Table S 6 Geometrical parameters of anhydrous theophylline dimer IV obtained from DFT and MP2 Calculations

Geometrical Parameters	B3LYP/6-311++G(d,p)	M06/6-311++G(d,p)	MP2/6-311++G(d,p)
N1-C2 (Å)	1.41230	1.40685	1.41648
C2-N3 (Å)	1.38668	1.38162	1.37986
N3-C4 (Å)	1.37331	1.36974	1.37571
C4-C5 (Å)	1.38455	1.37669	1.38903
C5-C6 (Å)	1.42162	1.41722	1.42884
C6-N1 (Å)	1.40580	1.40012	1.39768
C5-N7 (Å)	1.38269	1.37663	1.37156
N7-C8 (Å)	1.34848	1.34293	1.35863
C8-N9 (Å)	1.33195	1.32566	1.33843
N9-C4 (Å)	1.35500	1.34807	1.35870
N1-C10 (Å)	1.46933	1.45863	1.46707
C2-O11 (Å)	**1.21748**	**1.20900**	**1.22349**
N3-C12 (Å)	1.46563	1.45504	1.46203
C6-O13 (Å)	**1.23770**	**1.22839**	**1.23970**
N7-H (Å)	**1.03050**	**1.03137**	**1.03089**
C8-H (Å)	**1.07911**	**1.08127**	**1.08107**
C2-N1-C6 (deg)	126.39435	126.74741	127.17255
N1-C2-N3 (deg)	117.02810	116.73666	116.70904
C2-N3-C4 (deg)	119.79231	119.92903	120.08658
N3-C4-C5 (deg)	121.53124	121.41201	121.24566
C4-C5-C6 (deg)	123.06446	123.27932	123.13060
C5-C6-N1 (deg)	112.18954	111.89557	111.65556
N9-C4-C5 (deg)	111.60347	111.76568	111.80053
C4-C5-N7 (deg)	104.98955	105.08575	105.22832
C5-N7-C8 (deg)	106.06314	105.76848	106.19895
N7-C8-N9 (deg)	113.55588	113.85101	113.43725
C8-N9-C4 (deg)	103.78797	103.52907	103.33496
C6-N1-C10 (deg)	118.76751	118.73183	118.72003
C10-N1-C2 (deg)	114.83814	114.52076	114.10742
N1-C2-O11 (deg)	121.04549	121.12528	120.80087
O11-C2-N3 (deg)	121.92640	122.13807	122.49009
C2-N3-C12 (deg)	118.29280	118.21800	117.97306
C12-N3-C4 (deg)	121.91489	121.85296	121.94036
N3-C4-N9 (deg)	126.86530	126.82231	126.95381
N9-C8-H (deg)	124.38921	124.34537	124.71211
H-C8-N7 (deg)	122.05491	121.80362	121.85064
C5-N7-H (deg)	**127.84413**	**127.66570**	**127.91199**
	125.55696*	**125.42734***	
N7-C5-C6 (deg)	**131.94599**	**131.63493**	**131.64108**
	131.20457*	**130.77576***	
C5-C6-O13 (deg)	**126.56113**	**126.51180**	**126.25270**
	126.04142*	**126.04975***	
O13-C6-N1 (deg)	121.24933	121.59262	122.09174

***values corresponds to anhydrous Theophylline Monomer**

Table S 7 Geometrical parameters of the anhydrous theophylline dimer II obtained from DFT Calculations

Geometrical parameters	B3LYP/6-311++G(d,p)	M06/6-311++G(d,p)
N1-C2 (Å)	1.40612	1.40086
C2-N3 (Å)	1.39879	1.392.84
N3-C4 (Å)	1.37220	1.36739
C4-C5 (Å)	1.37462	1.36695
C5-C6 (Å)	1.43083	1.42700
C6-N1(Å)	1.41773	1.41309
C5-N7(Å)	1.38192	1.37727
N7-C8 (Å)	1.35013	1.34503
C8-N9 (Å)	1.33430	1.32786
N9-C4 (Å)	1.36491	1.35675
N1-C10(Å)	1.46973	1.45910
C2-O11(Å)	1.21685	1.20818
N3-C12(Å)	1.46219	1.45099
C6-O13 (Å)	1.22093	1.21242
N7-H (Å)	1.00939	1.01032
C8-H (Å)	1.08246	1.08476
C2-N1-C6 (deg)	126.90137	127.41104
N1-C2-N3 (deg)	116.97108	116.54794
C2-N3-C4 (deg)	119.47977	119.59758
N3-C4-C5 (deg)	121.71730	121.85978
C4-C5-C6 (deg)	123.89312	123.89098
C5-C6-N1(deg)	111.03735	110.69269
N9-C4-C5 (deg)	111.10107	111.31625
C4-C5-N7 (deg)	105.00127	104.99896
C5-N7-C8 (deg)	107.22469	106.97435
N7-C8-N9 (deg)	111.92630	112.12565
C8-N9-C4 (deg)	104.74666	104.58478
C6-N1-C10 (deg)	117.89919	117.81542
C10-N1-C2 (deg)	115.19944	114.77355
N1-C2-O11 (deg)	121.04788	121.28606
O11-C2-N3 (deg)	121.98104	122.16600
C2-N3-C12 (deg)	119.63349	120.01877
C12-N3-C4 (deg)	120.88674	120.38365
N3-C4-N9 (deg)	127.18163	126.82397
N9-C8-H (deg)	123.57049	123.18000
H-C8-N7 (deg)	124.50320	124.69435
C5-N7-H (deg)	125.58655	125.50222
N7-C5-C6 (deg)	131.10561	131.11006
C5-C6-O13 (deg)	125.99386	126.14439
O13-C6-N1 (deg)	122.96879	123.16292

Table S 8 Rotational constants (GHz) and zero-point vibrational energy (kJ mol^{-1}) of the two theophylline dimer Form IV, II and M

Rotational Constants/ZPVE	Calculated values at M06/6-311++G(d,p) level		
	Dimer Form IV	Dimer Form II	Dimer Form M
A (GHz)	1.965787	2.196841	1.261867
B (GHz)	0.390083	0.349766	0.317291
C (GHz)	0.326650	0.302273	0.254040
ZPVE*	843.44	842.69	969.59

*values obtained at B3LYP/6-311++G(d,p) level