Table 1S Populations of different motifs of bare (0W), monohydrated (1W) and dihydrated (2W) complexes of guanine···cytosine and their tautomers.

	Populations [%]								
	0W			1W			2W		
	HB [a]	ST [b]	TS [c]	HB	ST	TS	HB	ST	TS
G19C1	73.93	1.08	24.99	77.20	3.38	19.41	54.20	15.10	30.70
G17C1	80.13	0.00	19.86	80.55	3.05	16.40	78.03	6.03	15.56
G19C2A	77.72	0.01	22.27	98.53	0.18	1.29	93.27	1.85	4.88
G19C2B	70.22	10.72	19.05	68.55	9.81	21.64	56.93	14.40	28.66
G17C2A	54.79	1.76	43.44	64.25	1.74	34.00	63.01	5.00	31.97
G17C2B	60.98	7.96	31.06	61.88	8.68	29.44	54.75	10.60	34.47
G91C1	57.27	17.21	25.51	49.57	13.55	36.87	44.51	17.75	37.65
G92C1	36.07	9.61	54.32	39.55	16.80	43.65	36.23	24.91	38.52
G91C2A	51.16	7.69	41.14	43.73	8.62	47.65	38.40	13.82	47.78
G91C2B	70.42	4.57	25.01	47.94	8.62	43.44	40.64	13.86	45.45
G92C2A	38.64	0.00	61.36	33.72	11.03	55.25	28.88	20.55	50.57
G92C2B	47.10	7.39	45.50	45.47	16.16	38.37	34.81	22.76	42.42

[a] hydrogen bonded (HB)

[b] stacked (ST)

[c] T-shaped (TS)

Table 2aS Interaction energies of different motifs (HB, ST, TS) of bare (0W), monohydrated (1W) and dihydrated (2W) complexes of guanine···cytosine and their tautomers computed by DFT-D/cc-pVTZ method.

	Interaction energies [kcal/mol]								
	HB [a]			ST [b]			TS [c]		
	0W	1W	2W	0W	1W	2W	0W	1W	2W
G19C1	-32.23	-41.84	-58.24	-14.01	-27.66	-48.72		-37.06	-48.26
G17C1	-30.07	-43.47	-56.27		-32.58	-49.50		-34.90	-45.24
G19C2A	-24.16	-32.20	-46.45		-25.03	-40.31	-12.57	-27.36	-39.60
G19C2B	-15.80	-25.87	-33.00		-29.88	-44.58	-11.76		-40.23
G17C2A	-22.51	-36.18	-47.34		-22.08	-39.22	-12.15	-26.53	-39.58
G17C2B	-15.19	-29.84	-41.73	-11.20	-24.18	-41.26	-10.60	-25.38	-39.58
G91C1	-20.72	-37.72	-47.29	-12.36	-31.15	-48.00	-13.45	-31.51	-40.19
G92C1	-19.92	-32.48	-46.62	-12.76	-28.59	-43.75	-12.71	-28.18	-41.60
G91C2A	-17.90	-34.80	-38.58	-8.67	-26.38	-40.97	-9.04	-22.55	-34.13
G91C2B	-13.30	-33.10	-37.36	-9.59	-22.13	-42.91	-9.66		-42.14
G92C2A	-26.33	-34.47	-50.68	-8.83	-23.77	-44.24	-7.51	-27.84	-35.98
G92C2B	-24.60			-10.09		-39.47	-10.70		-40.91

[a] hydrogen bonded (HB)

[b] stacked (ST)

[a] T-shaped (TS)

Table 2bS BSSE corrected interaction energies of different motifs (HB, ST, TS) of bare (0W), monohydrated (1W) and dihydrated (2W) complexes of guanine···cytosine and their tautomers computed by RI-MP2/cc-pVTZ method.

	Interaction energies [kcal/mol]								
	HB[a]			ST[b]			TS[c]		
	0W	1W	2W	0W	1W	2W	0W	1W	2W
G19C1	-29.13	-38.12	-51.28	-17.71	-28.90	-46.97			-44.71
G17C1	-26.89	-39.35	-50.11		-33.94	-49.17		-31.33	
G19C2A	-16.62	-28.91	-42.26		-27.38	-32.57			
G19C2B	-15.27	-24.91	-43.81		-30.57	-40.44			
G17C2A	-20.29	-33.74	-34.23		-23.05	-42.57			
G17C2B	-14.69	-28.01	-34.10	-14.17	-30.51	-40.25	-11.46	-23.34	
G91C1	-20.74	-34.98	-37.88	-15.73	-31.83	-44.06			
G92C1	-19.80	-31.29	-43.17	-16.58	-30.45	-43.57	-15.92		
G91C2A	-17.40	-31.59	-40.64	-12.27	-26.81	-34.94			
G91C2B	-10.50	-28.24	-31.79	-12.20	-27.63	-42.11	-10.80		
G92C2A	-20.57	-29.22	-41.06	-11.59	-25.61	-42.11			
G92C2B	-19.45	-29.85	-44.81	-13.48	-24.01	-38.45			

[a] hydrogen bonded (HB)

[b] stacked (ST)

[a] T-shaped (TS)