Supporting information to "A modified interaction-strength interpolation method as an important step towards self-consistent calculations."

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May 28, 2020

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1 Correlation energies

TABLE S1: Correlation energies (in Hartree) corresponding to various methods. The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect CCSD(T) data. All calculation have been performed with fully uncontacted cc-pVTZ basis set. [a]) with TPSS-GL2

				@OEPx				
	CCSD(T)	MP2	OEP2-sc	GL2	ISI	mISI	mISI[a]	ACSC
He	-0.039660	-0.033722	-0.035329	-0.044363	-0.036901	-0.037880	-0.040245	-0.037672
Li	-0.037664	-0.032766	-0.033113	-0.039994	-0.036625	-0.037035	-0.049778	-0.047631
Be	-0.086245	-0.066051	-0.070758	-0.113802	-0.093827	-0.093499	-0.092874	-0.079241
B	-0.109096	-0.084970	-0.087452	-0.134885	-0.115990	-0.115052	-0.116710	-0.106086
C	-0.135090	-0.111021	-0.113715	-0.161406	-0.142634	-0.141143	-0.150590	-0.138592
N	-0.162907	-0.143243	-0.145089	-0.191277	-0.172266	-0.170403	-0.191004	-0.175988
O	-0.214710	-0.194242	-0.197836	-0.254493	-0.227236	-0.223866	-0.238568	-0.227975
F	-0.271139	-0.254910	-0.259624	-0.326869	-0.290263	-0.285027	-0.295228	-0.283542
Ne	-0.330714	-0.323416	-0.352746	-0.405145	-0.358920	-0.351796	-0.358131	-0.341914
BeH	-0.084363	-0.070275	-0.071935	-0.097580	-0.086060	-0.086295	-0.101597	-0.095427
OH	-0.271866	-0.251585	-0.258501	-0.340946	-0.293686	-0.287072	-0.287883	-0.274333
NH_2	-0.258278	-0.233404	-0.240494	-0.327428	-0.276327	-0.269931	-0.281918	-0.266560
NH	-0.209239	-0.184706	-0.190263	-0.254288	-0.221721	-0.218033	-0.233549	-0.220886
NO	-0.523898	-0.495963	-0.535165	-0.723276	-0.598771	-0.578557	-0.516251	-0.483107
PH_2	-0.341964	-0.308711	-0.321864	-0.422856	-0.399211	-0.395922	-0.636401	-0.593883
O_2	-0.560216	-0.538007	-0.577888	-0.770850	-0.644323	-0.623286	-0.553919	-0.521423
S_2	-0.701416	-0.655289	-0.689624	-0.918035	-0.865781	-0.857520	-1.307655	-1.210837
SiH_2	-0.288381	-0.261218	-0.272959	-0.347422	-0.330311	-0.328249	-0.561487	-0.530983
C_2H_5	-0.470713	-0.422178	-0.438171	-0.601636	-0.501556	-0.489244	-0.553714	-0.516717
CH_2	-0.216482	-0.184017	-0.167273	-0.278421	-0.230092	-0.225144	-0.235491	-0.216760
CH_3	-0.231320	-0.203690	-0.209144	-0.287709	-0.242062	-0.237557	-0.271475	-0.255391
CN	-0.428080	-0.368123	-0.467363	-0.541471	-0.454037	-0.441432	-0.416596	-0.384800
COH	-0.492578	-0.464970	-0.496161	-0.670735	-0.556537	-0.538997	-0.503263	-0.469853
CH_4	-0.273230	-0.244444	-0.251094	-0.348835	-0.286640	-0.280100	-0.316937	-0.296330
LiF	-0.379001	-0.368097	-0.383314	-0.487876	-0.418808	-0.409232	-0.386879	-0.366502
Li_2	-0.107293	-0.087716	-0.089084	-0.137012	-0.116623	-0.117194	-0.133762	-0.114303
F_2	-0.654028	-0.630457	-0.664311	-0.875886	-0.742294	-0.719318	-0.635842	-0.601346
CO	-0.476203	-0.452759	-0.367822	-0.659650	-0.544370	-0.526297	-0.472407	-0.438719
CH_2	-0.216482	-0.184017	-0.189700	-0.278421	-0.230092	-0.225144	-0.235491	-0.216760
H_2	-0.039681	-0.031980	-0.032188	-0.047537	-0.035415	-0.037274	-0.040603	-0.040486
CS	-0.558780	-0.519899	-0.556076	-0.781015	-0.711328	-0.699825	-0.852923	-0.781827
LiH	-0.073802	-0.060677	-0.061393	-0.086328	-0.073108	-0.073835	-0.083349	-0.076529
N_2	-0.491774	-0.471751	-0.496969	-0.708486	-0.572963	-0.551501	-0.485747	-0.446633
P_2	-0.660820	-0.621909	-0.653345	-0.910610	-0.852628	-0.843207	-1.211761	-1.112793
NaCl	-0.579755	-0.553435	-0.569281	-0.729401	-0.690595	-0.685110	-1.101806	-1.034700
H_2O	-0.329409	-0.314495	-0.323122	-0.433049	-0.361010	-0.350379	-0.340403	-0.321116
	MAE [mH]	24.39	18.43	95.35	42.28	34.85	85.91	73.58
	MARE [%]	10.47	8.73	26.57	10.35	8.28	21.44	16.91

2 Atomization energies

TABLE S2: Atomization energies (in kcal/mol) of 27 molecules from G2 set corresponding to various methods. The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect CCSD(T) data. All calculation have been performed with fully uncontacted cc-pVTZ basis set. [a]) with TPSS-GL2

				@OEPx				
	CCSD(T)	MP2	OEP2-sc	GL2	ISI	mISI	mISI[a]	ACSC
BeH	-49.15	-52.98	-53.01	-39.92	-42.19	-42.17	-52.6	-57.28
OH	-103.44	-103.55	-104.22	-121.36	-105.77	-103.37	-95.08	-93.23
NH_2	-176.28	-173.01	-174.07	-200.95	-174.72	-171.15	-166.6	-166.39
NH	-79.69	-76.64	-77.45	-89.6	-78.05	-76.54	-73.78	-75.26
NO	-144.92	-152.57	-162.43	-224.93	-175.84	-166.44	-105.19	-100.46
PH_2	-149.07	-142.01	-143.65	-159.39	-147.72	-146.1	-153.92	-147.3
O_2	-115.11	-126.86	-135.52	-195.27	-150.09	-141.11	-79.13	-72.04
S_2	-94.41	-100.09	-105.18	-142.12	-131.3	-129.07	-103.31	-85.56
SiH_2	-129.98	-126.63	-127.48	-132.36	-123.82	-122.8	-137.63	-134.82
C_2H5	-593.95	-593.7	-596	-641.03	-586.57	-578.9	-609.65	-601.49
CH_2	-187.46	-187.02	-187.52	-195.64	-182.36	-180.91	-195.93	-194.39
CH_3	-303	-300.77	-301.33	-321.41	-295.41	-292.43	-309.07	-306.51
CN	-171.62	-161.44	-194.38	-207.01	-175.86	-170.05	-135.61	-132.61
COH	-271.53	-282.15	-288	-340.18	-294.35	-286.03	-248.89	-242.1
CH_4	-414.67	-411.71	-412.49	-444.7	-405.27	-400.65	-419.56	-414.15
LiF	-134.05	-140.47	-142.08	-166.01	-147.76	-144.77	-116.35	-112.24
Li_2	-24	-17.87	-17.97	-39.56	-31	-30.84	-25.25	-15.73
F_2	-35.38	-40.96	-47.34	-101.76	-63.87	-56.03	9.16	16.14
CO	-253.5	-266.73	-271.54	-325.78	-282.32	-274.03	-225.06	-218.1
CH_2	-177.08	-171.81	-172.29	-198.61	-173.98	-171.08	-172.5	-168.28
H_2	-108.64	-103.8	-103.85	-113.57	-99.87	-100.31	-103.26	-103.19
CS	-164.1	-172.12	-178.58	-241.72	-220.75	-215.95	-151.98	-136.4
LiH	-56.86	-51.69	-51.75	-63.2	-53.97	-53.81	-52.21	-49.28
N_2	-218.75	-230.86	-235.74	-317.06	-255.88	-244.75	-177.64	-171.94
P_2	-104.75	-107.94	-112.76	-181.65	-163.77	-160.21	-103.6	-81.6
NaCl	-93.3	-96.27	-96.1	-111.04	-106.48	-105.61	-100.02	-92.66
H_2O	-226.28	-229.77	-230.7	-265.92	-231.73	-226.45	-211.82	-206.37
	MAE	5.51	8.01	37.75	16.43	14.51	14.83	17.76
	MARE[%]	5.19	7.32	31.59	15.03	13.00	13.22	17.16

TABLE S3: Atomization energies (in kcal/mol) of AE6 set corresponding to various methods. The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect CCSD(T) data. All calculation have been performed with fully uncontacted cc-pVTZ basis set. [a]) with TPSS-GL2

				@OEPx				
	CCSD(T)	MP2	OEP2-sc	GL2	ISI	mISI	mISI[a]	ACSC
SiH_4	318.8	310.2	311.2	332.5	324.6	323.7	333.2	318
SiO	182	194.7	201	255.1	230.1	225	149.9	142.5
S_2	94.3	100	104.9	140.6	130	127.9	104.9	86.9
C_3H_4	690.8	703.4	707.2	789.4	723.4	712.8	704.1	680.5
$C_2H_2O_2$	618.1	641.6	651.9	761.8	671.5	654.1	586.2	564
C_4H_8	-1130.5	-1140.7	-1145.6	-1247.9	-1150.7	-1132.3	-1181.1	-1144.8
	MAE	12.2	17.1	82.1	32.7	23.6	25.5	21
	MARE[%]	3.71	5.55	23.59	13.56	11.67	7.51	6.87

TABLE S4: Atomization energies (in kcal/mol) of small radicals set corresponding to various methods. The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect CCSD(T) data. All calculation have been performed with fully uncontacted cc-pVTZ basis set. [a]) with TPSS-GL2

				@OEPx				
	CCSD(T)	MP2	OEP2-sc	GL2	ISI	mISI	mISI[a]	ACSC
BH_2	-167.4	-167.9	-168.0	-169.2	-163.6	-162.8	-177.9	-174.5
BH^+	218.0	217.2	136.5	148.4	144.9	144.7	134.5	135.0
BO	-189.7	-204.1	-209.5	-240.0	-211.8	-206.3	-167.5	-164.4
C_2^-	-210.3	-222.3	-229.8	-317.3	-250.0	-238.6	-184.6	-175.9
CF	-129.6	-134.8	-140.2	-165.3	-143.5	-139.0	-110.2	-107.9
CH	-82.0	-78.4	-78.8	-91.1	-82.0	-80.7	-80.1	-77.7
CN	-174.0	-161.0	-194.4	-206.4	-175.3	-169.5	-135.3	-132.3
CO^+	66.2	62.1	45.2	17.9	48.2	53.9	103.0	107.3
F_2^+	321.0	310.8	298.7	214.7	270.3	281.2	390.1	395.4
FH^+	227.4	228.7	228.3	225.9	228.1	228.7	231.0	232.8
H_2O^+	60.1	61.0	60.5	44.7	57.8	60.2	65.4	70.5
HCO	-272.6	-282.2	-288.0	-340.0	-296.6	-288.3	-250.8	-242.4
HNF	-151.4	-152.5	-159.5	-202.1	-167.2	-160.1	-124.2	-119.3
HOO	-169.0	-170.8	-183.1	-234.4	-190.3	-181.4	-134.8	-125.8
N_2^+	136.3	121.4	109.0	-17.7	76.7	92.4	208.6	213.6
NH_2	-176.4	-173.1	-174.1	-200.6	-179.6	-176.2	-170.4	-166.9
NH^+	228.4	235.3	234.8	221.3	228.8	229.9	241.5	241.0
NO	-146.0	-152.7	-162.4	-225.0	-175.9	-166.4	-105.3	-100.6
O_2^+	158.6	141.4	132.9	38.7	106.2	118.8	217.9	226.3
OF	-49.9	-50.5	-63.0	-99.5	-68.6	-62.2	-12.6	-7.9
OH	-103.5	-103.6	-104.3	-121.2	-108.1	-105.7	-97.0	-93.5
	MAE	6.1	15.7	52.98	21.69	16.59	30.23	34.23
	MARE[%]	3.89	12.05	89.10	16.97	12.18	32.10	46.35

TABLE S5: Energies (in kcal/mol) of K9 set corresponding to various methods. The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect CCSD(T) data. All calculation have been performed with fully uncontacted cc-pVTZ basis set. [a]) with TPSS-GL2

				@OEPx				
	CCSD(T)	MP2	OEP2-sc	GL2	ISI	mISI	mISI[a]	ACSC
OH + CH_4 → TS	7.1	8.52	2.07	-7.04	2.95	5.69	18.48	20.25
TS → CH_3 + H_2O	18.25	23.74	17.3	14.05	18.97	20.49	24.78	25.79
H + OH → TS	50.1	55.43	13.35	13.9	13.93	13.65	13.59	15.06
TS→O+H_2	55.19	55.63	12.93	6.27	11.12	14.03	23.15	24.71
H+H_2S → TS	4.1	6.77	5.66	3.09	4.06	4.16	5.94	6.5
TS → H_2+SH	18.93	17.37	16.15	9.92	8.03	10.15	11.73	18.62
	MAE	2.82	14.89	18.91	16.01	15.01	15.92	14.82
	MARE[%]	22.46	46.45	75.88	45.52	37.87	68.32	68.63

3 Reaction energies

TABLE S6: Hydrogen transfer reactions energies (in kcal/mol) corresponding to various methods. The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect CCSD(T) data. All calculation have been performed with fully uncontacted cc-pVTZ basis set. [a]) with TPSS-GL2

				@OEPx				
	CCSD(T)	MP2	OEP2-sc	GL2	ISI	mISI	mISI[a]	ACSC
$CH_3 + H_2 \rightarrow CH_4 + H$	-3.03	-7.15	-7.34	-9.7	-6.96	-4.52	-6.02	-4.62
$C_2H + H_2 \rightarrow C_2H_2 + H$	-30.55	-51.11	-34.73	-71.78	-54.8	-49.79	-37.39	-35.1
$C_2H_3 + H_2 \rightarrow C_2H_4 + H$	-9.43	-20.85	-12.97	-29.19	-21.58	-18.16	-14.63	-12.76
$C(CH_3)_3 + H_2 \rightarrow HC(CH_3)_3 + H$	4.81	-0.29	0.9	-2.43	0.46	3.22	2.52	3.54
$C_6H_5 + H_2 \rightarrow C_6H_6 + H$	-12.05	-41.48	-12.79	-64.98	-42.71	-36.44	-15.78	-10.14
$C_2H + C_2H_4 \rightarrow C_2H_2 + C_2H_3$	-21.11	-30.27	-21.76	-42.6	-33.22	-31.63	-22.76	-22.34
$C\ (CH_3)_3 + C_2H_4 \rightarrow HC(CH_3)_3 + C_2H_3$	14.24	20.55	13.86	26.76	22.05	21.38	17.15	16.3
$C_6H_5 + C_2H_4 \rightarrow C_6H_6 + C_2H_3$	-2.62	-20.63	0.17	-35.79	-21.12	-18.28	-1.16	2.62
$C_2H + HC(CH_3)_3 \rightarrow C_2H_2 + C(CH_3)_3$	-35.36	-50.82	-35.63	-69.36	-55.26	-53	-39.91	-38.64
$C_6H_5 + HC(CH_3)_3 \rightarrow C_6H_6 + C(CH_3)_3$	-16.86	-41.19	-13.69	-62.55	-43.17	-39.66	-18.31	-13.68
$C_2H + C_6H_6 \rightarrow C_2H_2 + C_6H_5$	-18.5	-9.63	-21.94	-6.81	-12.09	-13.35	-21.6	-24.96
$C_2H + CH_4 \rightarrow C_2H_2 + CH_3$	-27.51	-43.96	-27.39	-62.08	-47.84	-45.27	-31.37	-30.48
$C_2H_3 + CH_4 \rightarrow C_2H_4 + CH_3$	-6.4	-13.69	-5.63	-19.48	-14.62	-13.64	-8.61	-8.13
$C(CH_3)_3 + CH_4 \rightarrow HC(CH_3)_3 + CH_3$	7.84	6.86	8.24	7.27	7.42	7.74	8.54	8.17
$C_6H_5 + CH_4 \rightarrow C_6H_6 + CH_3$	-9.02	-34.32	-5.45	-55.28	-35.75	-31.92	-9.77	-5.51
	MAE	13.52	2.15	25.39	14.8	12.16	2.91	2.84
	MARE[%]	143.54	32.56	259.46	150.18	118.95	29.49	33.94

TABLE S7: Isomerization/reaction energies (in kcal/mol) corresponding to various methods. The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect CCSD(T) data. All calculation have been performed with fully uncontacted cc-pVTZ basis set. [a]) with TPSS-GL2

				@OEPx				
	CCSD(T)	MP2	OEP2-sc	GL2	ISI	mISI	mISI[a]	ACSC
cis-2-butene→trans-2-butene	-1.331	-1.596	-1.671	-1.958	-1.658	-1.508	-0.838	-1.188
methylformate→acetic acid	-16.528	-17.43	-16.406	-15.848	-16.32	-16.398	-21.717	-20.579
Propanone → Propanal	-10.543	-10.823	-11.012	-9.651	-10.339	-10.404	-11.573	-11.506
Acetylenediol → Glioxal	46.733	44.385	46.563	47.096	46.985	47.191	37.038	39.885
Water-methanol 1 → Water-methanol 2	-0.098	-0.107	-0.121	-0.213	-0.133	-0.117	0.001	0.075
Methylenecyclopropene → Cumulene	15.788	12.248	15.362	26.964	20.04	18.997	-8.516	-4.995
Methylenecyclopropene → Cyclobutadiene	-58.429	-61.826	-58.475	77.872	25.305	16.937	-52.645	-50.569
Methylenecyclopropene → Vinylacetylene	23.81	23.772	23.798	35.923	29.722	28.809	2.495	6.395
Cumulene → Cyclobutadiene	-74.217	-74.075	-73.838	50.908	5.265	-2.06	-44.129	-45.574
Cumulene → Vinylacetylene	8.022	11.524	8.436	8.959	9.682	9.812	11.011	11.39
Cyclobutadiene → Vinylacetylene	82.239	85.599	82.274	-41.949	4.417	11.872	55.14	56.964
$(H_2O)_6$ book → $(H_2O)_6$ book2	-1.413	-1.449	-1.383	-0.872	-1.263	-1.297	-2.003	-2.032
$(H_2O)_6$ book → $(H_2O)_6$ ring	-2.138	-2.096	-2.762	-4.507	-2.963	-2.316	0.291	0.292
$(H_2O)_6$ book → $(H_2O)_6$ prism	2.226	2.076	2.82	4.558	3.109	2.362	-0.016	-0.226
$(H_2O)_6$ book → $(H_2O)_6$ cage	1.459	1.4	1.909	3.211	2.096	1.565	-0.291	-0.416
$(H_2O)_6$ book2 → $(H_2O)_6$ ring	-0.724	-0.647	-1.379	-3.635	-1.701	-1.018	2.295	2.324
$(H_2O)_6$ book2 → $(H_2O)_6$ prism	3.639	3.525	4.203	5.43	4.372	3.66	1.988	1.806
$(H_2O)_6$ book2 → $(H_2O)_6$ cage	2.872	2.849	3.292	4.083	3.359	2.862	1.713	1.616
$(H_2O)_6$ ring → $(H_2O)_6$ prism	4.363	4.172	5.582	9.065	6.072	4.678	-0.307	-0.517
$(H_2O)_6$ ring → $(H_2O)_6$ cage	3.596	3.496	4.671	7.718	5.06	3.881	-0.582	-0.708
$(H_2O)_6$ prism → $(H_2O)_6$ cage	-0.767	-0.676	-0.911	-1.347	-1.013	-0.797	-0.275	-0.19
	MAE	0.89	0.39	20.71	12.48	**10.97**	7.16	6.61
	MARE[%]	7.97	16.59	96.60	42.74	**23.87**	82.39	84.89

TABLE S8: Close-shell reaction energies (in kcal/mol) corresponding to various methods. The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect CCSD(T) data. All calculation have been performed with fully uncontacted cc-pVTZ basis set. [a]) with TPSS-GL2

				@OEPx				
	CCSD(T)	MP2	OEP2-sc	GL2	ISI	mISI	mISI[a]	ACSC
$F_2O+H_2 \rightarrow F_2+H_2O$	-66.83	-70.1	-62.78	-51.36	-61.53	-62.39	-84.4	-84.06
$H_2O_2+H_2 \rightarrow 2H_2O$	-84.27	-88.41	-85.31	-73	-81.29	-81.78	-100.43	-99.96
$CO+H_2 \rightarrow CH_2O$	-3.55	-4	-3.51	2.81	-0.66	0.76	-18.33	-15.88
$CO+3H_2 \rightarrow CH_4+H_2O$	-61.55	-63.33	-60.11	-43.96	-54.98	-52.04	-96.55	-92.87
$N_2+3H_2 \rightarrow 2NH_3$	-34.7	-34.22	-31.3	-3.05	-21.26	-19.43	-69.6	-68.37
$N_2O+H_2 \rightarrow N_2+H_2O$	-79.86	-75.13	-65.2	-45.43	-66.06	-68.82	-91.89	-94.06
$HNO_2+3H_2 \rightarrow 2H_2O+NH_3$	-117.82	-121.89	-108.26	-76.06	-102.67	-103.56	-158.59	-157.86
$C2H_2+H_2 \rightarrow C_2H_4$	-49.62	-47.55	-47.29	-33.33	-42.29	-41.8	-64.31	-64.49
CH_2-$CO+2H_2 \rightarrow CH_2O+CH_4$	-42.64	-40.51	-37.57	-28	-37.12	-36.09	-54.95	-55.98
$BH_3+3HF \rightarrow BF_3+3H_2$	-98.1	-99.17	-102.65	-99.81	-96.09	-98.95	-86.38	-91.4
$HCOOH \rightarrow CO_2+H_2$	1.17	-1.4	-3.19	-11.82	-5.03	-6.04	13.36	12.72
$CO+H_2O \rightarrow CO_2+H_2$	-7.67	-11.7	-16.35	-17.36	-12.03	-12.05	-8.49	-7.72
$C_2H_2+HF \rightarrow CH_2CHF$	-28.48	-25.51	-27.47	-13.37	-21.32	-21.43	-40.56	-39.42
$HCN+H_2O \rightarrow CO+NH_3$	-13.37	-10.46	-11.27	-0.05	-7.55	-8.88	-14.89	-17.11
$HCN+H_2O \rightarrow HCONH_2$	-21.73	-21.12	-23.89	-3.57	-13.72	-14.12	-40.07	-40.11
$HCONH_2+H_2O \rightarrow HCOOH+NH_3$	-0.49	0.35	-0.54	-2.02	-0.83	-0.77	3.33	2.56
$HCN+NH_3 \rightarrow N_2+CH_4$	-40.22	-39.57	-40.07	-40.97	-41.27	-41.49	-41.83	-41.61
$CO+CH_4 \rightarrow CH_3CHO$	4.56	3.3	2.62	7.41	6.54	7.74	-11.52	-7.5
$N_2+F_2 \rightarrow$trans-N_2F_2	17.32	18.62	11.03	20.87	19.96	21.35	10.35	12.14
$N_2+F_2 \rightarrow$cis-N_2F_2	18.83	19.51	16.19	29.07	24.62	25.33	8.61	10.21
$2BH_3 \rightarrow B_2H_6$	-43.09	-44.58	-46.07	-57.72	-48.1	-45.68	-51.06	-46.96
$CH_3ONO \rightarrow CH_3NO_2$	-0.89	-5.61	-1.7	-1.85	-2.11	-2.22	-8.16	-7.29
CH_2-$C \rightarrow C_2H_2$	-44.28	-51.94	-49.9	-59.35	-51.99	-50.74	-45.45	-44.31
Allene→propyne	-1.1	-4.43	-3.43	-7.61	-5.42	-5.16	-3.36	-2.86
Cyclopropene→propyne	-23.38	-23.68	-23.99	-28.09	-26.72	-26.64	-10.5	-13.49
Oxirane→CH3CHO	-26.73	-25.75	-26.63	-27.36	-27.74	-27.95	-16.78	-19.52
Vinlyalcohol→CH_3CHO	-9.79	-10	-10.8	-11.8	-11.11	-11.18	-9.38	-9.01
Cyclobutene→1,3-butadiene	-11.5	-9.41	-9.81	-11.44	-11.72	-11.95	3.4	1.5
$2NH_3 \rightarrow (NH_3)_2$	-3.84	-4	-4.25	-5.14	-4.58	-3.91	-2.99	-3.06
$2H_2O \rightarrow (H_2O)_2$	-5.67	-5.84	-6.26	-6.84	-6.3	-5.67	-4.83	-4.93
$2HF \rightarrow (HF)_2$	-4.19	-4.22	-4.81	-5.41	-4.77	-4.14	-3.03	-3.02
	MAE	2.04	2.98	10.57	4.66	4.63	11.4	10.32
	MARE[%]	46.79	34.53	106.30	54.47	57.34	144.77	125.72

TABLE S9: Open-shell reaction energies (in kcal/mol) corresponding to various methods. The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect CCSD(T) data. All calculation have been performed with fully uncontacted cc-pVTZ basis set. [a]) with TPSS-GL2

				@OEPx				
	CCSD(T)	MP2	OEP2-sc	GL2	ISI	mISI	mISI[a]	ACSC
$HCl+H \rightarrow Cl+H_2$	4.13	-1.17	-1.17	-6.1	-8.82	-6.75	-5.26	1.46
$H_2O+F_2 \rightarrow 2HF+O$	13.37	12.48	6.48	-42.22	-8.27	-1.4	49.25	54.13
$CH_4+OH \rightarrow CH_3+H_2O$	11.19	15.28	15.31	21.27	16.1	14.86	6.26	5.5
$CH_3+Cl_2 \rightarrow CH_3Cl+Cl$	31.32	31.85	30.75	23.28	31.5	31.39	39.76	37.69
$S+2HCl \rightarrow H_2S+Cl_2$	22.54	23.43	26.05	49.22	42.3	40.84	24.01	13.42
$N+O_2 \rightarrow NO+O$	29.8	25.71	26.91	29.66	25.75	25.33	26.05	28.42
$4HCl+O_2 \rightarrow 2H_2O+2Cl_2$	25.73	28.07	25.25	32.55	24.54	24.47	15.28	19.12
$2NO \rightarrow N_2+O_2$	44.03	52.59	46.4	62.47	54.29	52.99	46.39	43.05
$2H_2O_2 \rightarrow 2H_2O+O_2$	48.36	51.74	52.44	36.69	49.01	52.47	62.86	65.6
$Cl_2+H \rightarrow HCl+Cl$	51.36	47.33	45.3	32.29	35.7	36.57	56.1	59.07
$2SO_2+O_2 \rightarrow 2SO_3$	35.68	44.58	42.25	26.62	31.9	31.95	65.02	59.18
$H_2S+F_2 \rightarrow S+2HF$	61.25	66.52	60.81	22.48	36.75	40.55	72.44	86.28
$NO+N \rightarrow O+N_2$	73.83	78.29	73.31	92.13	80.04	78.31	72.45	71.48
$2SiH_3 \rightarrow Si_2H_6$	76.97	77.02	77.61	90.38	86.03	85.11	87.9	77.18
$CH_3+SH \rightarrow CH_3SH$	76.72	81.14	82.15	101.73	100.37	98.36	87.69	73.49
$CS+O \rightarrow CO+S$	89.4	94.62	92.98	84.05	61.57	58.08	73.08	81.7
$CH_3+Cl \rightarrow CH_3Cl$	84.47	89.49	90.46	110.72	110.61	108.73	95.53	80.34
$NH+H \rightarrow NH_2$	96.59	96.37	96.62	111.34	99.71	98.01	94.5	91.13
$Si+2H_2 \rightarrow SiH_4$	101.49	102.58	103.41	105.31	112.11	108.57	119.99	111.48
$2C_2H_4+O_2 \rightarrow 2CH_3CHO$	104.35	110.31	109.71	105.1	102.52	100.69	116.47	111.97
$NH_2+H \rightarrow NH_3$	113.4	115.24	115.22	129.55	116.75	114.87	113.58	108.54
$2H_2+O_2 \rightarrow 2H_2O$	120.18	125.07	118.19	109.32	113.58	111.1	138	134.33
$CO_2+C \rightarrow 2CO$	128.17	129.06	128.34	156.12	138.46	135.88	108	107.2
$N_2H_4+O_2 \rightarrow N_2+2H_2O$	132.86	139.43	134.48	151.22	139.48	138.47	119.64	116.77
$4NH_3+5O_2 \rightarrow 4NO+6H_2O$	203.1	201.58	199.16	196.92	189.65	188.46	182	180.15
$2NH_3+2NO+O \rightarrow 2N_2+3H_2O$	247.16	269.4	260.13	321.04	278.44	270.76	223.35	212.2
	MAE	4.33	3.43	18.29	11.52	10.38	12.16	11.61
	MARE [%]	12.28	12.65	51.53	34.64	29.47	35.61	30.20

4 Vertical ionization potentials

TABLE S10: Vertical ionization potential (in eV) corresponding to various methods calculated as an energy difference between the neutral and the ionic species ($VIP = E(N) - E(N-1)$) . The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect CCSD(T) data. The computational setup, namely basis sets and geometries in case of molecules, is identical as in J. Comput. Chem. 37 (2016), 2081. [a]) with TPSS-GL2

				@OEPx				
	CCSD(T)	MP2	OEP2-sc	GL2	ISI	mISI	mISI[a]	ACSC
Ar	-15.63	-15.77	-15.75	-16.31	-16.01	-15.94	-16.06	-15.76
Be	-9.31	-8.88	-8.88	-10.01	-9.51	-9.49	-9.14	-8.82
C_2H_2	-11.43	-11.6	-11.64	-12.94	-11.94	-11.77	-11.08	-10.91
C_2H_4	-10.63	-10.65	-10.7	-11.94	-11.01	-10.85	-10.11	-9.95
C_2H_6	-13.01	-13.11	-12.67	-12.74	-12.69	-12.68	-13.23	-13.19
CH_2CF_2	-10.61	-10.67	-10.5	-11.3	-10.83	-10.74	-10.77	-10.58
CH_3CN	-10.75	-11.33	-10.76	-12.47	-11.53	-11.36	-10.94	-10.72
CH_4	-14.37	-14.4	-14.41	-14.82	-14.46	-14.4	-14.52	-14.41
CHF_3	-14.59	-14.61	-14.23	-15.87	-15.93	-15.94	-16.95	-16.85
Cl_2	-11.45	-11.5	-11.37	-11.06	-11.09	-11.1	-12.54	-12.29
CO_2	-13.7	-14.63	-13.85	-15.51	-14.63	-14.46	-14.03	-13.79
CO	-13.94	-14.31	-13.71	-14.88	-14.35	-14.24	-14.32	-14.21
CS	-11.27	-11.63	-11.19	-14.52	-13.73	-13.58	-11.64	-11.23
FCN	-13.65	-13.85	-13.72	-16.33	-15.32	-15.13	-14.45	-14.29
H_2CO	-10.83	-11.26	-10.84	-12.2	-11.41	-11.27	-10.75	-10.59
H_2CS	-9.29	-9.57	-9.28	-10.73	-10.27	-10.19	-9.48	-9.16
H_2O	-12.5	-12.69	-12.7	-13.6	-12.86	-12.73	-12.16	-12
HCCF	-11.5	-11.54	-11.44	-12.49	-11.8	-11.67	-11.41	-11.2
HCl	-12.59	-12.67	-12.66	-13.27	-13.07	-13.03	-12.97	-12.65
HCN	-13.9	-14.72	-14.09	-16.44	-15.1	-14.86	-13.81	-13.66
He	-24.48	-24.54	-24.41	-24.74	-24.51	-24.54	-24.49	-24.34
He_2	-24.48	-24.54	-24.52	-24.7	-24.52	-24.54	-24.74	-24.8
HF	-15.96	-16.18	-16.2	-17.05	-16.38	-16.26	-15.57	-15.41
Mg	-7.57	-7.32	-7.33	-8.11	-7.97	-7.94	-7.44	-7.22
N_2	-15.51	-15.34	-15.06	-13.41	-14.59	-14.76	-16.66	-16.64
NCCN	-13.51	-14.85	-13.77	-12.62	-13.89	-14.08	-16.52	-16.49
Ne_2	-21.34	-21.42	-20.72	-22.37	-21.79	-21.67	-21.01	-20.86
Ne	-21.47	-21.67	-21.69	-22.39	-21.83	-21.72	-21.04	-20.88
NH_3	-10.78	-10.86	-10.88	-11.69	-11.01	-10.9	-10.52	-10.39
OCS	-11.18	-11.75	-11.27	-12.6	-12.1	-12	-11.69	-11.36
P_2	-10.66	-10.68	-10.55	-10.43	-10.4	-10.39	-11.41	-11.06
SiH_4	-12.78	-12.82	-12.72	-13.13	-13	-12.98	-12.99	-12.88
	MAE	0.24	0.17	1.17	0.61	0.52	0.42	0.39
	MARE [%]	2.37	1.21	9.03	4.90	4.26	3.83	3.55

5 Binding energies

TABLE S11: Binding energies (in kcal/mol) corresponding to various methods. The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect CCSD(T) data. All calculation have been performed with fully uncontacted cc-pVTZ basis set. [a]) with TPSS-GL2

				@OEPx				
	CCSD(T)	MP2	OEP2-sc	GL2	ISI	mISI	mISI[a]	ACSC
				WI				
He-Ne	-0.04	-0.03	-0.03	0.90	0.61	0.62	0.32	0.02
He-Ar	-0.06	-0.06	-0.06	0.83	0.72	0.72	0.51	0.05
Ne-Ne	-0.08	-0.06	-0.07	-0.10	-0.12	-0.05	0.03	0.04
Ne-Ar	-0.13	-0.12	-0.14	-0.23	-0.21	-0.16	0.64	0.08
CH_4-Ne	-0.22	-0.18	-0.21	-0.32	-0.31	-0.17	-4.99	0.05
C_6H_6-Ne	-0.45	-0.61	-0.71	-1.26	-0.97	-0.55	-6.78	0.18
CH_4-CH_4	-0.59	-0.56	-0.65	-1.25	-0.97	-0.53	0.22	0.24
				HB6				
HF-HF	-4.67	-4.59	-4.74	-4.17	-4.46	-4.15	-4.74	-4.80
H_2O-H_2O	-5.11	-5.10	-5.34	-5.15	-5.19	-4.71	-4.93	-5.04
NH_3-H_2O	-6.57	-6.66	-7.00	-7.13	-6.86	-6.26	-6.73	-6.49
HF-HCN	-7.58	-7.86	-8.20	-7.76	-7.77	-7.20	-8.35	-7.73
$(HCONH_2)_2$	-15.27	-15.83	-15.36	-14.91	-15.36	-14.39	-16.30	-16.77
$(HCOOH)_2$	-18.32	-18.18	-18.16	-15.50	-16.85	-16.01	-19.98	-20.51
				DI6				
HCl-HCl	-2.26	-2.56	-2.71	-3.74	-3.3	-3.15	-1.32	-1.28
H_2S-HCl	-3.67	-4.19	-4.41	-5.83	-5.19	-4.98	-3	-2.8
CH_3Cl-HCl	-3.98	-4.49	-4.81	-6.59	-5.67	-5.36	-3.6	-2.5
CH_3SH-HCN	-3.96	-4.25	-4.52	-5.46	-4.71	-4.29	-2.63	-3.28
CH_3SH-HCl	-5.34	-6.13	-6.56	-8.84	-7.61	-7.22	-5.79	-3.99
				DHB				
AlH-HF	-6.56	-6.85	-7.31	-7.71	-7.05	-6.77	-5.11	-4.98
LiH-HCl	-11.8	-13.28	-13.53	-14.93	-13.9	-13.6	-11.92	-12.41
LiH-HF	-14.1	-14.74	-15.22	-14.75	-14.37	-13.86	-14.12	-14.66
MgH_2-HCl	-5.08	-5.72	-5.93	-7.45	-6.7	-6.49	-3.78	-3.99
MgH_2-HF	-7.38	-7.66	-8.05	-8.21	-7.66	-7.39	-6.66	-6.58
BeH_2-Hcl	-2.44	-2.7	-2.81	-3.88	-3.33	-3.15	-1.35	-1.43
BeH_2-HF	-3.66	-3.72	-3.96	-4.2	-3.85	-3.45	-2.82	-2.94
				CT7				
C_2H_4-F_2	-1.19	-1.6	-2.05	-3.02	-1.93	-1.34	-4.79	0.43
NF_3-HNC	-1.85	-1.99	-2.46	-3.6	-2.57	-2.01	-1.42	-0.33
C_2H_4-Cl_2	-2.6	-4.14	-5.09	-8.96	-6.73	-6.1	-8.02	0.11
NH_3-F_2	-1.86	-2.06	-2.91	-3.1	-2.23	-1.66	-3.07	-0.21
NF_3-ClF	-2.03	-2.44	-3.43	-4.96	-3.54	-2.95	0.5	0.18
NF_3-HF	-2.26	-2.33	-2.75	-3.62	-2.77	-2.25	-0.97	-0.9
C_2H_2-ClF	-3.85	-5.04	-5.95	-7.73	-5.88	-5.18	-6.52	-2.18
HCN-ClF	-4.87	-5.8	-6.89	-7.93	-6.54	-5.95	-5.33	-3.3
NH_3-Cl_2	-5.13	-5.87	-7.18	-8.75	-7.47	-7.07	-7.63	-3.33
H_2O-ClF	-5.22	-5.69	-6.73	-6.3	-5.68	-5.28	-5.47	-4.57
NH_3-ClF	-2.03	-2.44	-3.43	-4.96	-7.7	-9.33	0.50	0.18
	MAE	0.38	0.74	1.63	1.05	0.91	1.37	1.03
	MARE[%]	12.58	24.28	154.22	113.00	103.06	195.24	58.49

6 GL2 correlation energies

TABLE S12: The GL2 correlation energies obtained from exact and various semi-local energy expression (in mHa) calculated on top of self-consistent OEPx otbitalas and orbital energies in fully uncontacted cc-pVTZ basis set. The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect exact GL2 data.

	GL2	PBE-GL2	TPSS-GL2	revTPSS-GL2
He	-44.363	-46.337	-47.903	-51.808
Li	-39.994	-57.796	-56.165	-60.445
Be	-113.802	-113.424	-112.781	-122.476
B	-134.885	-140.928	-137.370	-147.979
C	-161.406	-176.444	-174.906	-186.062
N	-191.277	-217.471	-219.668	-231.197
O	-254.493	-281.078	-275.198	-288.047
F	-326.869	-346.140	-341.461	-355.356
Ne	-405.145	-412.358	-414.265	-429.077
BeH	-97.580	-124.940	-119.303	-129.022
OH	-340.946	-349.456	-342.201	-357.811
NH_2	-327.428	-357.601	-347.322	-364.718
NH	-254.288	-286.955	-277.722	-292.113
NO	-723.276	-627.878	-619.234	-645.573
PH_2	-422.856	-746.632	-735.950	-764.970
O_2	-770.850	-666.881	-657.954	-684.674
S_2	-918.035	-1515.626	-1501.791	-1552.064
SiH_2	-347.422	-654.585	-643.779	-670.087
C_2H_5	-601.636	-738.081	-717.140	-753.425
CH_2	-226.830	-289.601	-277.095	-292.735
CH_3	-287.709	-359.285	-346.147	-365.085
CN	-541.471	-511.417	-500.899	-524.626
COH	-670.735	-619.500	-610.582	-638.332
CH_4	-348.835	-424.613	-417.151	-439.289
LiF	-487.876	-451.807	-453.721	-473.896
Li_2	-137.012	-164.787	-162.619	-177.591
F_2	-875.886	-744.616	-744.887	-773.967
CO	-659.650	-571.305	-568.931	-594.734
CH_2	-278.421	-300.300	-296.480	-313.730
H_2	-47.537	-57.984	-53.775	-58.555
CS	-781.015	-1000.806	-995.643	-1033.483
LiH	-86.328	-100.313	-100.918	-109.990
N_2	-708.486	-595.291	-592.501	-618.479
P_2	-910.610	-1403.130	-1396.442	-1445.947
NaCl	-729.401	-1265.590	-1264.610	-1307.531
H_2O	-433.049	-418.419	-416.233	-434.688
	MAE	105.03	101.89	110.28
	MARE [%]	21.99	15.45	18.87

7 Binding energies on top of HF orbitals

TABLE S13: Binding energies (in kcal/mol) from MP2, ISI and mISI methods calculated on top of HF orbitals. The last lines report the mean absolute error (MAE) and mean absolute relative error (MARE) calculated with respect CCSD(T) data. All calculation have been performed with fully uncontacted cc-pVTZ basis set.

	MP2	ISI	mISI
	WI		
He-Ne	-0.03	-0.05	-0.02
He-Ar	-0.06	-0.05	-0.04
Ne-Ne	-0.06	-0.08	-0.04
Ne-Ar	-0.12	-0.11	-0.09
CH_4-Ne	-0.18	-0.19	-0.12
C_6H_6-Ne	-0.61	-0.54	-0.34
CH_4-CH_4	-0.56	-0.49	-0.28
MAE	0.04	0.04	0.09
	HB6		
NH_3-NH_3	-3.25	-3.13	-2.87
HF-HF	-4.59	-4.68	-4.51
H_2O-H_2O	-5.10	-5.10	-4.85
NH_3-H_2O	-6.66	-6.54	-6.24
HF-HCN	-7.86	-7.83	-7.54
$(HCONH_2)_2$	-15.83	-15.80	-15.27
$(HCOOH)_2$	-18.18	-18.35	-17.84
MAE	0.17	0.14	0.23
	DI6		
H_2S-H_2S	-2.15	-1.98	-1.92
Hcl-Hcl	-2.56	-2.39	-2.33
H_2S-Hcl	-4.19	-3.94	-3.86
CH_3Cl-Hcl	-4.49	-4.13	-4.01
CH_3SH-HCN	-4.25	-3.98	-3.81
CH_3SH-Hcl	-6.13	-5.67	-5.52
MAE	0.45	0.17	0.11
	DHB		
AlH-HCl	-5.26	-4.92	-4.83
AlH-HF	-6.85	-6.54	-6.41
LiH-HCl	-13.28	-12.78	-12.65
LiH-HF	-14.74	-14.44	-14.16
MgH_2-HCl	-5.72	-5.4	-5.31
MgH_2-HF	-7.66	-7.38	-7.24
BeH_2-HCl	-2.7	-2.49	-2.42
BeH_2-HF	-3.72	-3.59	-3.39
MAE	0.54	0.27	0.25
	CT7		
NF_3-HCN	-1.24	-1.2	-1.00
C_2H_4-F_2	-1.6	-1.29	-1.03
NF_3-HNC	-1.99	-1.68	-1.44
C_2H_4-Cl_2	-4.14	-3.43	-3.22
NH_3-F_2	-2.06	-1.77	-1.51
NF_3-ClF	-2.44	-2.11	-1.88
NF_3-HF	-2.33	-2.04	-1.80
C_2H_2-ClF	-5.04	-4.47	-4.21
HCN-ClF	-5.8	-5.39	-5.16
NH_3-Cl_2	-5.87	-5.44	-5.29
H_2O-ClF	-5.69	-5.45	-5.27
NH_3-ClF	-2.44	-4.65	-5.68
MAE	0.55	0.48	0.57
MAE	0.38	0.25	0.29
MARE[%]	12.58	10.26	17.11