Supporting information for: A combined QM/MM investigation on the light-driven electron-induced repair of the (6-4) thymine dimer catalyzed by DNA photolyase

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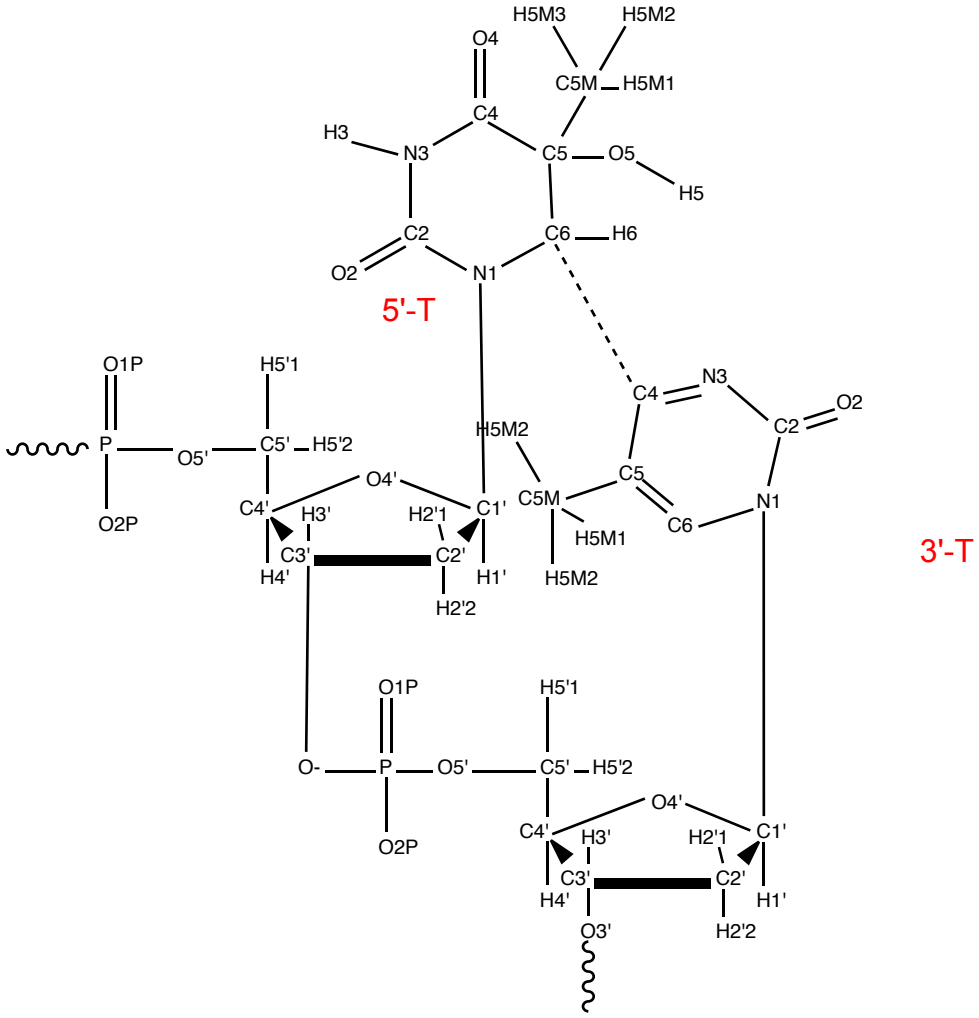

Figure S1: Atom labeling scheme of T(6-4)T-PP

Table S1: Amber atom types and charges for 3′ Thymine. RESP stands for restrained electrostatic potential derived charges.

Atom name	Atom type	3-′Thymine RESP
P	P	1.357204
O1P	O2	-0.891222
O2P	O2	-0.701483
O5’	OS	-0.573304
C5’	CT	0.099541
H5’1	H1	0.056165
H5’2	H1	0.041056
C4’	CT	0.433934
H4’	H1	-0.030180
O4’	OS	-0.517976
C1’	CT	0.173632
H1’	H2	0.118668
N1	N*	-0.225784
C6	CT	-0.288650
H6	H1	0.045069
C5	CT	0.394507
C5M	CT	-0.456855
H5M1	HC	0.119752
H5M2	HC	0.152372
H5M3	HC	0.158779
C4	C	0.819965
O4	O	-0.605398
N3	NA	-0.888421
H3	H	0.443692
C2	C	0.886495
O2	O	-0.676492
C3’	CT	0.612817
H3’	H1	-0.143936
C2’	CT	-0.243167
H2’1	HC	0.118770
H2’2	HC	0.085189
O3’	OS	-0.659577
O5	OH	-0.736047
H5	HO	0.461506

Table S2: Amber atom types and charges for 5′ Thymine. RESP stands for restrained electrostatic potential derived charges

Atom name	Atom type	5′-Thymine RESP
P	P	1.402640
O1P	O2	-0.824241
O2P	O2	-0.843788
O5’	OS	-0.589978
C5’	CT	0.088372
H5’1	H1	0.095771
H5’2	H1	0.033814
C4’	CT	0.270636
H4’	H1	-0.018549
O4’	OS	-0.536136
C1’	CT	0.475209
H1’	H2	0.051907
N1	N*	-0.582601
C6	CM	0.300920
H6	H4	0.137292
C5	CM	-0.359157
C5M	CT	-0.249970
H5M1	HC	0.096690
H5M2	HC	0.168649
H5M3	HC	0.097503
C4	CM	0.672092
N3	NC	-0.737938
C2	C	0.977512
O2	O	-0.689980
C3’	CT	0.615191
H3’	H1	-0.050049
C2’	CT	-0.302130
H2’1	HC	0.068873
H2’2	HC	0.121689
O3’	OS	-0.830864

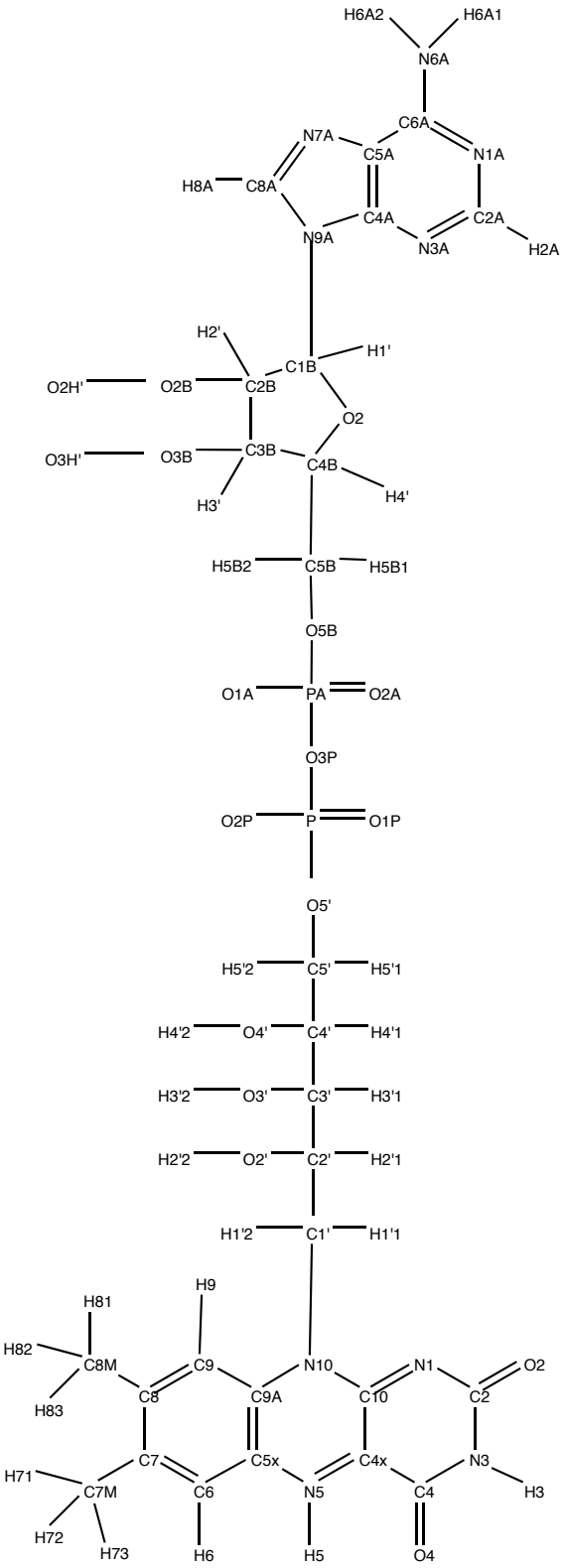

Figure S2: Atom labeling scheme of FADH

Table S3: Amber atom types and charges for $FADH^{\cdot}$ and $FADH^{-}$. RESP stands for restrained electrostatic potential derived charges.

Atom name	Atom type	$FADH^{\cdot}$ RESP	$FADH^{-}$ RESP
P	P	1.253618	1.251459
O1P	O2	-0.817104	-0.810741
O2P	O2	-0.829602	-0.816394
O3P	OS	-0.443156	-0.459921
PA	P	1.232827	1.237751
O1A	O2	-0.798282	-0.787938
O2A	O2	-0.859139	-0.855200
O5B	OS	-0.450030	-0.445070
C5B	CT	-0.014793	-0.032645
H5B1	H1	0.067898	0.081051
H5B2	H1	0.095104	0.101180
C4B	CT	0.254015	0.247580
H4'	H1	-0.038832	-0.029800
O4B	OS	-0.473372	-0.458106
C3B	CT	0.320749	0.360671
H3'	H1	0.087116	0.073251
O3B	OH	-0.625602	-0.628362
HO3'	HO	0.393633	0.393081
C2B	CT	0.133181	0.101579
H2'	H1	0.245860	0.233611
O2B	OH	-0.729672	-0.726716
HO2'	HO	0.422484	0.430845
C1B	CT	0.150370	0.123909
H1'	H2	0.108981	0.125058
N9A	N*	-0.257974	-0.265655
C8A	CK	0.439608	0.484669
H8A	H5	0.091818	0.078076
N7A	NB	-0.655906	-0.665206
C5A	CB	-0.215834	-0.230058
C6A	CA	0.680875	0.709818
N6A	N2	-1.084446	-1.063487
H6A1	H	0.430942	0.419246
H6A2	H	0.450882	0.444182
N1A	NC	-0.943747	-0.960351
C2A	CQ	0.601928	0.623055
H2A	H5	0.078945	0.073702
N3A	NC	-0.896015	-0.905331
C4A	CB	0.680875	0.709818
N1	N*	-0.844470	-0.774129
C2	C	0.988906	0.925587
O2	O	-0.725475	-0.624118

Table S3 cont.: Amber atom types and charges for $FADH^{\cdot}$ and $FADH^{-}$. RESP stands for restrained electrostatic potential derived charges.

Atom name	Atom type	$FADH^{\cdot}$ RESP	$FADH^{-}$ RESP
N3	NA	-0.813953	-0.779968
H3	H	0.390250	0.414936
C4	C	0.856898	0.816341
O4	O	-0.763743	-0.639363
C4x	CM	-0.467608	-0.223546
N5	N*	-0.407042	-0.323935
H5	H	0.323797	0.345137
C5x	CB	0.224603	0.156918
C6	CA	-0.534020	-0.445904
H6	HA	0.206485	0.222408
C7	CA	0.169184	0.157910
C7M	CT	-0.243494	-0.283547
H72	HC	0.059295	0.079810
H71	HC	0.063156	0.092788
H73	HC	0.045637	0.077964
C8	CA	0.067818	0.142616
C8M	CT	-0.241102	-0.326756
H82	HC	0.042490	0.085767
H81	HC	0.066358	0.107061
H83	HC	0.065693	0.101311
C9	CA	-0.447694	-0.367191
H9	HA	0.311474	0.287761
C9A	CB	0.173370	0.162293
N10	N*	-0.077641	-0.087947
C10	CM	0.532025	0.599379
C1’	CT	-0.263010	-0.246535
H1’1	H1	0.183267	0.140751
H1’2	H1	0.065544	0.090318
C2’	CT	0.287764	0.344396
H2’1	H1	0.007670	-0.001528
O2’	OH	-0.803080	-0.796467
H2’2	HO	0.490052	0.491518
C3’	CT	0.418257	0.430569
H3’1	H1	-0.006512	-0.036289
O3’	OH	-0.891452	-0.896015
H3’2	HO	0.473310	0.479218
C4’	CT	0.129975	0.142114
H4’1	H1	0.027467	0.031997
O4’	OH	-0.785140	-0.793168
H4’2	HO	0.479441	0.494045
C5’	CT	0.460101	0.460090
H5’1	H1	-0.007244	-0.010043
H5’2	H1	-0.089890	-0.091884
O5’	OS	-0.637139	-0.633684

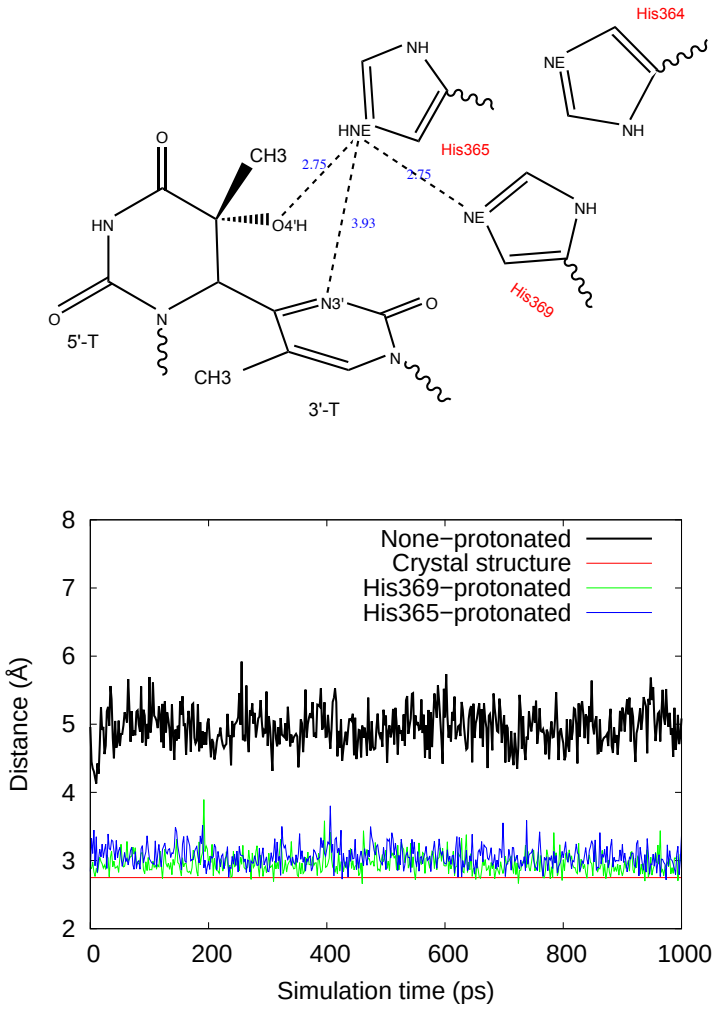

Figure S3: Structure of the covalently linked thymine bases of the T(6-4)-PP and three Histidine residues, His365, His369, and His364. The NE(His365)-NE(369) as a function of simulation time is plotted. The corresponding distance in the crystal structure is shown by the red line (2.75).

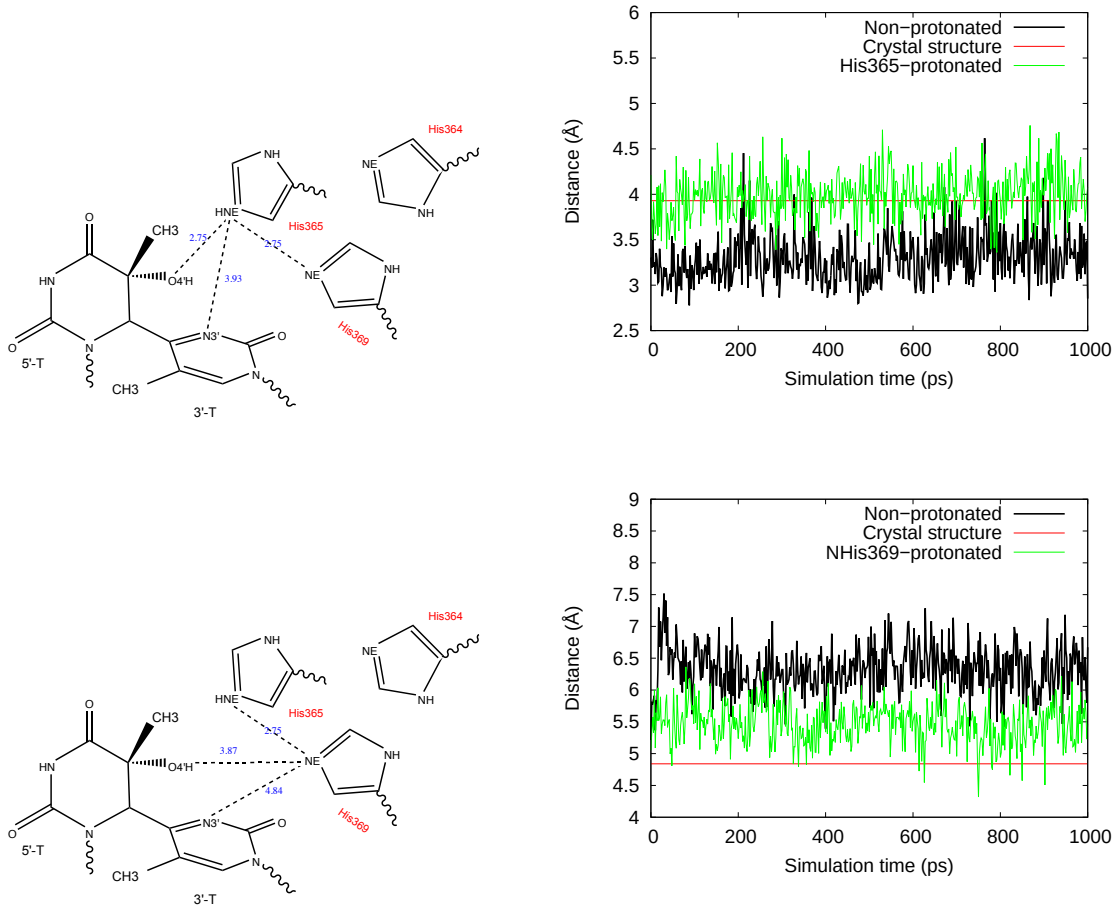

Figure S4: Time evolution of the NE(His365)-N3' and NE(His369)-N3' distances for protonated His365 and protonated His369, upper and lower panel, respectively.

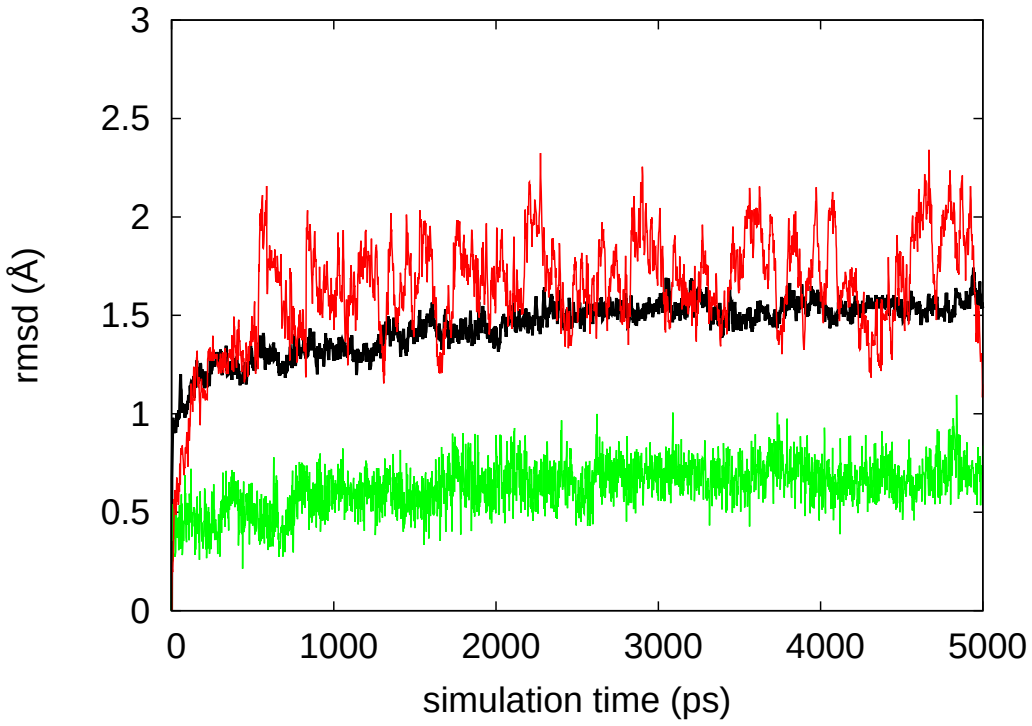

Figure S5: Root-mean-square deviation (RMSD) as a function of simulation time for DNA photolyase (black), DNA (red), and binding region defined in the text (green).

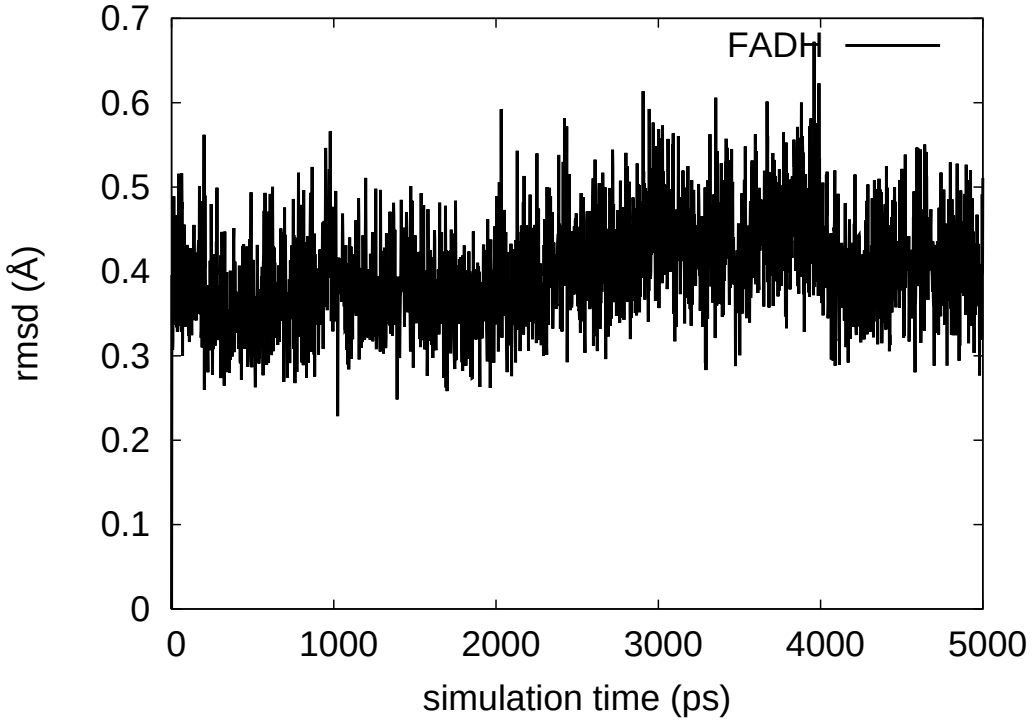

Figure S6: Root-mean-square deviation (RMSD) as a function of simulation time for FADH

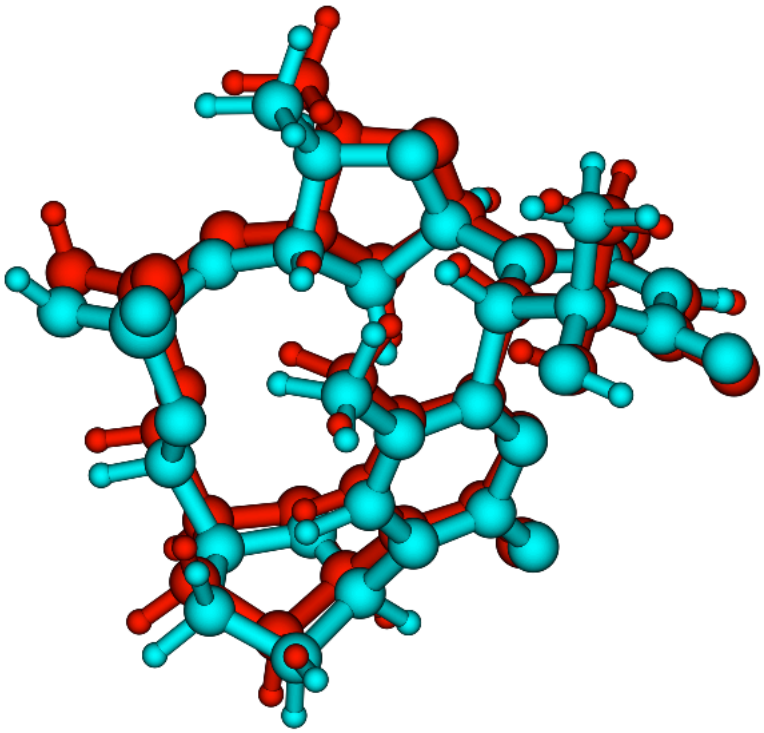

Figure S7: Superposition of the average structure of the T(6-4)T dimer and close by His365 after 5 ns productive MD run (cyan) and crystal structure (red)

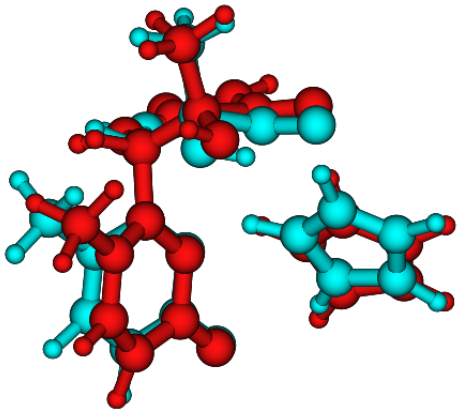

Figure S8: QMMM optimized structure of the QM region (cyan) vs. crystal structure (red).

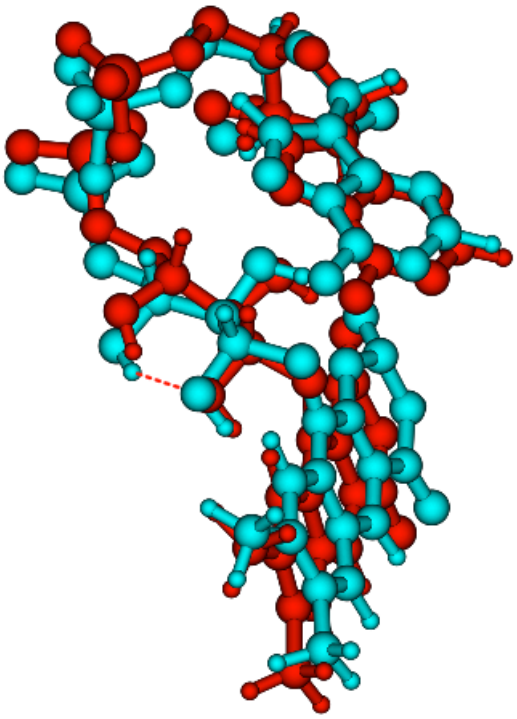

Figure S9: Superposition of the average structure of the FADH after 5 ns productive MD run (cyan) and crystal structure (red)