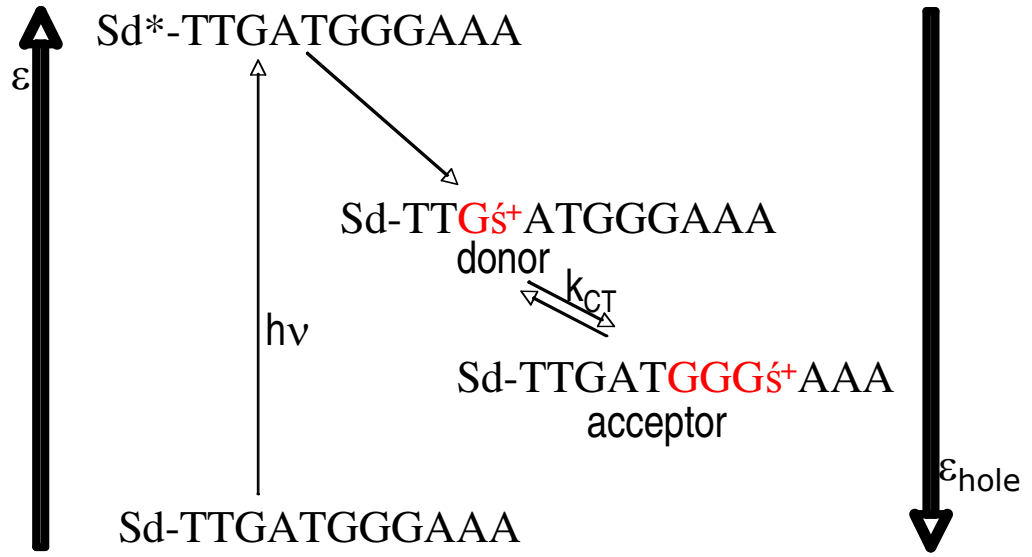

Figure S1. Hole formation and transfer between G bases of a DNA. Upon initial excitation due to light absorption, a hole is formed on a electron-donor linker, Sd, following by the hole moving to the single G base and then transferring to the G triplet. On the last stage of the process, the single G is the hole donor and the triple G is the hole acceptor (sink).

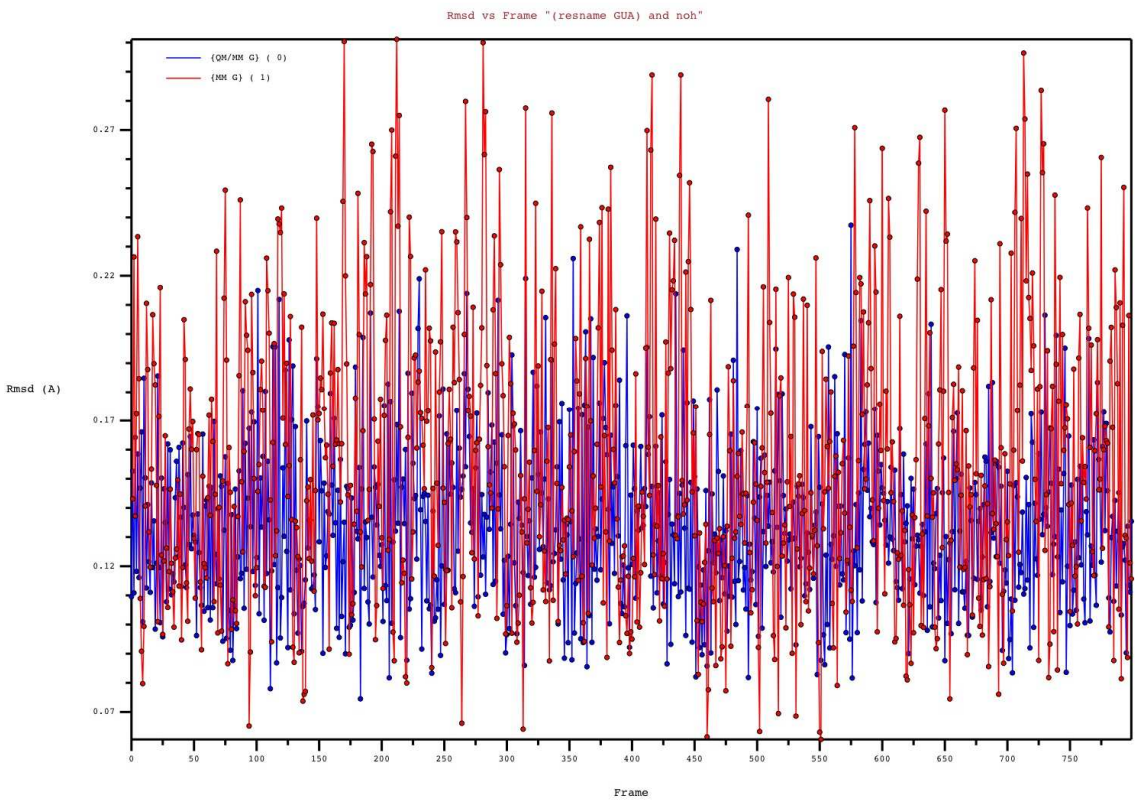

Figure S2. Dynamics of the **G** base RMSD from its initial geometry for 800 snapshots in the QM/MM (blue line) and the MM (red line) simulations.

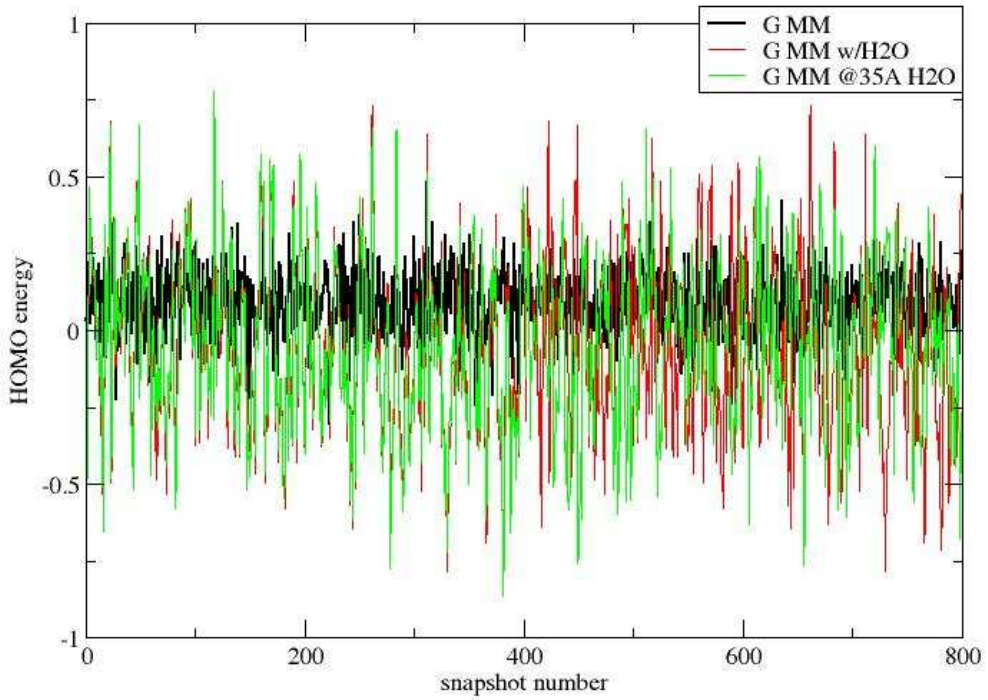

Figure S3. Time progression of the energy of the SOMO (or HOMO) for all 800 snapshots for G with and without water.

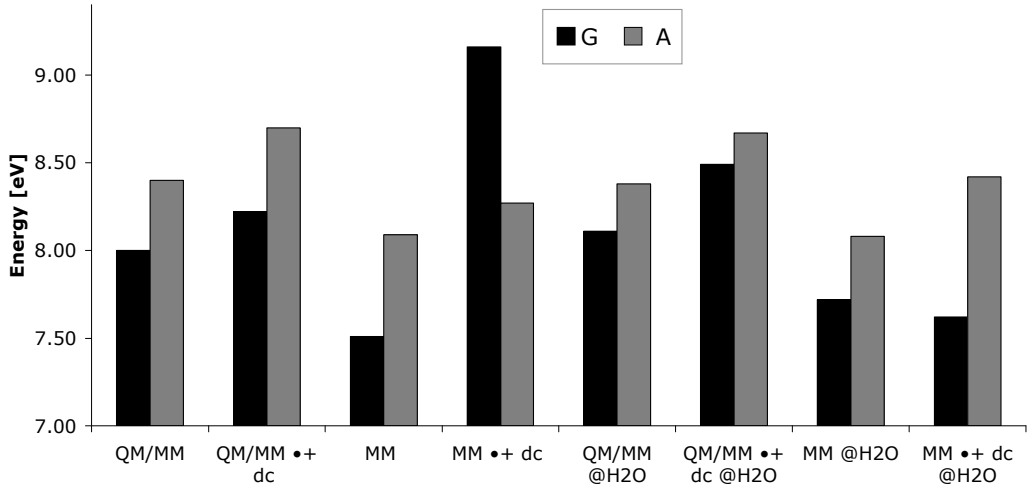

Figure S4. Comparing the SOMO (or HOMO) to LUMO energy difference between G and A, for all systems, with and without water shell.

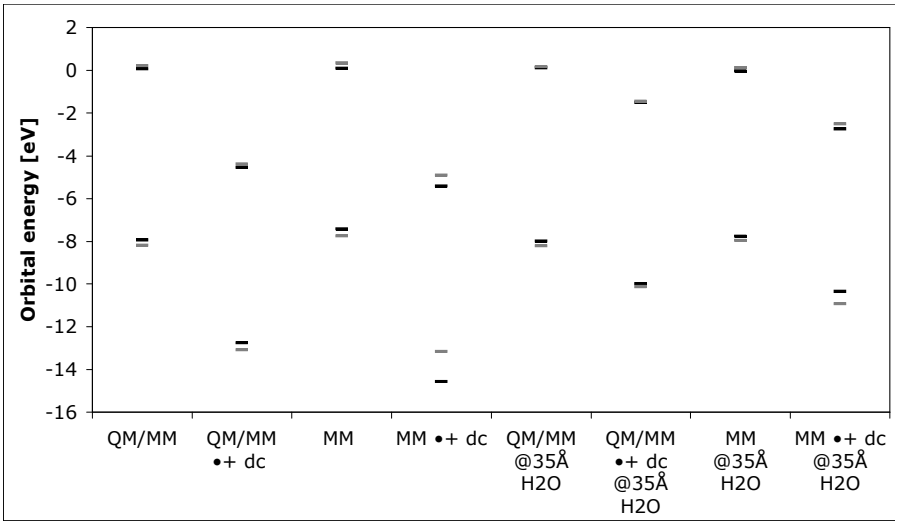

Figure S5. Plot of the SOMO/HOMO and LUMO energies for the 4 calculated systems, with and without water, for A (black) and G (grey).

Table S1. The average values (of 800 snapshots) and standard deviations of orbital energies for **G** and **A** bases. Water shell (if present) has a 35Å radius. All energies are eV.

	G HOMO/ SOMO energy	**G HOMO/ SOMO standard deviation**	**G LUMO energy**	**G LUMO standard deviation**	**A HOMO/ SOMO energy**	**A HOMO/ SOMO standard deviation**	**A LUMO energy**	**A LUMO standard deviation**
QMMM	-7.94	±0.08	0.06	±0.07	-8.19	±0.09	0.21	±0.06
QMMM @35Å H_2O	-7.99	±0.28	0.12	±0.28	-8.21	±0.28	0.17	±0.27
QMMM w/H_2O	-7.99	±0.29	0.11	±0.28	-8.20	±0.29	0.16	±0.28
QMMM dc	-12.76	±0.13	-4.54	±0.07	-13.09	±0.13	-4.39	±0.06
QMMM dc @35Å H_2O	-10.00	±0.29	-1.51	±0.28	-10.13	±0.30	-1.46	±0.28
QMMM dc w/H_2O	-10.01	±0.30	-1.52	±0.28	-10.75	±0.33	-1.41	±0.28
MM	-7.42	±0.14	0.09	±0.12	-7.75	±0.13	0.34	±0.09
MM @35Å H_2O	-7.76	±0.30	-0.04	±0.28	-7.96	±0.30	0.12	±0.28
MM w/H_2O	-7.73	±0.32	-0.05	±0.29	-7.93	±0.33	0.14	±0.29
MM dc	-14.58	±0.08	-5.42	±0.10	-13.17	±0.20	-4.90	±0.10
MM dc @35Å H_2O	-10.36	±0.25	-2.74	±0.26	-10.93	±0.35	-2.51	±0.29
MM dc w/H_2O	-10.36	±0.32	-2.75	±0.26	-10.08	±0.33	-2.45	± 0.29

Table S2. Average (of first 400 snapshots) and standard deviations of atomic charges for 3 cases: QM/MM Gdc with and without water shell, and MM G with water shell. Charges were calculated with INDO/s. Water shell (if present) has a 35Å radius, and water were described as point charges.

	QM/MM G dc No water		QM/MM G dc With water		QM/MM G dc (with water - without water)		MM G ground state With water		QM/MM G dc - MM G (with water)	
	Average	std. dev.	Average	std. dev.	Average difference	std. dev. difference	Average	std. dev.	Average difference	std. dev. difference
N9	-0.096	0.010	-0.093	0.012	0.002	0.002	-0.148	0.031	0.054	0.019
C4	0.295	0.007	0.297	0.008	0.002	0.001	0.168	0.019	0.129	0.010
H9	0.188	0.004	0.195	0.007	0.008	0.003	0.218	0.023	-0.022	0.016
N3	-0.339	0.009	-0.334	0.014	0.006	0.006	-0.434	0.016	0.101	0.002
C2	0.422	0.005	0.430	0.006	0.008	0.001	0.371	0.009	0.059	0.003
N1	-0.087	0.011	-0.073	0.012	0.014	0.001	-0.112	0.015	0.038	0.003
H1	-0.087	0.011	-0.073	0.012	0.014	0.001	-0.112	0.015	0.038	0.003
N2	0.187	0.005	0.203	0.007	0.016	0.003	0.171	0.006	0.031	-0.001
H21	-0.145	0.012	-0.136	0.016	0.009	0.004	-0.201	0.014	0.065	-0.002
H22	0.176	0.005	0.188	0.008	0.013	0.003	0.171	0.006	0.018	-0.002
C6	0.178	0.005	0.181	0.007	0.003	0.003	0.160	0.006	0.022	-0.001
O6	0.404	0.007	0.414	0.008	0.010	0.001	0.392	0.008	0.022	0.000
C5	-0.464	0.008	-0.496	0.016	-0.032	0.008	-0.640	0.019	0.144	0.003
N7	0.151	0.006	0.153	0.008	0.002	0.002	-0.020	0.018	0.173	0.010
C8	-0.257	0.008	-0.297	0.014	-0.040	0.006	-0.361	0.016	0.065	0.002
H8	0.277	0.006	0.265	0.010	-0.012	0.004	0.185	0.018	0.081	0.008

CHARMM charges for cations of GUA and ADE:

The method presented by MacKerell *et al*:

http://dogmans.umaryland.edu/~kenno/tutorial/

is used to parameterized CHARMM atomic charges. See also: "Parameterization of 2-thiouracil and 4-thiouracil in CHARMM all-atom empirical force field" J. Sarzyńska and T. Kuliński; *Comp. Meth. Sci. Tech.* 11(1), 49-55 (2005).

GUA with +1 charge:

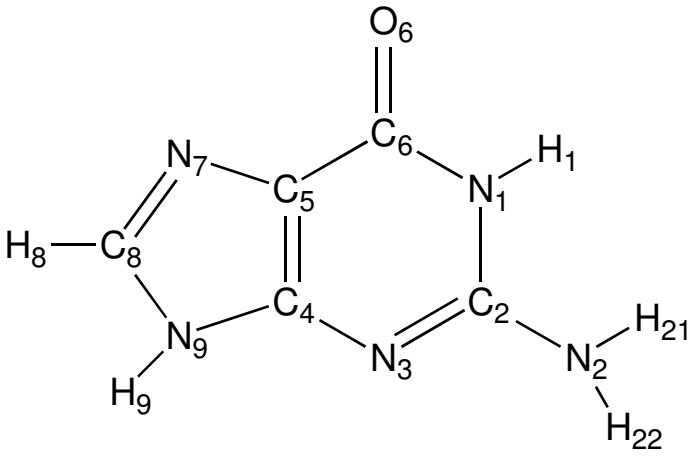

Table S3: New CHARMM partial atomic charge and atom type parameters for GUA +1 cation.

Atom name	Atom type	Atom charge
N9	NN2B	-0.17
C4	CN5	0.20
N2	NN1	-0.64
H21	HN1	0.40

H22	HN1	0.35
N3	NN3G	-0.55
C2	CN2	0.73
N1	NN2G	-0.34
H1	HN2	0.30
C6	CN1	0.52
O6	ON1	-0.30
C5	CN5G	0.01
N7	NN4	-0.66
C8	CN4	0.58
H8	HN3	0.32
H9	HN2	0.25

Table S4: Differences between *Ab initio* and empirical energies and distances between GUA +1 cation and water molecules (MM-QM).

Atom name	Distance difference [Å]	Energy difference [Kcal/mol]
H9	0.17	0.10
H1	0.16	-0.38
H21	0.10	-0.41
H22	0.15	0.03
N7	-0.24	-0.04
N3	-0.03	-0.20
O6	-0.22	-0.42

For ADE with +1 charge:

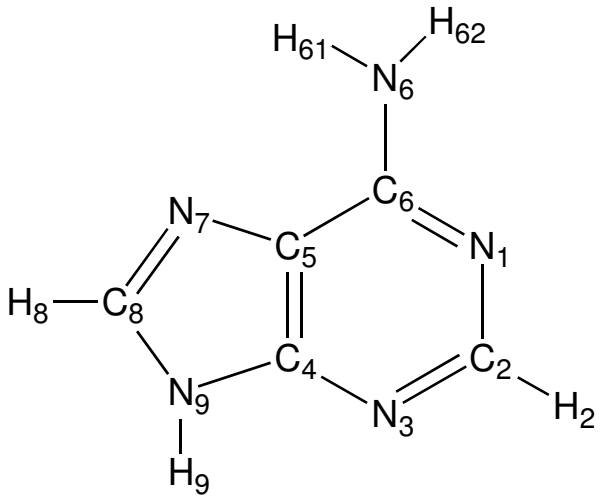

Table S5: New CHARMM partial atomic charge and atom type parameters for ADE +1 cation.

Atom name	Atom type	Atom charge
N9	NN2	-0.12
C5	CN5	-0.69
N7	NN4	-0.22
C8	CN4	-0.06
H8	HN3	0.50
N1	NN3A	-0.40
C2	CN4	0.18
H2	HN3	0.74
N3	NN3A	-0.61
C4	CN5	0.45
C6	CN2	0.52
N6	NN1	-0.50

H61	HN1	0.44
H62	HN1	0.42
H9	HN2	0.35

Table S6: Differences between *Ab initio* and empirical energies and distances between ADE +1 cation and water molecules (MM-QM).

Atom name	Distance difference [Å]	Energy difference [Kcal/mol]
H9	0.14	-0.25
H62	0.18	0.15
H61	0.17	0.04
N3	-0.08	0.37
N1	-0.20	0.17
N7	-0.11	0.42