Supplementary Information

The expectation values ($<S^2>$) for the doublet systems lie between 0.7617 and 0.7837 for both the optimization and energy calculations. This is somewhat above 0.75, which is the value expected for a radical system with no spin-contamination. It has been argued that spin-contamination is not well defined for the DFT approach and that $<S^2>$ should not be equal to $S_Z(S_Z+1)$. (J. A. Pople, P. M. W. Gill, N. C. Handy, *Int. J. Quantum Chem.*, **1995**, *56*, 303.

Table S1. The single point and zero point vibrational energies (ZPE) of the optimized complexes and single bases in hartrees (a.u.). The hydrogen bonded complexes are corrected for BSSE the values are given in kcal mol^{-1}. The molecular structures are shown in Schemes 2 and 3.

Systems	Energy/a.u.	ZPE/a.u. [a]	BSSE / kcal mol^{-1}
G	-542.61397	0.11473	x
G$^\bullet$	-541.96843	0.10061	x
G-N$_2$(-H)$^\bullet$	-541.96817	0.10137	x
G-O^{8}H-N^2(-H)$^\bullet$	-541.95689	0.10127	x
C	-394.96653	0.09669	x
C-N3(+H)$^+$	-395.33542	0.11054	x
C-N^4(-H)•	-394.30334	0.08301	x
C-N3(+H)-N^4(-H)	-394.96653	0.09669	x
G - C	-937.62262	0.21373	3.8
G$^{\bullet+}$ - C	-937.37676	0.21369	3.4
G$^\bullet$ - C(+H)+	-937.37432	0.21393	3.4
G - C-N^4(-H)-	-937.06788	0.19931	5.4
G-N^2(-H)- - C	-937.04511	0.19860	4.1
G - C-N^4(-H)•	-936.94797	0.19892	3.5
G-N^2(-H)• - C	-936.96710	0.20039	3.7
G$^\bullet$ - C-N3(+H)-N^4(-H)	-936.95762	0.20071	3.7
G$^\bullet$ - C (shifted)	-936.96522	0.19991	2.9
G - C + H$_2$O	-937.61659	0.21373	7.0
G$^{\bullet+}$ - C + H$_2$O	-937.37137	0.21369	6.7
G$^\bullet$ - C(+H)$^+$ + H$_2$O	-937.36896	0.21393	6.7
G$^\bullet$ - C (shifted) + H$_2$O	-936.96053	0.19991	7.2
H$_2$O	-76.42875	0.02095	x
H$_3$O$^+$	-76.69759	0.03375	x

[a] Corrected according to Wong.[30] x = not applicable.