Published in "Dyes and Pigments 92(1): 714–723, 2012"
which should be cited to refer to this work.

SUPPORTING INFORMATION

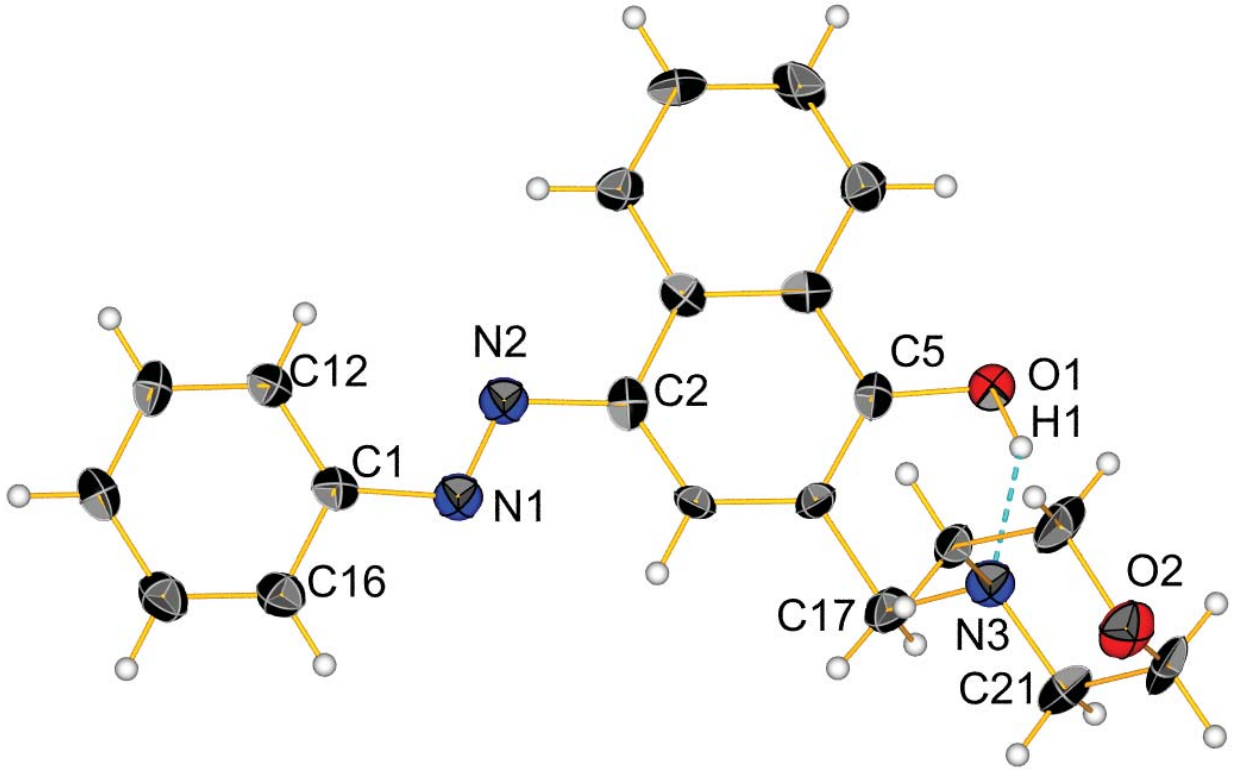


Figure S1. View of the molecular structure of **3** with hydrogen bond indicated as dashed blue line, 50% probability.

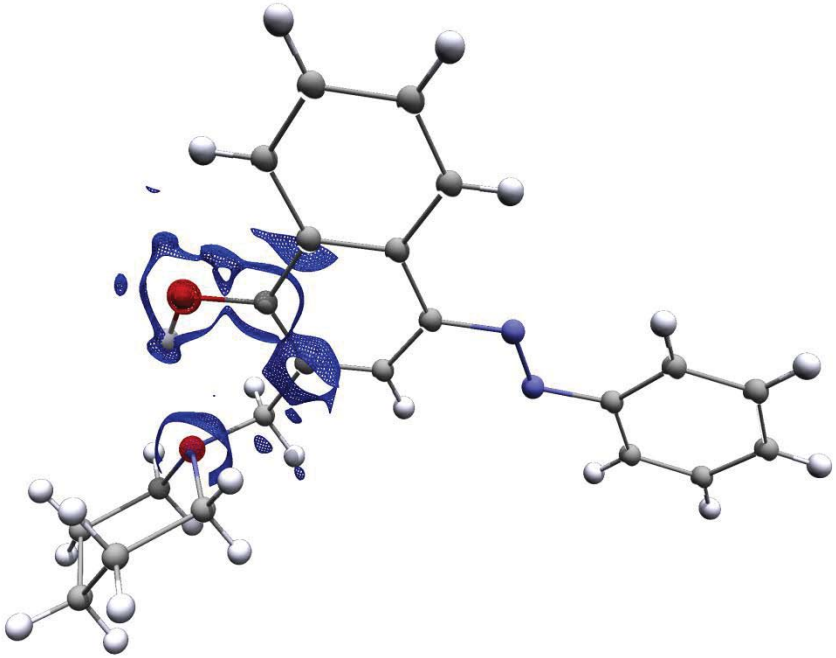


Figure S2. Fourier map of **3**.

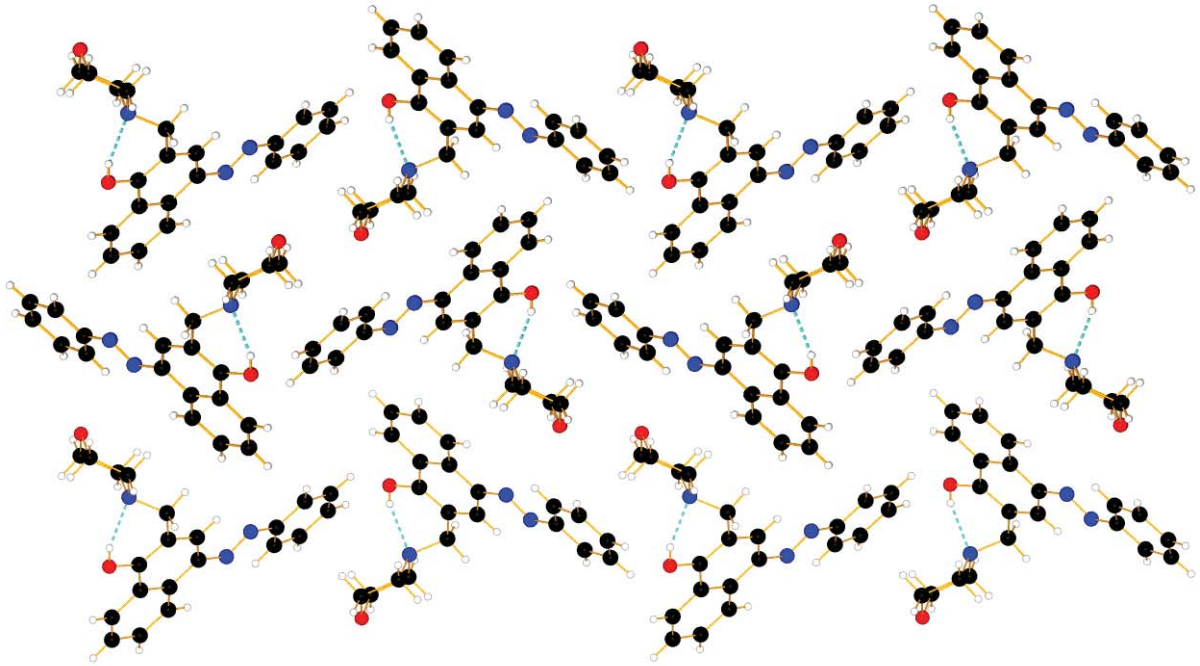


Figure S3. View along the *a* axis of the molecular structure of **3** with hydrogen bonds indicated as dashed blue lines.

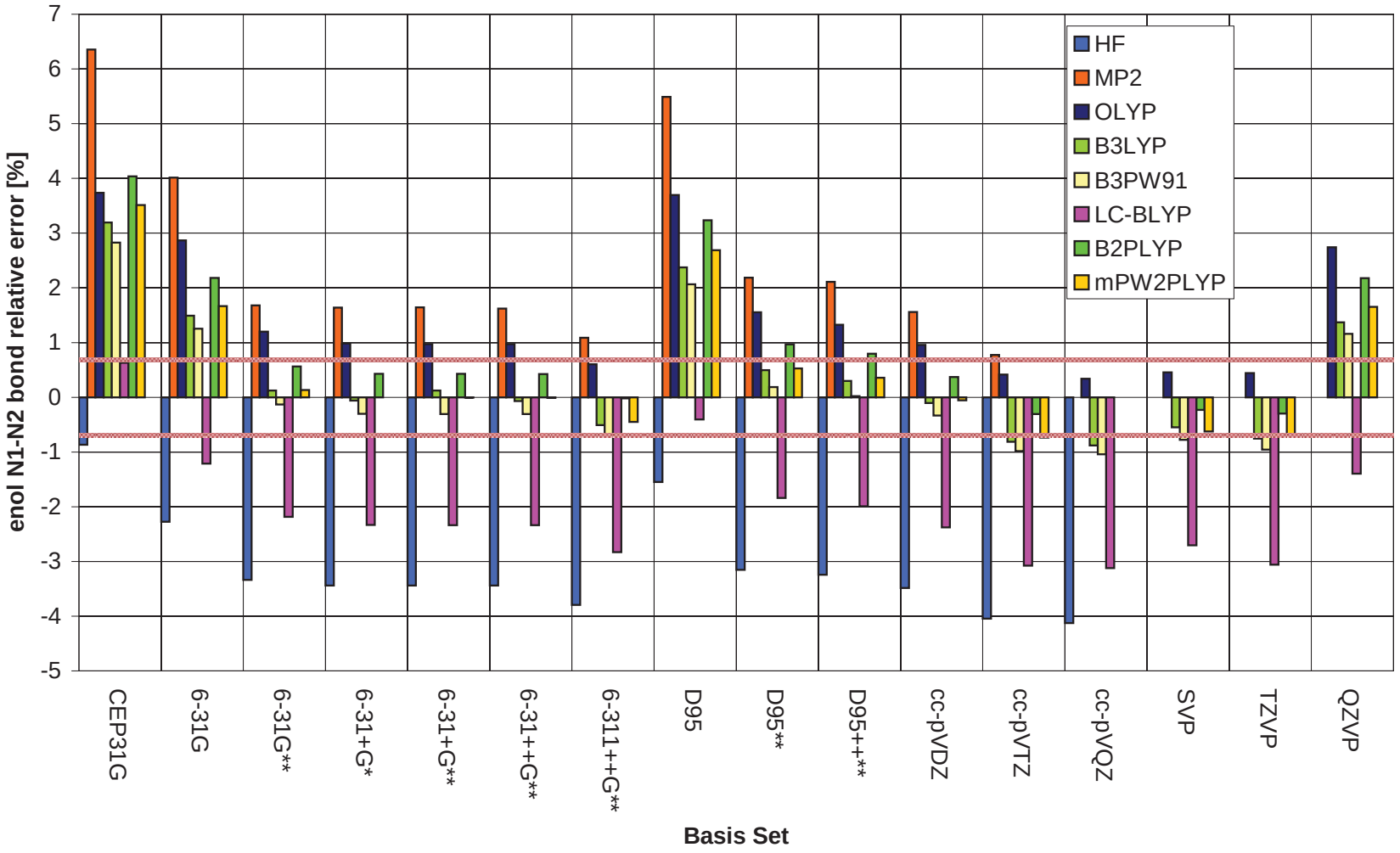


Figure S4. Relative deviations between the X-ray (Table 2) and computational values for the enol N1-N2 bond. The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The standard deviations of the X-ray determination are marked with red line.

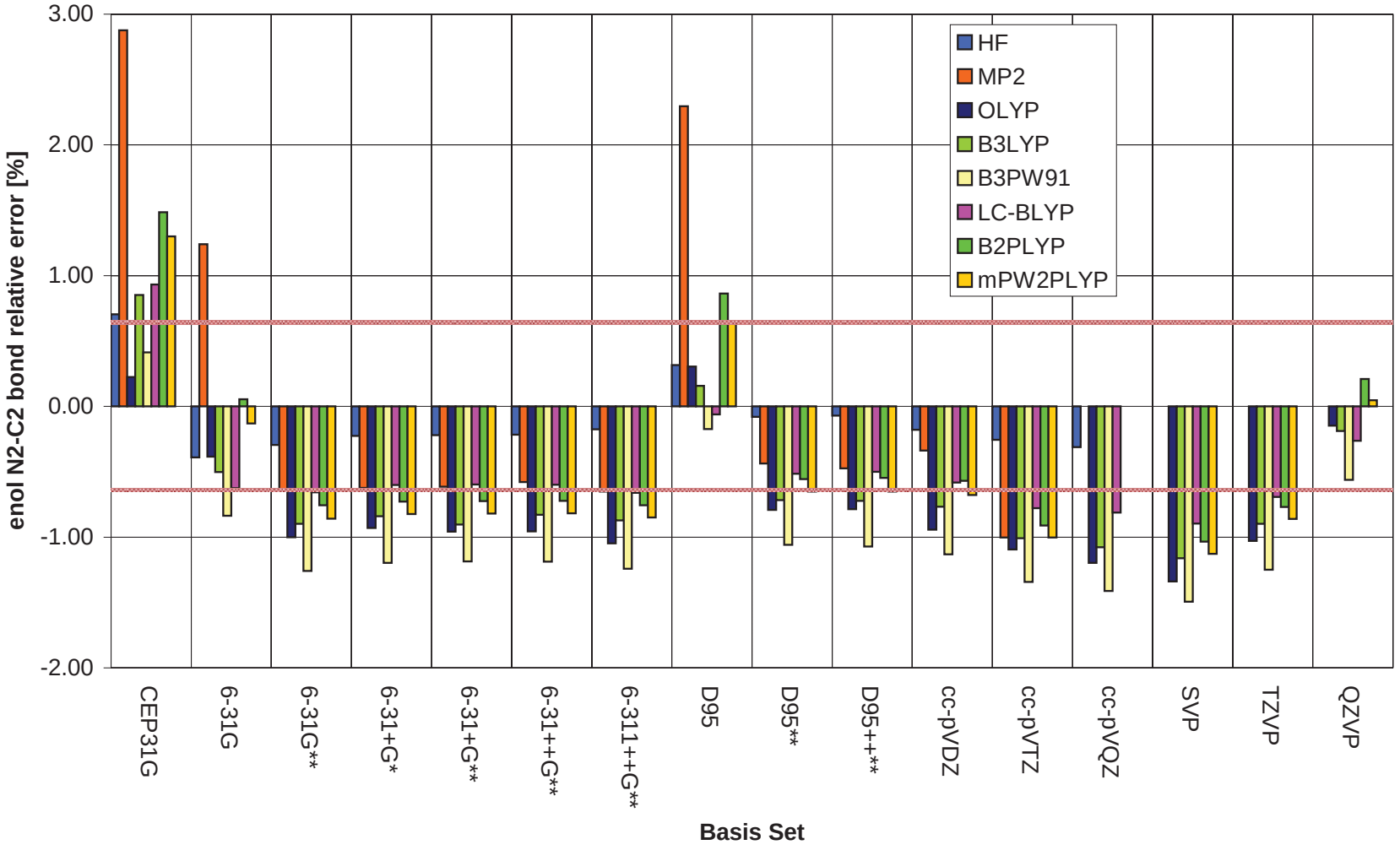


Figure S5. Relative deviations between the X-ray (Table 2) and computational values for the enol N2-C2 bond. The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The standard deviations of the X-ray determination are marked with red line.

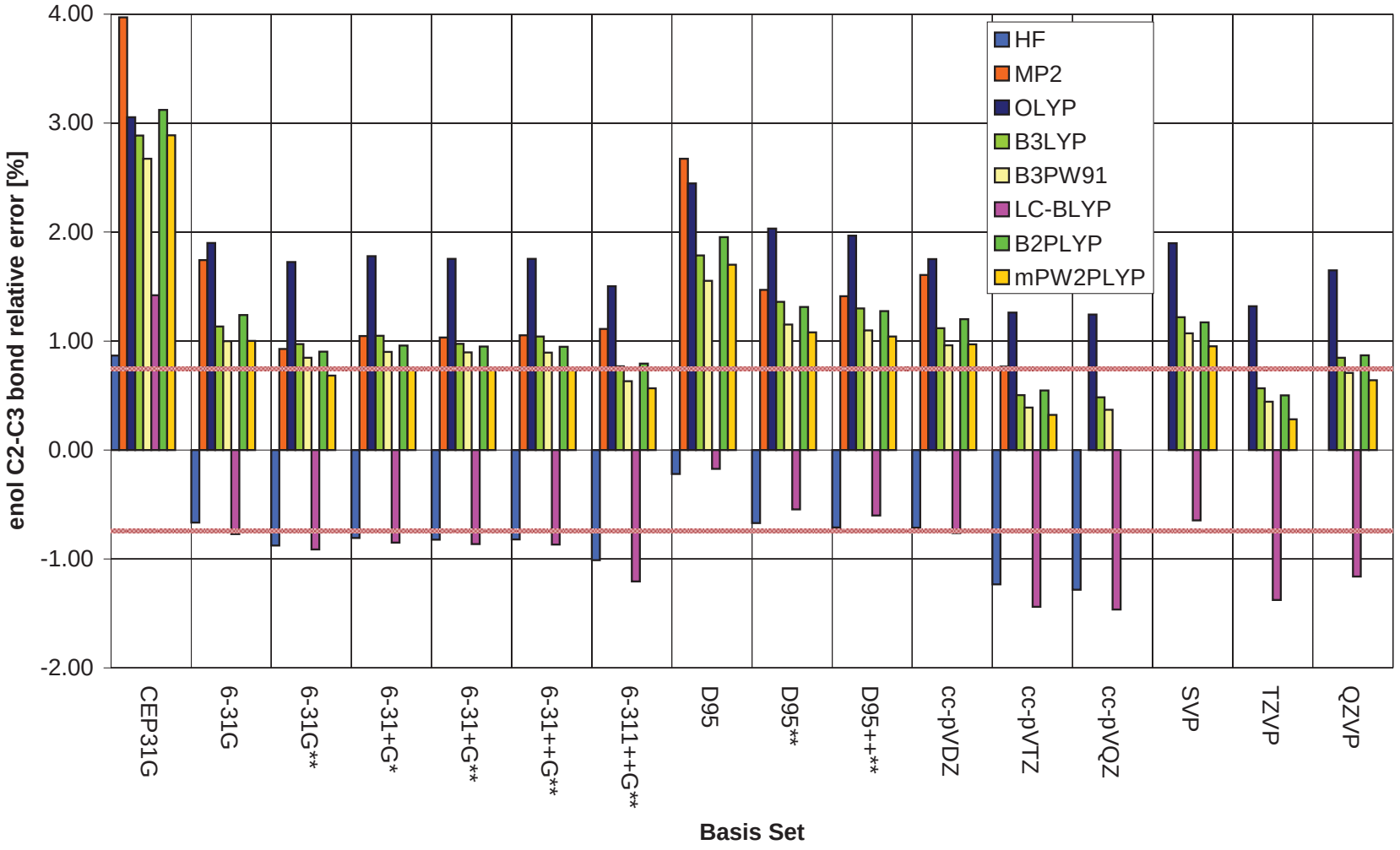


Figure S6. Relative deviations between the X-ray (Table 2) and computational values for the enol C2-C3 bond. The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The standard deviations of the X-ray determination are marked with red line.

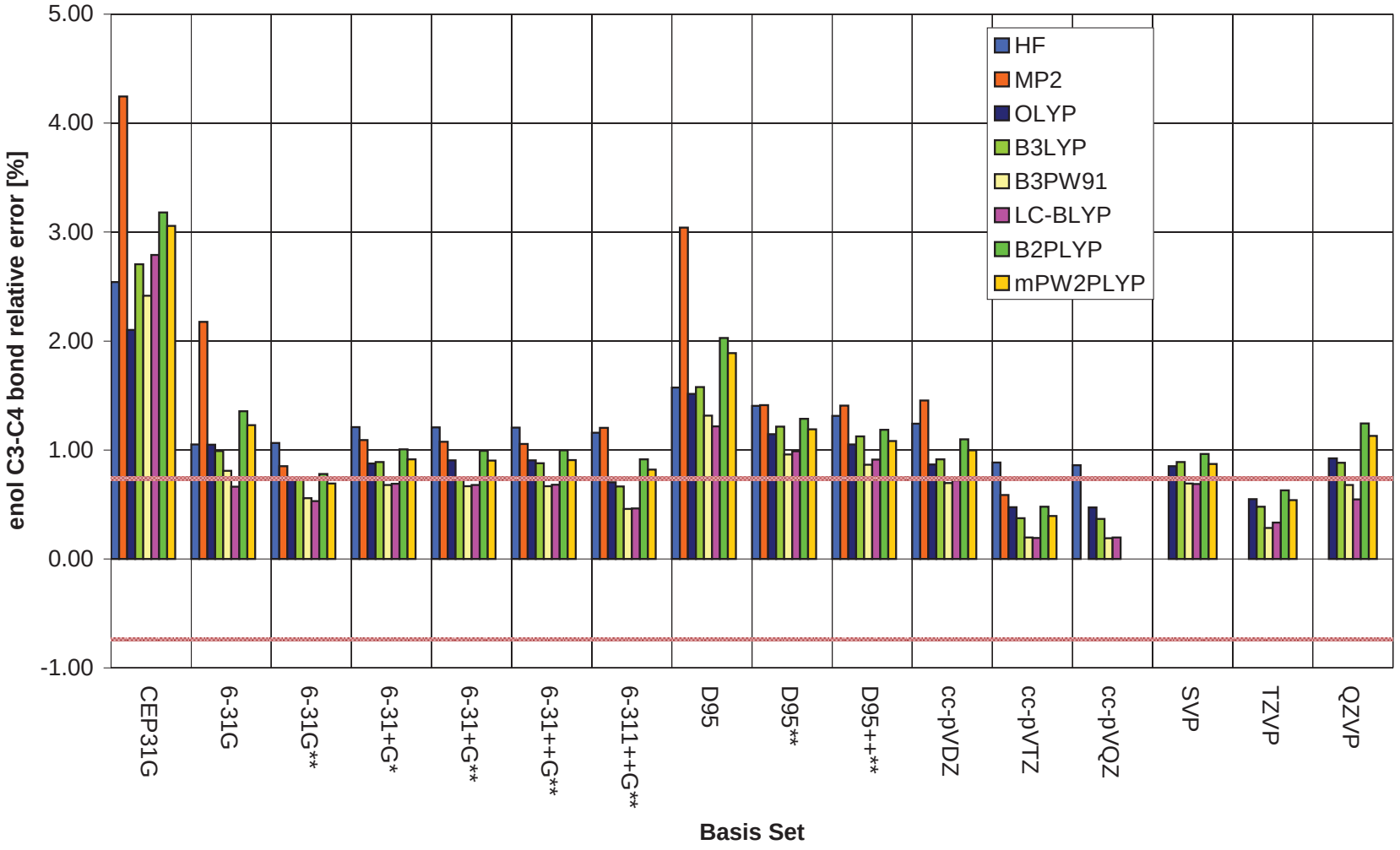


Figure S7. Relative deviations between the X-ray (Table 2) and computational values for the enol C3-C4 bond. The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The standard deviations of the X-ray determination are marked with red line.

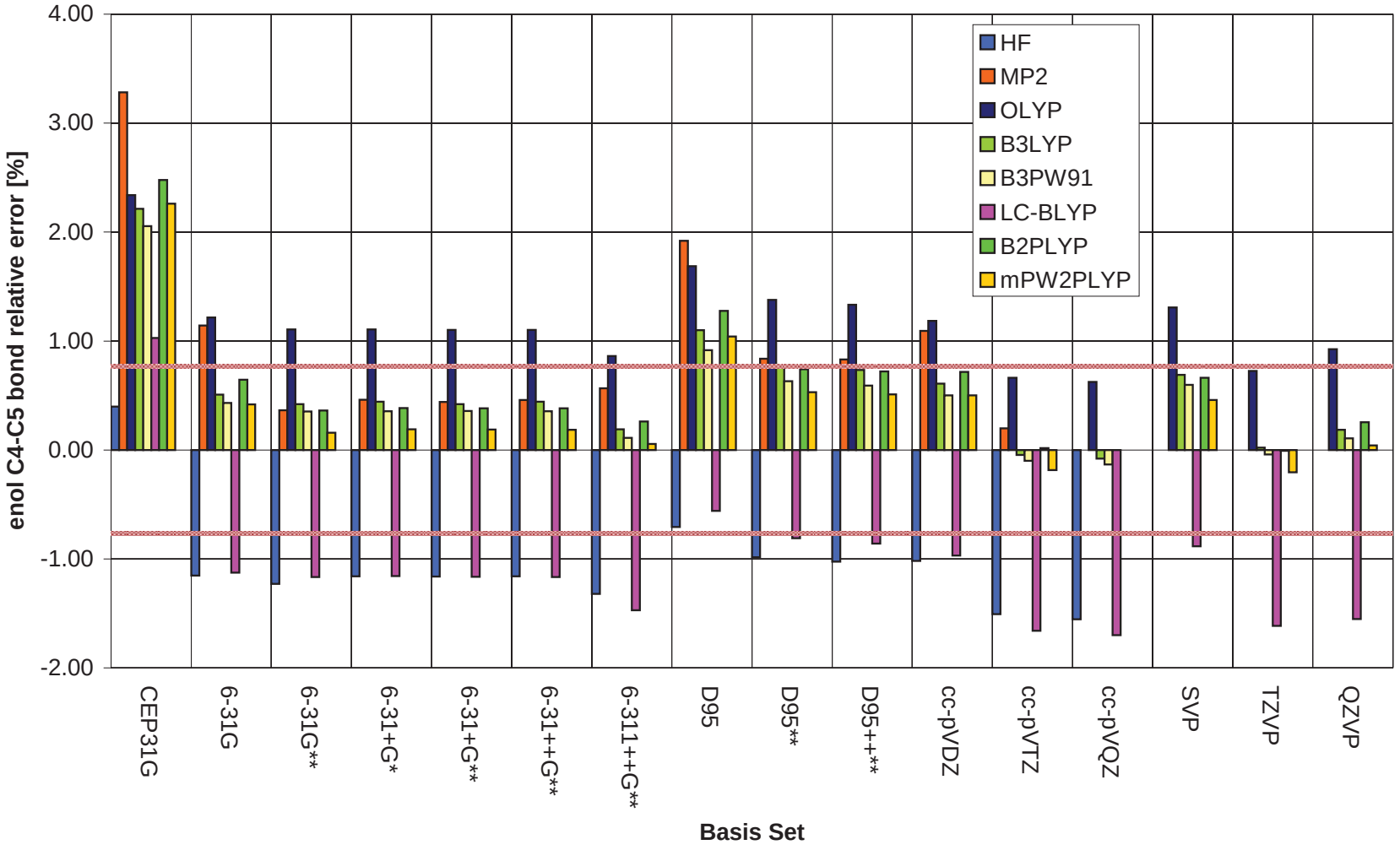


Figure S8. Relative deviations between the X-ray (Table 2) and computational values for the enol C4-C5 bond. The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The standard deviations of the X-ray determination are marked with red line.

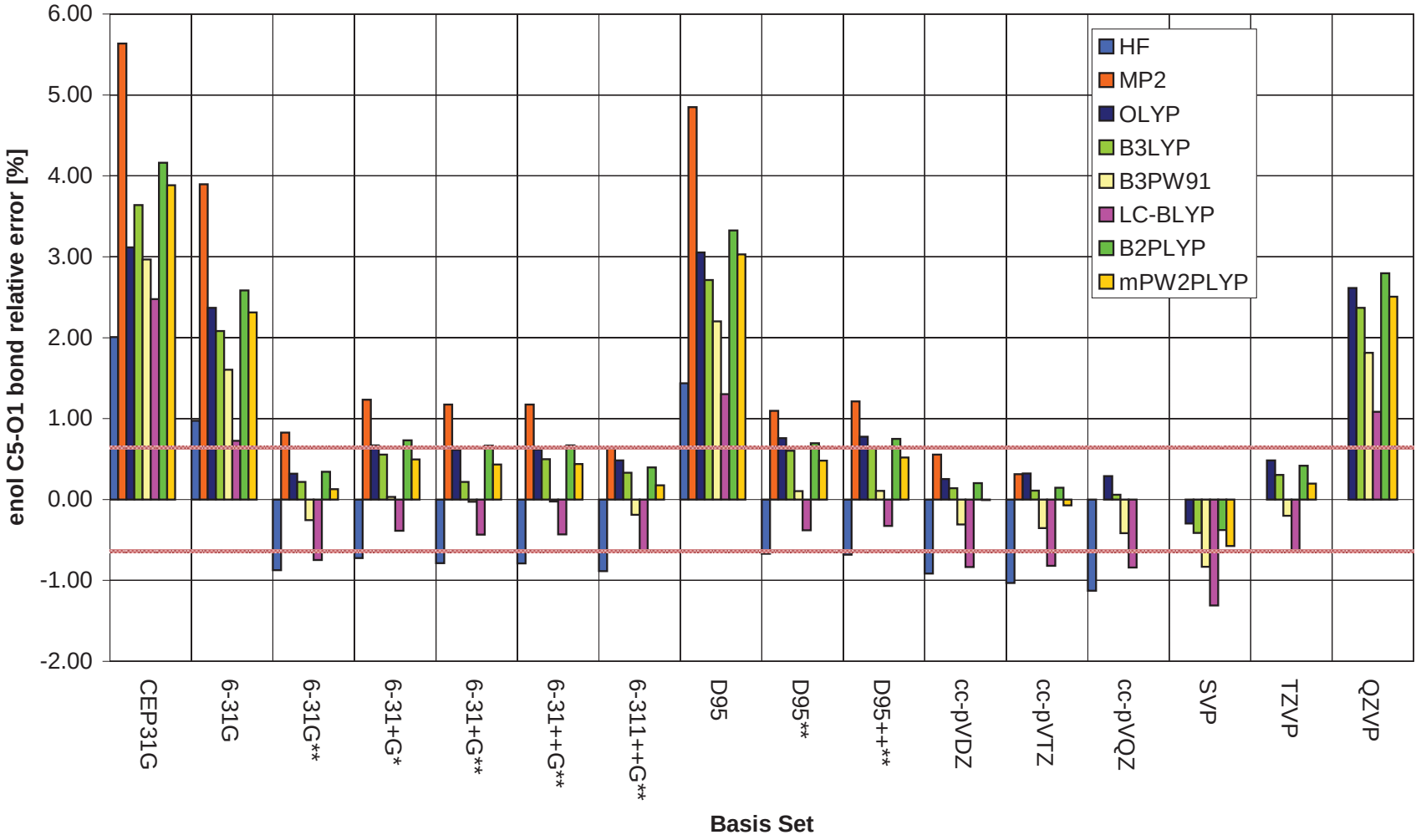


Figure S9. Relative deviations between the X-ray (Table 2) and computational values for the enol C5-O1 bond. The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The standard deviations of the X-ray determination are marked with red line.

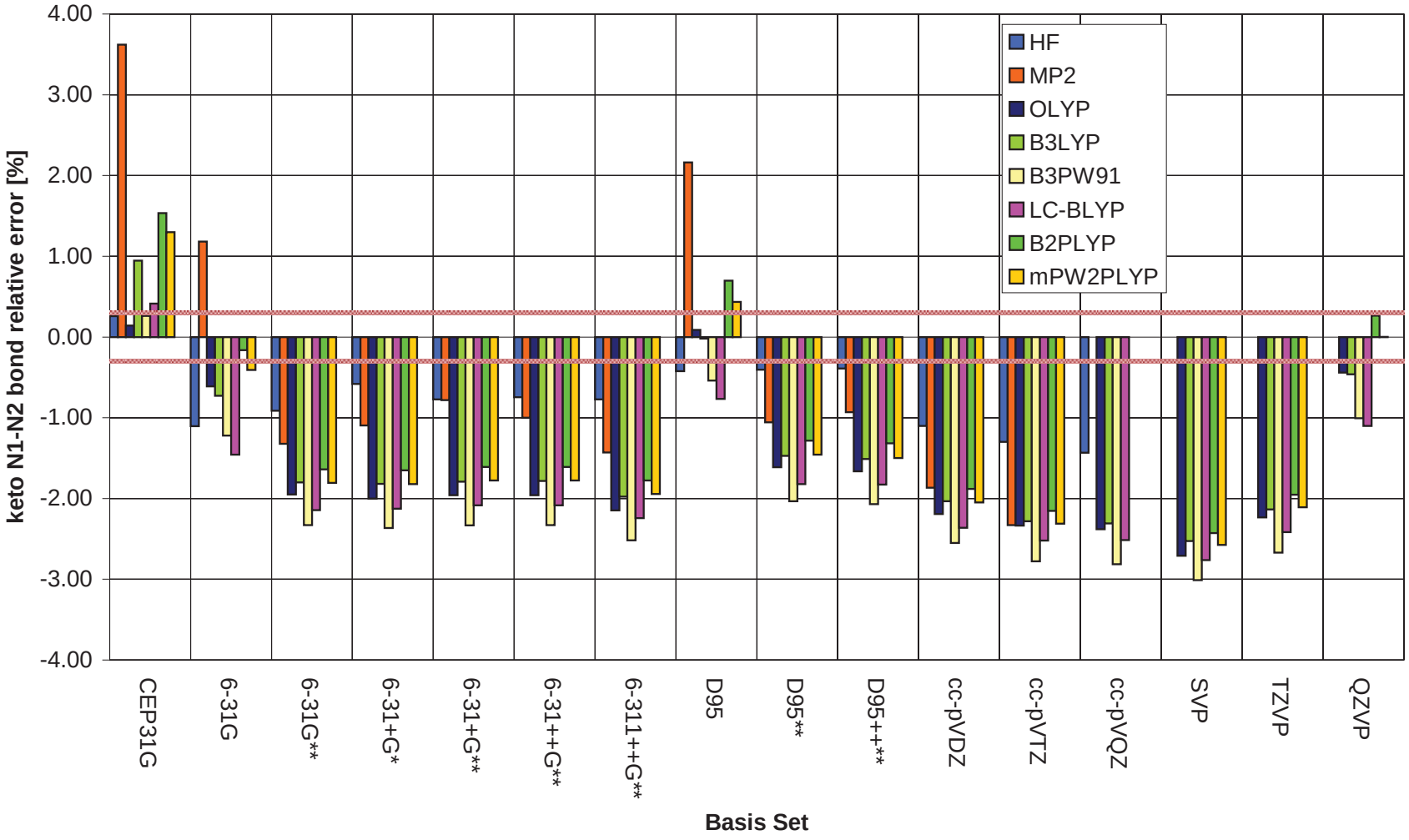


Figure S10. Relative deviations between the X-ray (Table 2) and computational values for the keto N1-N2 bond. The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The standard deviations of the X-ray determination are marked with red line.

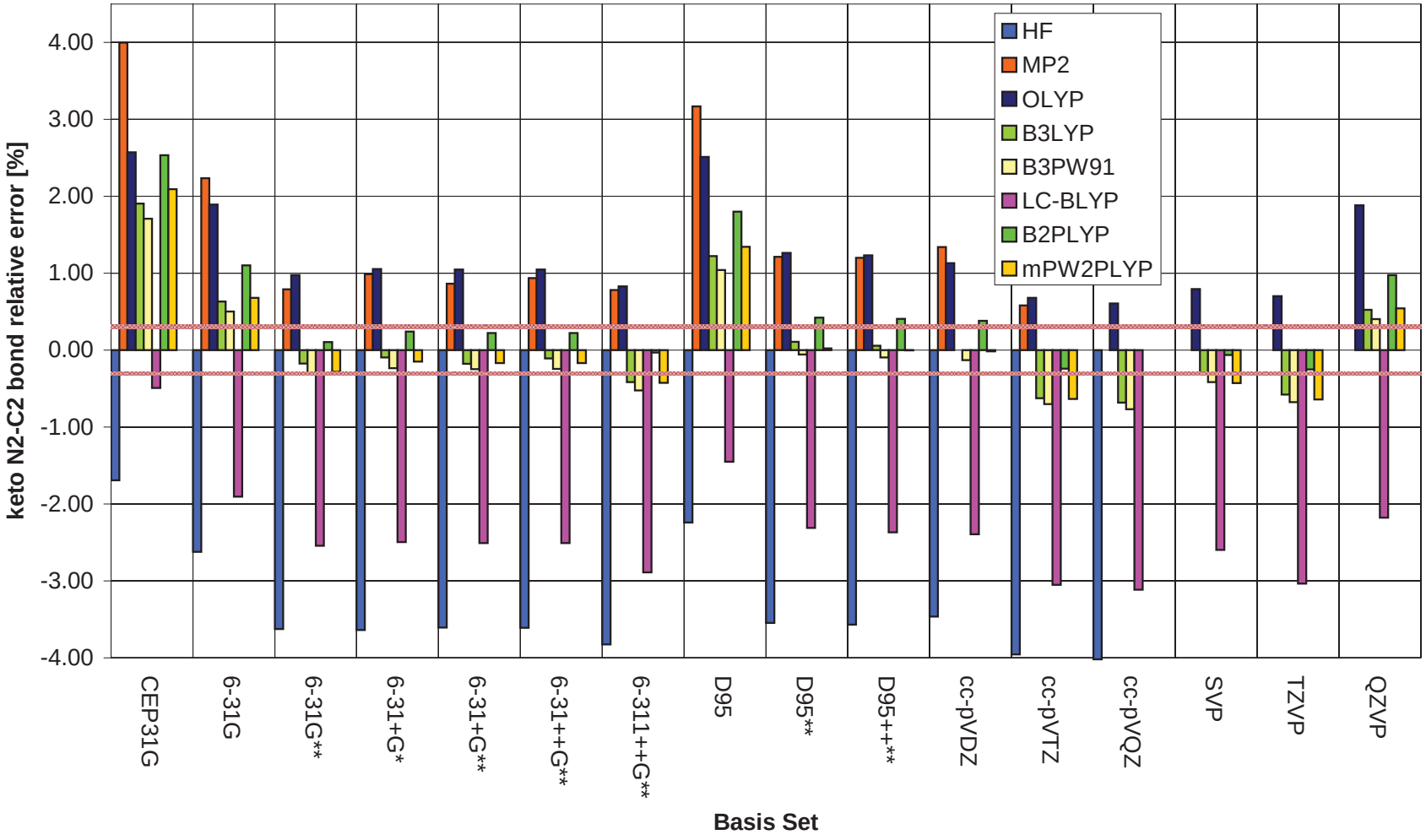


Figure S11. Relative deviations between the X-ray (Table 2) and computational values for the keto N2-C2 bond. The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The standard deviations of the X-ray determination are marked with red line.

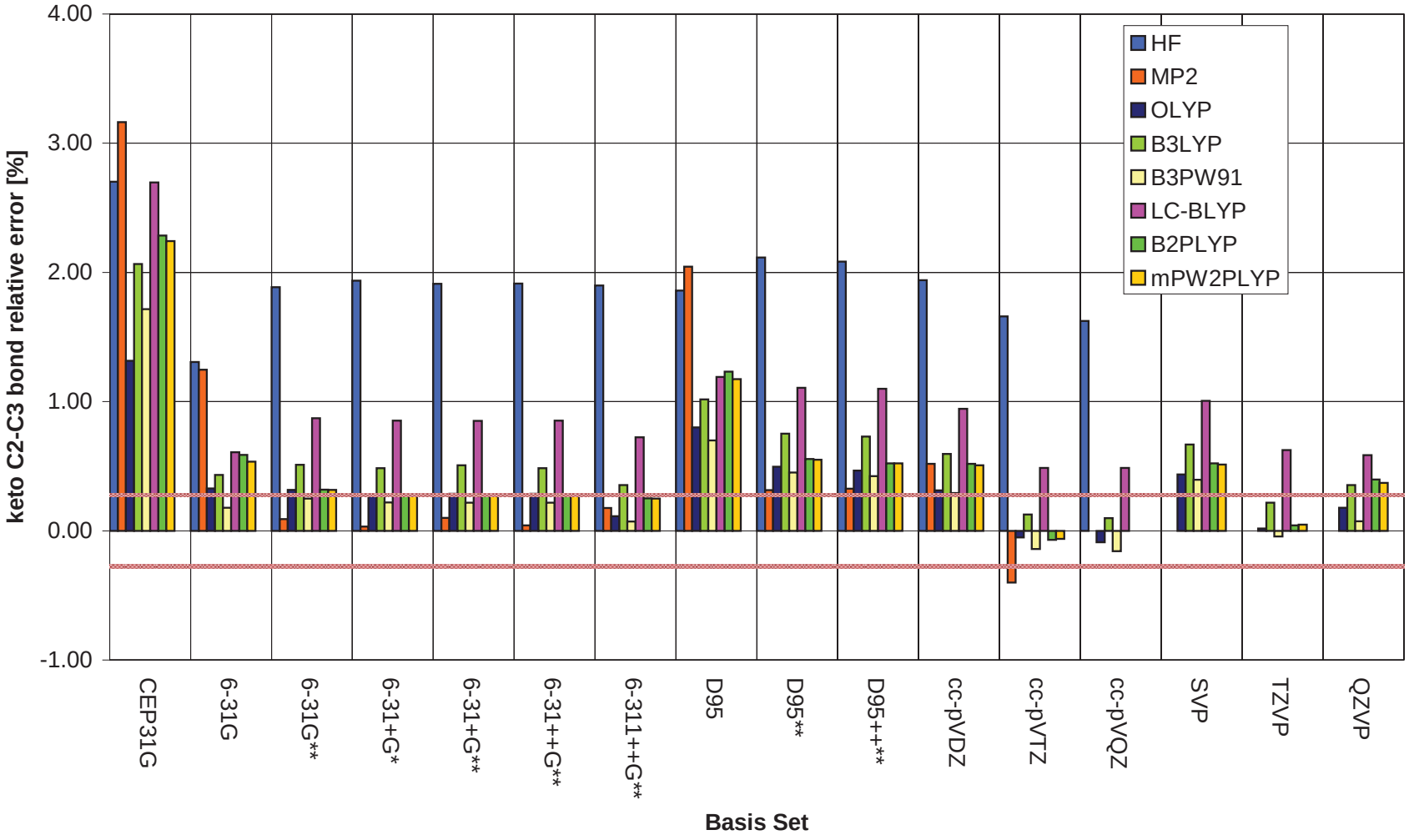


Figure S12. Relative deviations between the X-ray (Table 2) and computational values for the keto C2-C3 bond. The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The standard deviations of the X-ray determination are marked with red line.

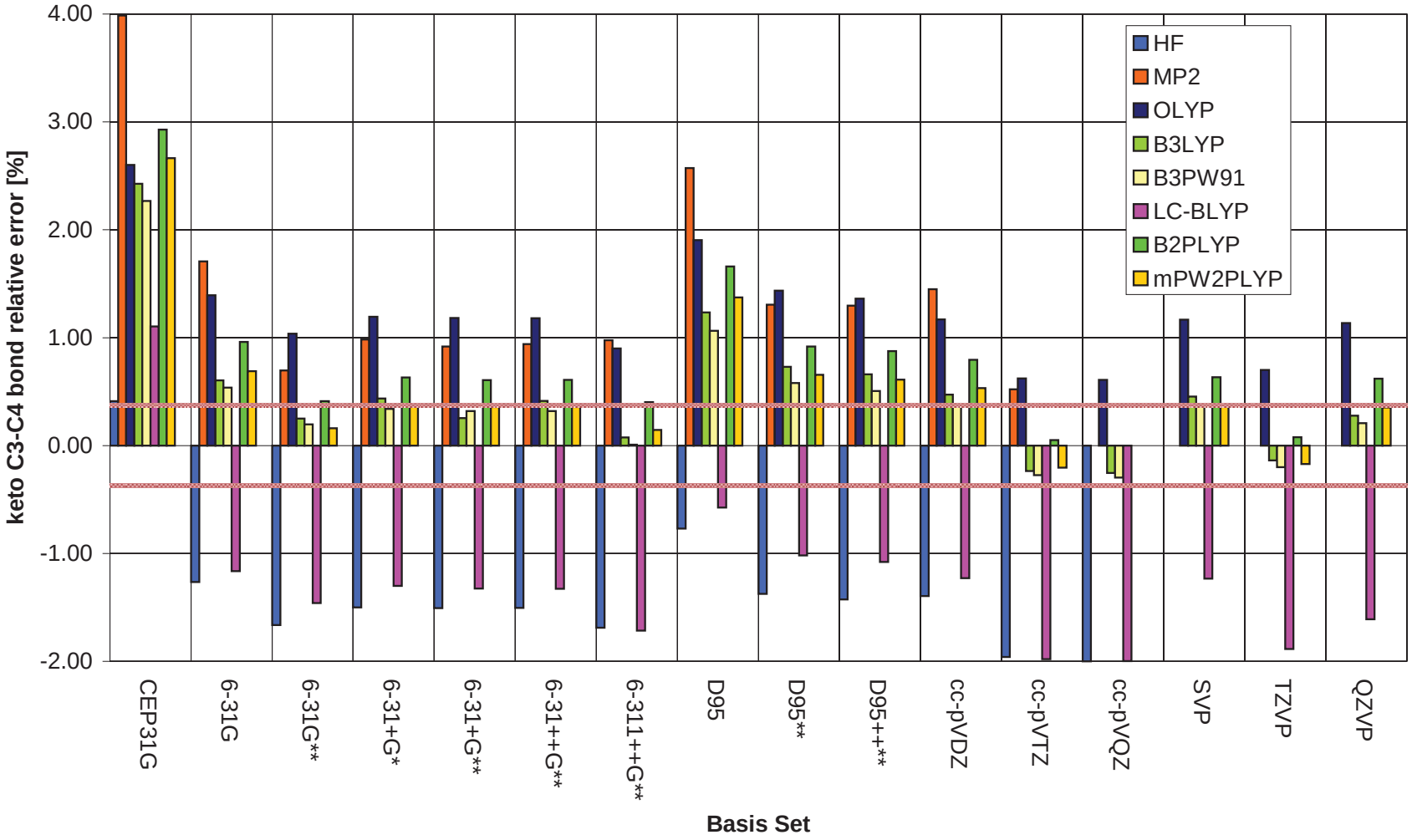


Figure S13. Relative deviations between the X-ray (Table 2) and computational values for the keto C3-C4 bond. The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The standard deviations of the X-ray determination are marked with red line.

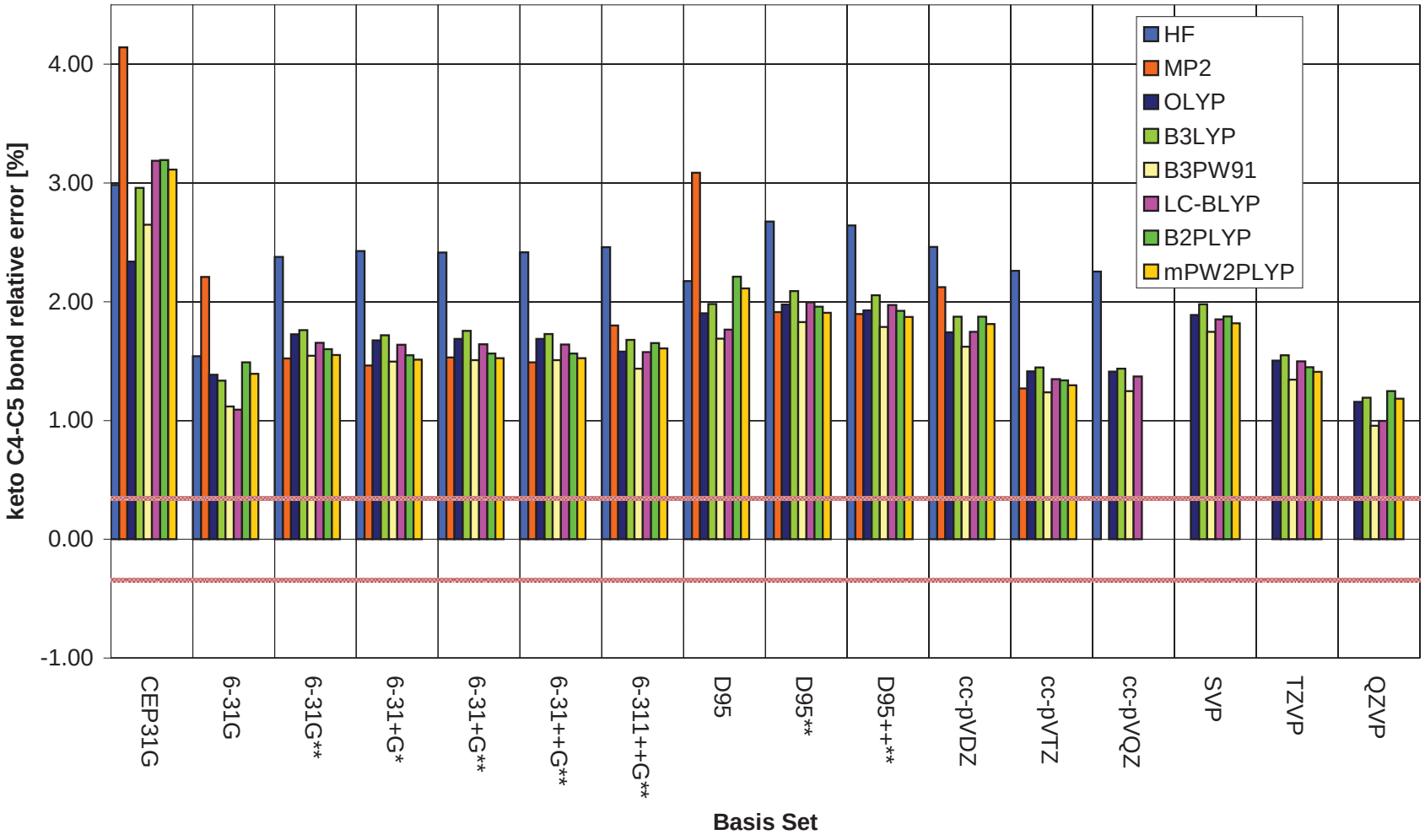


Figure S14. Relative deviations between the X-ray (Table 2) and computational values for the keto C4-C5 bond. The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The standard deviations of the X-ray determination are marked with red line.

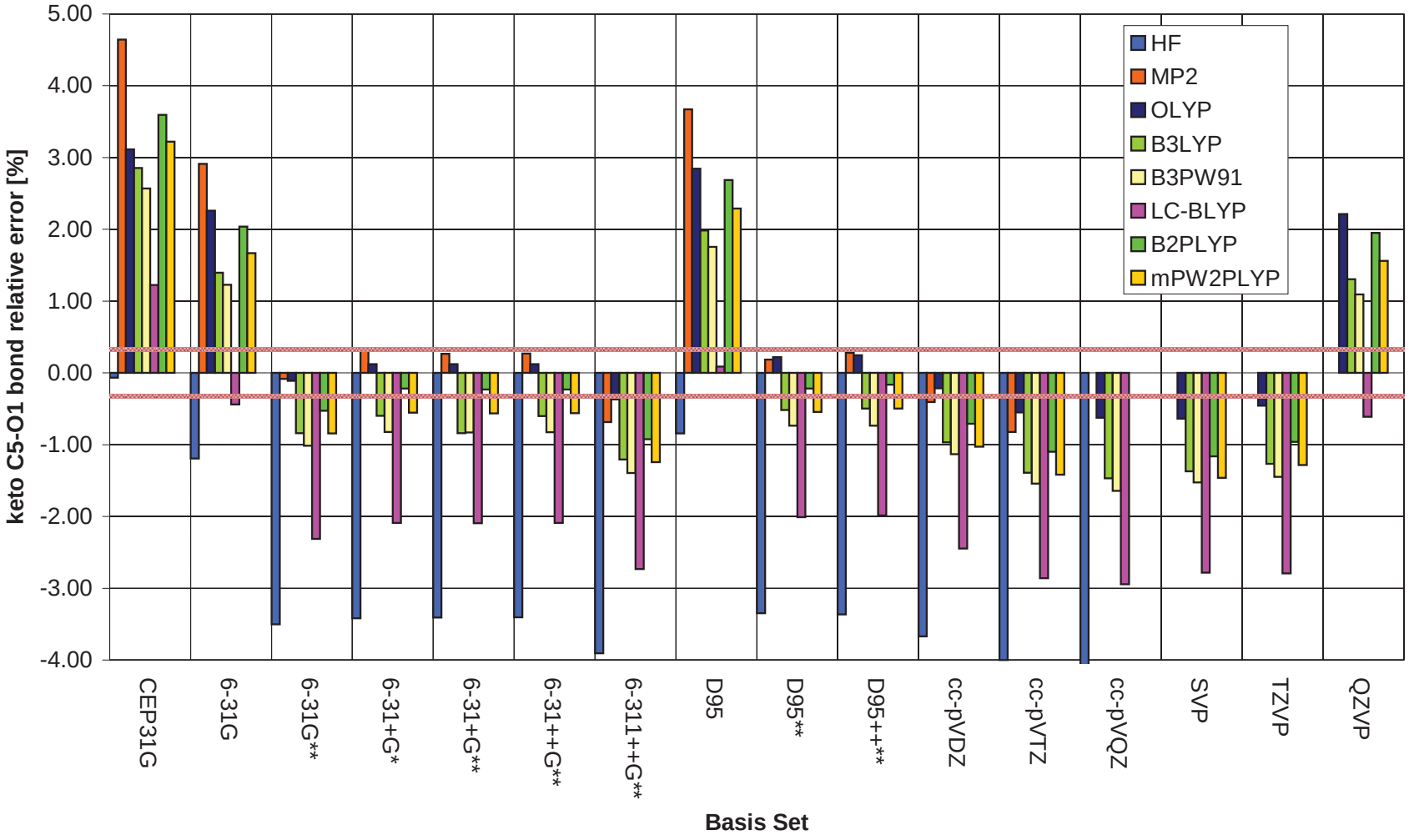


Figure S15. Relative deviations between the X-ray (Table 2) and computational values for the keto C5-O1 bond. The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The standard deviations of the X-ray determination are marked with red line.

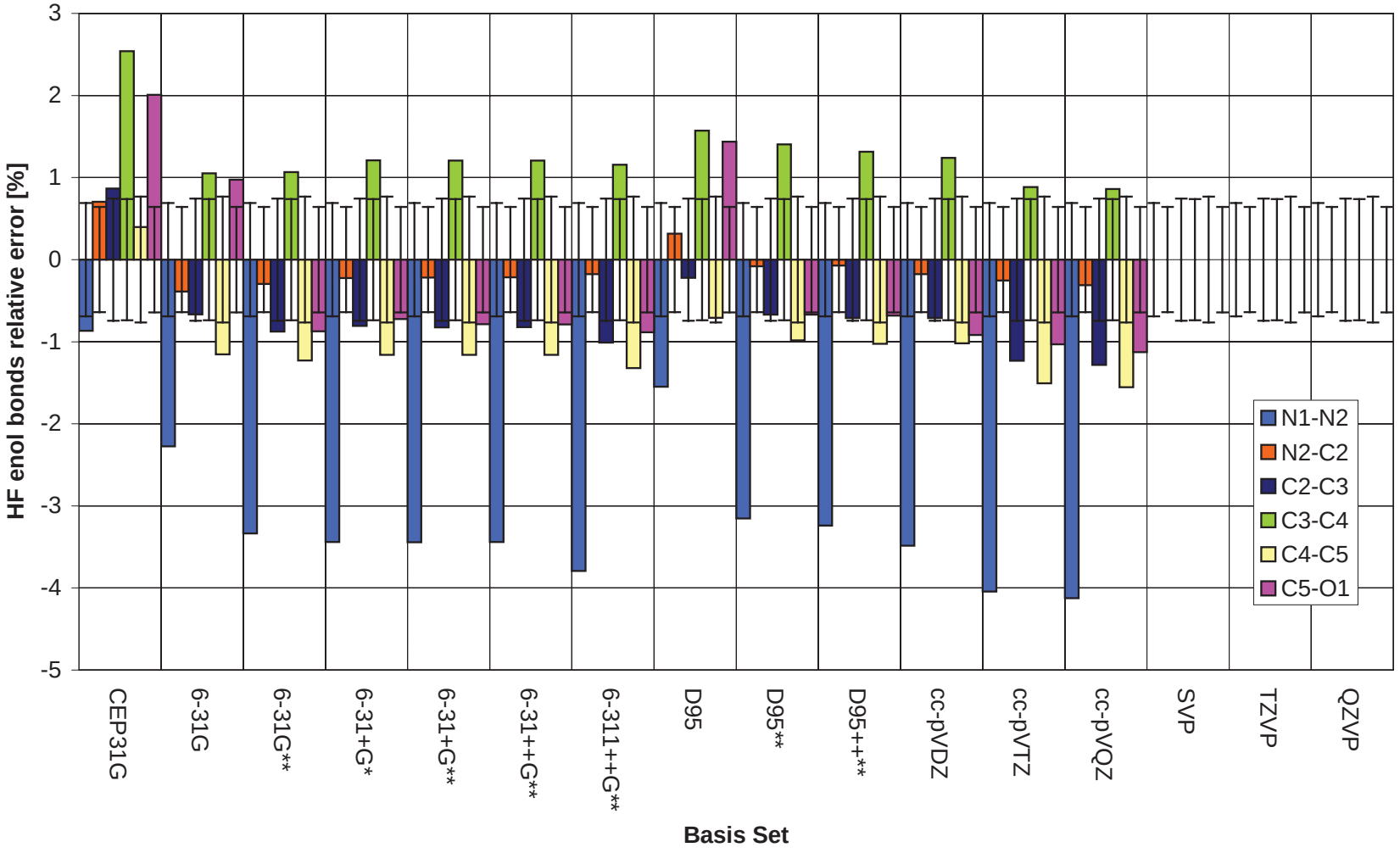


Figure S16. Relative deviations of the enol form bond lengths predicted by using HF method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

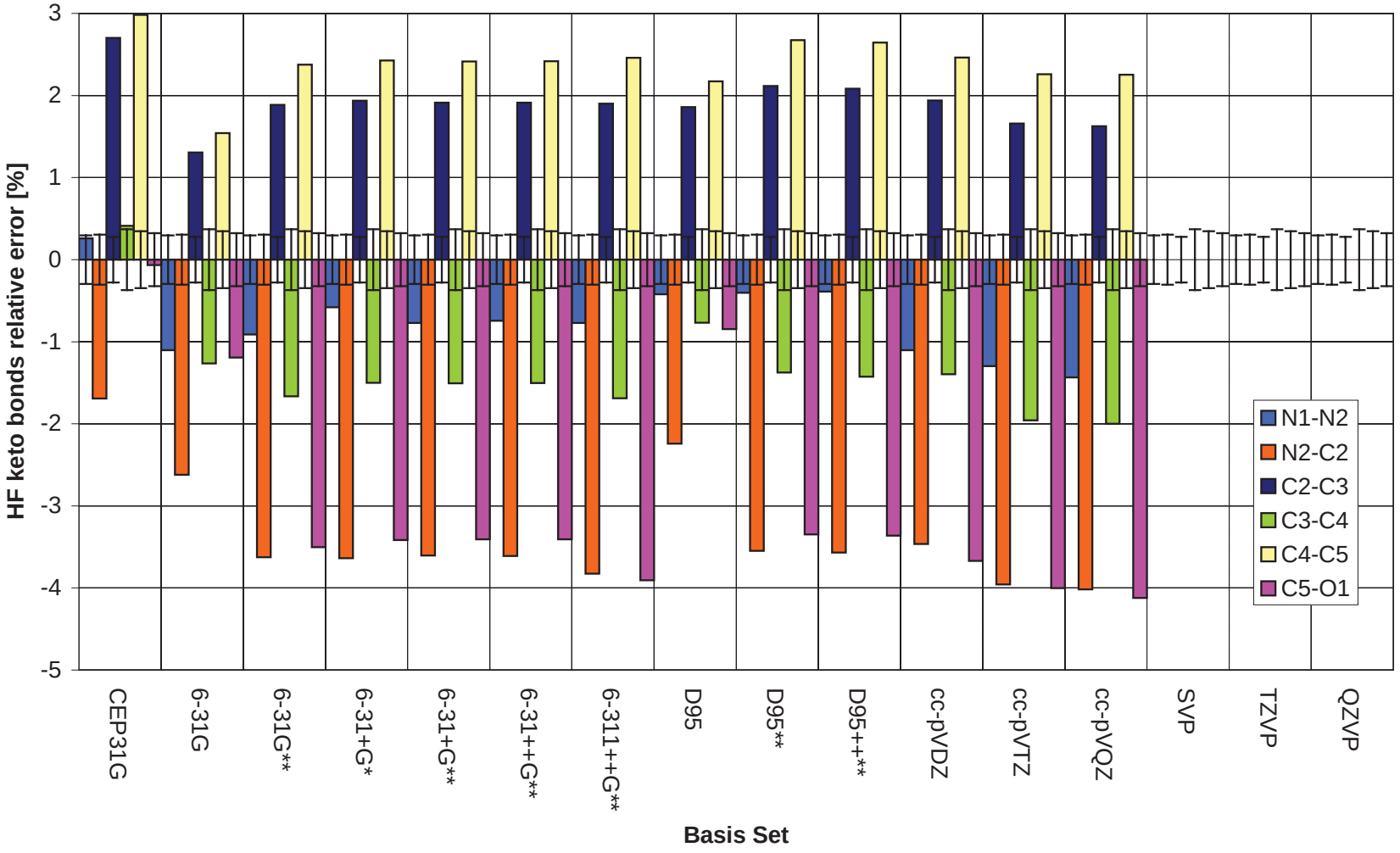


Figure S17. Relative deviations of the keto form bond lengths predicted by using HF method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

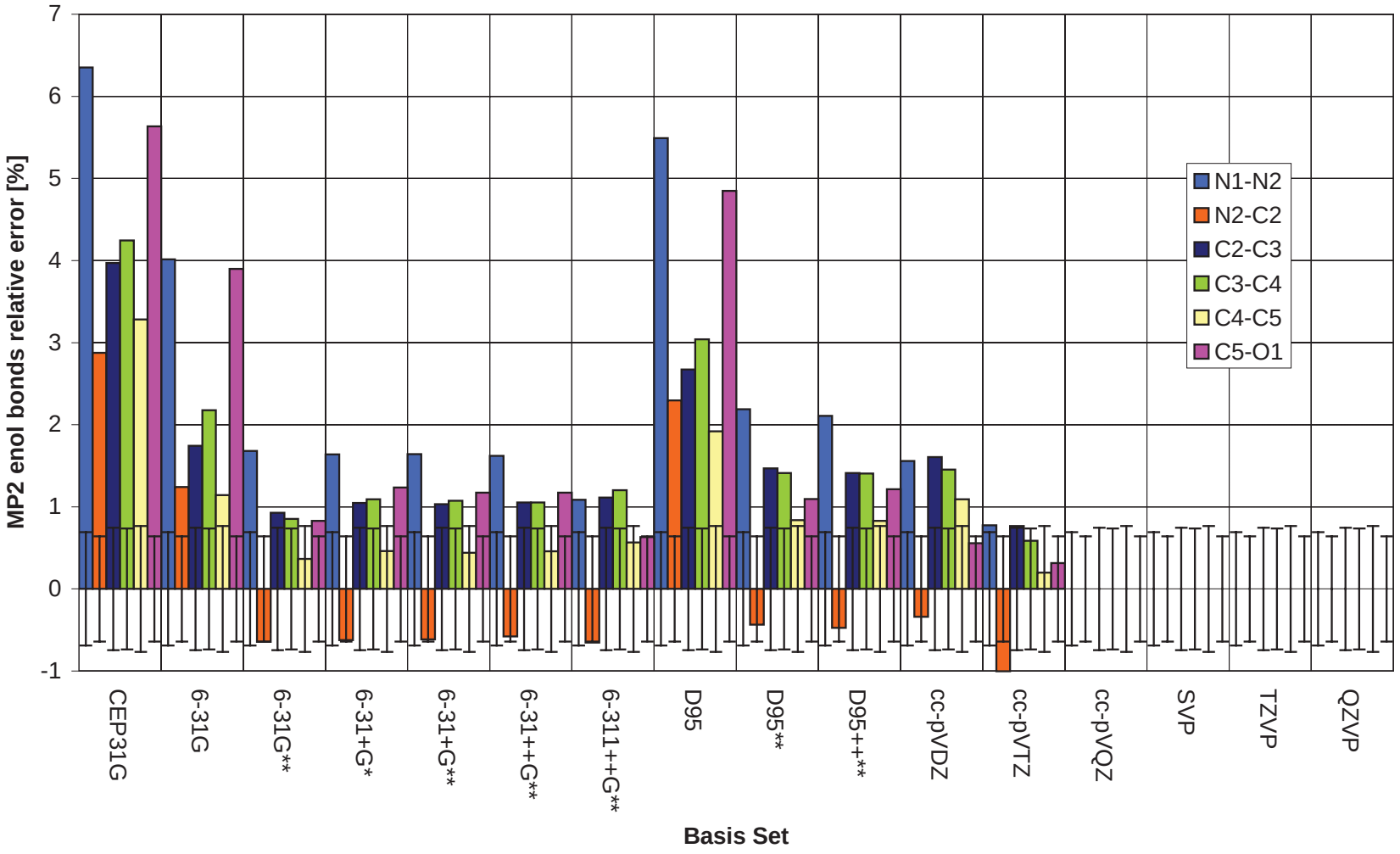


Figure S18. Relative deviations of the enol form bond lengths predicted by using MP2 method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

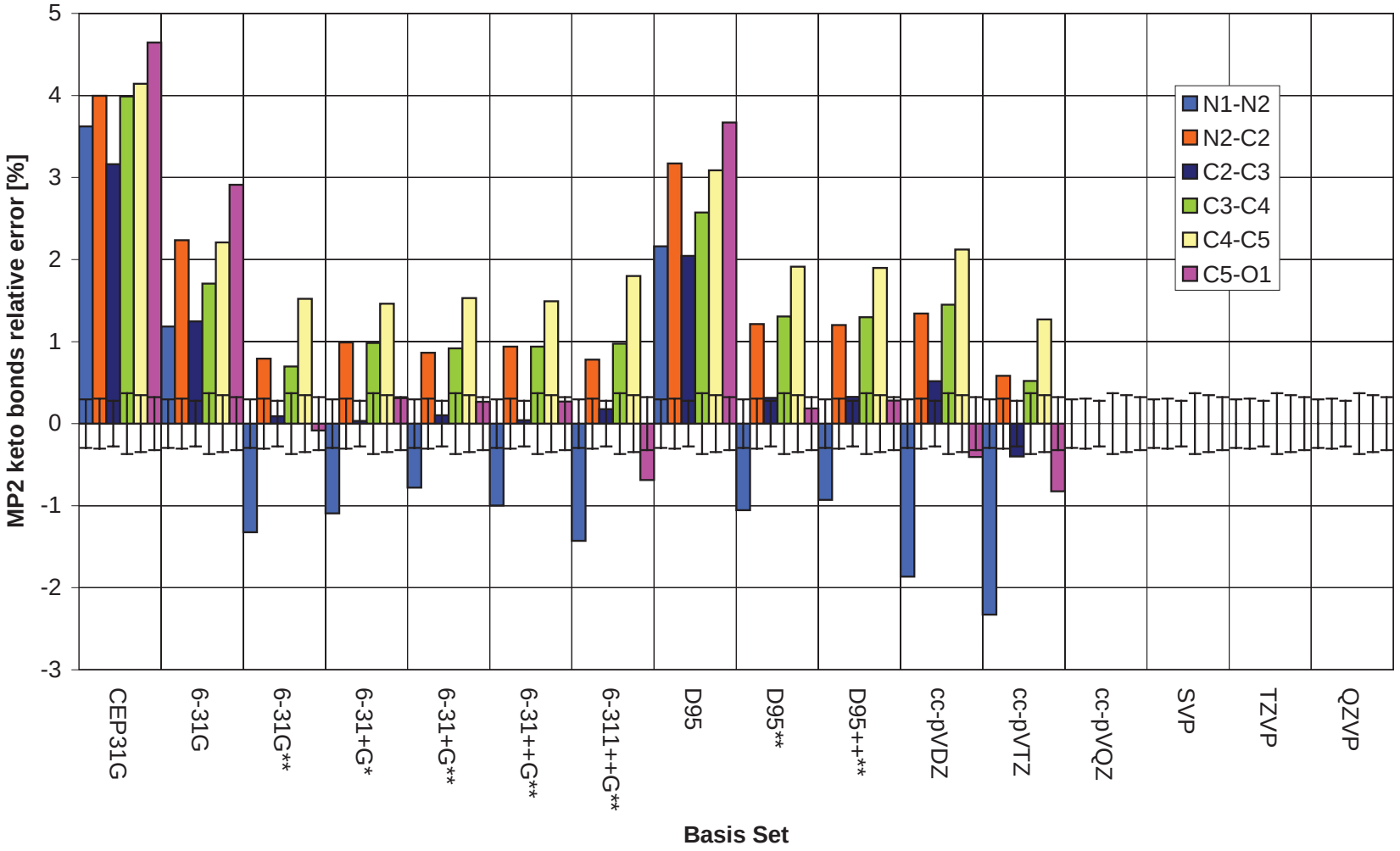


Figure S19. Relative deviations of the keto form bond lengths predicted by using MP2 method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

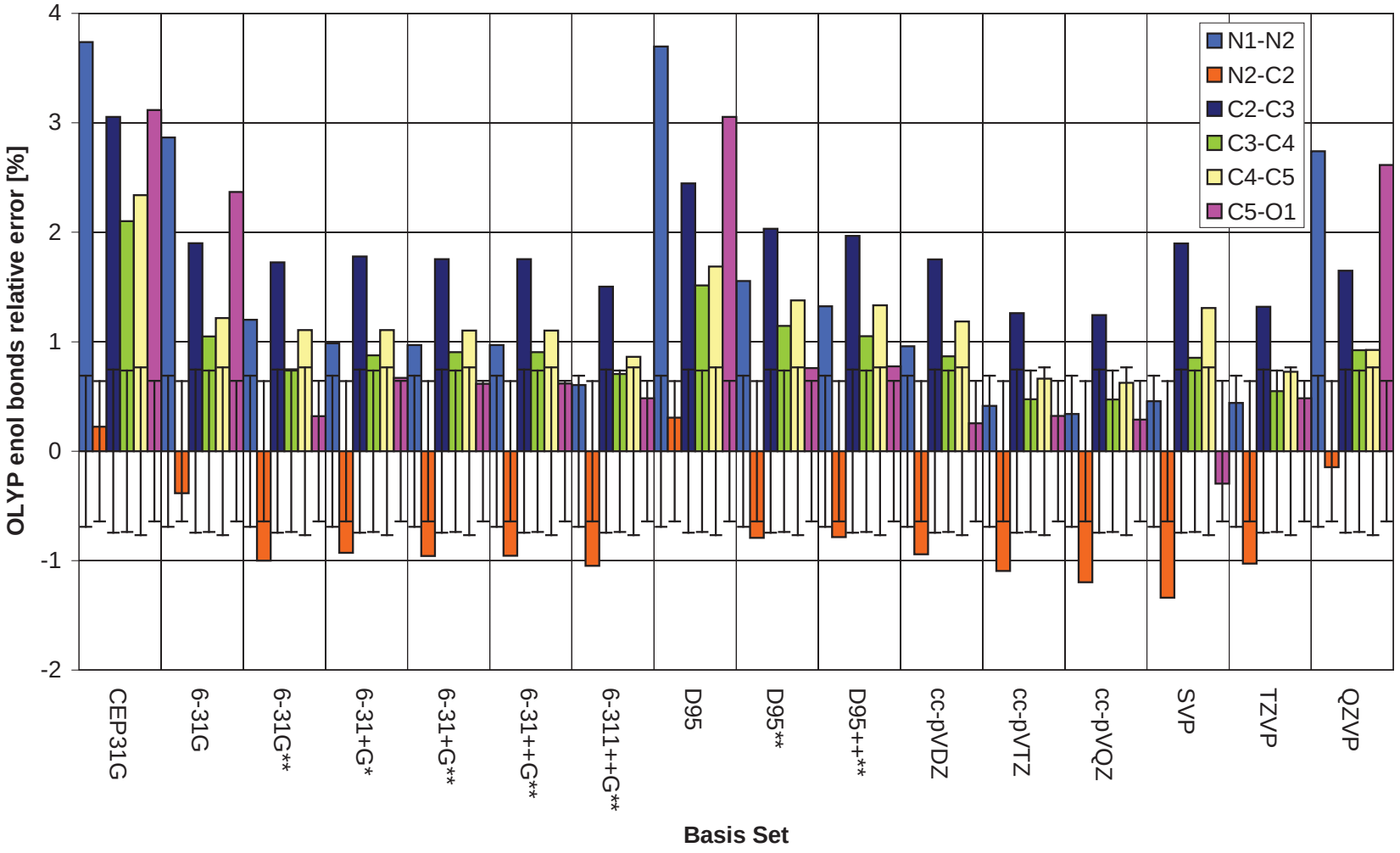


Figure S20. Relative deviations of the enol form bond lengths predicted by using OLYP method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

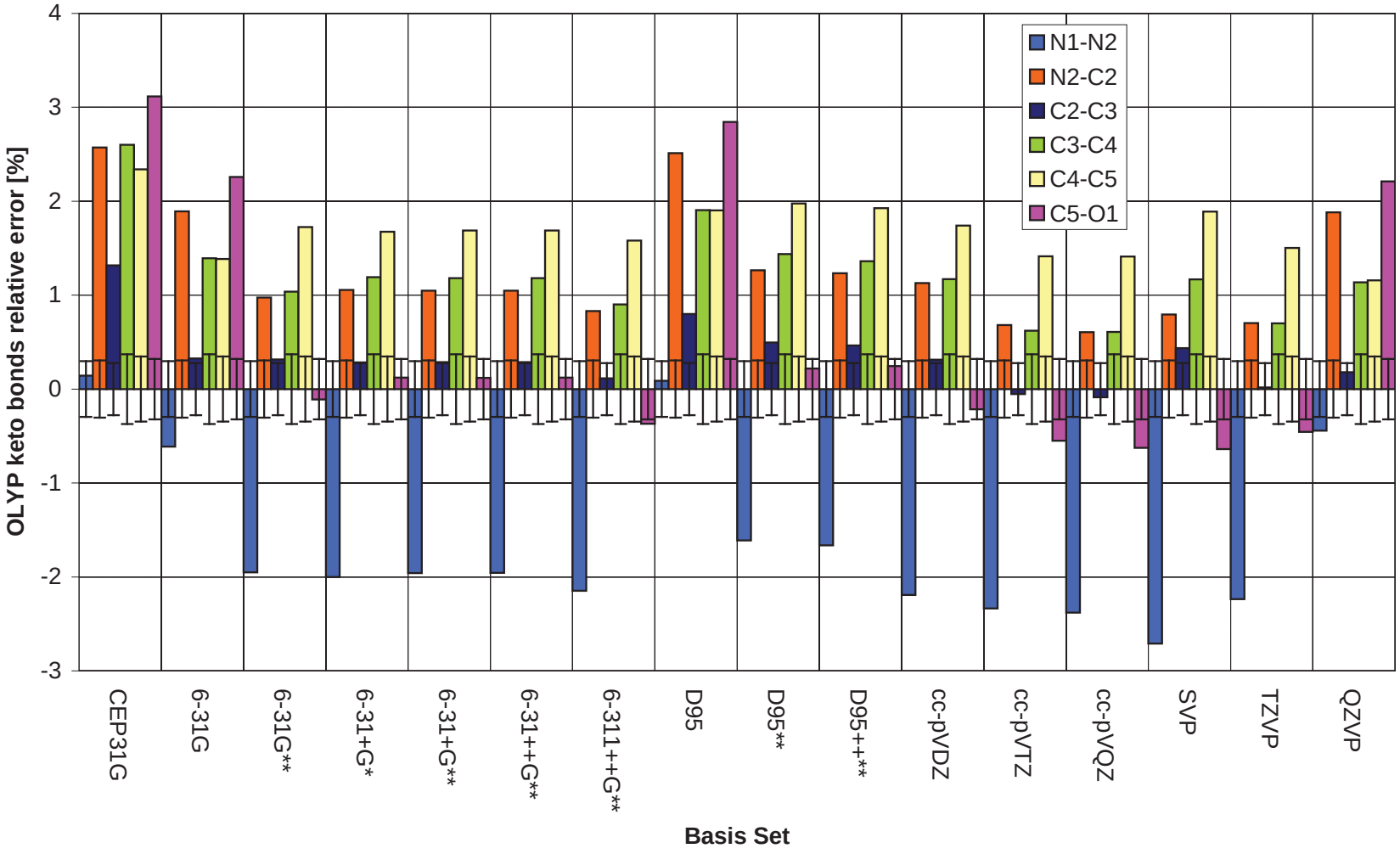


Figure S21. Relative deviations of the keto form bond lengths predicted by using OLYP method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

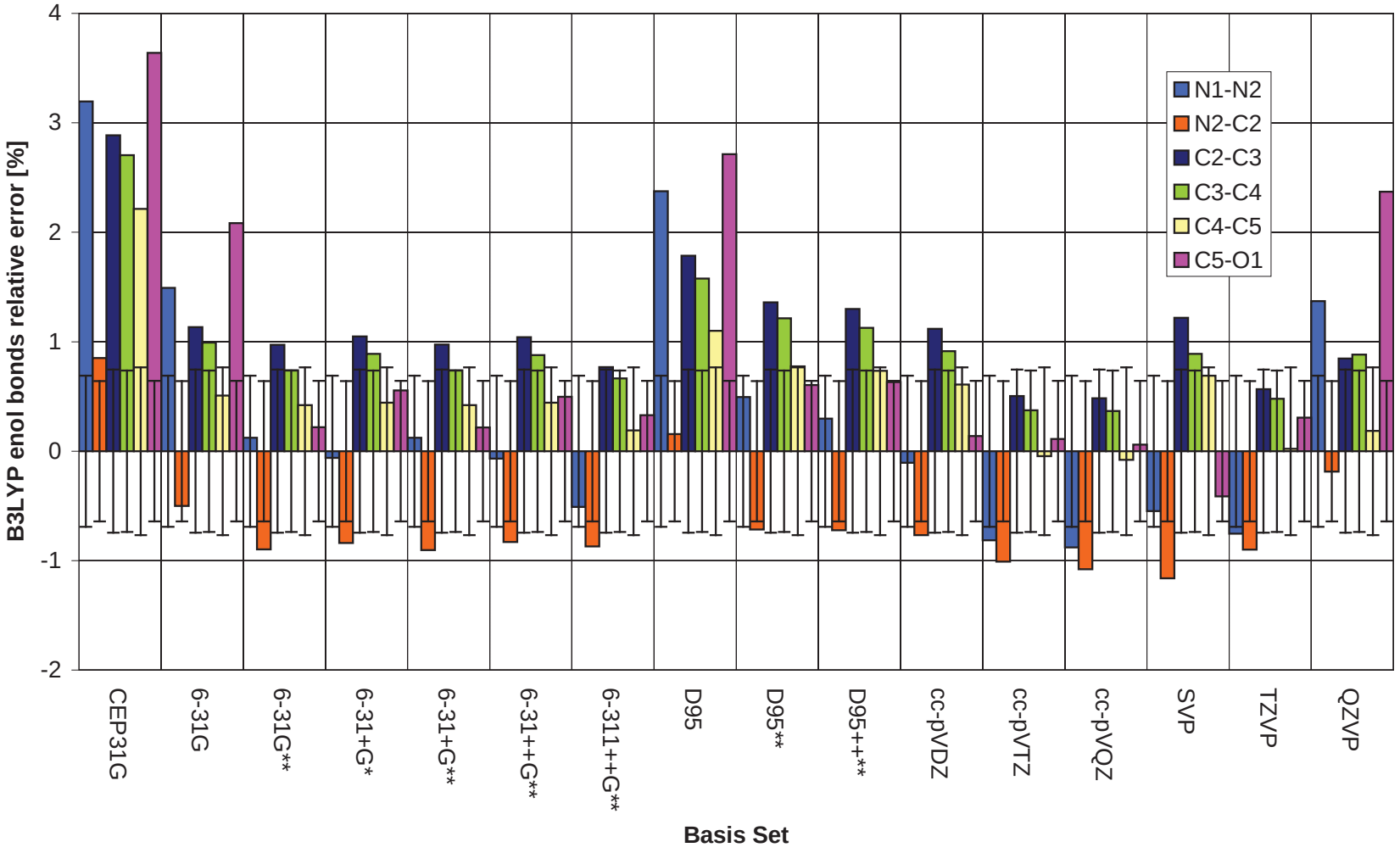


Figure S22. Relative deviations of the enol form bond lengths predicted by using B3LYP method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

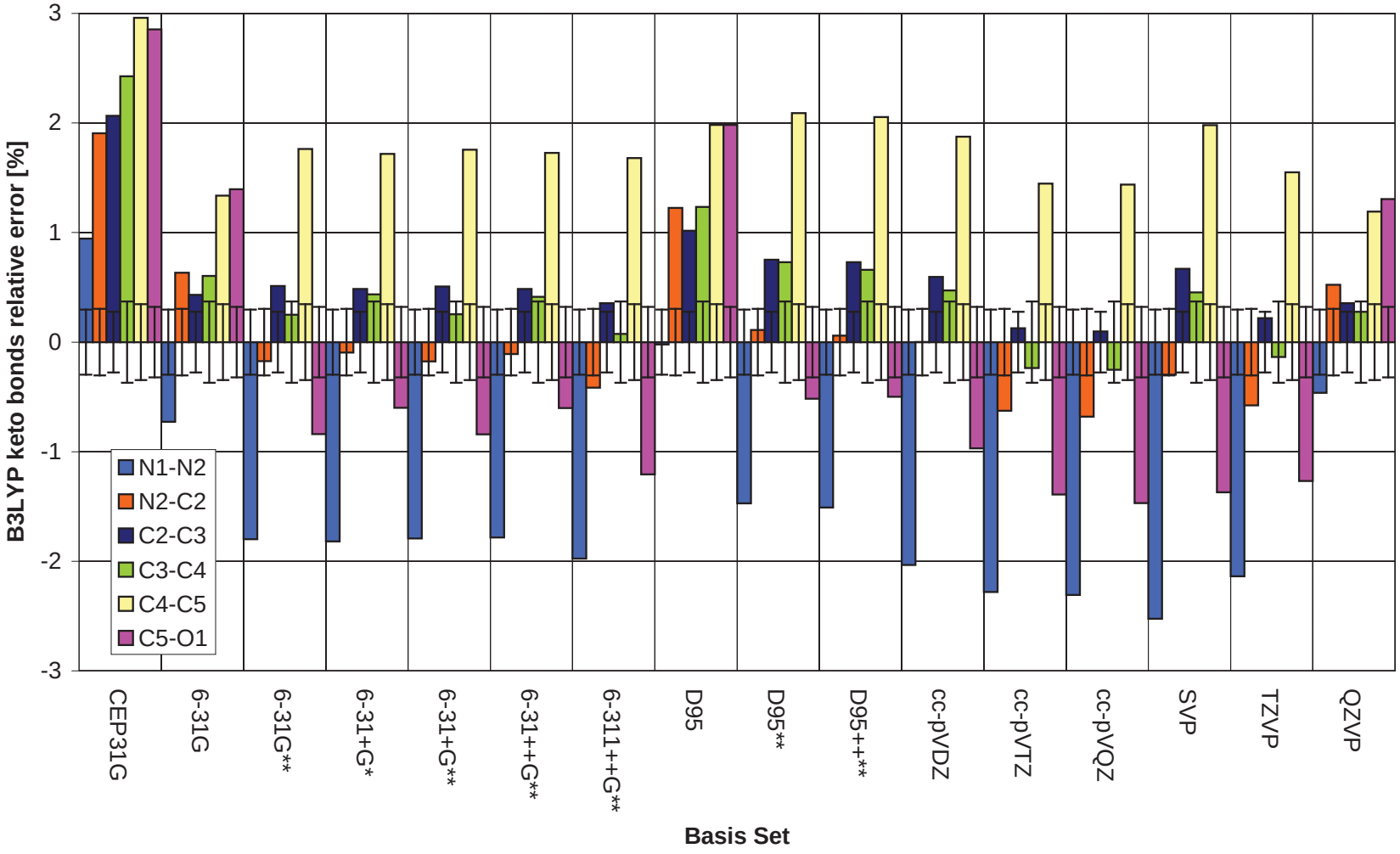


Figure S23. Relative deviations of the keto form bond lengths predicted by using B3LYP method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

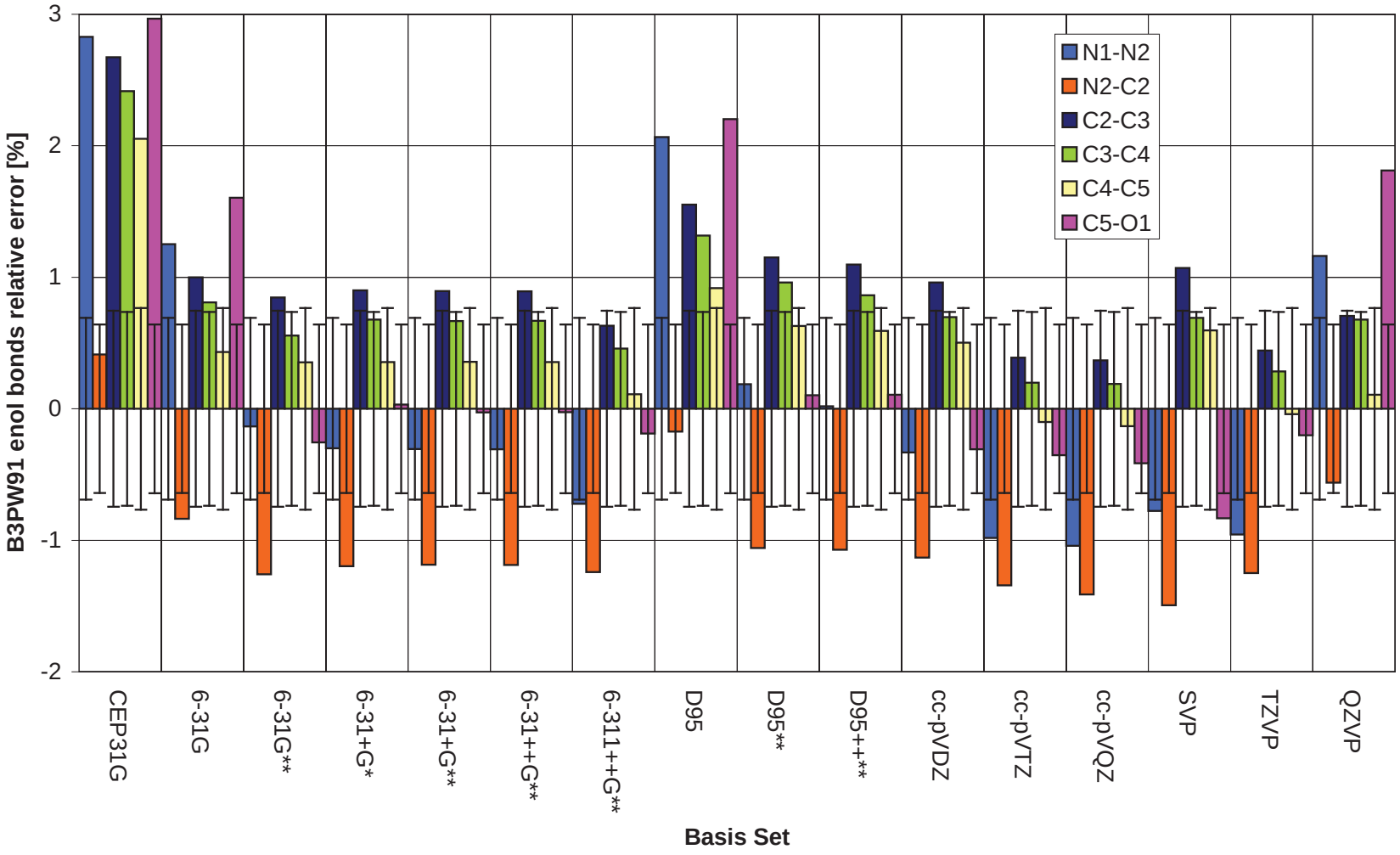


Figure S24. Relative deviations of the enol form bond lengths predicted by using B3PW91 method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

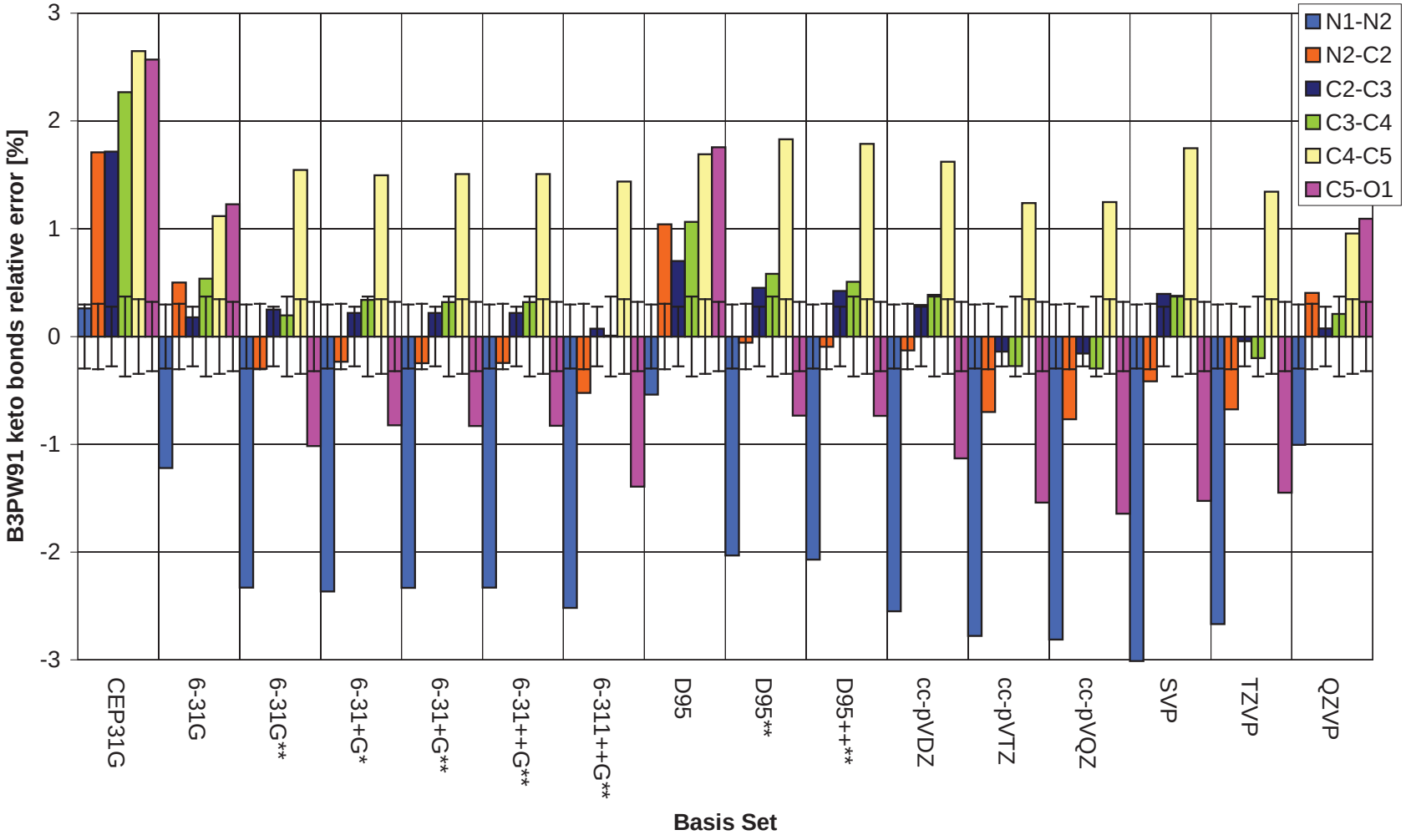


Figure S25. Relative deviations of the keto form bond lengths predicted by using B3PW91 method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

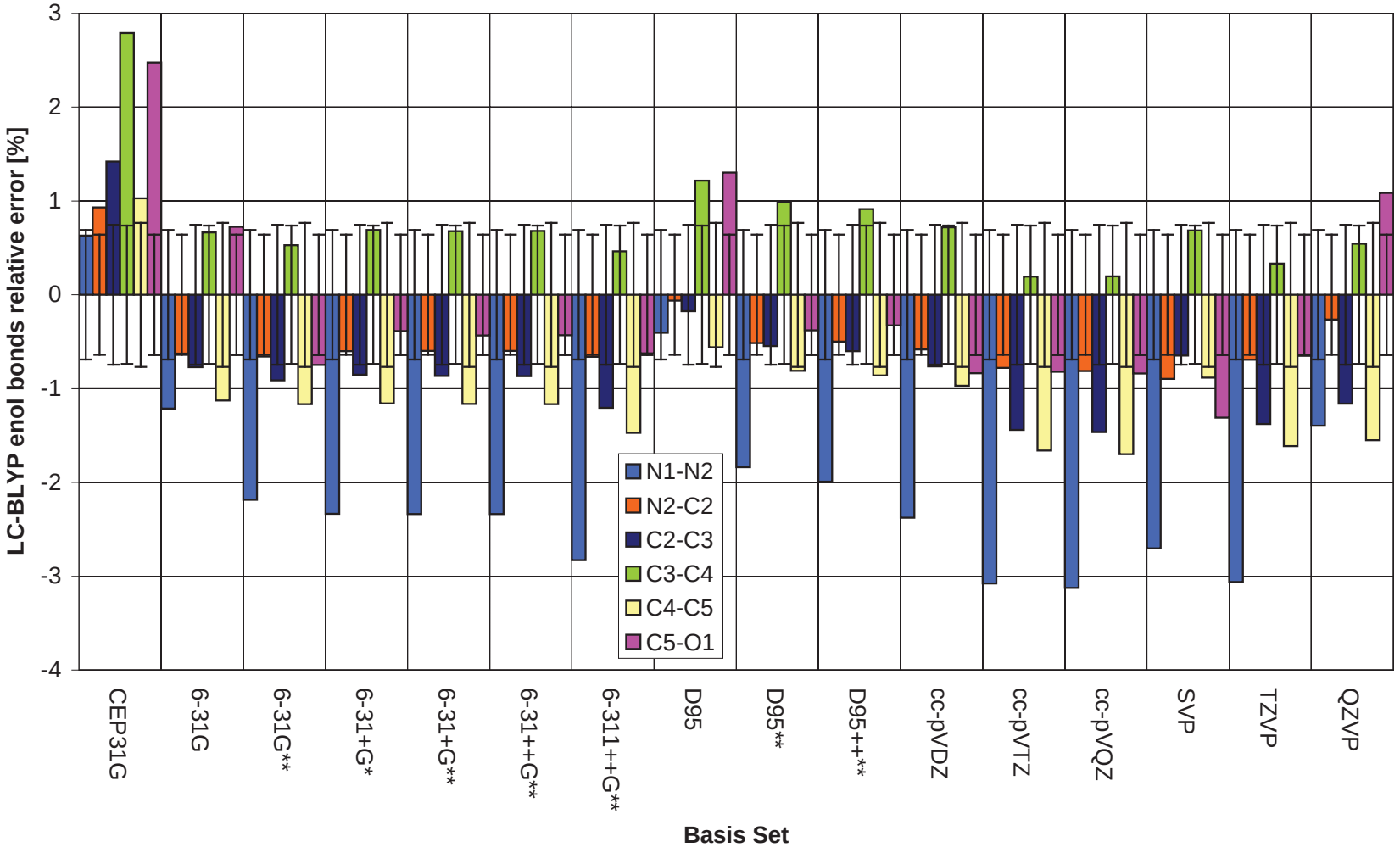


Figure S26. Relative deviations of the enol form bond lengths predicted by using LC-BLYP method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

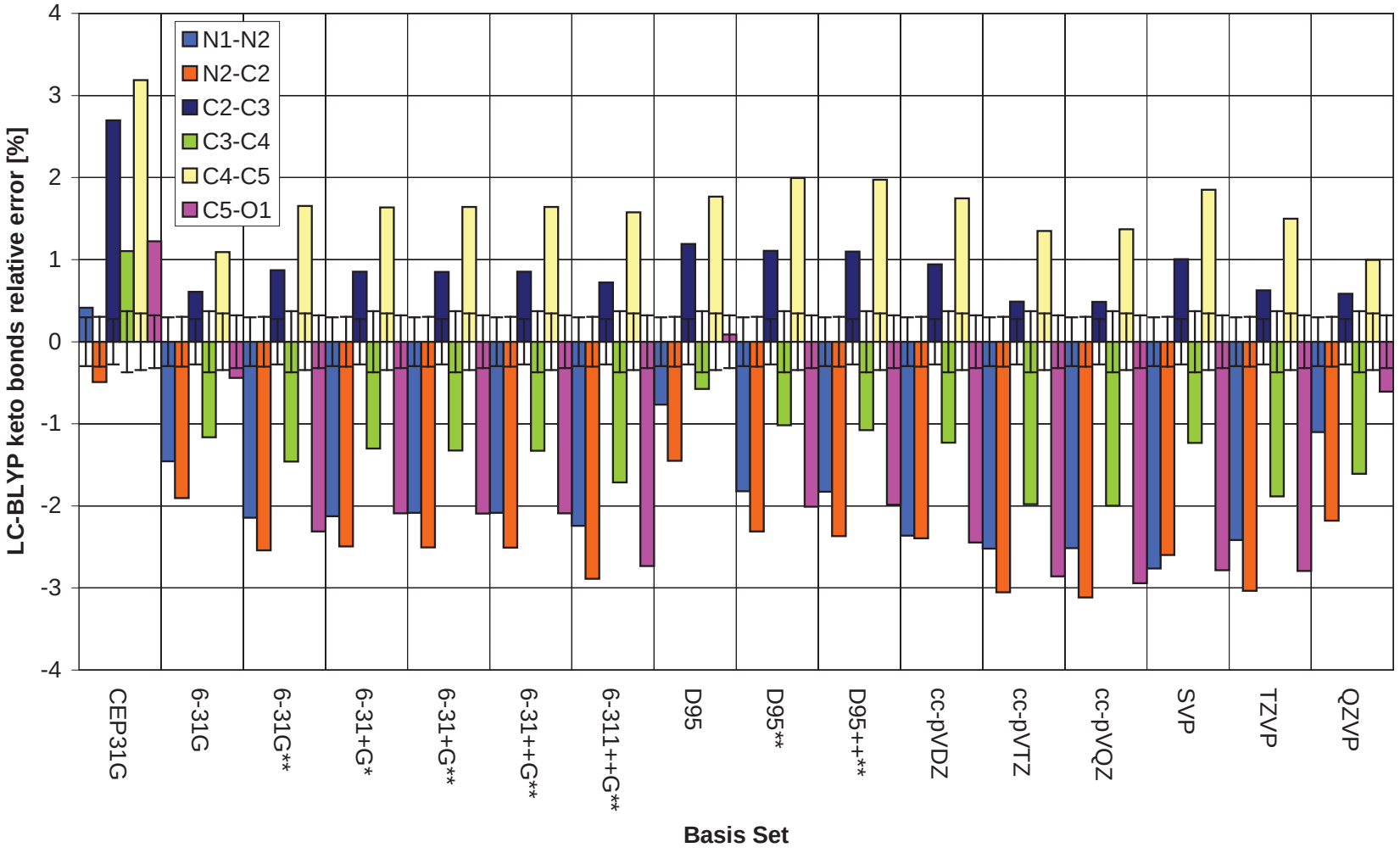


Figure S27. Relative deviations of the keto form bond lengths predicted by using LC-BLYP method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

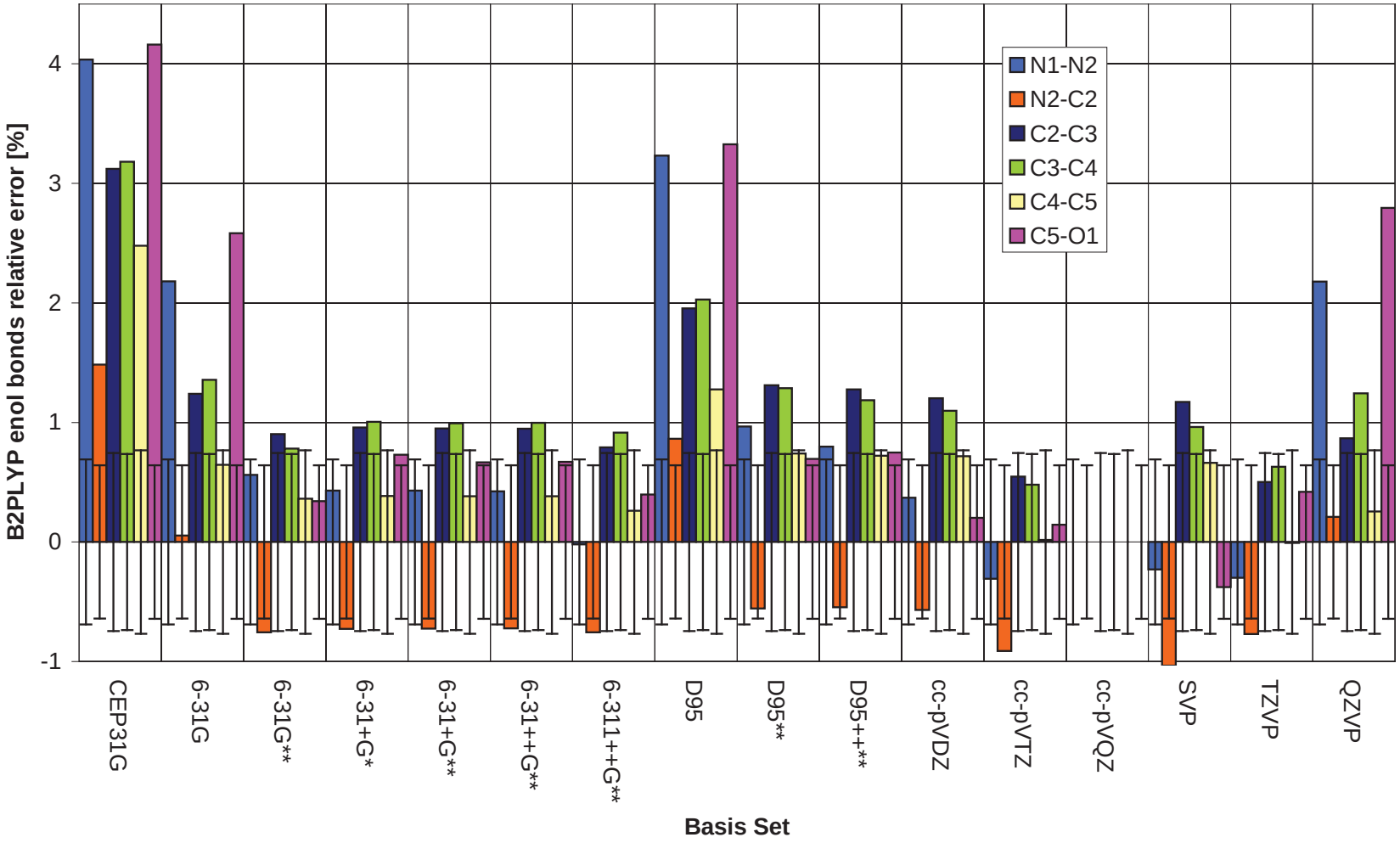


Figure S28. Relative deviations of the enol form bond lengths predicted by using B2PLYP method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

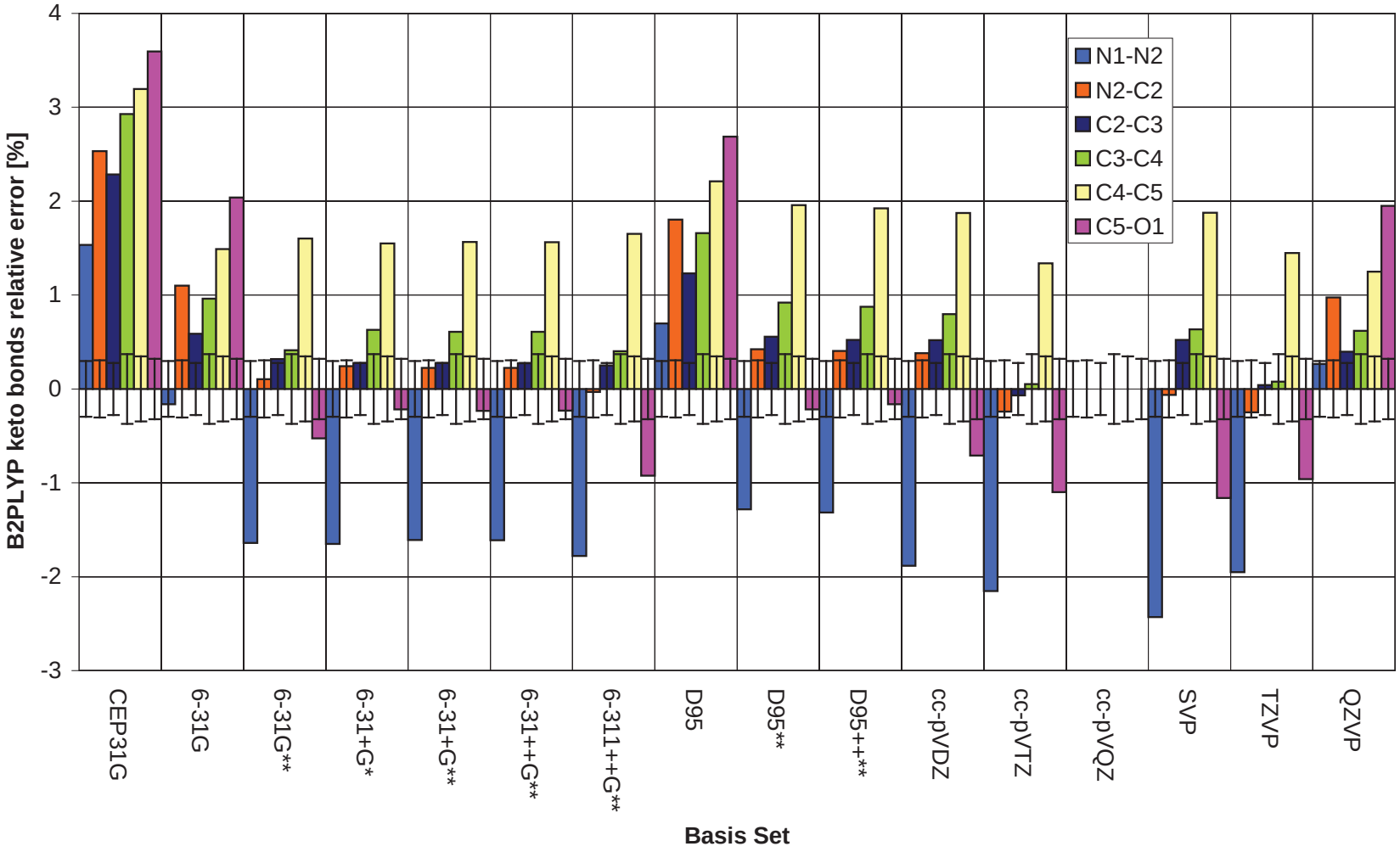


Figure S29. Relative deviations of the keto form bond lengths predicted by using B2PLYP method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

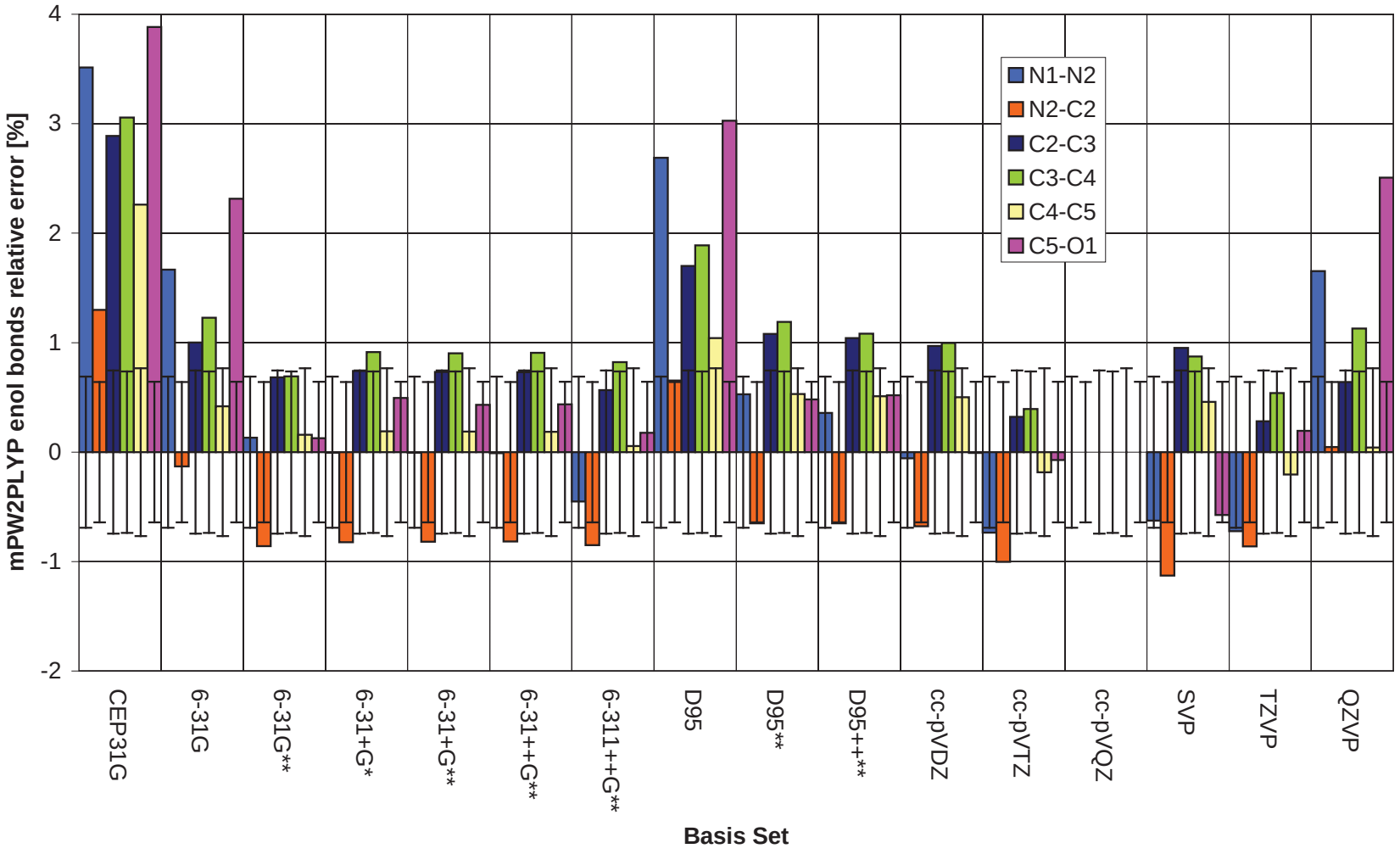


Figure S30. Relative deviations of the enol form bond lengths predicted by using mPW2PLYP method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.

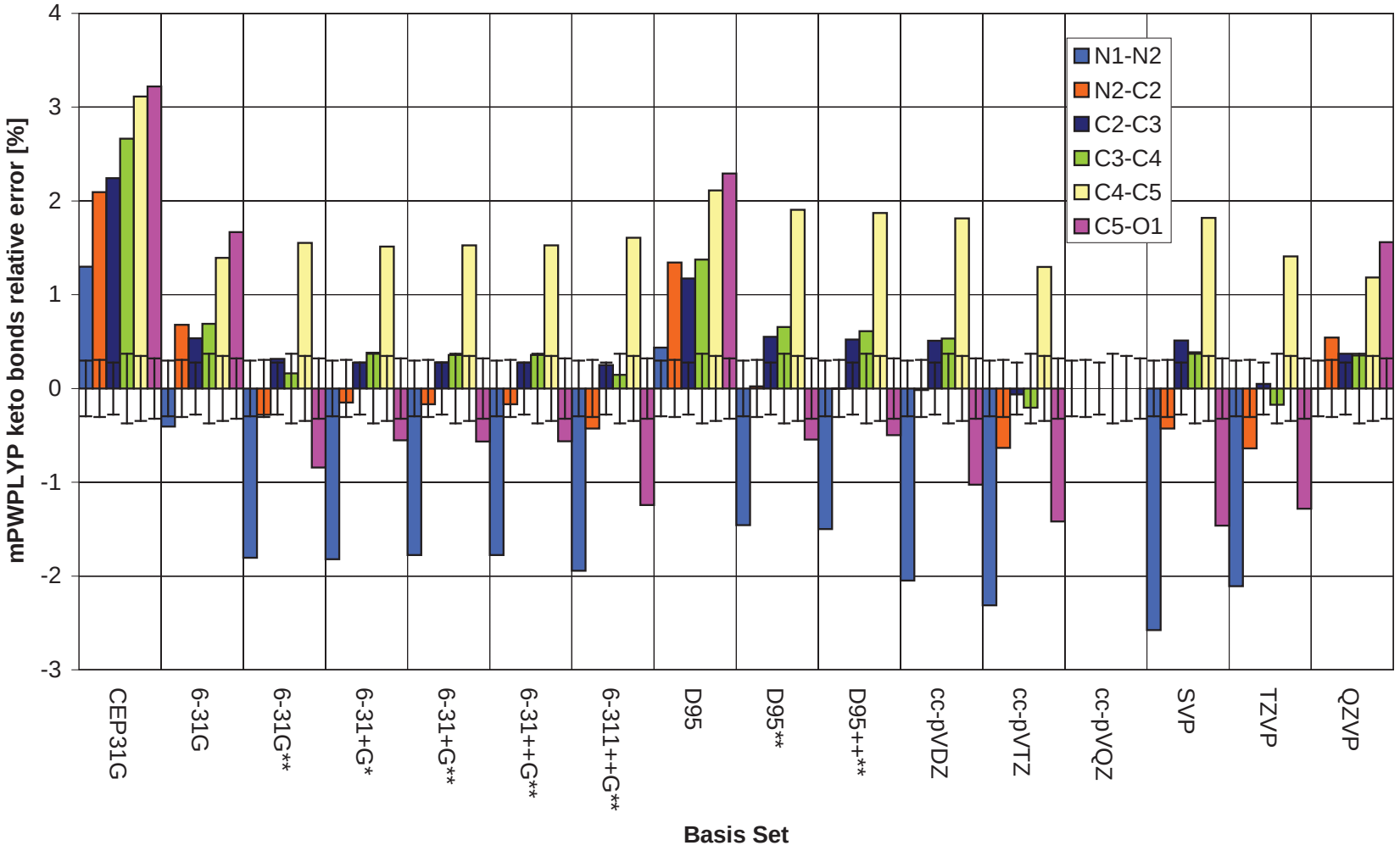
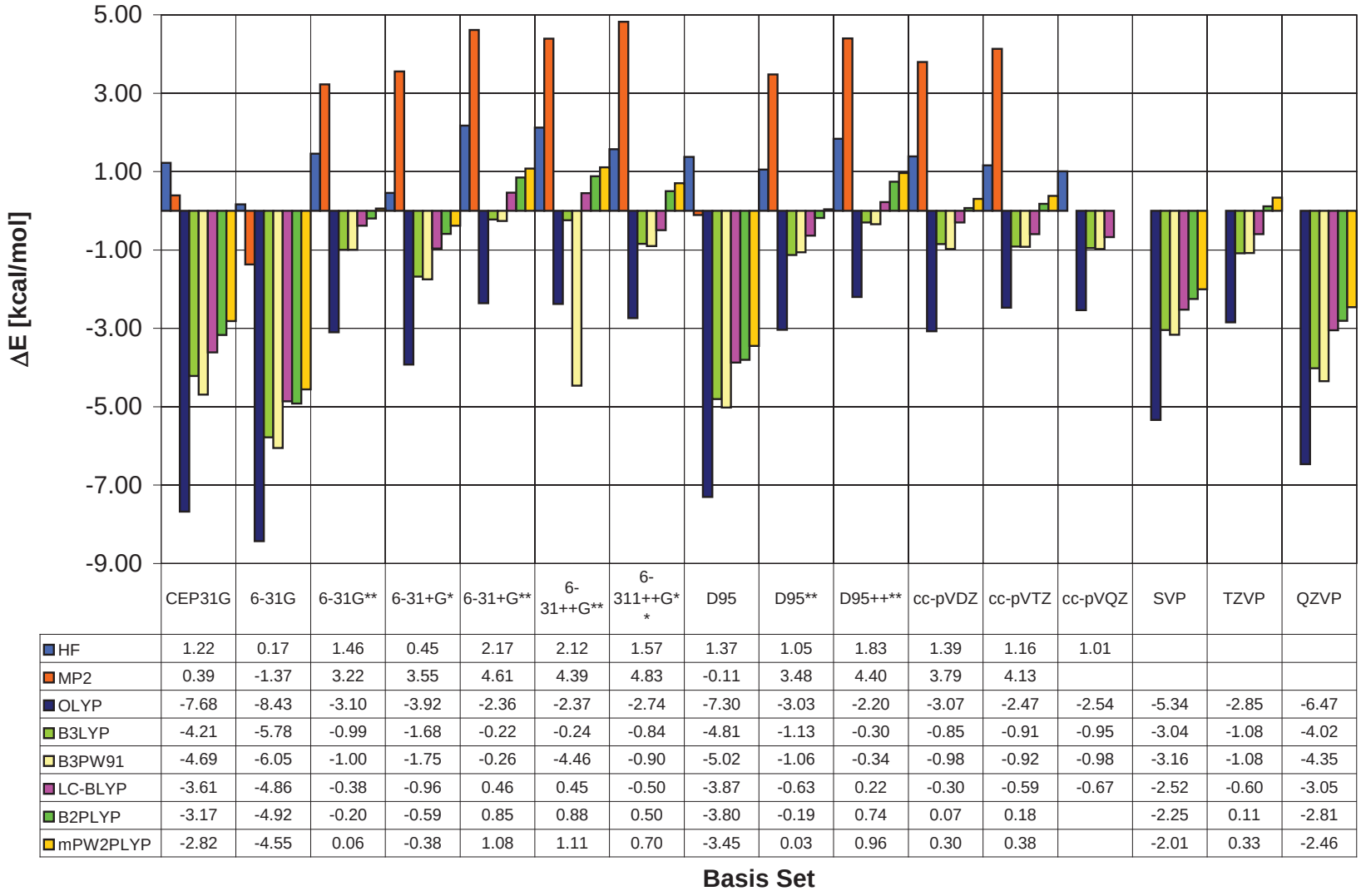
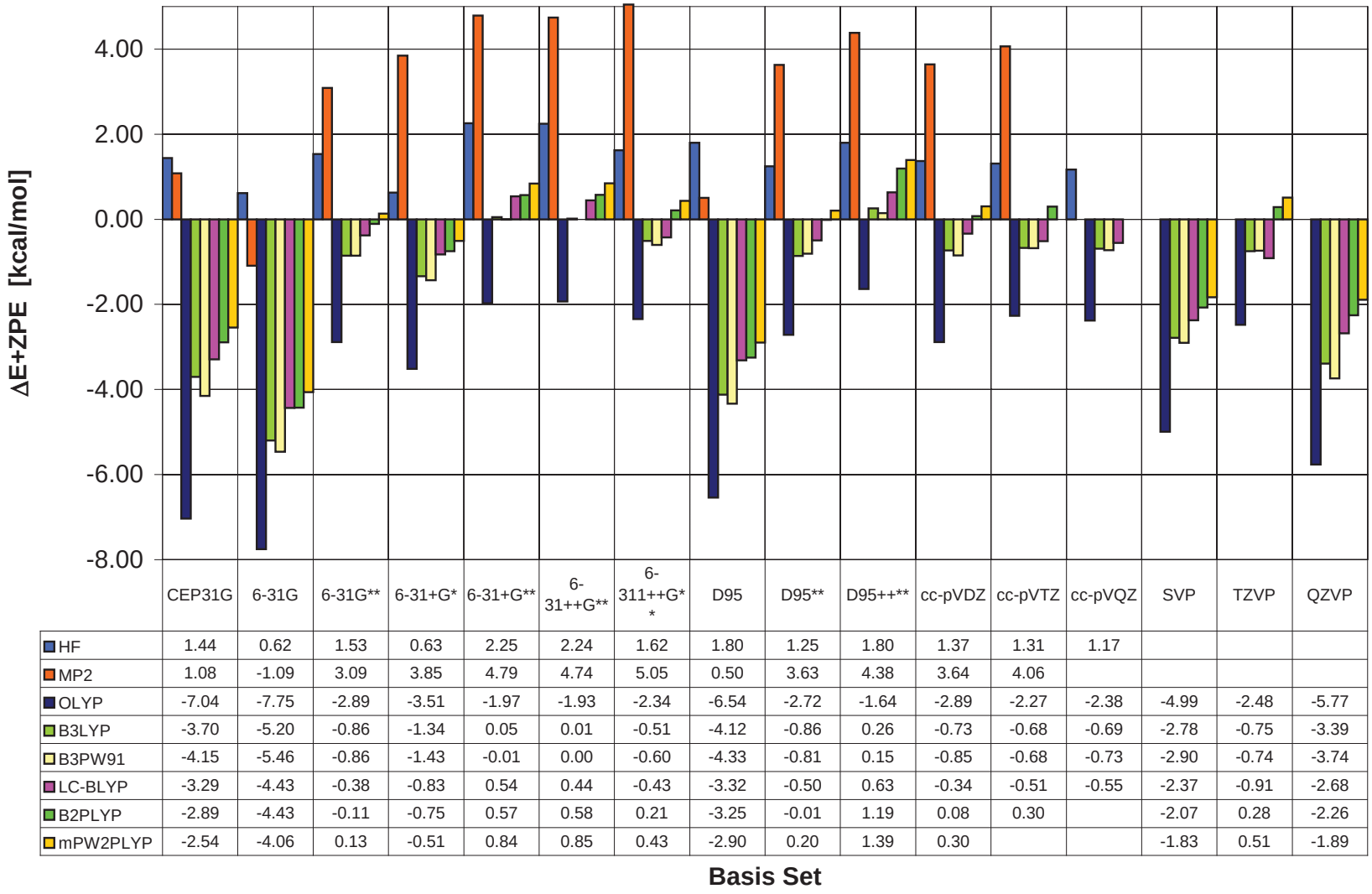


Figure S31. Relative deviations of the keto form bond lengths predicted by using mPW2PLYP method in respect of the X-ray values (Table 2). The negative value means that the bond is predicted to be shorter by the quantum-chemical calculation. The corresponding standard deviations of the X-ray values are given with error bars.



	CEP31G	6-31G	6-31G**	6-31+G*	6-31+G**	6-31++G**	6-311++G**	D95	D95**	D95++**	cc-pVDZ	cc-pVTZ	cc-pVQZ	SVP	TZVP	QZVP
HF	1.22	0.17	1.46	0.45	2.17	2.12	1.57	1.37	1.05	1.83	1.39	1.16	1.01			
MP2	0.39	-1.37	3.22	3.55	4.61	4.39	4.83	-0.11	3.48	4.40	3.79	4.13				
OLYP	-7.68	-8.43	-3.10	-3.92	-2.36	-2.37	-2.74	-7.30	-3.03	-2.20	-3.07	-2.47	-2.54	-5.34	-2.85	-6.47
B3LYP	-4.21	-5.78	-0.99	-1.68	-0.22	-0.24	-0.84	-4.81	-1.13	-0.30	-0.85	-0.91	-0.95	-3.04	-1.08	-4.02
B3PW91	-4.69	-6.05	-1.00	-1.75	-0.26	-4.46	-0.90	-5.02	-1.06	-0.34	-0.98	-0.92	-0.98	-3.16	-1.08	-4.35
LC-BLYP	-3.61	-4.86	-0.38	-0.96	0.46	0.45	-0.50	-3.87	-0.63	0.22	-0.30	-0.59	-0.67	-2.52	-0.60	-3.05
B2PLYP	-3.17	-4.92	-0.20	-0.59	0.85	0.88	0.50	-3.80	-0.19	0.74	0.07	0.18		-2.25	0.11	-2.81
mPW2PLYP	-2.82	-4.55	0.06	-0.38	1.08	1.11	0.70	-3.45	0.03	0.96	0.30	0.38		-2.01	0.33	-2.46

Figure S32. Estimated [illegible] values. Positive value corresponds to more stable enol form and *vice versa*.



	CEP31G	6-31G	6-31G**	6-31+G*	6-31+G**	6-31++G**	6-311++G**	D95	D95**	D95++**	cc-pVDZ	cc-pVTZ	cc-pVQZ	SVP	TZVP	QZVP
HF	1.44	0.62	1.53	0.63	2.25	2.24	1.62	1.80	1.25	1.80	1.37	1.31	1.17			
MP2	1.08	-1.09	3.09	3.85	4.79	4.74	5.05	0.50	3.63	4.38	3.64	4.06				
OLYP	-7.04	-7.75	-2.89	-3.51	-1.97	-1.93	-2.34	-6.54	-2.72	-1.64	-2.89	-2.27	-2.38	-4.99	-2.48	-5.77
B3LYP	-3.70	-5.20	-0.86	-1.34	0.05	0.01	-0.51	-4.12	-0.86	0.26	-0.73	-0.68	-0.69	-2.78	-0.75	-3.39
B3PW91	-4.15	-5.46	-0.86	-1.43	-0.01	0.00	-0.60	-4.33	-0.81	0.15	-0.85	-0.68	-0.73	-2.90	-0.74	-3.74
LC-BLYP	-3.29	-4.43	-0.38	-0.83	0.54	0.44	-0.43	-3.32	-0.50	0.63	-0.34	-0.51	-0.55	-2.37	-0.91	-2.68
B2PLYP	-2.89	-4.43	-0.11	-0.75	0.57	0.58	0.21	-3.25	-0.01	1.19	0.08	0.30		-2.07	0.28	-2.26
mPW2PLYP	-2.54	-4.06	0.13	-0.51	0.84	0.85	0.43	-2.90	0.20	1.39	0.30			-1.83	0.51	-1.89

Figure S33. Estimated ΔE+ZPE values. Positive value corresponds to more stable enol form and *vice versa*.