Conformational Interconversion of the *trans*-4-Hydroxynonenal-Derived (6*S*,8*R*,11*S*) 1,N^2-Deoxyguanosine Adduct When Mismatched with Deoxyadenosine in DNA

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Supporting Information

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Abbreviations: HNE, *trans*-4-hydroxynonenal; 1,N^2-HNE-dG, HNE derived 1,N^2-2'-deoxyguanosine adduct; PdG, 1,N^2-propano-2'-deoxyguanosine; M_1dG, 3-(2'-deoxy-β-D-erythro-pentofuranosyl)pyrimido[1,2-α]purin-10(3*H*)-one; NOESY, nuclear Overhauser effect spectroscopy; COSY, correlation spectroscopy; TOCSY, total correlation spectroscopy; DQF-COSY, double-quantum filtered COSY; NOE, nuclear Overhauser effect; rMD, restrained molecular dynamics

Table S1. Chemical shifts of the mismatched duplex at pH 5.5.

Residue	H6/H8	H2/H5/Me	H1′	H2′	H2′′	H3′
G^{1}	7.94		5.95	2.62	2.73	4.79
C^{2}	7.48	5.30	5.99	2.07	2.44	4.78
T^{3}	7.38	1.61	5.44	2.10	2.35	4.81
A^{4}	8.14		6.00	2.69	2.85	5.00
G^{5}	7.64		5.46	2.43	2.47	4.99
C^{6}	7.23	5.18	5.74	1.58	2.52	4.72
X^{7}	7.37		5.46	3.04	2.27	4.77
A^{8}	8.22	7.69	5.95	2.67	2.79	4.99
G^{9}	7.54		5.75	2.46	2.62	4.79
T^{10}	7.18	1.17	5.94	2.06	2.41	4.76
C^{11}	7.54	5.61	5.98	2.15	2.43	4.76
C^{12}	7.60	5.68	6.17	2.21	2.24	4.51
G^{13}	7.81		5.59	2.45	2.63	4.77
G^{14}	7.78		5.52	2.64	2.72	4.97
A^{15}	8.16	7.82	6.21	2.68	2.87	5.00
C^{16}	7.20	5.12	5.73	1.84	2.36	4.65
T^{17}	7.41	1.55	5.39	2.13	2.44	4.79
A^{18}	8.42	7.04	6.06	2.77	2.94	5.03
G^{19}	7.74		5.76	2.52	2.63	4.95
C^{20}	7.29	5.22	5.75	1.88	2.35	4.65
T^{21}	7.33	1.57	5.41	2.06	2.32	4.78
A^{22}	8.16		5.96	2.68	2.83	4.96
G^{23}	7.64		5.73	2.41	2.58	4.89
C^{24}	7.30	5.24	6.03	2.09	2.09	4.40

Table S2. Chemical shifts of the mismatched duplex at pH 8.9.

Residue	H6/H8	H2/H5/Me	H1′	H2′	H2′′	H3′
G^{1}	7.94		5.95	2.62	2.74	4.79
C^{2}	7.48	5.30	6.01	2.07	2.45	4.77
T^{3}	7.37	1.60	5.44	2.11	2.35	4.81
A^{4}	8.14	7.40	6.00	2.69	2.84	5.00
G^{5}	7.64		5.56	2.43	2.53	4.92
C^{6}	7.28	5.32	5.84	1.96	2.22	4.75
X^{7}	7.59		5.38	2.31	2.56	4.77
A^{8}	8.13	7.89	5.78	2.68	2.69	4.96
G^{9}	7.57		5.86	2.46	2.66	4.80
T^{10}	7.24	1.19	6.01	2.11	2.47	4.81
C^{11}	7.54	5.63	5.98	2.15	2.42	4.78
C^{12}	7.59	5.70	6.18	2.22	2.22	4.51
G^{13}	7.81		5.60	2.47	2.66	4.77
G^{14}	7.79		5.54	2.66	2.74	4.98
A^{15}	8.16	7.86	6.22	2.69	2.87	5.01
C^{16}	7.18	5.12	5.73	1.78	2.32	4.66
T^{17}	6.90	1.55	5.84	1.10	1.65	4.63
A^{18}	8.19	7.37	5.79	2.60	2.66	4.94
G^{19}	7.89		5.86	2.63	2.71	4.93
C^{20}	7.29	5.26	5.77	1.89	2.36	4.67
T^{21}	7.33	1.57	5.42	2.07	2.33	4.79
A^{22}	8.17	7.39	6.00	2.70	2.84	5.01
G^{23}	7.65		5.73	2.43	2.58	4.90
C^{24}	7.32	5.26	6.04	2.06	2.13	4.40

Table S3. NOE restraints utilized in the rMD calculation at pH 5.5.

Residue I No.	Residue I Name	Atom I Name	Residue II No.	Residue II Name	Atom II Name	Lower bond	Upper bond
1	DG5	H3′	1	DG5	H8	3.71	5.87
2	DC	H6	1	DG5	H8	3.47	5.39
2	DC	H5	1	DG5	H8	3.78	5.12
2	DC	H2′	2	DC	H5	3.32	4.32
3	DT	M7	2	DC	H5	2.31	3.13
8	DA	H8	7	DGH	H2′	4.50	5.50
9	DG	H1′	8	DA	H2	3.88	5.18
9	DG	H8	8	DA	H8	3.93	5.31
10	DT	M7	9	DG	H1′	3.81	5.15
10	DT	M7	9	DG	H8	2.30	3.10
10	DT	H2′	10	DT	M7	3.65	4.95
11	DC	H5	10	DT	H6	3.39	4.59
11	DC	H5	10	DT	M7	3.77	5.11
12	DC3	H6	12	DC3	H1′	2.85	3.75
12	DC3	H5	12	DC3	H1′	4.03	5.45
12	DC3	H3′	12	DC3	H1′	2.88	3.80
15	DA	H8	15	DA	H1′	2.95	3.89
15	DA	H2	15	DA	H1′	4.27	5.77
15	DA	H3′	15	DA	H1′	3.03	3.97
15	DA	H2′	15	DA	H1′	2.33	3.09
15	DA	H2′′	15	DA	H1′	2.13	2.87
16	DC	H1′	15	DA	H1′	3.28	4.44

16	DC	H1′	15	DA	H2	2.62	3.54
16	DC	H6	15	DA	H1′	2.55	3.45
16	DC	H6	15	DA	H8	3.86	4.89
16	DC	H6	15	DA	H3′	3.96	5.12
16	DC	H6	15	DA	H2′′	2.02	2.84
16	DC	H5	15	DA	H1′	3.53	4.57
16	DC	H5	15	DA	H8	2.76	3.74
16	DC	H5	15	DA	H2′	3.09	4.57
16	DC	H5	15	DA	H2′′	2.97	4.01
16	DC	H3′	15	DA	H1′	3.33	5.05
16	DC	H3′	16	DC	H5	4.53	6.37
16	DC	H2′	15	DA	H1′	3.72	4.91
16	DC	H2′	15	DA	H8	4.86	6.48
16	DC	H2′	16	DC	H6	2.00	2.78
16	DC	H2′	16	DC	H5	3.24	4.28
16	DC	H2′′	16	DC	H5	4.22	5.34
17	DT	H6	16	DC	H3′	4.53	5.76
17	DT	H6	16	DC	H2′	2.67	4.25
17	DT	M7	16	DC	H5	2.42	3.08
18	DAP	H2	8	DA	H2	3.50	4.50
18	DAP	H3′	18	DAP	H8	3.04	4.00
18	DAP	H2′	18	DAP	H8	1.84	2.50
18	DAP	H2′′	18	DAP	H8	2.83	3.63
19	DG	H8	18	DAP	H8	3.33	4.37
19	DG	H8	18	DAP	H2′′	2.10	2.90
20	DC	H6	19	DG	H3′	3.17	7.25

20	DC	H5	19	DG	H8	3.67	4.85
20	DC	H2′	20	DC	H5	2.51	4.95
21	DT	M7	20	DC	H5	2.80	3.48
25	HNE	H1	6	DC	H5	2.41	2.95
25	HNE	H22	6	DC	H5	3.16	4.28
25	HNE	H3	6	DC	H6	4.16	5.52
25	HNE	H3	25	HNE	H22	2.30	3.73
25	HNE	H4	6	DC	H6	4.26	5.66
25	HNE	H4	25	HNE	H22	2.33	3.27
1	DG5	H1′	1	DG5	H8	3.47	4.31
1	DG5	H1′	2	DC	H5	3.22	4.36
2	DC	H1′	2	DC	H5	4.09	5.43
2	DC	H1′	3	DT	M7	3.93	4.95
5	DG	H1′	6	DC	H6	3.17	5.25
5	DG	H8	6	DC	H6	4.23	5.61
7	DGH	H1′	7	DGH	H2′	2.70	3.66
7	DGH	H8	7	DGH	H2′	3.86	5.22
7	DGH	H8	8	DA	H2	3.15	4.27
8	DA	H3′	9	DG	H8	4.76	6.44
10	DT	H1′	10	DT	H6	2.84	3.70
10	DT	H1′	10	DT	M7	3.93	5.13
9	DG	H3′	10	DT	M7	3.19	4.31
9	DG	H2′	10	DT	M7	2.69	3.63
9	DG	H2′′	10	DT	M7	2.87	3.89
11	DC	H2′′	12	DC3	H1′	3.16	4.28
11	DC	H3′	12	DC3	H5	4.37	5.83

14	DG	H1′	15	DA	H1′	4.85	6.57
11	DC	H1′	15	DA	H2	3.03	4.03
17	DT	H1′	18	DAP	H8	4.22	6.20
18	DAP	H1′	18	DAP	H8	2.92	3.96
18	DAP	H1′	19	DG	H8	2.52	3.40
23	DG	H1′	23	DG	H3′	3.43	4.63
24	DC3	H1′	24	DC3	H5	4.65	6.54
2	DC	H5	1	DG5	H2′	2.68	3.62
2	DC	H5	1	DG5	H2′′	2.69	3.65
3	DT	H6	2	DC	H1′	2.90	3.76
5	DG	H2′	5	DG	H3′	2.28	3.26
9	DG	H8	8	DA	H2′′	2.01	2.71
16	DC	H5	16	DC	H1′	4.00	5.40
19	DG	H8	18	DAP	H3′	4.29	6.47
23	DG	H2′′	23	DG	H8	3.01	3.73
24	DC3	H6	23	DG	H8	3.57	4.69
24	DC3	H5	23	DG	H8	3.29	4.45
24	DC3	H5	23	DG	H2′	4.23	6.51
24	DC3	H3′	24	DC3	H6	3.02	3.90
25	HNE	Q5	25	HNE	H3	2.00	3.00
25	HNE	Q5	25	HNE	H4	2.72	3.86
1	DG5	H8	1	DG5	H2′	2.08	2.82
1	DG5	H8	1	DG5	H2′′	2.77	3.57
1	DG5	H1′	2	DC	H1′	3.27	4.43
2	DC	H6	2	DC	H2′	2.01	2.71
2	DC	H6	3	DT	M7	2.81	3.81

5	DG	H1′	5	DG	H8	3.87	4.85
5	DG	H1′	5	DG	H3′	3.41	4.55
7	DGH	H2′′	8	DA	H8	3.50	4.50
7	DGH	H1′	8	DA	H2′	2.96	5.20
9	DG	H1′	9	DG	H3′	2.86	3.86
10	DT	H6	10	DT	H2′	2.11	2.85
10	DT	H1′	11	DC	H5	3.15	4.27
11	DC	H3′	12	DC3	H6	3.44	4.29
14	DG	H1′	15	DA	H8	2.53	3.43
15	DA	H2′′	16	DC	H1′	3.96	5.18
16	DC	H6	16	DC	H3′	3.64	5.25
7	DGH	H8	18	DAP	H2	2.47	3.33
18	DAP	H1′	18	DAP	H3′	3.14	4.24
18	DAP	H1′	18	DAP	H2′′	2.11	2.85
18	DAP	H1′	19	DG	H3′	4.38	5.80
19	DG	H1′	19	DG	H8	2.95	3.89
21	DT	H6	21	DT	H3′	3.36	4.41
23	DG	H1′	23	DG	H8	2.87	3.89
24	DC3	H1′	24	DC3	H6	2.69	3.65
25	HNE	H1	25	HNE	H3	3.24	4.38
25	HNE	H1	25	HNE	H4	3.84	4.68
2	DC	H6	1	DG5	H2′′	2.29	3.21
2	DC	H2′′	2	DC	H6	2.88	3.66
3	DT	M7	2	DC	H2′′	2.91	3.93
6	DC	H3′	6	DC	H6	3.60	4.88
6	DC	H2′	6	DC	H3′	2.19	2.97

6	DC	H2′′	6	DC	H3′	2.11	2.85
19	DG	H1′	18	DAP	H2	3.42	4.52
19	DG	H2′	19	DG	H3′	2.27	3.07
20	DC	H3′	20	DC	H6	2.87	3.89
20	DC	H2′	20	DC	H6	2.03	2.75
21	DT	H6	20	DC	H2′	3.04	3.66
25	HNE	H1	6	DC	H6	2.49	3.51
25	HNE	H21	6	DC	H5	3.50	4.50
25	HNE	H22	25	HNE	H1	2.20	2.98
25	HNE	H4	6	DC	H3′	3.43	4.71
25	HNE	Q5	25	HNE	H1	4.43	5.93
25	HNE	Q6	25	HNE	H22	4.08	5.02
25	HNE	Q8	6	DC	H3′	3.71	5.01
25	HNE	Q8	25	HNE	H4	2.58	3.66
25	HNE	M9	25	HNE	Q8	2.55	3.45
1	DG5	H3′	1	DG5	H2′′	2.04	2.76
1	DG5	H2′	2	DC	H6	2.86	3.76
2	DC	H3′	3	DT	M7	3.33	4.51
6	DC	H1′	6	DC	H6	3.10	4.34
6	DC	H1′	6	DC	H2′	2.47	3.00
6	DC	H1′	25	HNE	H22	3.34	4.44
5	DG	H8	6	DC	H5	4.12	5.72
10	DT	H6	10	DT	H3′	3.26	4.24
11	DC	H1′	12	DC3	H6	2.89	3.77
11	DC	H1′	12	DC3	H5	3.93	5.31
11	DC	H2′′	12	DC3	H6	1.98	2.68

13	DG5	H1′	13	DG5	H3′	3.13	3.93
13	DG5	H1′	13	DG5	H2′′	1.82	2.46
14	DG	H1′	14	DG	H8	3.54	4.78
14	DG	H1′	14	DG	H2′	2.26	2.96
14	DG	H1′	14	DG	H2′′	2.12	2.86
16	DC	H1′	16	DC	H3′	2.92	3.84
7	DGH	H1′	18	DAP	H2	3.48	4.60
18	DAP	H1′	18	DAP	H2′	2.34	3.00
18	DAP	H3′	18	DAP	H2′′	2.62	3.35
19	DG	H8	19	DG	H3′	3.24	4.24
23	DG	H1′	24	DC3	H1′	3.16	4.20
24	DC3	H5	24	DC3	H2′	3.87	5.45
25	HNE	H3	25	HNE	Q8	3.83	5.03
2	DC	H3′	2	DC	H6	2.88	3.90
6	DC	H2′	6	DC	H5	3.22	4.28
8	DA	H3′	8	DA	H8	3.39	4.33
11	DC	H6	10	DT	H2′	3.12	3.85
11	DC	H5	10	DT	H2′	2.52	3.40
12	DC3	H6	11	DC	H2′	2.31	3.27
12	DC3	H5	11	DC	H2′	2.89	3.91
19	DG	H2′	19	DG	H8	2.34	3.16
25	HNE	H22	25	HNE	H21	1.59	2.27
1	DG5	H1′	2	DC	H6	2.33	3.15
2	DC	H1′	2	DC	H6	3.04	4.12
2	DC	H1′	2	DC	H2′	2.46	3.12
3	DT	H3′	3	DT	H2′′	2.82	3.83

5	DG	H3′	5	DG	H2′′	2.79	6.01
6	DC	H1′	6	DC	H5	4.67	6.33
6	DC	H1′	6	DC	H3′	3.06	4.14
6	DC	H6	6	DC	H2′′	2.40	3.38
6	DC	H6	25	HNE	H21	2.87	3.89
6	DC	H6	25	HNE	H22	2.74	3.84
7	DGH	H1′	8	DA	H8	3.50	4.50
9	DG	H1′	9	DG	H8	2.82	3.82
10	DT	H1′	10	DT	H2′	2.34	2.98
11	DC	H1′	11	DC	H2′	2.27	2.97
11	DC	H1′	12	DC3	H2′	3.14	4.14
11	DC	H2′′	12	DC3	H5	3.33	4.33
13	DG5	H1′	13	DG5	H2′	2.43	3.11
14	DG	H8	14	DG	H3′	3.90	5.28
14	DG	H8	14	DG	H2′′	3.00	3.64
16	DC	H1′	16	DC	H2′	2.40	3.10
16	DC	H1′	17	DT	H6	3.79	5.13
16	DC	H6	16	DC	H2′′	2.73	3.63
17	DT	H1′	17	DT	H2′′	2.17	2.93
18	DAP	H1′	19	DG	H1′	3.43	4.65
18	DAP	H2′	19	DG	H8	2.81	3.49
20	DC	H1′	20	DC	H2′	2.31	3.13
21	DT	H3′	21	DT	H2′′	2.82	3.63
24	DC3	H1′	24	DC3	H2′	2.26	3.06
24	DC3	H1′	24	DC3	H2′′	1.90	2.58
24	DC3	H3′	24	DC3	H2′	2.14	3.00

24	DC3	H3′	24	DC3	H2′′	2.60	3.58
24	DC3	H5	24	DC3	H2′′	4.01	5.31
25	HNE	H1	25	HNE	H21	2.23	3.01
25	HNE	H3	25	HNE	Q6	3.75	5.07
25	HNE	H4	25	HNE	Q6	3.12	4.04
1	DG5	H2′	1	DG5	H3′	1.95	2.63
2	DC	H6	1	DG5	H3′	3.92	5.16
2	DC	H2′	2	DC	H3′	1.97	2.67
2	DC	H2′′	2	DC	H3′	2.12	2.88
3	DT	H6	2	DC	H2′	2.71	3.37
3	DT	H6	2	DC	H2′′	2.00	3.00
3	DT	M7	2	DC	H2′	2.66	3.60
3	DT	H3′	3	DT	H6	3.21	4.35
3	DT	H2′	3	DT	M7	3.17	4.29
3	DT	H2′	3	DT	H3′	2.32	3.20
4	DA	H8	3	DT	H2′	3.25	4.49
4	DA	H8	3	DT	H2′′	2.76	3.74
4	DA	H2′	4	DA	H8	2.27	3.07
4	DA	H2′	4	DA	H3′	2.38	3.22
4	DA	H2′′	4	DA	H8	2.65	3.59
4	DA	H2′′	4	DA	H3′	2.62	3.54
5	DG	H8	4	DA	H2′	3.01	4.07
5	DG	H8	4	DA	H2′′	2.59	3.51
5	DG	H3′	5	DG	H8	3.40	4.12
5	DG	H2′	5	DG	H8	2.27	3.07
6	DC	H6	5	DG	H2′	2.91	3.93

6	DC	H6	5	DG	H2′′	2.35	3.29
6	DC	H5	5	DG	H2′	3.11	4.21
6	DC	H5	5	DG	H2′′	2.83	3.83
6	DC	H2′	6	DC	H6	2.05	2.77
6	DC	H2′′	6	DC	H5	3.99	5.21
8	DA	H2′	8	DA	H3′	1.86	2.52
8	DA	H2′′	8	DA	H8	2.59	3.33
8	DA	H2′′	8	DA	H3′	2.78	3.76
9	DG	H8	8	DA	H2′	2.83	3.60
9	DG	H2′	9	DG	H8	2.37	3.27
9	DG	H2′	9	DG	H3′	2.10	2.84
9	DG	H2′′	9	DG	H8	2.87	3.89
9	DG	H2′′	9	DG	H3′	2.34	3.16
10	DT	H6	9	DG	H8	3.59	4.71
10	DT	H6	9	DG	H3′	3.52	4.76
10	DT	H6	9	DG	H2′	2.90	3.76
10	DT	H6	9	DG	H2′′	2.33	3.33
10	DT	H2′	10	DT	H3′	1.90	2.56
10	DT	H2′′	10	DT	H6	2.62	3.54
10	DT	H2′′	10	DT	H3′	2.29	2.95
11	DC	H6	10	DT	H6	3.82	5.00
11	DC	H6	10	DT	H2′′	2.01	2.83
11	DC	H5	10	DT	H2′′	2.45	3.31
11	DC	H3′	11	DC	H6	2.96	4.00
11	DC	H3′	11	DC	H5	4.77	6.25
11	DC	H2′	11	DC	H3′	2.14	2.90

11	DC	H2′′	11	DC	H6	2.79	3.49
11	DC	H2′′	11	DC	H3′	2.36	2.99
12	DC3	H6	11	DC	H5	4.00	5.00
12	DC3	H5	11	DC	H6	3.43	4.65
12	DC3	H2′	11	DC	H6	4.50	5.50
12	DC3	H2′	12	DC3	H6	2.07	2.97
12	DC3	H2′	12	DC3	H5	3.21	4.33
12	DC3	H2′′	12	DC3	H6	2.62	3.54
12	DC3	H2′′	12	DC3	H5	3.63	4.73
13	DG5	H2′	13	DG5	H8	2.79	3.77
13	DG5	H2′	13	DG5	H3′	1.81	2.45
13	DG5	H2′′	13	DG5	H3′	2.46	3.32
14	DG	H8	13	DG5	H2′	2.96	4.00
14	DG	H8	13	DG5	H2′′	2.37	2.99
14	DG	H2′	14	DG	H8	2.06	2.78
15	DA	H8	14	DG	H2′	2.69	3.63
15	DA	H8	14	DG	H2′′	2.30	3.12
15	DA	H2′	15	DA	H8	1.97	2.54
15	DA	H2′	15	DA	H3′	2.33	3.25
15	DA	H2′′	15	DA	H8	2.89	3.85
15	DA	H2′′	15	DA	H3′	2.13	2.89
16	DC	H6	15	DA	H2′	3.10	3.87
17	DT	H6	16	DC	H6	3.69	4.93
17	DT	H6	16	DC	H2′′	2.40	3.24
17	DT	M7	16	DC	H6	2.48	3.36
17	DT	M7	16	DC	H2′	2.83	3.83

17	DT	M7	16	DC	H2′′	2.76	3.74
17	DT	H2′	17	DT	H6	2.28	3.08
17	DT	H2′	17	DT	M7	3.28	4.44
17	DT	H2′	17	DT	H3′	2.35	3.17
17	DT	H2′′	17	DT	H6	2.86	3.80
17	DT	H2′′	17	DT	H3′	2.17	2.79
20	DC	H6	19	DG	H2′	2.75	3.65
20	DC	H6	19	DG	H2′′	2.19	3.15
20	DC	H5	19	DG	H2′′	2.87	3.89
20	DC	H2′′	20	DC	H6	2.86	3.76
21	DT	H6	20	DC	H2′′	2.27	3.15
21	DT	M7	20	DC	H6	2.32	3.14
21	DT	M7	20	DC	H2′	2.42	3.28
21	DT	M7	20	DC	H2′′	2.90	3.92
21	DT	M7	21	DT	H1′	4.09	5.08
21	DT	H3′	21	DT	H1′	3.37	4.55
21	DT	H3′	21	DT	M7	4.07	5.41
21	DT	H2′	21	DT	H1′	2.60	3.52
21	DT	H2′	21	DT	M7	3.00	4.06
21	DT	H2′	21	DT	H3′	2.05	2.77
21	DT	H2′′	21	DT	H1′	1.88	2.54
21	DT	H2′′	21	DT	H6	2.65	3.47
22	DA	H8	21	DT	H1′	2.96	4.00
22	DA	H8	21	DT	H6	3.61	4.89
22	DA	H8	21	DT	H2′	3.62	4.90
22	DA	H8	21	DT	H2′′	2.23	3.27

22	DA	H2′	22	DA	H8	2.16	2.92
22	DA	H2′	22	DA	H3′	2.42	3.00
22	DA	H2′′	22	DA	H8	2.70	3.66
22	DA	H2′′	22	DA	H3′	2.46	3.32
23	DG	H8	22	DA	H8	4.05	5.35
23	DG	H8	22	DA	H2′	3.05	4.13
23	DG	H8	22	DA	H2′′	2.50	3.38
23	DG	H3′	23	DG	H8	3.45	4.51
23	DG	H2′	23	DG	H8	2.19	2.97
24	DC3	H6	23	DG	H2′	2.96	3.94
24	DC3	H6	23	DG	H2′′	2.38	3.32
24	DC3	H5	23	DG	H2′′	3.10	4.20
24	DC3	H2′	24	DC3	H6	2.02	2.74
24	DC3	H2′′	24	DC3	H6	2.85	3.71
25	HNE	H1	5	DG	H2′	4.00	5.00
25	HNE	H1	6	DC	H2′	2.64	3.56
25	HNE	H21	6	DC	H3′	3.67	4.83
25	HNE	H22	6	DC	H3′	2.76	3.74
25	HNE	H3	6	DC	H3′	5.50	6.50
25	HNE	H3	25	HNE	H21	1.98	2.68
25	HNE	H4	25	HNE	H21	1.95	2.63
25	HNE	Q5	6	DC	H6	4.00	5.42
25	HNE	Q5	6	DC	H3′	3.75	5.83
25	HNE	Q5	25	HNE	H22	2.87	3.89
25	HNE	Q6	6	DC	H3′	5.13	6.93
25	HNE	Q6	25	HNE	H1	5.00	6.00

25	HNE	Q6	25	HNE	Q5	2.49	3.37
25	HNE	Q7	6	DC	H3′	4.35	5.89
25	HNE	Q7	25	HNE	H3	3.24	4.28
25	HNE	Q7	25	HNE	H4	2.75	3.73
25	HNE	Q7	25	HNE	Q5	2.33	3.15
25	HNE	Q7	25	HNE	Q6	2.13	2.89
25	HNE	Q8	25	HNE	H1	4.87	6.47
25	HNE	Q8	25	HNE	Q5	3.06	4.14
25	HNE	Q8	25	HNE	Q7	2.62	3.54
1	DG5	H1′	1	DG5	H3′	3.01	4.07
1	DG5	H1′	1	DG5	H2′	2.45	3.31
1	DG5	H1′	1	DG5	H2′′	1.85	2.51
2	DC	H1′	2	DC	H2′′	1.91	2.59
3	DT	H1′	3	DT	H3′	3.55	4.79
3	DT	H1′	3	DT	H2′	2.23	3.01
3	DT	H1′	3	DT	H2′′	2.08	2.82
3	DT	H1′	4	DA	H8	2.97	4.03
4	DA	H1′	4	DA	H8	3.16	4.10
4	DA	H1′	4	DA	H3′	2.90	3.84
4	DA	H1′	4	DA	H2′	2.73	3.69
4	DA	H1′	4	DA	H2′′	2.07	2.79
4	DA	H1′	5	DG	H8	2.70	3.66
5	DG	H1′	5	DG	H2′	2.35	3.19
6	DC	H1′	6	DC	H2′′	1.88	2.54
6	DC	H1′	7	DGH	H8	3.50	4.50
6	DC	H1′	25	HNE	H21	5.00	6.00

7	DGH	H1′	7	DGH	H8	2.04	2.76
7	DGH	H1′	7	DGH	H3′	3.23	5.66
7	DGH	H1′	7	DGH	H2′′	2.36	3.22
8	DA	H1′	8	DA	H8	2.95	3.99
8	DA	H1′	8	DA	H3′	2.95	3.99
8	DA	H1′	8	DA	H2′	2.42	3.28
8	DA	H1′	8	DA	H2′′	2.04	2.76
9	DG	H1′	9	DG	H2′	2.39	3.23
9	DG	H1′	9	DG	H2′′	2.35	3.19
9	DG	H1′	10	DT	H6	3.26	4.20
10	DT	H1′	10	DT	H2′′	2.11	2.85
10	DT	H1′	11	DC	H6	2.27	3.07
11	DC	H1′	11	DC	H6	2.87	3.67
11	DC	H1′	11	DC	H2′′	1.94	2.62
12	DC3	H1′	12	DC3	H2′	2.27	3.07
12	DC3	H1′	12	DC3	H2′′	2.07	2.81
12	DC3	H3′	12	DC3	H2′	2.10	2.98
12	DC3	H3′	12	DC3	H2′′	2.72	3.68
13	DG5	H1′	13	DG5	H8	3.13	4.23
13	DG5	H1′	14	DG	H8	2.90	3.92
14	DG	H3′	14	DG	H2′	2.14	2.90
14	DG	H3′	14	DG	H2′′	1.95	2.65
16	DC	H1′	16	DC	H6	2.65	3.59
16	DC	H1′	16	DC	H2′′	2.01	2.71
16	DC	H3′	16	DC	H2′	2.07	2.79
16	DC	H3′	16	DC	H2′′	2.23	3.01

16	DC	H3′	17	DT	M7	3.21	4.35
17	DT	H1′	17	DT	H6	2.87	3.81
17	DT	H1′	17	DT	M7	3.88	5.14
17	DT	H1′	17	DT	H3′	3.34	4.52
17	DT	H1′	17	DT	H2′	2.34	3.16
18	DAP	H3′	18	DAP	H2′	2.27	3.07
19	DG	H1′	19	DG	H2′	2.31	3.03
19	DG	H1′	19	DG	H2′′	2.21	3.14
19	DG	H1′	20	DC	H6	2.77	3.65
19	DG	H1′	20	DC	H5	3.28	4.44
19	DG	H3′	19	DG	H2′′	2.02	2.74
20	DC	H1′	20	DC	H6	2.88	3.72
20	DC	H1′	20	DC	H2′′	2.04	2.76
20	DC	H1′	21	DT	H1′	3.50	4.74
20	DC	H1′	21	DT	H6	2.18	2.94
20	DC	H1′	21	DT	M7	3.50	4.74
20	DC	H3′	20	DC	H2′	2.05	2.77
20	DC	H3′	20	DC	H2′′	2.23	3.01
20	DC	H3′	21	DT	M7	3.30	4.46
22	DA	H1′	22	DA	H8	2.93	3.87
22	DA	H1′	22	DA	H2′	2.53	3.43
22	DA	H1′	22	DA	H2′′	2.44	3.30
22	DA	H1′	23	DG	H8	2.82	3.72
23	DG	H1′	23	DG	H2′	2.31	3.03
23	DG	H1′	23	DG	H2′′	2.07	2.79
23	DG	H1′	24	DC3	H6	2.75	3.65

23	DG	H1′	24	DC3	H5	3.62	4.78
24	DC3	H1′	24	DC3	H3′	3.01	3.97

M7 represents the methyl group of thymine; DGH represents HNE-dG adduct (X^7) without HNE moiety and the atoms are named as regular deoxyguanosine; DAP represents protonated A^{18} and the atoms are named as regular deoxyadenosine; HNE moiety is considered as a residue with residue No. 25, the atoms are named as following: X^7 H8: HNE H1; X^7 H7α(β): HNE H21(2); X^7 H6: HNE H3; X^7 H11: HNE H4; X^7 H12: HNE Q5; X^7 H13: HNE Q6; X^7 H14: HNE Q7; X^7 H15: HNE Q8; X^7 H16: HNE M9

Table S4. NOE restraints utilized in the rMD calculation at pH 8.9.

Residue I No.	Residue I Name	Atom I Name	Residue II No.	Residue II Name	Atom II Name	Lower bond	Upper bond
2	DC	H6	1	DG5	H8	3.57	5.78
2	DC	H5	1	DG5	H8	3.30	4.46
3	DT	M7	2	DC	H6	2.64	3.58
6	DC	H6	6	DC	H2′2	2.78	3.56
6	DC	H5	6	DC	H2′2	3.45	5.48
10	DT	M7	9	DG	H2′1	2.79	3.77
11	DC	H5	10	DT	H6	3.52	4.76
11	DC	H5	10	DT	M7	3.28	4.44
12	DC3	H6	12	DC3	H3′	2.21	2.99
12	DC3	H5	12	DC3	H3′	3.84	5.20
15	DA	H8	14	DG	H8	3.55	4.45
16	DC	H1′	15	DA	H2	2.98	4.02
16	DC	H2′1	16	DC	H1′	2.23	3.01
16	DC	H3′	16	DC	H2′1	2.04	2.90
16	DC	H6	15	DA	H2′1	2.63	3.55
16	DC	H6	15	DA	H2′2	2.06	2.92
16	DC	H6	15	DA	H3′	3.43	4.63
16	DC	H6	15	DA	H8	3.54	4.72
16	DC	H6	16	DC	H1′	2.89	3.91
16	DC	H6	16	DC	H2′1	2.05	2.87
16	DC	H6	16	DC	H3′	3.10	4.10
16	DC	H5	15	DA	H2′1	2.87	3.87
16	DC	H5	15	DA	H2′2	3.06	4.14

16	DC	H5	15	DA	H8	3.18	4.30
16	DC	H5	16	DC	H2′1	3.35	4.43
16	DC	H5	16	DC	H3′	5.50	6.50
17	DT	H6	16	DC	H1′	3.61	4.71
17	DT	H6	16	DC	H2′1	2.89	4.01
17	DT	H6	16	DC	H6	4.50	5.50
17	DT	M7	15	DA	H8	4.35	5.71
17	DT	M7	16	DC	H6	2.52	3.40
17	DT	M7	16	DC	H5	2.35	3.17
18	DA	H8	17	DT	H3′	5.00	6.00
18	DA	H8	17	DT	H6	4.50	5.50
19	DG	H8	18	DA	H8	5.00	6.00
20	DC	H3′	20	DC	H2′1	2.14	2.96
20	DC	H6	19	DG	H8	4.50	5.50
20	DC	H6	20	DC	H3′	2.72	4.46
20	DC	H5	20	DC	H2′1	3.23	4.29
20	DC	H5	20	DC	H3′	4.31	5.63
22	DA	H8	21	DT	H3′	5.00	6.00
24	DC3	H6	24	DC3	H1′	2.98	4.02
25	HNE	H3	25	HNE	H21	2.45	3.31
25	HNE	H3	25	HNE	H22	2.38	3.28
25	HNE	H4	25	HNE	H22	2.54	3.36
25	HNE	H4	25	HNE	H3	2.35	3.07
25	HNE	H52	18	DA	H8	5.00	6.00
25	HNE	Q6	25	HNE	H3	2.54	4.08
25	HNE	Q6	25	HNE	H4	2.69	3.85

1	DG5	H1′	1	DG5	H8	3.28	4.44
1	DG5	H1′	2	DC	H5	3.08	4.18
1	DG5	H3′	1	DG5	H8	2.81	3.69
1	DG5	H2′1	2	DC	H6	2.93	4.19
1	DG5	H2′2	2	DC	H6	2.21	2.95
2	DC	H2′1	2	DC	H6	1.99	2.69
2	DC	H2′1	2	DC	H5	3.31	4.27
2	DC	H2′2	2	DC	H6	2.77	3.57
2	DC	H2′2	3	DT	M7	2.64	4.01
2	DC	H1′	2	DC	H5	4.27	5.77
2	DC	H1′	3	DT	H1′	3.45	4.67
2	DC	H1′	3	DT	M7	3.92	5.30
3	DT	H3′	4	DA	H8	3.70	6.52
3	DT	H6	4	DA	H8	3.87	6.68
5	DG	H1′	5	DG	H8	2.82	3.82
5	DG	H1′	6	DC	H6	2.87	4.13
5	DG	H1′	6	DC	H5	3.02	4.08
6	DC	H1′	6	DC	H2′2	2.33	3.13
6	DC	H2′1	6	DC	H3′	2.35	2.89
6	DC	H2′1	6	DC	H5	3.19	4.31
6	DC	H2′1	7	DGH	H8	3.65	5.49
7	DGH	H1′	7	DGH	H2′2	2.19	2.97
7	DGH	H1′	7	DGH	H8	3.77	4.61
8	DA	H1′	8	DA	H8	3.00	4.00
9	DG	H1′	9	DG	H2′1	2.35	3.03
8	DA	H3′	9	DG	H8	3.49	5.19

9	DG	H2′2	10	DT	H6	2.02	2.63
10	DT	H1′	10	DT	H6	2.93	3.97
10	DT	H1′	10	DT	M7	3.86	5.22
10	DT	H2′1	10	DT	H6	1.95	2.63
10	DT	H2′1	10	DT	M7	2.91	3.75
12	DC3	H1′	12	DC3	H3′	2.83	3.83
12	DC3	H1′	12	DC3	H6	2.82	3.70
12	DC3	H1′	12	DC3	H5	4.80	6.50
14	DG	H3′	14	DG	H8	3.24	4.28
15	DA	H1′	15	DA	H2′1	2.39	3.23
15	DA	H1′	15	DA	H2′2	2.01	2.71
15	DA	H1′	15	DA	H3′	2.78	3.76
15	DA	H1′	15	DA	H8	3.00	3.98
15	DA	H1′	15	DA	H2	4.05	5.47
15	DA	H1′	16	DC	H1′	3.83	5.17
15	DA	H1′	16	DC	H3′	4.35	5.49
15	DA	H1′	16	DC	H6	2.53	3.43
15	DA	H1′	16	DC	H5	3.49	4.73
14	DG	H1′	15	DA	H8	2.48	3.22
11	DC	H1′	15	DA	H2	3.10	4.20
16	DC	H2′2	16	DC	H6	3.05	4.13
16	DC	H2′2	16	DC	H5	3.12	5.49
16	DC	H2′2	17	DT	H6	2.35	3.19
17	DT	H2′1	17	DT	H3′	2.06	2.78
17	DT	H2′1	17	DT	H6	1.89	2.67
17	DT	H2′1	18	DA	H8	3.68	4.98

17	DT	H1′	17	DT	H6	2.84	3.74
17	DT	H1′	25	HNE	H21	4.00	5.00
8	DA	H2	25	HNE	H21	3.00	4.00
8	DA	H2	25	HNE	H52	3.39	4.49
17	DT	H2′2	18	DA	H8	2.67	3.43
20	DC	H1′	20	DC	H2′1	2.29	3.03
20	DC	H1′	21	DT	H3′	4.31	5.47
21	DT	H6	22	DA	H8	2.77	6.40
23	DG	H1′	23	DG	H2′2	2.16	2.92
23	DG	H1′	23	DG	H8	2.83	3.83
23	DG	H1′	24	DC3	H1′	4.05	5.47
23	DG	H1′	24	DC3	H5	4.49	6.07
23	DG	H3′	24	DC3	H6	3.60	4.88
24	DC3	H3′	24	DC3	H6	2.73	3.69
24	DC3	H3′	24	DC3	H5	4.35	5.36
18	DA	H1′	25	HNE	H21	4.21	5.69
18	DA	H1′	25	HNE	H22	3.12	4.22
18	DA	H1′	25	HNE	H51	3.28	4.64
18	DA	H2	25	HNE	H21	3.50	4.50
18	DA	H2	25	HNE	H3	1.91	2.75
18	DA	H2	25	HNE	H4	4.00	5.00
18	DA	H2	25	HNE	Q6	5.00	6.00
25	HNE	H1	25	HNE	H21	1.50	2.50
25	HNE	H1	25	HNE	H22	2.19	2.89
25	HNE	H1	25	HNE	H4	2.86	4.04
25	HNE	H1	25	HNE	H52	3.50	4.50

25	HNE	H1	25	HNE	Q6	5.14	6.96
18	DA	H4′	25	HNE	H22	2.85	3.79
18	DA	H4′	25	HNE	H4	4.50	5.50
18	DA	H4′	25	HNE	M9	3.74	5.06
19	DG	H4′	25	HNE	H51	2.56	3.64
19	DG	H4′	25	HNE	Q6	3.05	4.23
19	DG	H4′	25	HNE	M9	4.48	6.06
18	DA	H5′2	25	HNE	Q8	2.66	3.26
9	DG	H5′1	25	HNE	M9	3.50	4.50
19	DG	H1′	25	HNE	M9	4.79	6.33
1	DG5	H8	1	DG5	H2′1	1.93	2.61
1	DG5	H8	1	DG5	H2′2	2.77	3.67
11	DC	H6	10	DT	H6	3.47	4.69
17	DT	H3′	17	DT	H2′2	2.20	2.98
17	DT	H6	17	DT	H2′2	2.88	3.72
19	DG	H8	19	DG	H1′	3.00	3.92
23	DG	H8	22	DA	H8	3.52	4.76
24	DC3	H6	23	DG	H8	3.92	4.96
24	DC3	H5	23	DG	H8	3.85	4.83
25	HNE	H4	25	HNE	H21	2.11	2.99
25	HNE	H51	25	HNE	H1	4.50	5.50
25	HNE	H51	25	HNE	H4	2.30	3.12
25	HNE	H52	25	HNE	H4	2.09	2.69
25	HNE	M9	25	HNE	Q6	3.22	4.68
1	DG5	H1′	1	DG5	H2′1	2.31	3.35
1	DG5	H1′	1	DG5	H2′2	1.73	2.35

2	DC	H5	3	DT	M7	2.55	3.45
3	DT	H1′	4	DA	H8	3.46	4.60
3	DT	H2′1	4	DA	H8	3.50	4.50
4	DA	H1′	4	DA	H8	2.94	3.80
6	DC	H1′	6	DC	H2′1	2.62	3.16
6	DC	H3′	6	DC	H6	3.01	4.07
5	DG	H8	6	DC	H5	3.12	4.22
7	DGH	H2′1	7	DGH	H8	2.37	3.57
7	DGH	H1′	8	DA	H8	4.00	5.00
9	DG	H1′	10	DT	H6	2.98	3.68
9	DG	H8	10	DT	M7	2.55	3.45
11	DC	H1′	12	DC3	H6	2.64	3.56
13	DG5	H1′	13	DG5	H2′2	1.78	2.40
13	DG5	H1′	13	DG5	H3′	3.75	4.75
14	DG	H1′	14	DG	H2′1	2.36	3.12
14	DG	H1′	14	DG	H2′2	2.27	3.07
17	DT	H1′	17	DT	H2′2	2.04	2.76
16	DC	H1′	17	DT	M7	3.48	4.58
18	DA	H2	19	DG	H1′	3.50	4.52
18	DA	H2	25	HNE	H1	4.00	5.00
18	DA	H2	25	HNE	H22	2.52	3.22
18	DA	H2	25	HNE	H51	3.61	4.89
18	DA	H2	25	HNE	H52	4.00	5.00
20	DC	H1′	20	DC	H6	3.00	4.00
20	DC	H1′	21	DT	M7	3.42	4.62
20	DC	H2′1	21	DT	H6	2.02	2.72

20	DC	H3′	21	DT	H6	3.58	4.84
21	DT	H1′	21	DT	H6	3.00	4.00
21	DT	H1′	22	DA	H8	3.26	4.42
20	DC	H5	21	DT	M7	2.41	3.27
21	DT	H2′1	22	DA	H8	3.00	4.00
21	DT	H2′2	22	DA	H8	2.50	3.50
22	DA	H1′	22	DA	H8	3.50	4.50
24	DC3	H1′	24	DC3	H3′	3.06	4.00
23	DG	H2′2	24	DC3	H6	2.10	2.80
23	DG	H2′1	24	DC3	H5	3.09	4.29
24	DC3	H2′1	24	DC3	H5	3.23	4.19
18	DA	H1′	25	HNE	H1	5.00	6.00
18	DA	H4′	25	HNE	H21	3.31	4.41
18	DA	H4′	25	HNE	Q6	2.75	3.83
8	DA	H2	25	HNE	H22	5.00	6.00
25	HNE	H3	25	HNE	H51	1.95	2.63
18	DA	H5′2	25	HNE	Q7	3.13	4.23
25	HNE	Q8	25	HNE	M9	2.18	2.96
10	DT	H6	9	DG	H8	3.51	4.75
12	DC3	H5	11	DC	H6	3.10	4.20
17	DT	H6	17	DT	H3′	2.47	3.35
20	DC	H6	20	DC	H2′2	3.06	3.71
21	DT	M7	20	DC	H6	2.25	3.05
25	HNE	M9	25	HNE	Q7	2.50	3.50
1	DG5	H1′	2	DC	H6	2.30	3.12
2	DC	H1′	2	DC	H6	2.94	3.67

2	DC	H3′	2	DC	H6	3.00	3.77
1	DG5	H2′1	2	DC	H5	2.38	3.22
2	DC	H2′2	3	DT	H6	2.38	3.65
3	DT	H3′	3	DT	H6	2.92	3.96
4	DA	H8	5	DG	H8	3.11	4.21
6	DC	H2′2	6	DC	H3′	2.25	3.05
5	DG	H2′2	6	DC	H6	2.36	3.05
5	DG	H2′2	6	DC	H5	2.64	3.58
7	DGH	H1′	7	DGH	H2′1	2.73	3.43
7	DGH	H2′2	7	DGH	H8	3.50	4.50
7	DGH	H2′2	8	DA	H8	2.50	3.50
8	DA	H1′	9	DG	H8	2.46	3.32
10	DT	H3′	10	DT	H6	2.97	4.01
11	DC	H3′	11	DC	H6	3.50	4.50
11	DC	H2′1	12	DC3	H5	3.02	4.52
11	DC	H2′2	12	DC3	H5	2.76	3.74
13	DG5	H3′	13	DG5	H8	3.65	4.97
14	DG	H1′	14	DG	H3′	2.81	3.81
16	DC	H1′	16	DC	H2′2	1.96	2.64
16	DC	H1′	16	DC	H3′	2.87	3.89
17	DT	H1′	17	DT	H2′1	2.30	3.00
17	DT	H1′	17	DT	H3′	2.90	3.82
18	DA	H1′	18	DA	H8	2.93	3.79
18	DA	H1′	18	DA	H2	3.20	4.34
18	DA	H1′	19	DG	H8	2.55	3.45
18	DA	H2′1	18	DA	H8	2.06	2.78

18	DA	H2′2	18	DA	H8	3.00	4.00
18	DA	H3′	18	DA	H8	3.50	4.50
19	DG	H3′	19	DG	H8	3.50	4.50
20	DC	H1′	20	DC	H2′2	1.83	2.47
20	DC	H1′	20	DC	H3′	2.93	3.97
19	DG	H2′2	20	DC	H6	2.19	2.97
19	DG	H2′2	20	DC	H5	2.67	3.39
19	DG	H1′	20	DC	H5	3.79	5.13
21	DT	H3′	21	DT	H6	3.03	5.10
20	DC	H2′1	21	DT	M7	2.75	3.79
23	DG	H1′	23	DG	H2′1	2.42	3.16
23	DG	H2′2	23	DG	H8	2.75	3.57
24	DC3	H2′1	24	DC3	H3′	2.00	2.70
24	DC3	H2′2	24	DC3	H3′	2.23	3.01
24	DC3	H2′2	24	DC3	H5	4.43	5.99
18	DA	H4′	25	HNE	H51	2.61	3.35
18	DA	H5′1	25	HNE	Q6	2.80	3.80
19	DG	H5′1	25	HNE	Q7	3.19	4.31
19	DG	H5′1	25	HNE	M9	4.55	5.66
6	DC	H6	5	DG	H8	3.84	5.20
7	DGH	H8	6	DC	H6	5.00	6.00
20	DC	H5	19	DG	H8	3.07	4.15
25	HNE	H3	25	HNE	H1	3.00	4.00
25	HNE	H52	25	HNE	H3	2.19	2.81
1	DG5	H1′	1	DG5	H3′	3.15	4.11
1	DG5	H2′2	2	DC	H5	2.62	3.54

3	DT	H2′2	4	DA	H8	2.56	3.46
6	DC	H1′	6	DC	H3′	2.89	3.83
5	DG	H2′1	6	DC	H6	3.08	4.16
6	DC	H2′1	6	DC	H6	1.85	2.57
7	DGH	H2′1	8	DA	H8	3.00	4.00
8	DA	H1′	9	DG	H2′1	3.50	4.50
9	DG	H1′	9	DG	H3′	3.30	3.99
9	DG	H1′	10	DT	M7	2.94	3.98
9	DG	H2′2	10	DT	M7	2.84	3.84
10	DT	H2′1	11	DC	H5	2.99	3.69
10	DT	H2′2	11	DC	H5	2.25	3.05
14	DG	H1′	14	DG	H8	2.86	3.86
16	DC	H2′2	16	DC	H3′	2.08	2.82
18	DA	H1′	18	DA	H2′1	2.48	3.22
20	DC	H2′2	20	DC	H3′	2.24	2.96
19	DG	H2′1	20	DC	H6	2.91	3.93
19	DG	H2′1	20	DC	H5	2.76	3.74
20	DC	H2′1	20	DC	H6	2.19	2.97
20	DC	H1′	21	DT	H6	3.63	4.91
21	DT	H2′1	21	DT	H6	1.96	2.64
24	DC3	H1′	24	DC3	H2′1	2.45	3.01
24	DC3	H1′	24	DC3	H2′2	1.91	2.59
23	DG	H1′	24	DC3	H6	3.08	4.16
23	DG	H2′1	24	DC3	H6	2.96	4.00
23	DG	H2′2	24	DC3	H5	3.14	4.26
8	DA	H2	25	HNE	H1	3.00	4.00

25	HNE	H22	25	HNE	H51	2.45	3.03
25	HNE	H22	25	HNE	H52	1.95	2.63
18	DA	H4′	25	HNE	Q7	2.85	3.85
19	DG	H4′	25	HNE	Q7	4.34	5.88
25	HNE	H4	25	HNE	Q7	2.50	3.50
2	DC	H3′	2	DC	H2′1	2.25	2.89
2	DC	H3′	2	DC	H2′2	2.26	3.06
3	DT	H6	2	DC	H2′1	2.52	3.21
3	DT	H6	2	DC	H3′	3.77	4.99
3	DT	H6	3	DT	H2′1	1.81	2.45
3	DT	M7	2	DC	H2′1	2.77	3.75
3	DT	M7	2	DC	H3′	3.54	4.78
3	DT	M7	3	DT	H2′1	3.06	4.14
3	DT	M7	3	DT	H3′	3.68	4.98
4	DA	H3′	4	DA	H2′1	1.91	2.59
4	DA	H3′	4	DA	H2′2	2.56	3.46
4	DA	H8	4	DA	H2′1	2.04	2.76
4	DA	H8	4	DA	H2′2	2.75	3.57
4	DA	H8	4	DA	H3′	2.97	3.91
5	DG	H3′	5	DG	H2′1	2.34	3.38
5	DG	H3′	5	DG	H2′2	2.48	3.36
5	DG	H8	4	DA	H2′1	3.22	4.04
5	DG	H8	4	DA	H2′2	2.42	3.34
5	DG	H8	4	DA	H3′	3.99	4.93
5	DG	H8	5	DG	H2′1	2.00	2.70
5	DG	H8	5	DG	H3′	3.86	5.22

6	DC	H6	5	DG	H3′	4.16	5.62
6	DC	H5	5	DG	H2′1	2.70	3.64
8	DA	H3′	8	DA	H2′1	2.20	2.98
8	DA	H3′	8	DA	H2′2	2.47	3.35
8	DA	H8	7	DGH	H8	4.50	5.50
8	DA	H8	8	DA	H2′2	2.98	3.54
8	DA	H8	8	DA	H3′	3.50	4.50
9	DG	H8	8	DA	H2′1	2.99	4.05
9	DG	H8	8	DA	H2′2	2.60	3.66
9	DG	H8	9	DG	H2′2	2.62	3.54
10	DT	M7	10	DT	H3′	4.50	5.50
11	DC	H3′	11	DC	H2′2	2.50	3.38
11	DC	H6	10	DT	H2′1	2.71	3.63
11	DC	H6	10	DT	H3′	3.58	4.84
11	DC	H6	10	DT	M7	3.84	4.98
11	DC	H6	11	DC	H2′1	2.13	3.03
11	DC	H6	11	DC	H2′2	3.08	4.16
12	DC3	H3′	12	DC3	H2′1	2.33	3.23
12	DC3	H3′	12	DC3	H2′2	2.58	3.28
12	DC3	H6	11	DC	H2′1	2.88	3.80
12	DC3	H6	11	DC	H2′2	2.41	3.41
12	DC3	H6	11	DC	H5	5.00	6.00
12	DC3	H6	12	DC3	H2′1	3.02	3.90
13	DG5	H3′	13	DG5	H2′1	2.21	2.99
13	DG5	H8	13	DG5	H2′1	2.17	3.07
14	DG	H8	13	DG5	H2′1	2.83	3.83

17	DT	M7	17	DT	H3′	3.41	4.61
18	DA	H3′	18	DA	H2′2	2.25	2.95
19	DG	H3′	19	DG	H2′1	2.14	3.00
19	DG	H3′	19	DG	H2′2	2.66	3.35
19	DG	H8	18	DA	H2′2	2.94	4.09
19	DG	H8	19	DG	H2′1	2.05	2.77
20	DC	H6	19	DG	H3′	4.28	5.80
21	DT	H3′	21	DT	H2′1	1.72	2.32
21	DT	H3′	21	DT	H2′2	2.85	3.85
21	DT	M7	20	DC	H3′	2.94	3.98
21	DT	M7	21	DT	H2′1	2.97	4.01
21	DT	M7	21	DT	H3′	3.80	5.04
22	DA	H2′1	22	DA	H1′	2.70	3.64
22	DA	H2′2	22	DA	H1′	2.09	2.83
22	DA	H3′	22	DA	H1′	3.08	4.16
22	DA	H3′	22	DA	H2′1	1.96	2.66
22	DA	H3′	22	DA	H2′2	2.37	3.21
22	DA	H8	22	DA	H2′1	2.14	2.71
22	DA	H8	22	DA	H2′2	2.81	3.67
22	DA	H8	22	DA	H3′	3.50	4.50
23	DG	H8	22	DA	H1′	2.83	3.83
23	DG	H8	22	DA	H2′1	3.38	4.42
23	DG	H8	22	DA	H2′2	2.67	3.61
23	DG	H8	22	DA	H3′	3.97	5.37
23	DG	H8	23	DG	H3′	3.15	4.92
24	DC3	H6	24	DC3	H2′1	1.95	2.63

25	HNE	Q6	25	HNE	H51	1.85	2.51
25	HNE	Q6	25	HNE	H52	2.32	3.14
25	HNE	Q7	25	HNE	Q6	2.57	3.47
25	HNE	Q8	25	HNE	Q6	2.10	2.84
1	DG5	H1′	2	DC	H2′1	3.11	4.21
1	DG5	H1′	2	DC	H2′2	3.98	5.24
2	DC	H1′	2	DC	H2′1	2.53	3.43
2	DC	H1′	2	DC	H2′2	2.22	2.72
2	DC	H1′	3	DT	H6	3.08	4.16
1	DG5	H3′	2	DC	H6	3.79	5.07
3	DT	H1′	3	DT	H2′1	2.25	3.05
3	DT	H1′	3	DT	H2′2	1.77	2.39
3	DT	H1′	3	DT	H3′	2.98	3.96
3	DT	H1′	3	DT	H6	3.05	4.13
3	DT	H1′	3	DT	M7	4.50	5.50
4	DA	H1′	4	DA	H2′1	2.68	3.62
4	DA	H1′	4	DA	H2′2	1.96	2.64
4	DA	H1′	4	DA	H3′	2.92	3.96
4	DA	H1′	5	DG	H8	2.80	3.72
5	DG	H1′	5	DG	H2′1	2.23	3.01
5	DG	H1′	5	DG	H2′2	2.36	3.13
5	DG	H1′	5	DG	H3′	3.37	4.55
6	DC	H1′	6	DC	H6	2.82	3.64
6	DC	H1′	6	DC	H5	4.06	5.48
6	DC	H1′	7	DGH	H8	3.25	4.41
6	DC	H3′	6	DC	H5	4.92	6.44

6	DC	H3′	7	DGH	H8	2.75	3.73
6	DC	H2′2	7	DGH	H8	1.81	2.45
7	DGH	H3′	7	DGH	H8	3.26	4.40
8	DA	H1′	8	DA	H2′1	2.47	3.35
8	DA	H1′	8	DA	H2′2	2.26	3.16
8	DA	H1′	8	DA	H3′	2.89	3.91
9	DG	H1′	9	DG	H2′2	1.72	2.32
9	DG	H1′	9	DG	H8	2.81	3.81
9	DG	H2′1	9	DG	H8	1.84	2.50
9	DG	H2′1	10	DT	H6	3.00	4.00
9	DG	H3′	9	DG	H8	3.50	4.50
9	DG	H3′	10	DT	H6	3.83	5.17
9	DG	H3′	10	DT	M7	2.92	3.94
10	DT	H1′	10	DT	H2′1	2.42	3.14
10	DT	H1′	10	DT	H2′2	2.21	2.99
10	DT	H1′	10	DT	H3′	3.00	3.92
10	DT	H1′	11	DC	H6	2.10	2.84
10	DT	H1′	11	DC	H5	3.86	5.22
11	DC	H1′	11	DC	H2′1	2.41	3.27
11	DC	H1′	11	DC	H2′2	2.46	3.19
11	DC	H1′	12	DC3	H5	3.33	4.51
12	DC3	H1′	12	DC3	H2′1	2.18	2.94
12	DC3	H1′	12	DC3	H2′2	2.31	3.13
13	DG5	H1′	13	DG5	H2′1	2.32	3.00
13	DG5	H1′	13	DG5	H8	3.08	4.18
13	DG5	H2′2	13	DG5	H3′	2.36	3.20

13	DG5	H2′2	14	DG	H8	2.57	3.27
14	DG	H2′1	14	DG	H3′	2.22	3.10
14	DG	H2′1	14	DG	H8	2.21	2.99
14	DG	H2′1	15	DA	H8	2.80	3.78
14	DG	H2′2	14	DG	H3′	2.19	2.81
15	DA	H2′1	15	DA	H3′	2.19	2.79
15	DA	H2′1	15	DA	H8	2.12	2.81
15	DA	H2′2	15	DA	H3′	2.31	3.13
15	DA	H2′2	15	DA	H8	2.95	3.81
16	DC	H2′1	17	DT	H3′	4.00	5.20
16	DC	H2′1	17	DT	M7	2.65	3.59
16	DC	H2′2	17	DT	H3′	2.90	3.92
16	DC	H2′2	17	DT	M7	2.29	3.09
16	DC	H3′	17	DT	H6	3.29	4.83
16	DC	H3′	17	DT	M7	2.64	3.56
17	DT	H1′	17	DT	M7	3.90	5.16
17	DT	H1′	18	DA	H8	3.50	4.50
17	DT	H1′	25	HNE	H1	4.00	5.00
17	DT	H2′1	17	DT	M7	3.12	4.22
18	DA	H1′	18	DA	H2′2	1.98	2.68
18	DA	H1′	18	DA	H3′	2.87	3.82
18	DA	H1′	25	HNE	H52	3.05	4.13
18	DA	H2′1	18	DA	H3′	1.98	2.68
18	DA	H2′1	19	DG	H8	3.52	4.56
19	DG	H1′	19	DG	H2′1	2.22	3.42
19	DG	H1′	19	DG	H2′2	2.05	2.77

19	DG	H1′	19	DG	H3′	2.94	4.12
19	DG	H1′	20	DC	H3′	4.50	5.50
19	DG	H1′	20	DC	H6	2.51	3.39
21	DT	H1′	21	DT	H2′1	2.65	3.59
21	DT	H1′	21	DT	H2′2	1.82	2.46
21	DT	H1′	21	DT	H3′	2.82	3.82
21	DT	H1′	21	DT	M7	3.85	5.21
20	DC	H2′2	21	DT	H6	2.07	2.81
20	DC	H2′2	21	DT	M7	2.32	3.14
23	DG	H1′	23	DG	H3′	3.79	5.13
23	DG	H2′1	23	DG	H8	2.11	2.67
18	DA	H5′1	25	HNE	H51	4.00	5.00
18	DA	H5′1	25	HNE	H52	3.13	4.23
18	DA	H5′1	25	HNE	Q7	3.35	4.53
18	DA	H5′1	25	HNE	Q8	2.89	3.92
19	DG	H5′1	25	HNE	H51	2.37	3.21
19	DG	H5′1	25	HNE	H52	3.31	4.40
19	DG	H5′1	25	HNE	Q8	3.03	4.09
18	DA	H4′	25	HNE	H52	2.40	3.20
18	DA	H4′	25	HNE	Q8	2.48	3.36
19	DG	H4′	25	HNE	H52	3.23	4.15
19	DG	H4′	25	HNE	Q8	3.49	4.55
25	HNE	H21	25	HNE	H52	1.70	2.70
25	HNE	H21	25	HNE	Q6	3.25	4.19
25	HNE	H22	25	HNE	Q6	3.30	4.46
25	HNE	H22	25	HNE	Q7	3.46	4.50

18	DA	H2	25	HNE	Q7	4.66	6.12
25	HNE	H3	25	HNE	Q7	3.91	5.29

M7 represents the methyl group of thymine; DGH represents HNE-dG adduct (X^7) without HNE moiety and the atoms are named as regular deoxyguanosine; HNE moiety is considered as a residue with residue No. 25, the atoms are named as following: X^7 H8: HNE H1; X^7 H7α(β): HNE H21(2); X^7 H6: HNE H3; X^7 H11: HNE H4; X^7 H12α(β): HNE H51(2); X^7 H13: HNE Q6; X^7 H14: HNE Q7; X^7 H15: HNE Q8; X^7 H16: HNE M9

Table S5. Backbone torsion angles of the rMD structure at pH 5.5.

Residue	Alpha	Beta	Gamma	Delta	Epsilon	Zeta	Chi
G^{1}			-175.2	150.9	178.3	-98.9	-100.9
C^{2}	-65.9	178.9	56.0	126.4	-175.3	-98.4	-123.6
T^{3}	-65.3	-179.3	53.6	133.8	-173.4	-95.0	-115.8
A^{4}	-69.6	-176.6	47.0	128.4	-177.7	-104.1	-115.3
G^{5}	-66.7	-172.8	51.3	127.6	-166.4	-96.4	-122.1
C^{6}	-69.5	178.4	53.3	138.3	-163.8	-117.3	-97.0
X^{7}	-78.3	-174.6	49.6	153.0	-88.5	178.0	114.3
A^{8}	-82.7	146.8	47.6	146.4	174.2	-100.6	-75.7
G^{9}	-67.2	-179.2	55.5	125.2	-170.6	-93.0	-122.0
T^{10}	-66.9	179.5	50.1	127.7	175.3	-102.5	-112.0
C^{11}	-60.0	-172.5	52.7	139.2	-166.8	-85.1	-113.6
C^{12}	-67.5	170.8	44.9	133.7			-104.5
G^{13}			179.9	149.5	-167.9	-110.5	-123.6
G^{14}	-69.7	-172.0	54.4	144.8	178.4	-100.6	-100.8
A^{15}	-65.8	-173.0	50.8	138.4	-179.5	-94.4	-105.3
C^{16}	-64.5	175.2	56.1	123.7	-177.0	-94.6	-123.5
T^{17}	-62.8	-179.2	53.8	116.9	-164.1	-93.5	-135.3
A^{18}	-65.6	-174.9	52.8	137.7	179.2	-98.4	-96.0
G^{19}	-64.1	176.4	54.7	126.1	-172.9	-107.8	-113.9
C^{20}	-68.2	-179.7	54.1	135.1	177.6	-100.9	-110.3
T^{21}	-64.6	-179.9	55.5	130.2	-173.8	-95.9	-125.7
A^{22}	-66.1	-172.6	48.0	134.5	-176.4	-102.8	-117.5
G^{23}	-68.2	-163.4	44.8	137.1	-175.2	-97.5	-102.3
C^{24}	-67.1	169.7	59.7	117.6			-126.5

alpha: O3′(i-1)-P-O5′-C5′; beta: P-O5′-C5′-C4′; gamma: O5′-C5′-C4′-C3′; delta: C5′-C4′-C3′-O3′; epsilon: C4′-C3′-O3′-P(i+1); zeta: C3′-O3′-P(i+1)-O5′(i+1); chi for pyrimidines: O4′-C1′-N1-C2; chi for purines: O4′-C1′-N9-C4

Table S6. Backbone torsion angles of the rMD structure at pH 8.9.

Residue	Alpha	Beta	Gamma	Delta	Epsilon	Zeta	Chi
G^{1}			-172.6	150.9	172.2	-92.7	-94.0
C^{2}	-63.7	174.6	55.2	120.2	-175.0	-89.8	-118.7
T^{3}	-65.0	165.3	57.9	100.5	-177.7	-93.8	-135.4
A^{4}	-61.0	-175.2	53.9	138.8	-171.2	-92.6	-102.9
G^{5}	-73.2	175.4	47.8	133.3	-170.6	-121.2	-100.4
C^{6}	-68.8	166.8	59.6	138.7	-84.1	-176.6	-110.9
X^{7}	-70.0	130.0	56.9	129.5	-175.6	-106.8	-122.1
A^{8}	-67.3	-176.5	52.5	134.7	-180.0	-103.6	-118.8
G^{9}	-68.6	-170.7	48.5	135.0	-177.9	-99.2	-113.3
T^{10}	-66.8	-179.8	52.1	132.3	177.9	-99.4	-109.4
C^{11}	-64.6	177.1	55.2	131.3	-174.8	-95.4	-121.3
C^{12}	-78.2	-179.7	43.0	81.9			-142.7
G^{13}			-169.3	145.2	-173.5	-95.7	-131.4
G^{14}	-69.7	-167.1	44.8	136.6	179.4	-98.7	-110.7
A^{15}	-65.4	-174.5	48.8	131.1	-175.2	-92.5	-112.4
C^{16}	-66.2	170.1	56.1	124.8	-165.7	-80.6	-118.5
T^{17}	-74.4	169.8	48.8	82.2	55.9	70.8	-118.5
A^{18}	-157.8	-160.8	52.6	131.3	-86.9	140.1	-90.8
G^{19}	-78.8	132.9	54.3	128.7	179.6	-101.8	-118.8
C^{20}	-65.5	-177.0	52.8	118.5	-174.9	-89.6	-122.5
T^{21}	-62.4	166.0	57.2	108.6	179.8	-93.3	-125.8
A^{22}	-59.0	176.6	56.5	135.0	-176.2	-98.6	-108.1
G^{23}	-70.4	-177.7	48.1	122.8	-168.3	-89.4	-113.0
C^{24}	-71.2	163.7	52.5	88.0			-128.4

alpha: O3′(i-1)-P-O5′-C5′; beta: P-O5′-C5′-C4′; gamma: O5′-C5′-C4′-C3′; delta: C5′-C4′-C3′-O3′; epsilon: C4′-C3′-O3′-P(i+1); zeta: C3′-O3′-P(i+1)-O5′(i+1); chi for pyrimidines: O4′-C1′-N1-C2; chi for purines: O4′-C1′-N9-C4

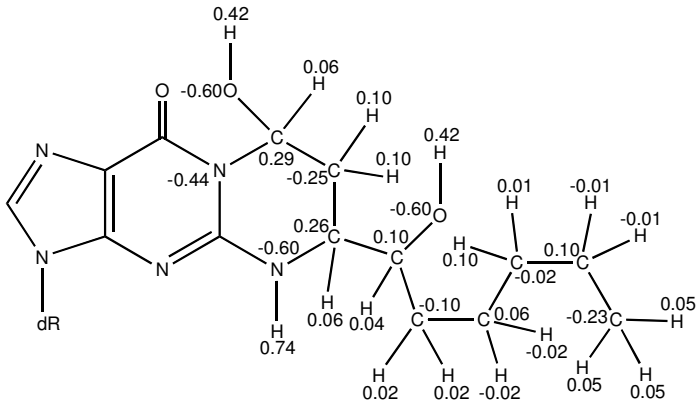

Figure S1. Force field parameters for the HNE-dG adduct obtained from DFT calculation.

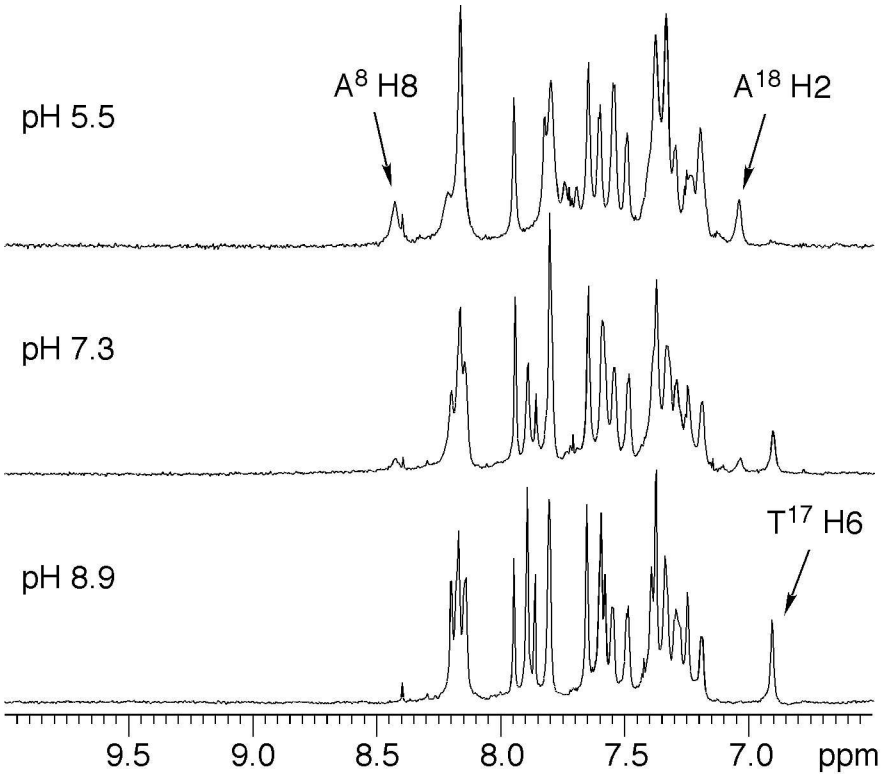

Figure S2. ^{1}H NMR spectra of the mismatched 5′-CpX-3′ duplex at different pHs indicating two conformations were in equilibrium. One was favored in acidic solution and the other was favored in basic solution.

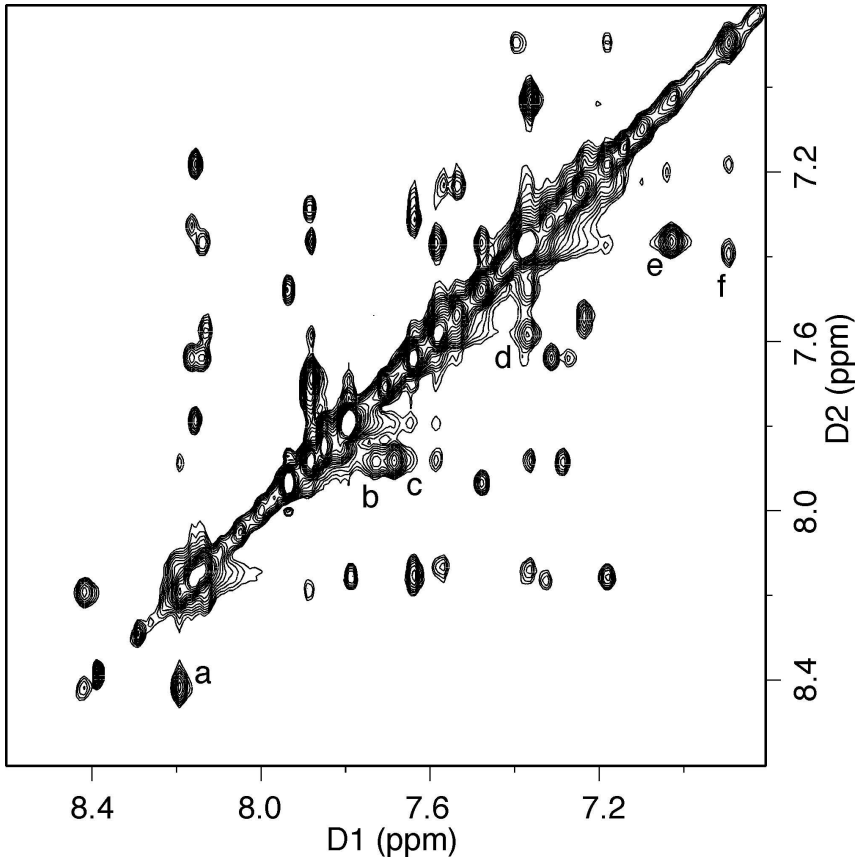

Figure S3. Two sets of resonances exhibiting exchange NOE interactions at pH 7.3, indicating the intercoversion of the two conformations. The assigned exchange crosspeaks are designated the resonances at the adduct region: (a) A^{18} H8, (b) G^{19} H8, (c) A^{8} H2, (d) X^{7} H2, (e) A^{18} H2, and (f) T^{17} H6.

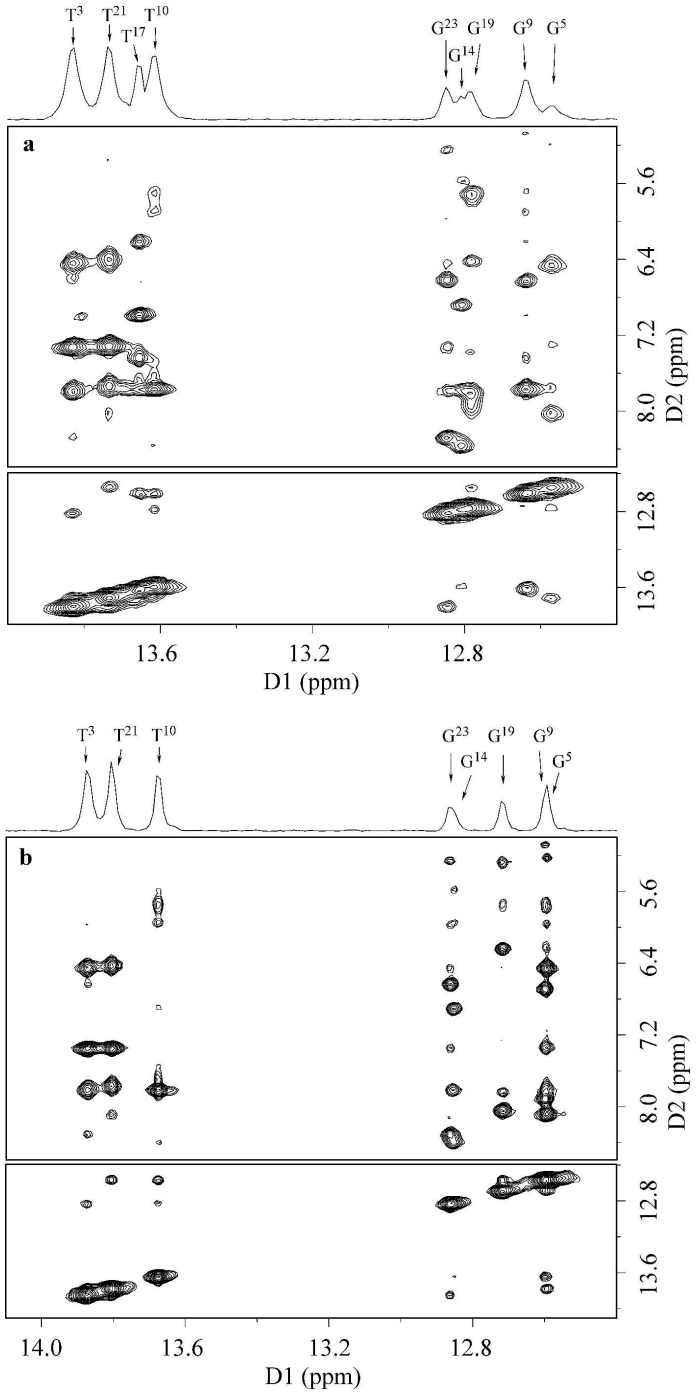

Figure S4. NOE cross peaks of the imino protons supporting the Watson-Crick hydrogen bonding: (a) At pH 5.5; (b) At pH 8.9.

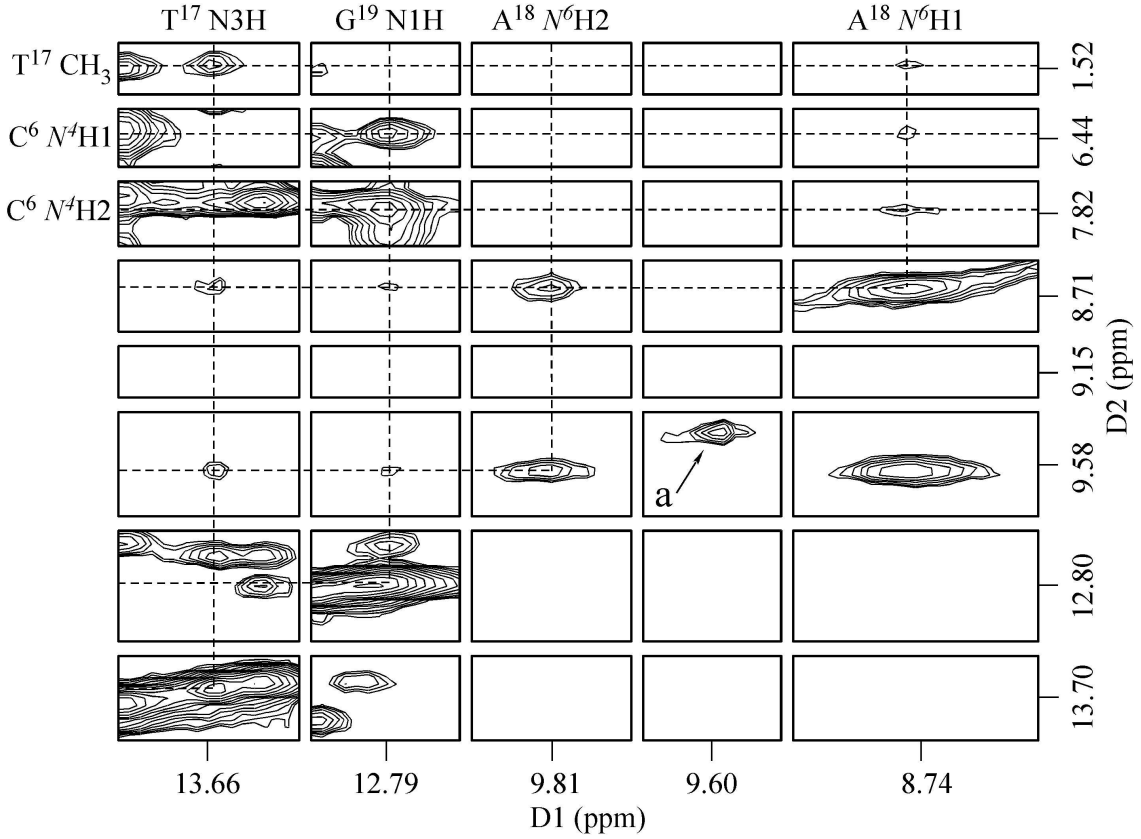

Figure S5. NOE cross peaks of the protonated A^{18} amino protons at pH 5.5. Extra resonance at 9.61ppm (peak a) was assigned to N^4H2 of partially protonated C^6.

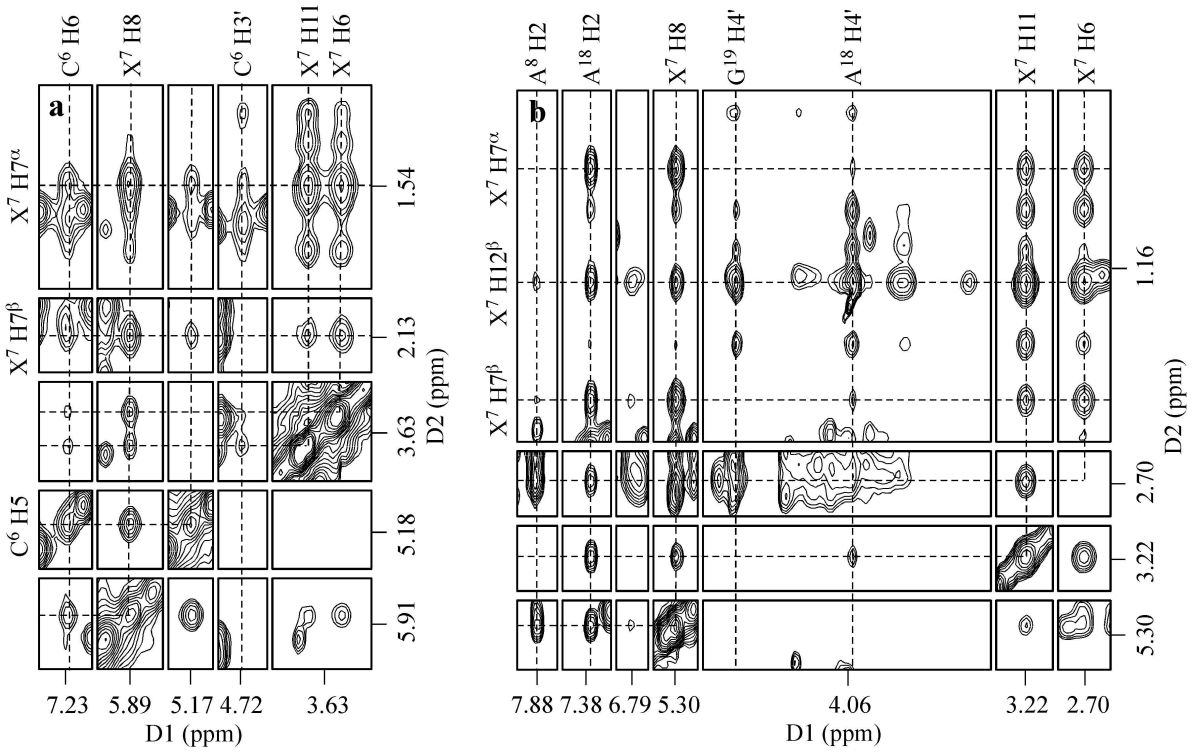

Figure S6. Some NOE crosspeaks associated with HNE protons: (a) At pH 5.5, HNE was located in the major groove; (b) At pH 8.9, HNE was located in the minor groove.

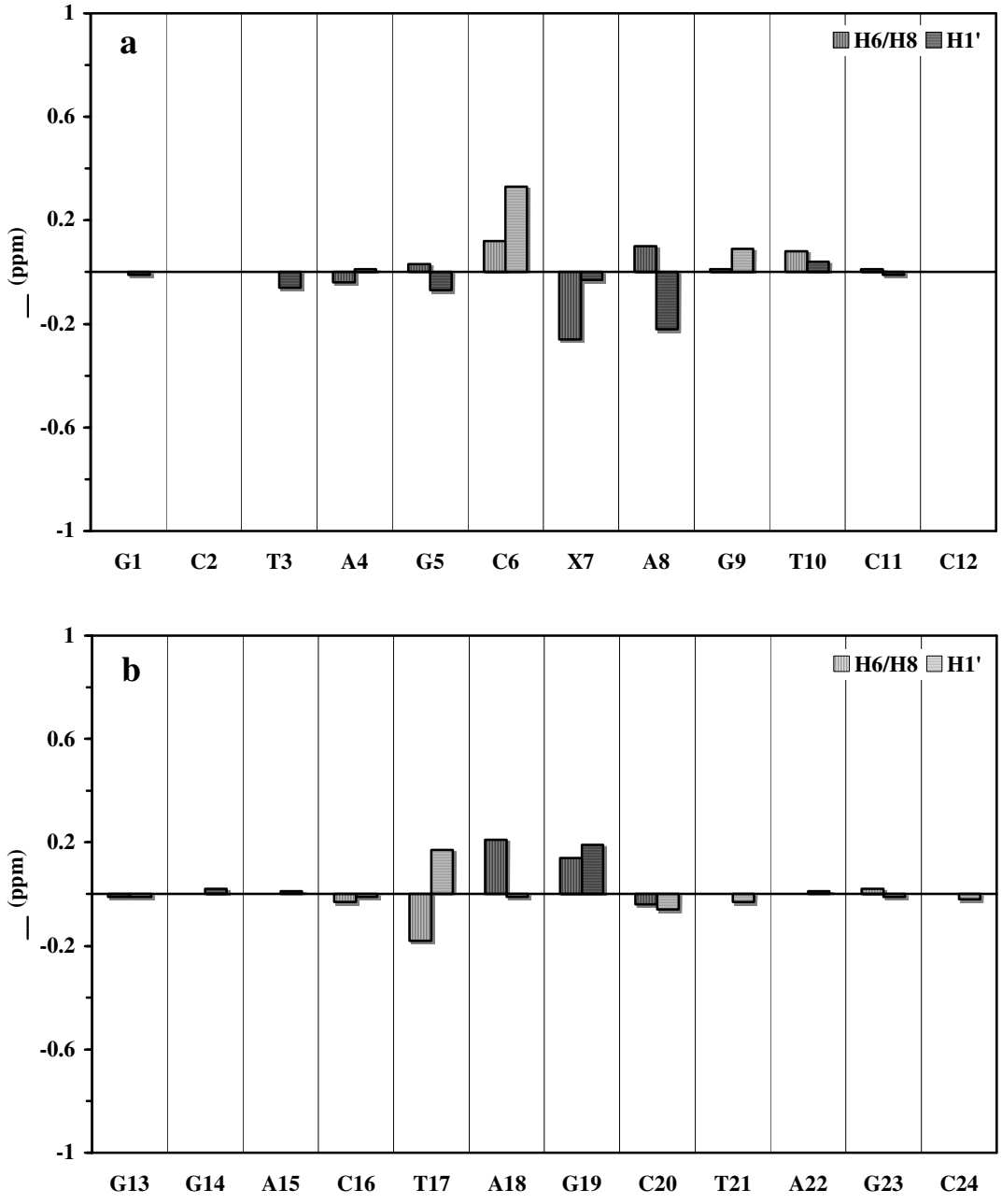

Figure S7. Chemical shift perturbations of the mismatched duplex at pH 8.9: (a) Adduct strand; (b) Complementary strand.

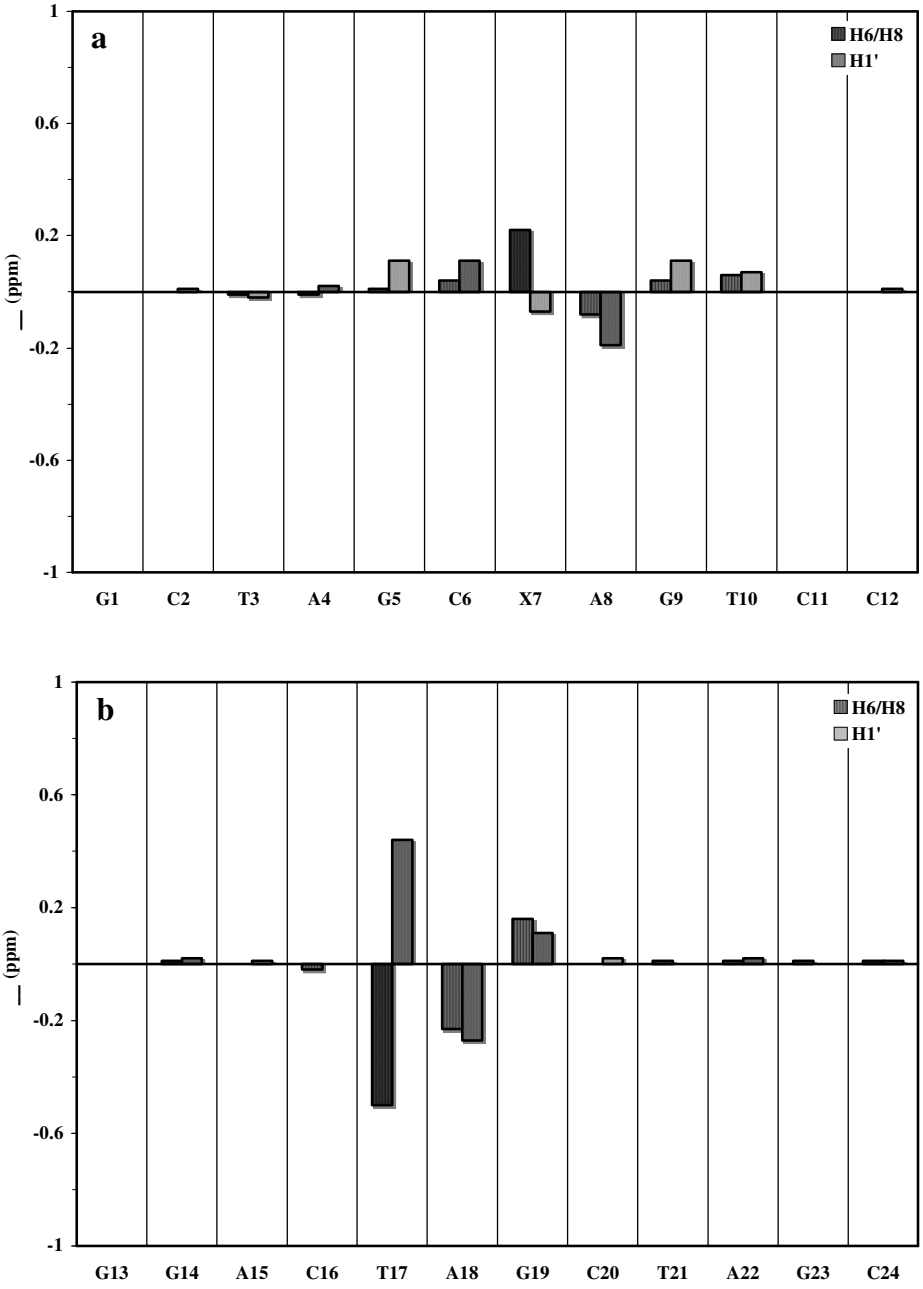

Figure S8. Chemical shift comparisons of the mismatched duplex between at pH 5.5 and 8.9: (a) Adduct strand; (b) Complementary strand.

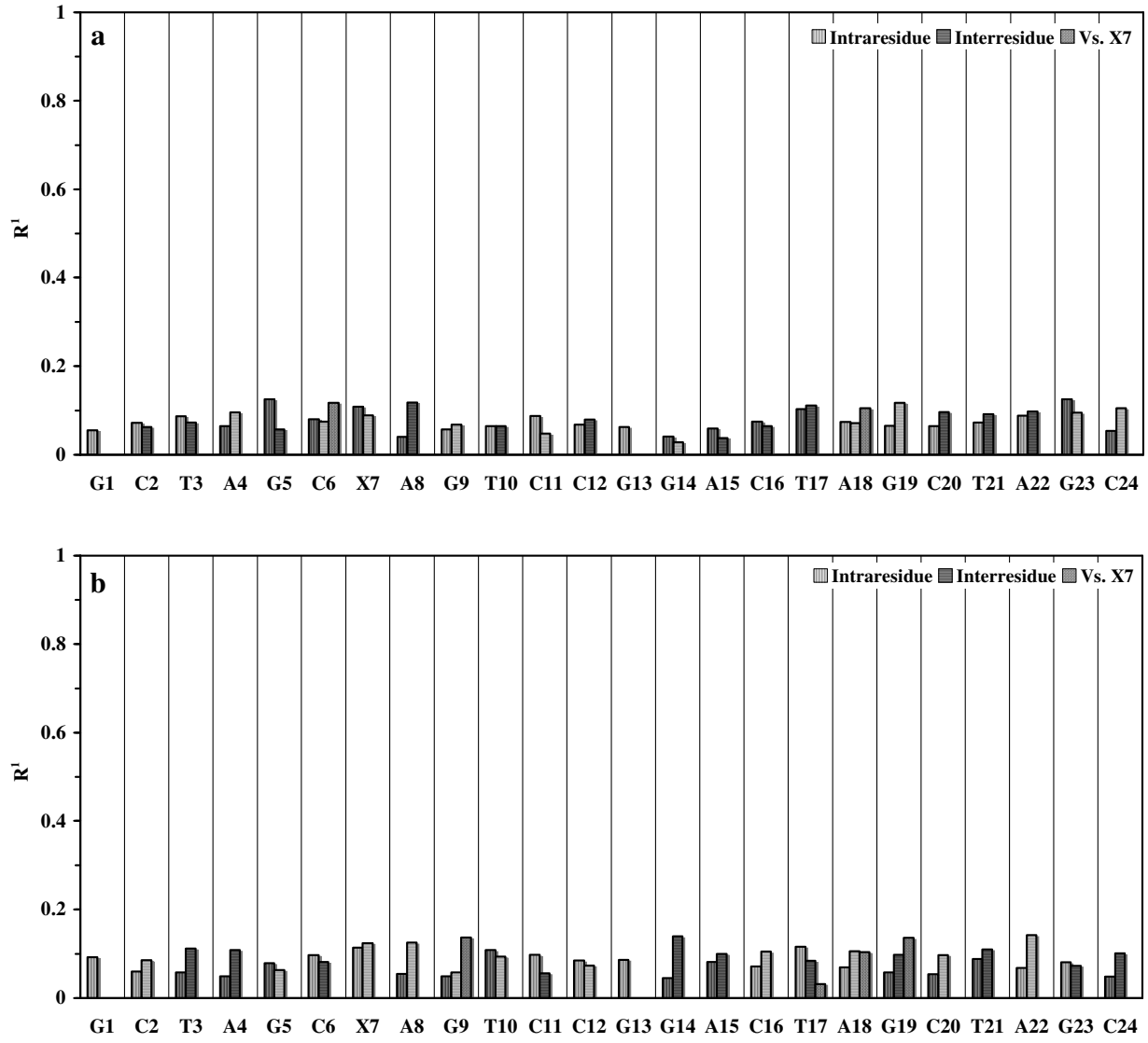

Figure S9. Residue-by-residue sixth root residuals (R_1^x): (a) At pH 5.5; (b) At pH 8.9.

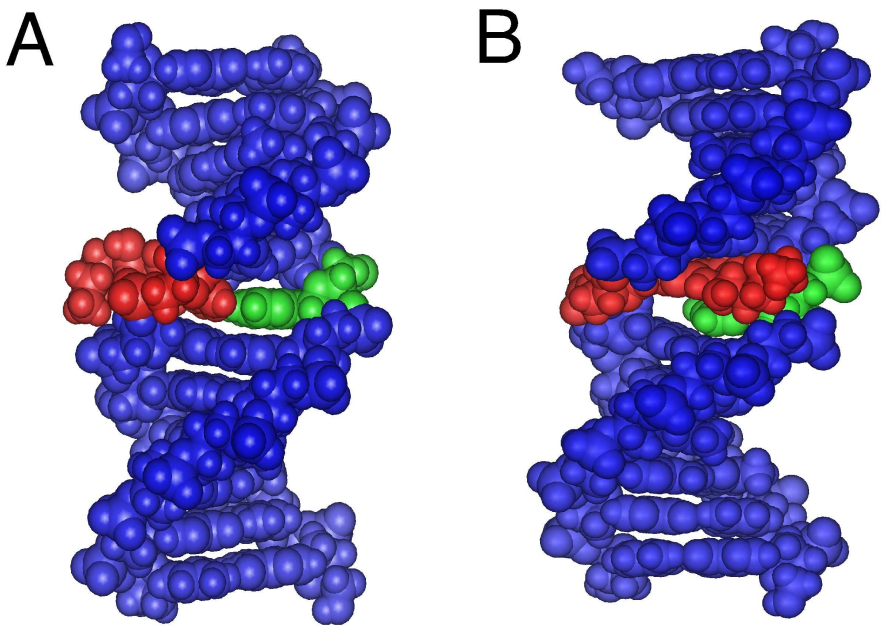

Figure S10. CPK models of average rMD structures of the mismatched 5′-CpX-3′ duplex. Red, green, and blue boxes represent X^7, HNE, and other nucleotides, respectively. (A) At pH 5.5. HNE was located in the major groove. (B) At pH 8.9. HNE was oriented towards the minor groove.