3	-1.8	-93.2	-2.8
I72	-92.2	-1.6	-92.7	-1.8
G73	-95.9	-0.8	-95.4	-1.9
T74	-92.0	-1.4	-92.1	-1.3
V75	-93.4	3.6	-93.4	3.3
L76	-92.5	2.2	-92.7	1.9
V77	-92.0	0.8	-	-
G78	-94.1	-0.6	-94.8	-1.2
T80	-91.6	3.5	-91.5	4.1
V82	-91.4	-2.5	-91.3	-3.1
N83	-93.3	-2.6	-93.1	-2.6
I84	-92.7	1.1	-92.8	1.7
I85	-92.3	2.3	-92.2	3.4
G86	-94.1	3.7	-93.7	4.3
R87	-	-	-93.3	0.1
N88	-95.5	-0.8	-95.1	-0.5
L90	-93.6	-1.2	-93.5	1.7
T91	-93.7	-1.4	-93.4	-1.2
Q92	-92.5	-2.2	-92.6	-1.7
I93	-92.1	0.3	-92.0	-2.4
G94	-93.9	-0.8	-93.7	0.1
A95	-93.8	0.1	-94.2	-1.4
T96	-93.9	-2.5	-93.8	1.5
L97	-91.4	-2.1	-91.2	-3.2
N98	-92.6	-3.4	-92.5	-2.1
F99	-93.5	-2.7	-93.4	-2.1

Table S2. $^1J_{\text{NH}}$ couplings and $^1D_{\text{NH}}$ RDCs measured for the inhibitor-free PR20 at 293 K. The average experimental error is 0.05 Hz in the measured $^1J_{\text{NH}}$ and 0.15 Hz in the measured $^1D_{\text{NH}}$.

	Inhibitor-free PR20 $^1J_{\text{NH}}$ (Hz)	Inhibitor-free PR20 $^1D_{\text{NH}}$ (Hz)
Q2	-91.7	-2.6
I3	-93.0	-2.4
T4	-93.2	-2.2
L5	-93.1	1.1
W6	-92.6	-1.2
K7	-91.4	-0.8
R8	-93.2	-2.2
F10	-92.0	0.7
V11	-93.2	-1.5
T12	-92.5	-1.4
V13	-93.5	-2.0
K14	-92.1	-2.3
I15	-94.0	-2.7
G16	-94.7	-2.0
G17	-93.5	-0.7
Q18	-92.6	-2.4
L19	-92.1	-1.1
K20	-93.8	-2.2
E21	-92.3	-1.8
A22	-93.4	-1.5
L23	-91.7	-1.5
L24	-92.4	-1.3
N25	-94.5	4.0
T26	-93.3	0.6
G27	-92.2	-2.7
A28	-94.7	-2.0
D29	-93.0	-0.8
N30	-92.4	0.9
T31	-93.1	1.8
I32	-91.1	2.0
F33	-92.4	1.5
E34	-92.7	0.1
D35	-92.2	-1.9
N37	-93.1	-2.1
L38	-93.1	-0.9
G40	-93.7	0.1
R41	-92.2	0.0
K43	-92.4	1.0
K45	-93.2	-2.7
M46	-92.6	-2.3
V47	-92.9	-2.5
G48	-94.0	-1.9
G49	-93.6	-1.4
I50	-92.3	-1.0
G51	-93.5	-0.8
G52	-93.8	-0.1
F53	-93.1	0.2
L54	-92.4	-0.7
K55	-92.3	-1.9
V56	-93.6	-2.5
R57	-91.2	-1.1
E58	-92.3	-1.1
Y59	-93.3	3.0
D60	-92.7	2.4
V62	-92.9	2.2
I64	-93.5	-2.4
E65	-92.1	-2.7
I66	-92.6	-1.7

A67	-93.8	-2.1
G68	-93.2	-1.3
H69	-91.9	1.1
K70	-92.9	-1.7
A71	-93.2	-2.0
I72	-92.3	-2.0
G73	-95.8	-1.7
T74	-92.1	0.0
V75	-93.4	3.8
V77	-92.4	-0.8
G78	-94.2	-1.2
T80	-91.6	-1.7
V82	-91.3	-2.9
V84	-92.9	1.6
I85	-92.1	2.7
G86	-94.2	4.0
R87	-93.2	-0.3
D88	-95.4	-1.8
T89	-94.0	-0.8
M90	-93.9	-0.9
T91	-93.8	-1.5
Q92	-92.5	-1.6
I93	-92.2	0.1
G94	-93.5	-1.3
A95	-93.7	0.8
T96	-93.8	-2.9
L97	-91.4	-2.9
N98	-92.3	-2.9
F99	-93.2	-2.3