Table S2: Proton Chemical Shifts in the d(GGGAAAAACGG)·d(CCGTTTTTCCC)-SN6999 Complex at pH 7.0, 28 ^{o}C (1 ^{o}C)[a]

DNA Chemical Shifts (ppm)

residue	N1H, N3H	NH_2	2H, 5H, $5CH_3$	6H,8H	1′H	2′H	2″H	3′H	4′H	5′H / 5″H[b]
G_1	13.21[c]		—	7.78	5.56	2.38	2.55	4.75	4.10	3.60, 3.60
G_2	13.07		—	7.72	5.50	2.57	2.65	4.93	4.29	3.98, 4.06
G_3	12.78 (12.83,12.79)		—	7.69	5.41	2.51	2.64	4.96	4.31	4.06, 4.11
A_4	—		7.50	8.03	5.81	2.56	2.78	5.01	4.37	4.11, 4.16
A_5	—		7.30	8.02	5.56	2.54	2.69	4.96	4.12	
A_6	—		7.53	7.86	5.56 (5.49, 5.43)	2.33	2.64	4.85	3.94	
A_7	—		7.58	7.73	5.23	2.19	2.55	4.78	4.03	3.64, 3.48
A_8	—		7.84 (7.95, 7.84)	7.78 (7.80, 7.78)	5.31	2.19	2.42	4.75	3.97	3.49, 3.78
C_9	—	7.93, 6.12	4.93	6.91 (6.89, 6.94)	5.13	1.51	1.88	4.58		
G_{10}	13.09		—	7.67	5.61	2.52	2.65	4.88	4.13	3.96, 3.79[d]

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DNA Chemical Shifts (ppm)

residue	N1H, N3H	NH_2	2H, 5H, $5CH_3$	6H,8H	1′H	2′H	2″H	3′H	4′H	5′H / 5″H[b]
G_{11}	12.95[c]		—	7.76	6.13	2.51	2.33	4.64	4.21	
C_{12}	—	7.85[e], 6.95[e]	5.87	7.67	5.94	1.98	2.46	4.63	4.08	
C_{13}	—	8.52, 6.80	5.61	7.46	5.55	2.05	2.37	4.82	4.10	3.98, 4.01
G_{14}	13.00		—	7.96	6.06	2.70	2.81	5.00	4.40	4.07, 4.11
T_{15}	14.37 (14.50, 14.43)		1.52	7.27	6.08	2.14	2.59	4.83	4.29	4.07, 4.17
T_{16}	14.48, 14.32[f] (14.59, 14.41)		1.60	7.37	5.76	2.06	2.37	4.66	4.05	
T_{17}	13.87, 14.05 (14.14, 13.90)[g]		1.54	7.24	5.48	1.87	2.36	4.59	3.91	
T_{18}	13.78 (13.95, 13.86)		1.54	7.20	5.25	1.78	2.27	4.63		
T_{19}	13.52		1.53	7.19	5.29	1.95	2.25	4.70	4.00	3.70, 3.44
C_{20}	—	8.32, 6.65	5.56	7.40	5.71	2.06	2.33	4.71		
C_{21}	—	8.52, 6.84	5.66	7.52	5.99 (5.87, 5.93)	2.16	2.43	4.80	4.10	

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DNA Chemical Shifts (ppm)										
residue	N1H, N3H	NH_2	2H, 5H, $5CH_3$	6H,8H	1′H	2′H	2″H	3′H	4′H	5′H / 5″H[b]
C_{22}	—	8.28[e], 7.16[e]	5.82	7.67	6.23	2.26	2.26	4.54	4.03	

Drug Chemical Shifts (ppm)		
Q-2H 8.81	BQ-2,6H 8.23	Q/BQ NH 10.41 (10.48, 10.53)
Q-3H 8.36	BQ-3,5H 8.02	BQ/BP NH 9.59 (9.62, 9.75)
Q-5H 8.67 (8.66, 8.62)	BP-2,6H 7.82 (7.85, 7.74)	P/BP NH 10.18 (10.31, 10.25)
Q-6H 8.01	BP-3,5H 8.00 (8.02, 7.84)	
Q-7H 8.22 (8.21, 8.18)	P-2,6H 8.16 (8.23, 8.10, 8.16)[h]	
Q-8H 8.34 (8.38, 8.31)	P-3,5H 7.17[i] (7.14, 8.01, 7.13)[h]	
Q-CH_3 4.28	P-CH_3 4.03 (4.04, 3.98)	

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DNA Chemical Shifts (ppm)

residue	N1H, N3H	NH_2	2H, 5H, $5CH_3$	6H,8H	1′H	2′H	2″H	3′H	4′H	5′H / 5″H[b]

[a]Chemical shifts in parentheses correspond to resonances at 1°C which are doubled at this temperature . The major is listed first, followed by the minor. All others are obtained at 28 °C unless otherwise noted. Measured from a 1 mM solution containing 50 mM phosphate buffer, 100 mM NaCl, 0.2 mM EDTA, and 0.003%NaN_3. Chemical shifts for the drug and for all exchangeable protons, including the A2H protons, were obtained with a drug-DNA ratio of 1:1. All other DNA chemical shifts were obtained with a slight excess of drug. The chemical shifts are referenced to 2,2-dimethyl-2-silapentane-5-sulfonate using the HO^2H resonance and a calibration curve (67,68). [b]Stereoassignment of 5′H / 5″H is based on relative intensity of d_i(3′, 5″) and d_i(3′, 5′) NOEs in 50 ms NOESY. [c]Observed only at 1°C . Because of spectral overlap and lack of sequential NOEs, the relative chemical shifts of G_1 and G_{11} imino protons were determined by the observation that 5′ G imino resonances are generally downfield from 3′ G imino resonances. [d]Observed at 37°C with slight drug excess. [e]Observed only at 1°C. [f]Assigned on the basis of one exchange peak only. [g]Relative intensities not certain. [h]One major and two minor, with the second minor less than the first. [i]Tentative assignment based solely on a very broad P-2,6H / P-3,5H crosspeak in the ^1H_2O NOESY spectrum in the region of NOE of C_{22} amino proton pair as observed at 1 °C.