Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting information for: Semi-empirical or non-empirical double-hybrid density functionals: which are more robust?

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SI.1 Damping parameters for the DFT-D3 dispersion correction

While details on the exact form of the three DFT-D3 variants are given elsewhere,[S1–S3] it suffices to know that one needs to empirically determine three damping parameters for DFT-D3(BJ), two for DFT-D3(0), and one for DFT-D3(CSO). In addition, all corrections use the same global scale parameter for the description of long-range interactions, called 's_6'. In conventional density functional approximations (DFAs), this is fixed to unity to ensure a proper asymptotic behaviour of the dispersion energy, however, due to their non-local second-order perturbative (PT2) contribution, s_6 needs to be individually determined for each DHDF. We did this according to the instructions outlined in Ref. S4, which recommended to compute the scaled perturbative dispersion energies in three rare-gas dimers at non-equilibrium geometries [E_{disps}(PT2)] and to relate them to those of coupled cluster singles doubles with perturbative triples treatments [E_{disp} (CCSD(T))]. For each dimer, a ratio between the two energy contributions was calculated and their average subtracted from unity, thus giving the s_6 for the DHDF. After having determined s_6, the remaining damping parameters in all three DFT-D3 variants were determined in a least-squares fit on the combined S66x8,[S5] S22x5,[S6] and NCIBLIND[S7] training sets with a total of 718 data points that include non-covalently bound dimers in their equilibrium and non-equilibrium internuclear distances.
Note that for PBE-QIDH-D3(BJ) and SOS1-PBE-QIDH-D3(BJ) the authors presented an s_6 that was determined in a free fit on their own training set. For consistency reasons, we keep the same DFT-D3(BJ) parameters. However, for the determination of the new DFT-D3(0) and -D3(CSO) damping parameters, we decided to follow the previously outlined strategy to determine s_6.

SI.1.1 Damping parameters for the DFT-D3 correction with Becke-Johnson damping [DFT-D3(BJ)]

DFT-D3(BJ) parameters for all tested DHDFs are shown in Tab. S1; a description of DFT-D3(BJ) and its parameters is given in Ref. S2. For those parameters that were determined in this work, s_6 had been determined according to instructions outlined in Ref. S4 before the remaining parameters were fitted.
Any work using DFT-D3(BJ) should cite Refs. S1 and S2 as well as the work that presented the respective functional parameters for the first time (see last column of Tab. S1).

Table S1: Parameters for the DFT-D3 correction with Becke-Johnson damping [DFT-D3(BJ)].

Functional	s_6	a_1	s_8	a_2	cite Ref.
B2NC-PLYP	0.363	0.609	0.616	5.354	this work
mPW2NC-PLYP	0.385	−0.493	0.000	9.529	this work
ωB97X-2	0.547	3.520	0.000	7.795	this work
B2PPW91	0.852	1.025	2.936	1.806	this work
mSD-PBEPBE	0.097	0.202	0.087	1.434	this work
LS1-DH	0.508	0.387	1.073	6.747	this work
LS1-TPSS	0.305	1.941	0.660	0.053	this work
PBE0-DH	0.880	0.000	0.165	6.385	S8
revPBE0-DH	0.835	0.412	1.089	4.186	this work
PBE0-2	0.540	0.000	0.515	8.345	S8
PBE-QIDH	0.61	0.114	0.566	7.538	S9
PBE-CIDH	0.781	0.000	1.228	7.455	this work
SOS0-PBE0-2	0.613	0.573	0.167	3.572	this work
SOS0-PBE0-DH	0.892	0.000	1.044	6.930	this work
SOS0-PBE-CIDH	0.856	0.000	1.011	6.957	this work
SOS1-PBE-QIDH	0.75	0.002	0.200	6.794	S9
SOS0-PBE0-2(b)	0.421	2.498	6.241	−0.138	this work
SOS0-PBE0-DH(b)	0.839	0.000	0.975	7.139	this work
SOS0-PBE-QIDH(b)	0.597	0.316	1.226	6.712	this work
SOS0-PBE-CIDH(b)	0.785	0.000	1.004	7.327	this work
TPSS0-DH	0.837	0.039	1.189	6.570	this work
TPSS-QIDH	0.601	0.099	0.808	6.691	this work
TPSS-CIDH	0.784	0.000	1.111	6.843	this work
MP2	−0.04	0.980	−2.887	3.945	S10
SCS-MP2	0.18	0.307	0.060	1.499	S10
SOS-MP2	0.30	0.141	0.139	2.583	S10

SI.1.2 Damping parameters for the DFT-D3 correction with zero damping [DFT-D3(0)]

DFT-D3(0) parameters for all tested DHDFs are shown in Tab. S2; a description of DFT-D3(0) and its parameters is given in Ref. S1. For those parameters that were determined in this work, s_6 had been determined according to instructions outlined in Ref. S4 before the remaining parameters were fitted.
Any work using DFT-D3(0) should cite Ref. S1 as well as the work that presented the respective functional parameters for the first time (see last column of Tab. S2).

Table S2: Parameters for the DFT-D3 correction with zero damping [DFT-D3(0)].

Functional	s_6	$s_{r,6}$	s_8	cite Ref.
B2NC-PLYP	0.363	1.321	0.092	this work
mPW2NC-PLYP	0.385	1.682	0.290	this work
ωB97X-2	0.547	4.733	−0.109	this work
B2PPW91	0.852	1.199	1.671	this work
mSD-PBEPBE	0.097	1.958	0.237	this work
LS1-DH	0.508	1.261	0.000	this work
LS1-TPSS	0.305	1.320	0.000	this work
PBE0-DH	0.880	1.128	0.000	this work
revPBE0-DH	0.835	7.260	1.931	this work
PBE0-2	0.540	1.243	−0.228	this work
PBE-QIDH	0.400	1.114	0.054	this work
PBE-CIDH	0.781	1.303	0.477	this work
SOS0-PBE0-2	0.613	1.441	0.679	this work
SOS0-PBE0-DH	0.892	1.356	0.855	this work
SOS0-PBE-CIDH	0.856	1.360	0.814	this work
SOS1-PBE-QIDH	0.732	1.084	0.000	this work
SOS0-PBE0-2(b)	0.421	1.382	0.000	this work
SOS0-PBE0-DH(b)	0.839	1.331	0.626	this work
SOS0-PBE-QIDH(b)	0.597	1.213	0.000	this work
SOS0-PBE-CIDH(b)	0.785	1.325	0.526	this work
TPSS0-DH	0.837	1.317	0.944	this work
TPSS-QIDH	0.601	1.094	0.000	this work
TPSS-CIDH	0.784	1.310	0.814	this work
MP2	−0.04	0.913	−0.183	S10
SCS-MP2	0.18	0.495	0.048	S10
SOS-MP2	0.30	0.605	0.147	S10

SI.1.3 Damping parameters for the DFT-D3 correction with 'C-six-only' damping [DFT-D3(CSO)]

DFT-D3(CSO) parameters for all tested DHDFs are shown in Tab. S3; a description of DFT-D3(CSO) and its parameters is given in Ref. S3. For those parameters that were determined in this work, s_6 had been determined according to instructions outlined in Ref. S4 before the remaining parameter was fitted.
Any work using DFT-D3(CSO) should cite Ref. S3 and Ref. S1 as well as the work that presented the respective functional parameters for the first time (see last column of Tab. S3).

Table S3: Parameters for the DFT-D3 correction with 'C-six-only' damping [DFT-D3(CSO)].

Functional	s_6	a	cite Ref.
ωB97X-2	0.547	−0.738	this work
B2GPPLYP	0.56	0.719	this work
DSD-BLYP	0.50	0.835	this work
DSD-PBEB95	0.61	0.201	this work
DSD-PBEP86	0.418	0.629	this work
mPW2PLYP	0.66	0.441	this work
PWPB95	0.82	0.048	this work
B2NC-PLYP	0.363	0.344	this work
mPW2NC-PLYP	0.385	0.319	this work
B2PPW91	0.852	1.435	this work
mSD-PBEPBE	0.097	1.532	this work
B2PLYP	0.64	0.949	this work
LS1-DH	0.508	0.115	this work
LS1-TPSS	0.305	0.194	this work
PBE0-DH	0.880	0.123	this work
revPBE0-DH	0.835	0.926	this work
PBE0-2	0.540	−0.200	this work
PBE-QIDH	0.400	0.789	this work
PBE-CIDH	0.781	0.225	this work
SOS0-PBE0-2(b)	0.421	−0.030	this work
SOS0-PBE0-DH(b)	0.839	0.216	this work
SOS0-PBE-CIDH(b)	0.785	0.220	this work
SOS0-PBE-QIDH(b)	0.597	0.159	this work
SOS0-PBE0-2	0.613	0.552	this work
SOS0-PBE0-DH	0.892	0.275	this work
SOS0-PBE-CIDH	0.856	0.303	this work
SOS1-PBE-QIDH	0.732	0.406	this work
TPSS0-DH	0.837	0.532	this work
TPSS-CIDH	0.784	0.531	this work
TPSS-QIDH	0.601	0.511	this work
SCS-MP2	0.18	0.898	this work
SOS-MP2	0.30	0.336	this work

SI.2 Dispersion-corrected vs. -uncorrected results for selected methods

SI.2.1 WTMADs for four different SOS1/SOS0-DHDF type of functionals

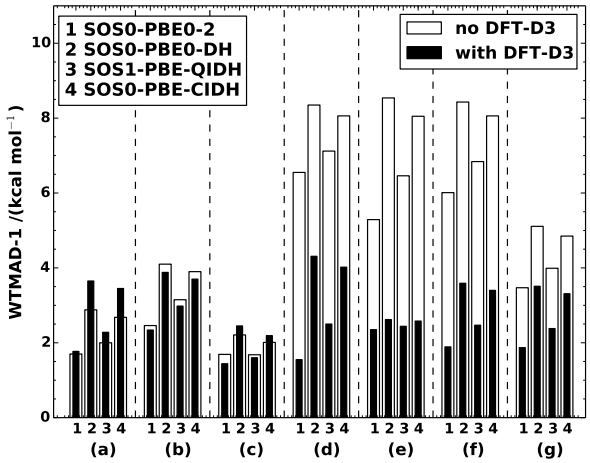

Figure S1: The effect of dispersion corrections on WTMAD-1 values (kcal/mol). The values are displayed for the various categories of GMTKN55, namely basic properties and reactions of small systems (a), isomerisations and reactions of large systems (b), barrier heights (c), intermolecular noncovalent interactions (d), intramolecular noncovalent interactions (e), and all noncovalent interactions (f). Values are also shown for the entire GMTKN55 database (g).

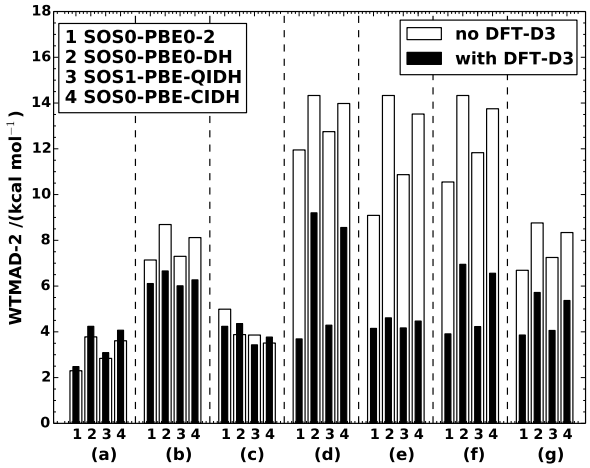

Figure S2: The effect of dispersion corrections on WTMAD-2 values (kcal/mol). The values are displayed for the various categories of GMTKN55, namely basic properties and reactions of small systems (a), isomerisations and reactions of large systems (b), barrier heights (c), intermolecular noncovalent interactions (d), intramolecular noncovalent interactions (e), and all noncovalent interactions (f). Values are also shown for the entire GMTKN55 database (g).

Table S4: WTMAD-1 and WTMAD-2 values (in parenthesis) for four different SOS1/SOS0-DHDF type of functionals with and without the DFT-D3(BJ) dispersion correction. All values are in kcal/mol

Functional	no DFT-D3	with DFT-D3
basic properties and reaction energies for small systems		
SOS0-PBE0-2	1.70 (2.30)	1.77 (2.48)
SOS0-PBE0-DH	2.88 (3.78)	3.65 (4.24)
SOS1-PBE-QIDH	2.00 (2.84)	2.28 (3.09)
SOS0-PBE-CIDH	2.68 (3.61)	3.45 (4.07)
reaction energies for large systems and isomerisation reactions		
SOS0-PBE0-2	2.46 (7.14)	2.34 (6.11)
SOS0-PBE0-DH	4.10 (8.69)	3.88 (6.66)
SOS1-PBE-QIDH	3.15 (7.30)	2.98 (6.01)
SOS0-PBE-CIDH	3.90 (8.12)	3.70 (6.27)
reaction barrier heights		
SOS0-PBE0-2	1.69 (4.99)	1.44 (4.24)
SOS0-PBE0-DH	2.21 (3.88)	2.45 (4.36)
SOS1-PBE-QIDH	1.68 (3.86)	1.60 (3.43)
SOS0-PBE-CIDH	2.01 (3.51)	2.19 (3.77)
intermolecular noncovalent interactions		
SOS0-PBE0-2	6.55 (11.95)	1.55 (3.69)
SOS0-PBE0-DH	8.35 (14.33)	4.31 (9.20)
SOS1-PBE-QIDH	7.12 (12.75)	2.50 (4.29)
SOS0-PBE-CIDH	8.06 (13.98)	4.02 (8.56)
intramolecular noncovalent interactions		
SOS0-PBE0-2	5.29 (9.09)	2.35 (4.15)
SOS0-PBE0-DH	8.54 (14.33)	2.62 (4.61)
SOS1-PBE-QIDH	6.46 (10.87)	2.44 (4.17)
SOS0-PBE-CIDH	8.05 (13.52)	2.58 (4.47)
all noncovalent interactions		
SOS0-PBE0-2	6.01 (10.55)	1.89 (3.91)
SOS0-PBE0-DH	8.43 (14.33)	3.59 (6.95)
SOS1-PBE-QIDH	6.84 (11.83)	2.47 (4.23)
SOS0-PBE-CIDH	8.06 (13.75)	3.40 (6.56)
GMTKN55		
SOS0-PBE0-2	3.47 (6.69)	1.87 (3.86)
SOS0-PBE0-DH	5.11 (8.76)	3.51 (5.72)
SOS1-PBE-QIDH	3.99 (7.25)	2.38 (4.06)
SOS0-PBE-CIDH	4.85 (8.34)	3.31 (5.37)

SI.2.2 WTMADs for four different SOS0-DHDF(b) type of functionals

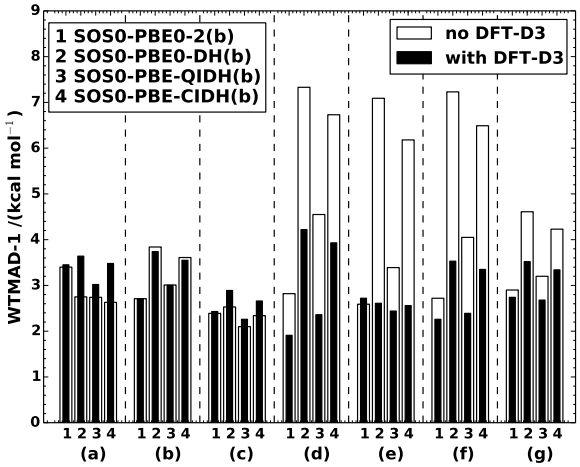

Figure S3: The effect of dispersion corrections on WTMAD-1 values (kcal/mol). The values are displayed for the various categories of GMTKN55, namely basic properties and reactions of small systems (a), isomerisations and reactions of large systems (b), barrier heights (c), intermolecular noncovalent interactions (d), intramolecular noncovalent interactions (e), and all noncovalent interactions (f). Values are also shown for the entire GMTKN55 database (g).

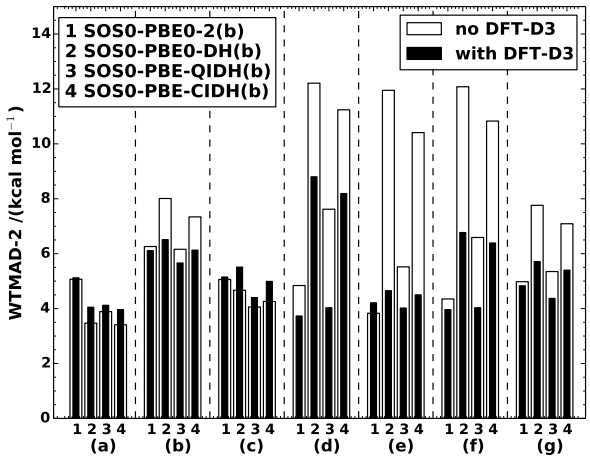

Figure S4: The effect of dispersion corrections on WTMAD-2 values (kcal/mol). The values are displayed for the various categories of GMTKN55, namely basic properties and reactions of small systems (a), isomerisations and reactions of large systems (b), barrier heights (c), intermolecular noncovalent interactions (d), intramolecular noncovalent interactions (e), and all noncovalent interactions (f). Values are also shown for the entire GMTKN55 database (g).

Table S5: WTMAD-1 and WTMAD-2 values (in parenthesis) for four different SOS0-DHDF(b) type of functionals with and without the DFT-D3(BJ) dispersion correction. All values are in kcal/mol.

Functional	no DFT-D3	with DFT-D3
basic properties and reaction energies for small systems		
SOS0-PBE0-2(b)	3.40 (5.07)	3.45 (5.12)
SOS0-PBE0-DH(b)	2.75 (3.47)	3.64 (4.05)
SOS0-PBE-QIDH(b)	2.74 (3.89)	3.02 (4.12)
SOS0-PBE-CIDH(b)	2.63 (3.41)	3.48 (3.96)
reaction energies for large systems and isomerisation reactions		
SOS0-PBE0-2(b)	2.71 (6.26)	2.71 (6.11)
SOS0-PBE0-DH(b)	3.84 (8.01)	3.74 (6.51)
SOS0-PBE-QIDH(b)	3.01 (6.16)	3.00 (5.66)
SOS0-PBE-CIDH(b)	3.61 (7.34)	3.55 (6.13)
reaction barrier heights		
SOS0-PBE0-2(b)	2.39 (5.06)	2.43 (5.15)
SOS0-PBE0-DH(b)	2.53 (4.67)	2.89 (5.51)
SOS0-PBE-QIDH(b)	2.10 (4.06)	2.26 (4.40)
SOS0-PBE-CIDH(b)	2.34 (4.26)	2.66 (4.99)
intermolecular noncovalent interactions		
SOS0-PBE0-2(b)	2.82 (4.84)	1.91 (3.73)
SOS0-PBE0-DH(b)	7.33 (12.21)	4.22 (8.80)
SOS0-PBE-QIDH(b)	4.55 (7.62)	2.36 (4.03)
SOS0-PBE-CIDH(b)	6.73 (11.24)	3.93 (8.19)
intramolecular noncovalent interactions		
SOS0-PBE0-2(b)	2.59 (3.83)	2.72 (4.21)
SOS0-PBE0-DH(b)	7.09 (11.95)	2.61 (4.65)
SOS0-PBE-QIDH(b)	3.39 (5.52)	2.44 (4.02)
SOS0-PBE-CIDH(b)	6.18 (10.41)	2.56 (4.50)
all noncovalent interactions		
SOS0-PBE0-2(b)	2.72 (4.35)	2.26 (3.96)
SOS0-PBE0-DH(b)	7.23 (12.08)	3.53 (6.77)
SOS0-PBE-QIDH(b)	4.05 (6.59)	2.39 (4.03)
SOS0-PBE-CIDH(b)	6.49 (10.83)	3.35 (6.39)
GMTKN55		
SOS0-PBE0-2(b)	2.90 (4.98)	2.74 (4.83)
SOS0-PBE0-DH(b)	4.61 (7.76)	3.52 (5.71)
SOS0-PBE-QIDH(b)	3.20 (5.35)	2.68 (4.37)
SOS0-PBE-CIDH(b)	4.23 (7.09)	3.34 (5.40)

SI.2.3 WTMADs for the B2NC-PLYP and mPW2NC-PLYP functionals

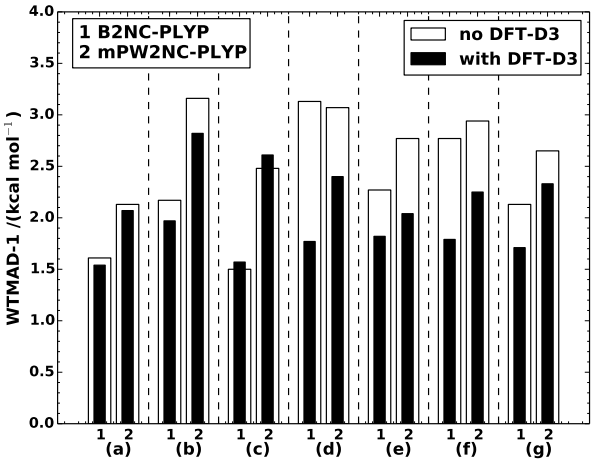

Figure S5: The effect of dispersion corrections on WTMAD-1 values (kcal/mol). The values are displayed for the various categories of GMTKN55, namely basic properties and reactions of small systems (a), isomerisations and reactions of large systems (b), barrier heights (c), intermolecular noncovalent interactions (d), intramolecular noncovalent interactions (e), and all noncovalent interactions (f). Values are also shown for the entire GMTKN55 database (g).

Table S6: WTMAD-1 and WTMAD-2 values (in parenthesis) for the B2NC-PLYP and mPW2NC-PLYP functionals with and without the DFT-D3(BJ) dispersion correction. All values are in kcal/mol.

Functional	no DFT-D3	with DFT-D3
basic properties and reaction energies for small systems		
B2NC-PLYP	1.61 (2.03)	1.54 (1.99)
mPW2NC-PLYP	2.13 (2.67)	2.07 (2.59)
reaction energies for large systems and isomerisation reactions		
B2NC-PLYP	2.17 (5.30)	1.97(4.73)
mPW2NC-PLYP	3.16 (6.09)	2.82 (5.15)
reaction barrier heights		
B2NC-PLYP	1.50 (3.73)	1.57 (3.84)
mPW2NC-PLYP	2.48 (5.33)	2.61 (5.61)
intermolecular noncovalent interactions		
B2NC-PLYP	3.13 (5.62)	1.77 (3.53)
mPW2NC-PLYP	3.07 (5.03)	2.40 (4.74)
intramolecular noncovalent interactions		
B2NC-PLYP	2.27 (3.75)	1.82 (3.17)
mPW2NC-PLYP	2.77 (4.58)	2.04 (3.53)
all noncovalent interactions		
B2NC-PLYP	2.77 (4.70)	1.79 (3.35)
mPW2NC-PLYP	2.94 (4.81)	2.25 (4.15)
GMTKN55		
B2NC-PLYP	2.13 (3.83)	1.71 (3.21)
mPW2NC-PLYP	2.65 (4.41)	2.33 (4.01)

SI.3 Performance of mSD-PBEPBE on the GMTKN55 database

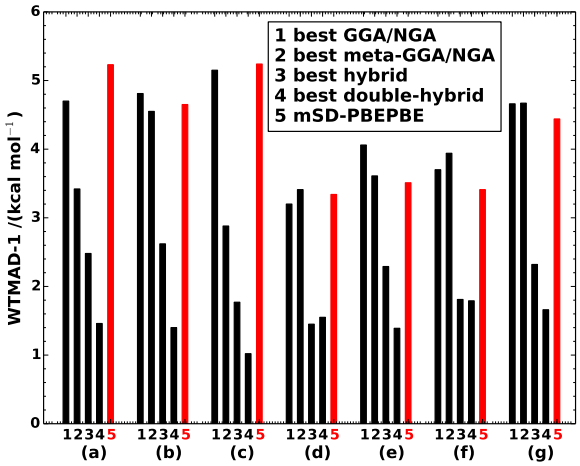

Figure S6: WTMAD-1 values (kcal/mol) of mSD-PBEPBE (5) compared with those of the best DFAs in rungs 2-5 on Jacob's Ladder (see this study and ref. S11). The values are displayed for basic properties and reactions of small systems (a), isomerisations and reactions of large systems (b), barrier heights (c), intermolecular noncovalent interactions (d), intramolecular noncovalent interactions (e), all noncovalent interactions (f), and for the entire GMTKN55 database (g).

SI.4 Performance of B2PPW91-D3(BJ) on the GMTKN55 database

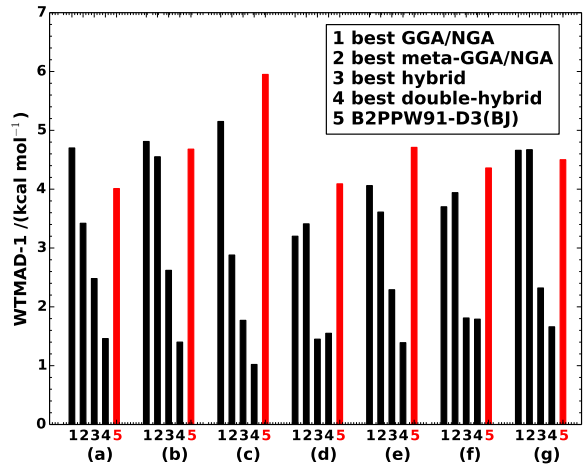

Figure S7: WTMAD-1 values (kcal/mol) of B2PPW91-D3(BJ) (5) compared with those of the best DFAs in rungs 2-5 on Jacob's Ladder (see this study and ref. S11). The values are displayed for basic properties and reactions of small systems (a), isomerisations and reactions of large systems (b), barrier heights (c), intermolecular noncovalent interactions (d), intramolecular noncovalent interactions (e), all noncovalent interactions (f), and for the entire GMTKN55 database (g).

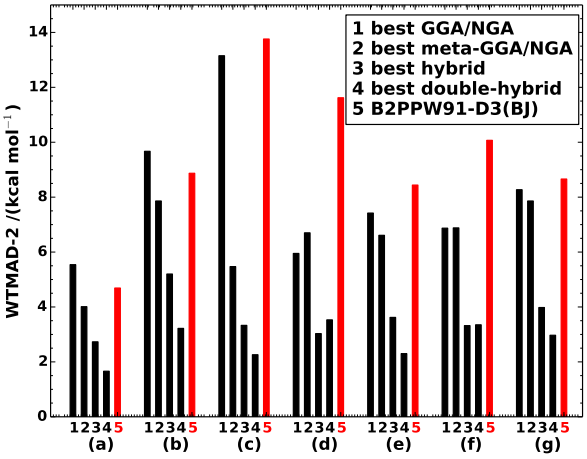

Figure S8: WTMAD-2 values (kcal/mol) of B2PPW91-D3(BJ) (5) compared with those of the best DFAs in rungs 2-5 on Jacob's Ladder (see this study and ref. S11). The values are displayed for basic properties and reactions of small systems (a), isomerisations and reactions of large systems (b), barrier heights (c), intermolecular noncovalent interactions (d), intramolecular noncovalent interactions (e), all noncovalent interactions (f), and for the entire GMTKN55 database (g).

SI.5 Analysis of best and worst mean absolute deviations

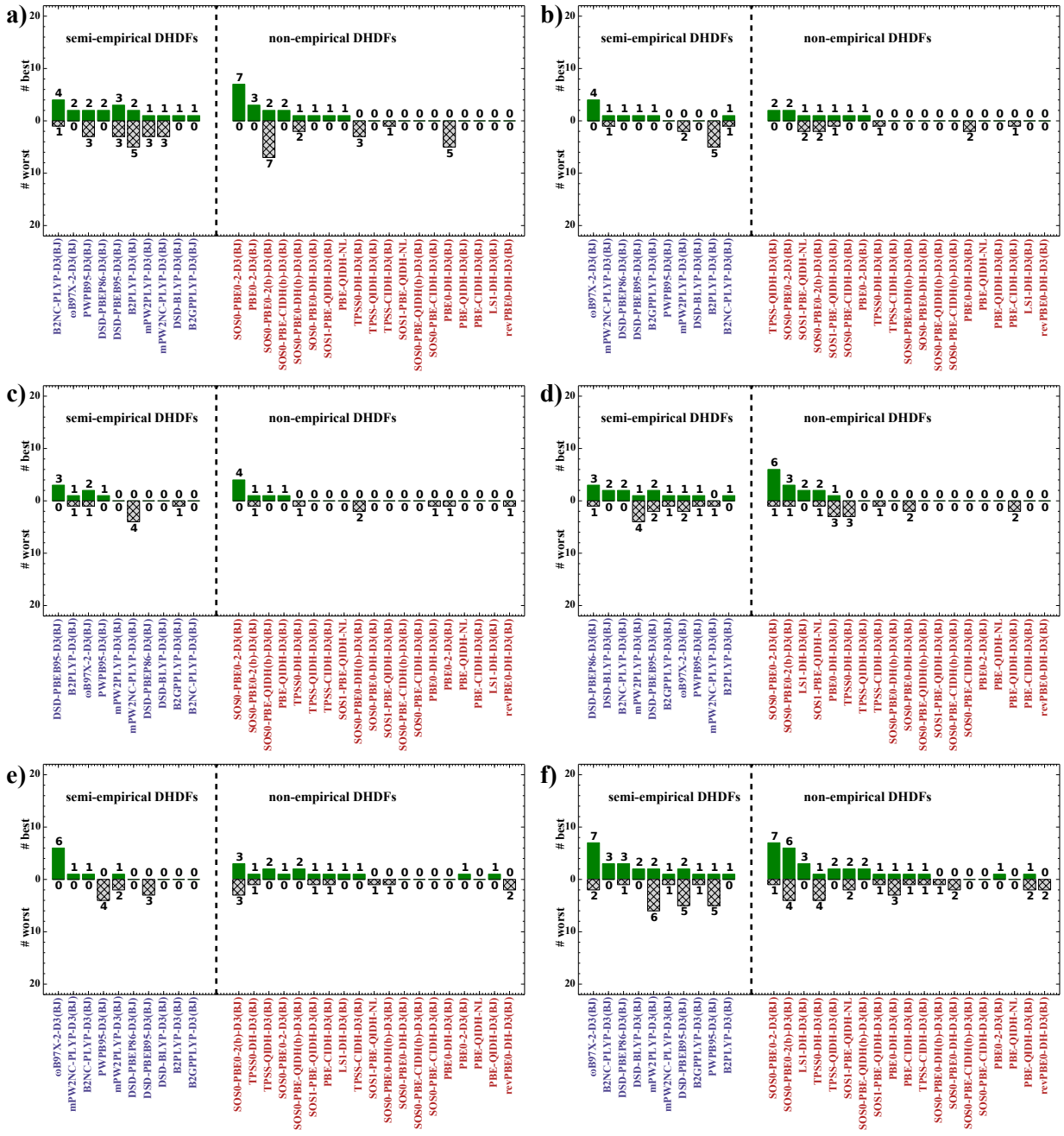

Figure S9: Analysis of how many times a double hybrid yields the worst and best MAD for basic properties and reactions of small systems (a), isomerisations and reactions of large systems (b), barrier heights (c), intermolecular noncovalent interactions (d), intramolecular noncovalent interactions (e), and all noncovalent interactions (f). The analysis was carried out separately for semi-empirical (left section in each image) and non-empirical (right section in each image) DHDFs.

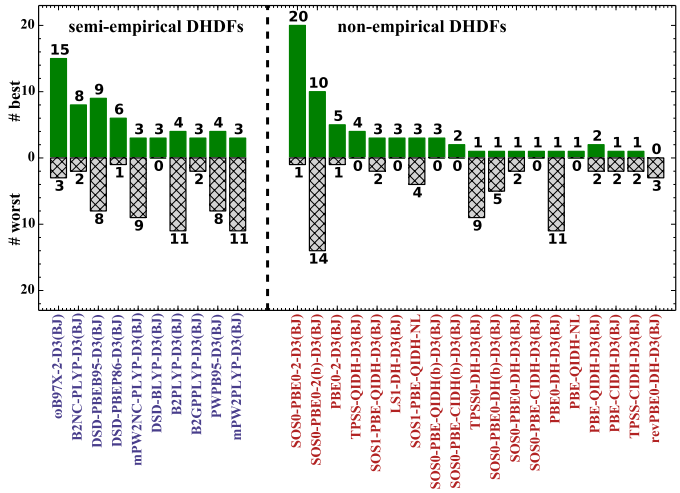

Figure S10: Analysis of how many times a double hybrid yields the worst and best MAD for the entire GMTKN55 database. The analysis was carried out separately for semi-empirical (left section) and non-empirical (right section) DHDFs.

Table S7: The best DHDF for each benchmark set based on mean absolute deviations (kcal/mol). The results are analysed separately for each of the semi-empirical and non-empirical classes of DHDFs before the best of all dispersion-corrected DHDFs is shown. The last column provides an update to the original GMTKN55 study[S11] and reports the best method out of 105 dispersion-corrected DFAs tested on the GMTKN55 database.

	semi-empirical	non-empirical	all DHDFs	all DFAs
W4-11	PWPB95-D3(BJ) (1.86)	PBE0-2-D3(BJ) (3.49)	PWPB95-D3(BJ) (1.86)	PWPB95-D3(BJ) (1.86)
G21EA	B2PLYP-D3(BJ) (1.29)	SOS0-PBE-CIDH(b)-D3(BJ) (1.21)	SOS0-PBE-CIDH(b)-D3(BJ) (1.21)	SOS0-PBE-CIDH(b)-D3(BJ) (1.21)
G21IP	B2NC-PLYP-D3(BJ) (1.82)	SOS0-PBE0-2-D3(BJ) (1.95)	B2NC-PLYP-D3(BJ) (1.82)	B2NC-PLYP-D3(BJ) (1.82)
DIPCS10	mPW2NC-PLYP-D3(BJ) (3.07)	SOS0-PBE0-DH(b)-D3(BJ) (2.52)	SOS0-PBE0-DH(b)-D3(BJ) (2.52)	SOS0-PBE0-DH(b)-D3(BJ) (2.52)
PA26	B2NC-PLYP-D3(BJ) (0.96)	PBE0-2-D3(BJ) (1.48)	B2NC-PLYP-D3(BJ) (0.96)	B2NC-PLYP-D3(BJ) (0.96)
SIE4x4	B2NC-PLYP-D3(BJ) (2.67)	PBE0-2-D3(BJ) (1.81)	PBE0-2-D3(BJ) (1.81)	PBE0-2-D3(BJ) (1.81)
ALKBDE10	ωB97X-2-D3(BJ) (2.52)	SOS0-PBE-CIDH(b)-D3(BJ) (2.81)	ωB97X-2-D3(BJ) (2.52)	ωB97X-2-D3(BJ) (2.52)
YBDE18	B2GPPLYP-D3(BJ) (0.93)	SOS0-PBE0-DH-D3(BJ) (1.87)	B2GPPLYP-D3(BJ) (0.93)	B2GPPLYP-D3(BJ) (0.93)
				M052X-D3(0) (0.93)
AL2X6	DSD-PBEP86-D3(BJ) (0.31)	SOS0-PBE0-2-D3(BJ) (0.42)	DSD-PBEP86-D3(BJ) (0.31)	DSD-PBEP86-D3(BJ) (0.31)
HEAVYSB11	PWPB95-D3(BJ) (0.73)	PBE-QIDH-NL (0.58)	PBE-QIDH-NL (0.58)	MPWB1K-D3(BJ) (0.45)
NBPRC	B2NC-PLYP-D3(BJ) (0.73)	SOS0-PBE0-2-D3(BJ) (1.60)	B2NC-PLYP-D3(BJ) (0.73)	B2NC-PLYP-D3(BJ) (0.73)
	DSD-PBEB95-D3(BJ) (0.73)		DSD-PBEB95-D3(BJ) (0.73)	DSD-PBEB95-D3(BJ) (0.73)
ALK8	B2PLYP-D3(BJ) (1.07)	SOS0-PBE0-2-D3(BJ) (1.64)	B2PLYP-D3(BJ) (1.07)	ωB97X-V (0.95)
RC21	mPW2PLYP-D3(BJ) (1.05)	SOS0-PBE0-2-D3(BJ) (1.96)	mPW2PLYP-D3(BJ) (1.05)	mPW2PLYP-D3(BJ) (1.05)
G2RC	DSD-BLYP-D3(BJ) (1.03)	SOS0-PBE0-2(b)-D3(BJ) (2.49)	DSD-BLYP-D3(BJ) (1.03)	DSD-BLYP-D3(BJ) (1.03)
BH76RC	ωB97X-2-D3(BJ) (0.71)	SOS0-PBE0-2-D3(BJ) (1.45)	ωB97X-2-D3(BJ) (0.71)	ωB97X-2-D3(BJ) (0.71)
FH51	DSD-PBEB95-D3(BJ) (0.69)	SOS0-PBE0-2(b)-D3(BJ) (1.54)	DSD-PBEB95-D3(BJ) (0.69)	DSD-PBEB95-D3(BJ) (0.69)
TAUT15	DSD-PBEB95-D3(BJ) (0.41)	SOS1-PBE-QIDH-D3(BJ) (0.46)	DSD-PBEB95-D3(BJ) (0.41)	DSD-PBEB95-D3(BJ) (0.41)
DC13	DSD-PBEP86-D3(BJ) (2.55)	SOS0-PBE0-2-D3(BJ) (4.95)	DSD-PBEP86-D3(BJ) (2.55)	DSD-PBEP86-D3(BJ) (2.55)
MB16-43	mPW2NC-PLYP-D3(BJ) (4.91)	SOS1-PBE-QIDH-D3(BJ) (10.72)	mPW2NC-PLYP-D3(BJ) (4.91)	mPW2NC-PLYP-D3(BJ) (4.91)
DARC	B2NC-PLYP-D3(BJ) (0.87)	TPSS-QIDH-D3(BJ) (3.43)	B2NC-PLYP-D3(BJ) (0.87)	B2NC-PLYP-D3(BJ) (0.87)
RSE43	B2GPPLYP-D3(BJ) (0.41)	SOS0-PBE-CIDH-D3(BJ) (0.25)	SOS0-PBE-CIDH-D3(BJ) (0.25)	SOS0-PBE-CIDH-D3(BJ) (0.25)
BSR36	ωB97X-2-D3(BJ) (1.18)	PBE0-2-D3(BJ) (0.62)	PBE0-2-D3(BJ) (0.62)	BMK-D3(BJ) (0.39)
CDIE20	ωB97X-2-D3(BJ) (0.27)	SOS0-PBE0-2(b)-D3(BJ) (0.31)	ωB97X-2-D3(BJ) (0.27)	ωB97X-2-D3(BJ) (0.27)
ISO34	DSD-PBEP86-D3(BJ) (0.41)	SOS0-PBE0-2-D3(BJ) (0.83)	DSD-PBEP86-D3(BJ) (0.41)	DSD-PBEP86-D3(BJ) (0.41)
ISOL24	DSD-PBEB95-D3(BJ) (0.96)	TPSS-QIDH-D3(BJ) (1.56)	DSD-PBEB95-D3(BJ) (0.96)	DSD-PBEB95-D3(BJ) (0.96)
C60ISO	ωB97X-2-D3(BJ) (3.44)	SOS1-PBE-QIDH-NL (2.02)	SOS1-PBE-QIDH-NL (2.02)	PW6B95-D3(BJ) (1.65)
PArel	ωB97X-2-D3(BJ) (0.36)	SOS0-PBE0-2-D3(BJ) (0.43)	ωB97X-2-D3(BJ) (0.36)	ωB97X-2-D3(BJ) (0.36)
BH76	DSD-PBEB95-D3(BJ) (1.03)	PBE-QIDH-D3(BJ) (1.22)	DSD-PBEB95-D3(BJ) (1.03)	M08HX-D3(0) (0.99)
BHPERI	PWPB95-D3(BJ) (0.80)	SOS0-PBE0-2-D3(BJ) (1.13)	PWPB95-D3(BJ) (0.80)	PWPB95-D3(BJ) (0.80)
BHDIV10	ωB97X-2-D3(BJ) (0.76)	SOS0-PBE0-2-D3(BJ) (0.90)	ωB97X-2-D3(BJ) (0.76)	ωB97X-2-D3(BJ) (0.76)
INV24	B2PLYP-D3(BJ) (0.69)	SOS0-PBE-QIDH(b)-D3(BJ) (0.78)	B2PLYP-D3(BJ) (0.69)	B2PLYP-D3(BJ) (0.69)
BHROT27	ωB97X-2-D3(BJ) (0.15)	SOS0-PBE0-2(b)-D3(BJ) (0.13)	SOS0-PBE0-2(b)-D3(BJ) (0.13)	SOS0-PBE0-2(b)-D3(BJ) (0.13)
PX13	DSD-PBEB95-D3(BJ) (0.43)	SOS0-PBE0-2-D3(BJ) (0.52)	DSD-PBEB95-D3(BJ) (0.43)	DSD-PBEB95-D3(BJ) (0.43)
WCPT18	DSD-PBEB95-D3(BJ) (0.58)	SOS0-PBE0-2-D3(BJ) (1.04)	DSD-PBEB95-D3(BJ) (0.58)	DSD-PBEB95-D3(BJ) (0.58)
RG18	B2PLYP-D3(BJ) (0.15)	PBE0-DH-D3(BJ) (0.09)	PBE0-DH-D3(BJ) (0.09)	BHLYP-D3(BJ) (0.06)
	DSD-PBEP86-D3(BJ) (0.15)			revTPSSh-D3(BJ) (0.06)
				revTPSS-D3(BJ) (0.06)
ADIM6	DSD-PBEP86-D3(BJ) (0.06)	SOS1-PBE-QIDH-NL (0.07)	DSD-PBEP86-D3(BJ) (0.06)	BHLYP-D3(BJ) (0.05)

	semi-empirical	non-empirical	all DHDFs	all DFAs
S22	B2GPPLYP-D3(BJ) (0.14)	SOS0-PBE0-2-D3(BJ) (0.19)	B2GPPLYP-D3(BJ) (0.14)	B2GPPLYP-D3(BJ) (0.14)
S66	DSD-BLYP-D3(BJ) (0.17)	SOS0-PBE0-2-D3(BJ) (0.13)	SOS0-PBE0-2-D3(BJ) (0.13)	ωB97X-V(0.12)
HEAVY28	mPW2PLYP-D3(BJ) (0.12)	LS1-DH-D3(BJ) (0.13)	mPW2PLYP-D3(BJ) (0.12)	mPW2PLYP-D3(BJ) (0.12)
		SOS0-PBE0-2(b)-D3(BJ) (0.13)		
		SOS1-PBE-QIDH-NL (0.13)		
WATER27	DSD-BLYP-D3(BJ) (0.94)	SOS0-PBE0-2-D3(BJ) (0.89)	SOS0-PBE0-2-D3(BJ) (0.89)	SOS0-PBE0-2-D3(BJ) (0.89)
CARBHB12	DSD-PBEB95-D3(BJ) (0.22)	SOS0-PBE0-2-D3(BJ) (0.30)	DSD-PBEB95-D3(BJ) (0.22)	DSD-PBEB95-D3(BJ) (0.22)
	PWPB95-D3(BJ) (0.22)		PWPB95-D3(BJ) (0.22)	PWPB95-D3(BJ) (0.22)
				M11-D3(BJ) (0.22)
PNICO23	B2NC-PLYP-D3(BJ) (0.13)	SOS0-PBE0-2-D3(BJ) (0.15)	B2NC-PLYP-D3(BJ) (0.13)	B2NC-PLYP-D3(BJ) (0.13)
HAL59	B2NC-PLYP-D3(BJ) (0.23)	SOS0-PBE0-2(b)-D3(BJ) (0.31)	B2NC-PLYP-D3(BJ) (0.23)	B2NC-PLYP-D3(BJ) (0.23)
AHB21	DSD-PBEB95-D3(BJ) (0.20)	SOS0-PBE0-2-D3(BJ) (0.46)	DSD-PBEB95-D3(BJ) (0.20)	DSD-PBEB95-D3(BJ) (0.20)
CHB6	ωB97X-2-D3(BJ) (0.83)	SOS0-PBE0-2(b)-D3(BJ) (0.98)	ωB97X-2-D3(BJ) (0.83)	MN15-D3(BJ) (0.32)
IL16	DSD-PBEP86-D3(BJ) (0.23)	LS1-DH-D3(BJ) (0.26)	DSD-PBEP86-D3(BJ) (0.23)	DSD-PBEP86-D3(BJ) (0.23)
IDISP	B2NC-PLYP-D3(BJ) (0.57)	TPSS0-DH-D3(BJ) (1.45)	B2NC-PLYP-D3(BJ) (0.57)	B2NC-PLYP-D3(BJ) (0.57)
ICONF	ωB97X-2-D3(BJ) (0.13)	SOS0-PBE-QIDH(b)-D3(BJ) (0.13)	SOS0-PBE-QIDH(b)-D3(BJ) (0.13)	SOS0-PBE-QIDH(b)-D3(BJ) (0.13)
			ωB97X-2-D3(BJ) (0.13)	ωB97X-2-D3(BJ) (0.13)
ACONF	ωB97X-2-D3(BJ) (0.02)	PBE-CIDH-D3(BJ) (0.08)	ωB97X-2-D3(BJ) (0.02)	ωB97X-2-D3(BJ) (0.02)
AMINO20x4	ωB97X-2-D3(BJ) (0.10)	SOS0-PBE0-2(b)-D3(BJ) (0.14)	ωB97X-2-D3(BJ) (0.10)	ωB97X-2-D3(BJ) (0.10)
PCONF21	ωB97X-2-D3(BJ) (0.19)	SOS0-PBE0-2-D3(BJ) (0.22)	ωB97X-2-D3(BJ) (0.19)	ωB97X-2-D3(BJ) (0.19)
MCONF	mPW2PLYP-D3(BJ) (0.10)	SOS1-PBE-QIDH-D3(BJ) (0.11)	mPW2PLYP-D3(BJ) (0.10)	mPW2PLYP-D3(BJ) (0.10)
SCONF	ωB97X-2-D3(BJ) (0.05)	TPSS-QIDH-D3(BJ) (0.06)	ωB97X-2-D3(BJ) (0.05)	ωB97X-2-D3(BJ) (0.05)
UPU23	mPW2NC-PLYP-D3(BJ) (0.37)	SOS0-PBE0-2(b)-D3(BJ) (0.35)	SOS0-PBE0-2(b)-D3(BJ) (0.35)	revTPSS-D3(BJ) (0.33)
BUT14DIOL	ωB97X-2-D3(BJ) (0.03)	LS1-DH-D3(BJ) (0.06)	ωB97X-2-D3(BJ) (0.03)	ωB97X-2-D3(BJ) (0.03)
		PBE-QIDH-D3(BJ) (0.06)		
		PBE0-2-D3(BJ) (0.06)		
		SOS0-PBE-QIDH(b)-D3(BJ) (0.06)		
		SOS0-PBE0-2(b)-D3(BJ) (0.06)		
		TPSS-CIDH-D3(BJ) (0.06)		
		TPSS-QIDH-D3(BJ) (0.06)		

Table S8: The worst DHDFs for each benchmark set based on mean absolute deviations (kcal/mol). The results are analysed separately for each of the semi-empirical and non-empirical class of DHDFs before the worst of all dispersion-corrected DHDFs is shown.

	semi-empirical	non-empirical	all DHDFs
W4-11	mPW2NC-PLYP-D3(BJ) (6.03)	SOS0-PBE0-2(b)-D3(BJ) (28.52)	SOS0-PBE0-2(b)-D3(BJ) (28.52)
G21EA	DSD-PBEB95-D3(BJ) (2.89)	SOS0-PBE0-2(b)-D3(BJ) (5.41)	SOS0-PBE0-2(b)-D3(BJ) (5.41)
G21IP	DSD-PBEB95-D3(BJ) (2.58)	SOS0-PBE0-2(b)-D3(BJ) (5.72)	SOS0-PBE0-2(b)-D3(BJ) (5.72)
DIPCS10	DSD-PBEB95-D3(BJ) (6.03)	SOS0-PBE0-2(b)-D3(BJ) (6.79)	SOS0-PBE0-2(b)-D3(BJ) (6.79)
PA26	PWPB95-D3(BJ) (1.90)	TPSS0-DH-D3(BJ) (4.09)	TPSS0-DH-D3(BJ) (4.09)
SIE4x4	B2PLYP-D3(BJ) (10.08)	SOS0-PBE0-DH(b)-D3(BJ) (8.43)	B2PLYP-D3(BJ) (10.08)
ALKBDE10	mPW2NC-PLYP-D3(BJ) (5.19)	SOS0-PBE0-2(b)-D3(BJ) (9.86)	SOS0-PBE0-2(b)-D3(BJ) (9.86)
YBDE18	B2PLYP-D3(BJ) (2.12)	SOS0-PBE0-2(b)-D3(BJ) (9.32)	SOS0-PBE0-2(b)-D3(BJ) (9.32)
AL2X6	mPW2PLYP-D3(BJ) (2.66)	TPSS0-DH-D3(BJ) (4.61)	TPSS0-DH-D3(BJ) (4.61)
HEAVYSB11	mPW2PLYP-D3(BJ) (2.44)	SOS0-PBE0-2(b)-D3(BJ) (6.80)	SOS0-PBE0-2(b)-D3(BJ) (6.80)
NBPRC	mPW2PLYP-D3(BJ) (1.71)	PBE0-DH-D3(BJ) (3.51)	PBE0-DH-D3(BJ) (3.51)
ALK8	PWPB95-D3(BJ) (4.23)	TPSS-CIDH-D3(BJ) (10.13)	TPSS-CIDH-D3(BJ) (10.13)
RC21	B2NC-PLYP-D3(BJ) (2.83)	PBE0-DH-D3(BJ) (4.79)	PBE0-DH-D3(BJ) (4.79)
BH76RC	mPW2NC-PLYP-D3(BJ) (1.50)	TPSS0-DH-D3(BJ) (3.59)	TPSS0-DH-D3(BJ) (3.59)
G2RC	PWPB95-D3(BJ) (2.17)	PBE0-DH-D3(BJ) (6.63)	PBE0-DH-D3(BJ) (6.63)
FH51	B2PLYP-D3(BJ) (1.50)	PBE0-DH-D3(BJ) (3.12)	PBE0-DH-D3(BJ) (3.12)
TAUT15	B2PLYP-D3(BJ) (0.72)	SOS0-PBE0-DH(b)-D3(BJ) (0.86)	SOS0-PBE0-DH(b)-D3(BJ) (0.86)
DC13	B2PLYP-D3(BJ) (6.77)	PBE0-DH-D3(BJ) (9.58)	PBE0-DH-D3(BJ) (9.58)
MB16-43	mPW2PLYP-D3(BJ) (22.08)	PBE-CIDH-D3(BJ) (21.01)	mPW2PLYP-D3(BJ) (22.08)
DARC	B2PLYP-D3(BJ) (5.03)	SOS1-PBE-QIDH-NL (8.29)	SOS1-PBE-QIDH-NL (8.29)
RSE43	B2NC-PLYP-D3(BJ) (1.15)	SOS0-PBE0-2(b)-D3(BJ) (1.58)	SOS0-PBE0-2(b)-D3(BJ) (1.58)
BSR36	mPW2PLYP-D3(BJ) (3.76)	SOS1-PBE-QIDH-D3(BJ) (3.30)	mPW2PLYP-D3(BJ) (3.76)
CDIE20	B2PLYP-D3(BJ) (0.75)	TPSS0-DH-D3(BJ) (0.95)	TPSS0-DH-D3(BJ) (0.95)
ISO34	B2PLYP-D3(BJ) (1.13)	PBE0-DH-D3(BJ) (1.44)	PBE0-DH-D3(BJ) (1.44)
ISOL24	B2PLYP-D3(BJ) (3.83)	SOS1-PBE-QIDH-NL (3.28)	B2PLYP-D3(BJ) (3.83)
C60ISO	mPW2NC-PLYP-D3(BJ) (13.28)	SOS0-PBE0-2(b)-D3(BJ) (13.13)	mPW2NC-PLYP-D3(BJ) (13.28)
PArel	B2PLYP-D3(BJ) (0.74)	PBE0-DH-D3(BJ) (1.21)	PBE0-DH-D3(BJ) (1.21)
BH76	B2PLYP-D3(BJ) (2.59)	TPSS0-DH-D3(BJ) (2.60)	TPSS0-DH-D3(BJ) (2.60)
BHPERI	mPW2NC-PLYP-D3(BJ) (3.89)	PBE0-2-D3(BJ) (3.78)	mPW2NC-PLYP-D3(BJ) (3.89)
BHDIV10	mPW2NC-PLYP-D3(BJ) (2.68)	SOS0-PBE0-DH(b)-D3(BJ) (2.99)	SOS0-PBE0-DH(b)-D3(BJ) (2.99)
INV24	ωB97X-2-D3(BJ) (1.02)	revPBE0-DH-D3(BJ) (1.64)	revPBE0-DH-D3(BJ) (1.64)
BHROT27	B2GPPLYP-D3(BJ) (0.33)	PBE0-DH-D3(BJ) (0.59)	PBE0-DH-D3(BJ) (0.59)
PX13	mPW2NC-PLYP-D3(BJ) (3.66)	SOS0-PBE0-DH(b)-D3(BJ) (4.38)	SOS0-PBE0-DH(b)-D3(BJ) (4.38)
WCPT18	mPW2NC-PLYP-D3(BJ) (2.96)	SOS0-PBE0-2(b)-D3(BJ) (2.75)	mPW2NC-PLYP-D3(BJ) (2.96)
RG18	mPW2PLYP-D3(BJ) (0.27)	SOS0-PBE0-2(b)-D3(BJ) (0.19)	mPW2PLYP-D3(BJ) (0.27)
ADIM6	DSD-PBEB95-D3(BJ) (0.49)	PBE-QIDH-D3(BJ) (0.73)	PBE-QIDH-D3(BJ) (0.73)
S22	DSD-PBEB95-D3(BJ) (0.51)	SOS1-PBE-QIDH-NL (0.46)	DSD-PBEB95-D3(BJ) (0.51)
S66	ωB97X-2-D3(BJ) (0.33)	PBE-QIDH-D3(BJ) (0.36)	PBE-QIDH-D3(BJ) (0.36)
HEAVY28	ωB97X-2-D3(BJ) (0.31)	TPSS0-DH-D3(BJ) (0.89)	TPSS0-DH-D3(BJ) (0.89)
		TPSS-CIDH-D3(BJ) (0.89)	TPSS-CIDH-D3(BJ) (0.89)
		SOS0-PBE0-DH-D3(BJ) (0.89)	SOS0-PBE0-DH-D3(BJ) (0.89)
WATER27	mPW2PLYP-D3(BJ) (4.99)	PBE0-DH-D3(BJ) (4.11)	mPW2PLYP-D3(BJ) (4.99)
CARBHB12	mPW2PLYP-D3(BJ) (0.75)	PBE0-DH-D3(BJ) (1.18)	PBE0-DH-D3(BJ) (1.18)
PNICO23	DSD-PBEP86-D3(BJ) (0.40)	TPSS0-DH-D3(BJ) (1.05)	TPSS0-DH-D3(BJ) (1.05)
HAL59	mPW2NC-PLYP-D3(BJ) (0.57)	TPSS0-DH-D3(BJ) (0.82)	TPSS0-DH-D3(BJ) (0.82)
AHB21	mPW2PLYP-D3(BJ) (0.63)	PBE0-DH-D3(BJ) (1.38)	PBE0-DH-D3(BJ) (1.38)
CHB6	B2GPPLYP-D3(BJ) (1.81)	SOS0-PBE0-DH-D3(BJ) (2.78)	SOS0-PBE0-DH-D3(BJ) (2.78)
IL16	PWPB95-D3(BJ) (1.25)	SOS0-PBE0-2-D3(BJ) (0.55)	PWPB95-D3(BJ) (1.25)
IDISP	mPW2PLYP-D3(BJ) (2.78)	SOS0-PBE0-2(b)-D3(BJ) (3.49)	SOS0-PBE0-2(b)-D3(BJ) (3.49)
ICONF	mPW2PLYP-D3(BJ) (0.25)	TPSS0-DH-D3(BJ) (0.32)	TPSS0-DH-D3(BJ) (0.32)
ACONF	DSD-PBEB95-D3(BJ) (0.37)	SOS0-PBE0-2(b)-D3(BJ) (0.32)	DSD-PBEB95-D3(BJ) (0.37)
AMINO20x4	PWPB95-D3(BJ) (0.23)	revPBE0-DH-D3(BJ) (0.26)	revPBE0-DH-D3(BJ) (0.26)
PCONF21	DSD-PBEB95-D3(BJ) (0.68)	SOS0-PBE0-DH(b)-D3(BJ) (0.61)	DSD-PBEB95-D3(BJ) (0.68)
		PBE-CIDH-D3(BJ) (0.61)	
MCONF	DSD-PBEB95-D3(BJ) (0.57)	SOS1-PBE-QIDH-NL (0.55)	DSD-PBEB95-D3(BJ) (0.57)
SCONF	PWPB95-D3(BJ) (0.18)	SOS0-PBE0-2(b)-D3(BJ) (0.30)	SOS0-PBE0-2(b)-D3(BJ) (0.30)
UPU23	PWPB95-D3(BJ) (0.53)	SOS1-PBE-QIDH-D3(BJ) (0.61)	SOS1-PBE-QIDH-D3(BJ) (0.61)
BUT14DIOL	PWPB95-D3(BJ) (0.36)	revPBE0-DH-D3(BJ) (0.20)	PWPB95-D3(BJ) (0.36)

Table S9: Ratios of the mean absolute deviations and averaged mean absolute deviations to compare the performance of semi-empirical with that of non-empirical DHDFs for each benchmark set; see Section 2.4 in the main text for details.

	$\frac{\mathrm{MAD}^{\mathrm{best}}_{\mathrm{semi\text{-}emp.}}}{\mathrm{MAD}^{\mathrm{best}}_{\mathrm{non\text{-}emp.}}}$	$\frac{\lvert\mathrm{MAD}_{\mathrm{semi\text{-}emp.}}\rvert}{\lvert\mathrm{MAD}_{\mathrm{non\text{-}emp.}}\rvert}$
W4-11	0.53	0.37
G21EA	1.07	0.62
G21IP	0.93	0.74
DIPCS10	1.22	1.20
PA26	0.65	0.45
SIE4x4	1.48	1.31
ALKBDE10	0.90	0.57
YBDE18	0.50	0.49
AL2X6	0.74	0.54
HEAVYSB11	1.26	0.65
NBPRC	0.46	0.36
ALK8	0.65	0.41
RC21	0.54	0.51
BH76RC	0.49	0.50
G2RC	0.41	0.32
FH51	0.45	0.45
TAUT15	0.89	0.78
DC13	0.52	0.53
MB16-43	0.46	0.69
DARC	0.25	0.39
RSE43	1.64	1.07
BSR36	1.90	0.82
CDIE20	0.87	0.81
ISO34	0.49	0.61
ISOL24	0.62	0.94
C60ISO	1.70	1.60
PArel	0.84	0.62
BH76	0.84	0.94
BHPERI	0.71	0.81
BHDIV10	0.84	0.77
INV24	0.88	0.59
BHROT27	1.15	0.58
PX13	0.83	0.73
WCPT18	0.56	0.74
RG18	1.67	1.28
ADIM6	0.86	0.75
S22	0.74	0.90
S66	1.31	0.88
HEAVY28	0.92	0.42
WATER27	1.06	1.19
CARBHB12	0.73	0.60
PNICO23	0.87	0.37
HAL59	0.74	0.73
AHB21	0.43	0.39
CHB6	0.85	0.75
IL16	0.88	1.64
IDISP	0.39	0.74
ICONF	1.00	0.77
ACONF	0.25	1.16
AMINO20x4	0.71	0.74
PCONF21	0.86	0.74
MCONF	0.91	0.98
SCONF	0.83	0.65
UPU23	1.06	0.89
BUT14DIOL	0.50	1.49

SI.6 Weighted total mean absolute deviations

In the original study of GMTKN55,[S11] authors defined a weighted total mean absolute deviation (WTMAD) to combine all obtained mean absolute deviations (MADs) for each subset into one final number for a tested method. In the WTMAD-1 scheme (see Eq. (1)), each of the 55 MAD values was weighted by factor w that aims at aligning benchmark sets with largely differing averaged absolute relative energy ($\overline{|\Delta E|}$). The MAD of a test set was scaled up by a factor of w=10 if the set's $\overline{|\Delta E|}$ < 7.5 kcal/mol. The MAD of a test set was scaled down by a factor of w=10 if the set's $\overline{|\Delta E|}$ > 7.5 kcal/mol. w was set to unity for the remaining benchmark sets. WTMAD-1 was then calculated as an average value over the 55 sets:

$$\text{WTMAD-1} = \frac{\sum_i^{55} w_i \cdot \text{MAD}_i}{55}. \tag{1}$$

In the WTMAD-2 scheme (Eq. (2)) the weight factor w was defined as the ratio between average of all 55 $\overline{|\Delta E|}$ values (56.84 kcal/mol) and the $\overline{|\Delta E|}$ for the respective benchmark set. This value was then scaled by the number of relative energies N in that particular set before it was combined with the MAD. The WTMAD-2 was calculated as the sum over all 55 weighted MADs and divided by the total number of relative energies in GMTKN55 (1505 data points):

$$\text{WTMAD-2} = \frac{1}{\sum_i^{55} N_i} \cdot \sum_i^{55} N_i \cdot \frac{56.84\text{kcal/mol}}{\overline{|\Delta E|}_i} \cdot MAD_i. \tag{2}$$

SI.6.1 WTMAD-1 diagram for all assessed methods

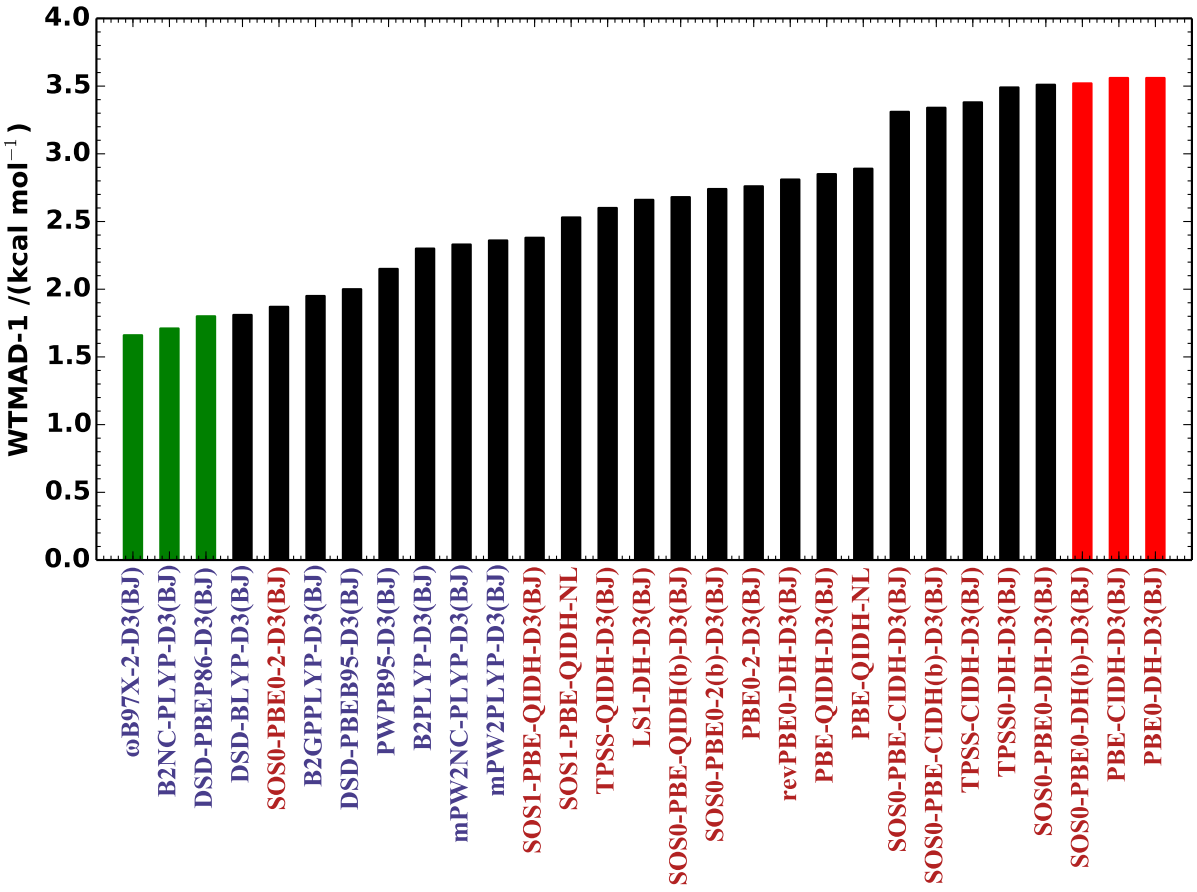

Figure S11: Final WTMAD-1 values over the entire GMTKN55database for all assessed dispersion-corrected DHDFs (kcal/mol). The green bars indicate the three best approaches and red bars indicate the three worst approaches.

SI.6.2 Averaged WTMADs for each of the semi- and non-empirical double-hybrid density functional theory approximations

Table S10: Averaged WTMAD-1 and WTMAD-2 values for each of the semi- and non-empirical classes of double-hybrid density functionals (kcal/mol).

	WTMAD-1	WTMAD-2
basic properties and reactions of small systems		
semi-empirical	1.82	2.09
non-empirical	3.03	3.86
isomerisations and reactions of large systems		
semi-empirical	2.37	4.77
non-empirical	3.45	6.18
barrier heights		
semi-empirical	1.68	3.73
non-empirical	2.34	4.56
intermolecular noncovalent interactions		
semi-empirical	2.08	4.24
non-empirical	3.20	6.19
intramolecular noncovalent interactions		
semi-empirical	2.20	3.90
non-empirical	2.56	4.45
all noncovalent interactions		
semi-empirical	2.13	4.07
non-empirical	2.93	5.34
GMTKN55		
semi-empirical	2.01	3.52
non-empirical	2.97	4.91

SI.6.3 The worst double-hybrid density functional theory approximations according to WTMADs

Table S11: The three worst dispersion-corrected DHDFs for basic properties and reactions of small systems according to WTMAD-1 and WTMAD-2 values (kcal/mol).

double hybrids	WTMAD-1	WTMAD-2
semi-empirical	mPW2PLYP-D3(BJ) (2.24)	mPW2NC-PLYP-D3(BJ) (2.59)
	B2PLYP-D3(BJ) (2.24)	B2PLYP-D3(BJ) (2.51)
	PWPB95-D3(BJ) (2.08)	mPW2PLYP-D3(BJ) (2.46)
non-empirical	TPSS0-DH-D3(BJ) (3.69)	SOS0-PBE0-2(b)-D3(BJ) (5.12)
	SOS0-PBE0-DH-D3(BJ) (3.65)	TPSS0-DH-D3(BJ) (4.48)
	SOS0-PBE0-DH(b)-D3(BJ) (3.64)	PBE0-DH-D3(BJ) (4.41)
all DHDFs	TPSS0-DH-D3(BJ) (3.69)	SOS0-PBE0-2(b)-D3(BJ) (5.12)
	SOS0-PBE0-DH-D3(BJ) (3.65)	TPSS0-DH-D3(BJ) (4.48)
	SOS0-PBE0-DH(b)-D3(BJ) (3.64)	PBE0-DH-D3(BJ) (4.41)

Table S12: The three worst dispersion-corrected DHDFs for isomerisation and reactions of large systems according to WTMAD-1 and WTMAD-2 values (kcal/mol).

double hybrids	WTMAD-1	WTMAD-2
semi-empirical	mPW2PLYP-D3(BJ) (3.36)	mPW2PLYP-D3(BJ) (6.76)
	B2PLYP-D3(BJ) (3.36)	B2PLYP-D3(BJ) (6.28)
	mPW2NC-PLYP-D3(BJ) (2.82)	PWPB95-D3(BJ) (5.41)
non-empirical	PBE0-DH-D3(BJ) (4.26)	PBE0-DH-D3(BJ) (6.98)
	PBE-CIDH-D3(BJ) (4.16)	PBE-CIDH-D3(BJ) (6.79)
	SOS0-PBE0-DH-D3(BJ) (3.88)	SOS0-PBE0-DH-D3(BJ) (6.66)
all DHDFs	PBE0-DH-D3(BJ) (4.26)	PBE0-DH-D3(BJ) (6.98)
	PBE-CIDH-D3(BJ) (4.16)	PBE-CIDH-D3(BJ) (6.79)
	SOS0-PBE0-DH-D3(BJ) (3.88)	mPW2PLYP-D3(BJ) (6.76)

Table S13: The three worst dispersion-corrected DHDFs for barrier heights according to WTMAD-1 and WTMAD-2 values (kcal/mol).

double hybrids	WTMAD-1	WTMAD-2
semi-empirical	mPW2NC-PLYP-D3(BJ) (2.61)	mPW2NC-PLYP-D3(BJ) (5.61)
	B2PLYP-D3(BJ) (2.02)	B2PLYP-D3(BJ) (4.90)
	DSD-PBEP86-D3(BJ) (1.80)	mPW2PLYP-D3(BJ) (4.22)
non-empirical	PBE0-DH-D3(BJ) (3.11)	PBE0-DH-D3(BJ) (5.56)
	SOS0-PBE0-DH(b)-D3(BJ) (2.89)	SOS0-PBE0-DH(b)-D3(BJ) (5.51)
	PBE-CIDH-D3(BJ) (2.79)	TPSS0-DH-D3(BJ) (5.33)
all DHDFs	PBE0-DH-D3(BJ) (3.11)	mPW2NC-PLYP-D3(BJ) (5.61)
	SOS0-PBE0-DH(b)-D3(BJ) (2.89)	PBE0-DH-D3(BJ) (5.56)
	PBE-CIDH-D3(BJ) (2.79)	SOS0-PBE0-DH(b)-D3(BJ) (5.51)

Table S14: The three worst dispersion-corrected DHDFs for intermolecular noncovalent interactions according to WTMAD-1 and WTMAD-2 values (kcal/mol).

double hybrids	WTMAD-1	WTMAD-2
semi-empirical	ωB97X-2-D3(BJ) (2.44)	ωB97X-2-D3(BJ) (5.26)
	mPW2NC-PLYP-D3(BJ) (2.40)	mPW2PLYP-D3(BJ) (4.75)
	mPW2PLYP-D3(BJ) (2.23)	mPW2NC-PLYP-D3(BJ) (4.74)
non-empirical	TPSS0-DH-D3(BJ) (4.43)	TPSS0-DH-D3(BJ) (9.22)
	TPSS-CIDH-D3(BJ) (4.37)	SOS0-PBE0-DH-D3(BJ) (9.20)
	SOS0-PBE0-DH-D3(BJ) (4.31)	TPSS-CIDH-D3(BJ) (9.17)
all DHDFs	TPSS0-DH-D3(BJ) (4.43)	TPSS0-DH-D3(BJ) (9.22)
	TPSS-CIDH-D3(BJ) (4.37)	SOS0-PBE0-DH-D3(BJ) (9.20)
	SOS0-PBE0-DH-D3(BJ) (4.31)	TPSS-CIDH-D3(BJ) (9.17)

Table S15: The three worst dispersion-corrected DHDFs for intramolecular noncovalent interactions according to WTMAD-1 and WTMAD-2 values (kcal/mol).

double hybrids	WTMAD-1	WTMAD-2
semi-empirical	DSD-PBEB95-D3(BJ) (3.47)	DSD-PBEB95-D3(BJ) (6.70)
	PWPB95-D3(BJ) (2.95)	PWPB95-D3(BJ) (5.98)
	mPW2PLYP-D3(BJ) (2.23)	B2PLYP-D3(BJ) (3.78)
non-empirical	SOS1-PBE-QIDH-NL (2.81)	revPBE0-DH-D3(BJ) (5.14)
	PBE0-DH-D3(BJ) (2.76)	PBE0-DH-D3(BJ) (5.02)
	SOS0-PBE0-2(b)-D3(BJ) (2.72)	PBE-CIDH-D3(BJ) (4.81)
all DHDFs	DSD-PBEB95-D3(BJ) (3.47)	DSD-PBEB95-D3(BJ) (6.70)
	PWPB95-D3(BJ) (2.95)	PWPB95-D3(BJ) (5.98)
	SOS1-PBE-QIDH-NL (2.81)	revPBE0-DH-D3(BJ) (5.14)

Table S16: The three worst dispersion-corrected DHDFs for all noncovalent interactions according to WTMAD-1 and WTMAD-2 values (kcal/mol).

double hybrids	WTMAD-1	WTMAD-2
semi-empirical	DSD-PBEB95-D3(BJ) (2.67)	DSD-PBEB95-D3(BJ) (5.27)
	PWPB95-D3(BJ) (2.27)	PWPB95-D3(BJ) (4.98)
	mPW2NC-PLYP-D3(BJ) (2.25)	mPW2PLYP-D3(BJ) (4.24)
non-empirical	TPSS0-DH-D3(BJ) (3.67)	TPSS0-DH-D3(BJ) (6.98)
	TPSS-CIDH-D3(BJ) (3.60)	SOS0-PBE0-DH-D3(BJ) (6.95)
	SOS0-PBE0-DH-D3(BJ) (3.59)	PBE-CIDH-D3(BJ) (6.92)
all DHDFs	TPSS0-DH-D3(BJ) (3.67)	TPSS0-DH-D3(BJ) (6.98)
	TPSS-CIDH-D3(BJ) (3.60)	SOS0-PBE0-DH-D3(BJ) (6.95)
	SOS0-PBE0-DH-D3(BJ) (3.59)	PBE-CIDH-D3(BJ) (6.92)

Table S17: The three worst dispersion-corrected DHDFs for GMTKN55 according to WTMAD-1 and WTMAD-2 values (kcal/mol).

double-hybrids	WTMAD-1	WTMAD-2
semi-empirical	mPW2PLYP-D3(BJ) (2.36)	mPW2PLYP-D3(BJ) (4.08)
	mPW2NC-PLYP-D3(BJ) (2.33)	mPW2NC-PLYP-D3(BJ) (4.01)
	B2PLYP-D3(BJ) (2.30)	PWPB95-D3(BJ) (3.98)
non-empirical	PBE0-DH-D3(BJ) (3.56)	TPSS0-DH-D3(BJ) (5.86)
	PBE-CIDH-D3(BJ) (3.56)	PBE-CIDH-D3(BJ) (5.77)
	SOS0-PBE0-DH(b)-D3(BJ) (3.52)	SOS0-PBE0-DH-D3(BJ) (5.72)
all DHDFs	PBE0-DH-D3(BJ) (3.56)	TPSS0-DH-D3(BJ) (5.86)
	PBE-CIDH-D3(BJ) (3.56)	PBE-CIDH-D3(BJ) (5.77)
	SOS0-PBE0-DH(b)-D3(BJ) (3.52)	SOS0-PBE0-DH-D3(BJ) (5.72)

SI.6.4 WTMADs for dispersion-corrected double-hybrid density functional theory approximations

Table S18: WTMAD-1 values (kcal/mol) for dispersion-corrected DHDFs for GMTKN55 and its categories (basic properties and reactions of small systems, isomerisations and reactions of large systems, barrier heights, intermolecular noncovalent interactions (NCIs), intramolecular NCIs, and all NCIs). This table only shows WTMADs of newly tested functionals. For values of the seven previously tested DHDFs, see the ESI of Ref. S11.

	basic + small	iso. + large	barriers	intermol. NCIs	intramol. NCIs	all NCIs	GMTKN55
ωB97X-2-D3(BJ)	1.47	1.40	1.49	2.44	1.39	1.99	1.66
B2NC-PLYP-D3(BJ)	1.54	1.97	1.57	1.77	1.82	1.79	1.71
mPW2NC-PLYP-D3(BJ)	2.07	2.82	2.61	2.40	2.04	2.25	2.33
B2PPW91-D3(BJ)	4.01	4.68	5.95	4.09	4.71	4.36	4.50
LS1-DH-D3(BJ)	2.45	3.56	2.45	2.63	2.39	2.53	2.66
PBE0-DH-D3(BJ)	3.60	4.26	3.11	3.84	2.76	3.37	3.56
PBE0-2-D3(BJ)	2.50	3.41	2.54	2.90	2.62	2.78	2.76
PBE-QIDH-D3(BJ)	2.66	3.78	2.43	3.02	2.43	2.77	2.85
PBE-CIDH-D3(BJ)	3.53	4.16	2.79	4.30	2.64	3.59	3.56
SOS0-PBE0-2(b)-D3(BJ)	3.45	2.71	2.43	1.91	2.72	2.26	2.74
SOS0-PBE0-DH(b)-D3(BJ)	3.64	3.74	2.89	4.22	2.61	3.53	3.52
SOS0-PBE-CIDH(b)-D3(BJ)	3.48	3.55	2.66	3.93	2.56	3.35	3.34
SOS0-PBE-QIDH(b)-D3(BJ)	3.02	3.00	2.26	2.36	2.44	2.39	2.68
SOS0-PBE0-2-D3(BJ)	1.77	2.34	1.44	1.55	2.35	1.89	1.87
SOS0-PBE0-DH-D3(BJ)	3.65	3.88	2.45	4.31	2.62	3.59	3.51
SOS0-PBE-CIDH-D3(BJ)	3.45	3.70	2.19	4.02	2.58	3.40	3.31
SOS1-PBE-QIDH-D3(BJ)	2.28	2.98	1.60	2.50	2.44	2.47	2.38
TPSS0-DH-D3(BJ)	3.69	3.52	2.37	4.43	2.65	3.67	3.49
TPSS-CIDH-D3(BJ)	3.56	3.40	2.23	4.37	2.56	3.60	3.38
TPSS-QIDH-D3(BJ)	2.52	2.92	1.91	3.09	2.31	2.75	2.60
PBE-QIDH-NL	2.70	3.88	2.57	2.91	2.46	2.72	2.89
SOS1-PBE-QIDH-NL	2.47	3.29	1.81	2.27	2.81	2.50	2.53
revPBE0-DH-D3(BJ)	3.06	3.56	2.33	2.26	2.66	2.43	2.81
MP2-D3(BJ)	2.99	3.33	3.22	2.67	3.42	2.99	3.08
SCS-MP2-D3(BJ)	3.25	2.98	2.30	2.13	3.48	2.71	2.88
SOS-MP2-D3(BJ)	4.27	3.58	2.09	2.35	3.75	2.95	3.37

Table S19: WTMAD-2 values (kcal/mol) for dispersion-corrected DHDFs for GMTKN55 and its categories (basic properties and reactions of small systems, isomerisations and reactions of large systems, barrier heights, intermolecular noncovalent interactions (NCIs), intramolecular NCIs, and all NCIs). This table only shows WTMADs of newly tested functionals. For values of the seven previously tested DHDFs, see the ESI of Ref. S11.

	basic + small	iso. + large	barriers	intermol. NCIs	intramol. NCIs	all NCIs	GMTKN55
ωB97X-2-D3(BJ)	1.66	3.22	3.25	5.26	2.30	3.81	2.97
B2NC-PLYP-D3(BJ)	1.99	4.73	3.84	3.53	3.17	3.35	3.21
mPW2NC-PLYP-D3(BJ)	2.59	5.15	5.61	4.74	3.53	4.15	4.01
B2PPW91-D3(BJ)	4.69	8.87	13.76	11.62	8.44	10.07	8.66
LS1-DH-D3(BJ)	3.27	6.13	4.57	4.41	4.14	4.28	4.30
PBE0-DH-D3(BJ)	4.41	6.98	5.56	6.65	5.02	5.85	5.54
PBE0-2-D3(BJ)	3.18	6.08	5.14	5.36	4.37	4.88	4.57
PBE-QIDH-D3(BJ)	3.52	6.31	4.12	4.91	4.26	4.59	4.47
PBE-CIDH-D3(BJ)	4.22	6.79	4.77	8.93	4.81	6.92	5.77
SOS0-PBE0-2(b)-D3(BJ)	5.12	6.11	5.15	3.73	4.21	3.96	4.83
SOS0-PBE0-DH(b)-D3(BJ)	4.05	6.51	5.51	8.80	4.65	6.77	5.71
SOS0-PBE-CIDH(b)-D3(BJ)	3.96	6.13	4.99	8.19	4.50	6.39	5.40
SOS0-PBE-QIDH(b)-D3(BJ)	4.12	5.66	4.40	4.03	4.02	4.03	4.37
SOS0-PBE0-2-D3(BJ)	2.48	6.11	4.24	3.69	4.15	3.91	3.86
SOS0-PBE0-DH-D3(BJ)	4.24	6.66	4.36	9.20	4.61	6.95	5.72
SOS0-PBE-CIDH-D3(BJ)	4.07	6.27	3.77	8.56	4.47	6.56	5.37
SOS1-PBE-QIDH-D3(BJ)	3.09	6.01	3.43	4.29	4.17	4.23	4.06
TPSS0-DH-D3(BJ)	4.48	6.22	5.33	9.22	4.64	6.98	5.86
TPSS-CIDH-D3(BJ)	4.29	5.90	5.00	9.17	4.47	6.87	5.66
TPSS-QIDH-D3(BJ)	3.43	5.48	4.31	5.80	4.06	4.95	4.47
PBE-QIDH-NL	3.65	6.26	4.35	4.75	4.26	4.51	4.50
SOS1-PBE-QIDH-NL	3.54	5.95	3.54	4.07	4.61	4.34	4.24
revPBE0-DH-D3(BJ)	4.17	5.77	4.11	3.84	5.14	4.48	4.54
MP2-D3(BJ)	4.02	9.54	9.37	4.81	5.60	5.19	6.07
SCS-MP2-D3(BJ)	4.03	8.15	7.31	4.20	5.76	4.96	5.49
SOS-MP2-D3(BJ)	4.63	8.68	7.38	4.55	5.95	5.23	5.88

SI.6.5 WTMADs for dispersion-uncorrected double-hybrid density functional theory approximations

Table S20: WTMAD-1 values (kcal/mol) for dispersion-uncorrected DHDFs for GMTKN55 and its categories (basic properties and reactions of small systems, isomerisations and reactions of large systems, barrier heights, intermolecular noncovalent interactions (NCIs), intramolecular NCIs, and all NCIs). This table only shows WTMADs of newly tested functionals. For values of the seven previously tested DHDFs, see the ESI of Ref. S11.

	basic + small	iso. + large	barriers	intermol. NCIs	intramol. NCIs	all NCIs	GMTKN55
ωB97X-2	1.47	1.40	1.49	2.44	1.39	1.99	1.66
B2NC-PLYP	1.61	2.17	1.50	3.13	2.27	2.77	2.13
mPW2NC-PLYP	2.13	3.16	2.48	3.07	2.77	2.94	2.65
B2PPW91	4.50	6.72	4.92	17.15	13.27	15.49	9.11
mSD-PBEPBE	5.23	4.65	5.24	3.34	3.51	3.41	4.44
LS1-DH	2.31	3.58	2.36	4.08	3.09	3.66	3.04
PBE0-DH	3.04	4.34	2.63	7.47	7.42	7.45	4.88
PBE0-2	2.23	3.37	2.44	3.37	2.58	3.04	2.75
PBE-QIDH	2.46	3.79	2.31	4.95	4.01	4.55	3.46
PBE-CIDH	2.89	4.20	2.50	6.86	6.55	6.73	4.52
SOS0-PBE0-2(b)	3.40	2.71	2.39	2.82	2.59	2.72	2.90
SOS0-PBE0-DH(b)	2.75	3.84	2.53	7.33	7.09	7.23	4.61
SOS0-PBE-CIDH(b)	2.63	3.61	2.34	6.73	6.18	6.49	4.23
SOS0-PBE-QIDH(b)	2.74	3.01	2.10	4.55	3.39	4.05	3.20
SOS0-PBE0-2	1.70	2.46	1.69	6.55	5.29	6.01	3.47
SOS0-PBE0-DH	2.88	4.10	2.21	8.35	8.54	8.43	5.11
SOS0-PBE-CIDH	2.68	3.90	2.01	8.06	8.05	8.06	4.85
SOS1-PBE-QIDH	2.00	3.15	1.68	7.12	6.46	6.84	3.99
TPSS0-DH	3.07	4.38	2.17	9.35	9.01	9.21	5.51
TPSS-CIDH	2.87	4.09	2.01	8.52	8.16	8.36	5.06
TPSS-QIDH	2.31	3.25	1.70	5.98	5.42	5.74	3.69
revPBE0-DH	3.04	4.29	1.86	12.38	11.57	12.03	6.53
MP2	3.02	3.58	3.29	3.15	5.17	4.01	3.52
SCS-MP2	2.48	2.06	1.94	3.87	2.96	3.48	2.73
SOS-MP2	2.99	3.10	3.12	6.75	4.27	5.69	4.05

Table S21: WTMAD-2 values (kcal/mol) for dispersion-uncorrected DHDFs for GMTKN55 and its categories (basic properties and reactions of small systems, isomerisations and reactions of large systems, barrier heights, intermolecular noncovalent interactions (NCIs), intramolecular NCIs, and all NCIs). This table only shows WTMADs of newly tested functionals. For values of the seven previously tested DHDFs, see the ESI of Ref. S11.

	basic + small	iso. + large	barriers	intermol. NCIs	intramol. NCIs	all NCIs	GMTKN55
ωB97X-2	1.66	3.23	3.25	5.26	2.30	3.81	2.97
B2NC-PLYP	2.03	5.30	3.73	5.62	3.75	4.70	3.83
mPW2NC-PLYP	2.67	6.09	5.33	5.03	4.58	4.81	4.41
B2PPW91	5.15	14.21	11.15	32.93	22.46	27.81	16.35
mSD-PBEPBE	7.62	9.18	10.66	5.89	6.82	6.34	7.76
LS1-DH	3.15	6.48	4.42	6.81	5.10	5.97	4.97
PBE0-DH	3.95	8.61	4.47	12.45	12.44	12.45	8.13
PBE0-2	3.01	6.30	4.98	5.70	4.19	4.96	4.57
PBE-QIDH	3.37	6.75	3.90	8.27	6.69	7.50	5.61
PBE-CIDH	3.81	7.94	4.09	11.46	11.02	11.24	7.45
SOS0-PBE0-2(b)	5.07	6.26	5.06	4.84	3.83	4.35	4.98
SOS0-PBE0-DH(b)	3.47	8.01	4.67	12.21	11.95	12.08	7.76
SOS0-PBE-CIDH(b)	3.41	7.34	4.26	11.24	10.41	10.83	7.09
SOS0-PBE-QIDH(b)	3.89	6.16	4.06	7.62	5.52	6.59	5.35
SOS0-PBE0-2	2.30	7.14	4.99	11.95	9.09	10.55	6.69
SOS0-PBE0-DH	3.78	8.69	3.88	14.33	14.33	14.33	8.76
SOS0-PBE-CIDH	3.61	8.12	3.51	13.98	13.52	13.75	8.34
SOS1-PBE-QIDH	2.84	7.30	3.86	12.75	10.87	11.83	7.25
TPSS0-DH	4.26	9.56	5.03	16.33	15.24	15.80	9.77
TPSS-CIDH	4.01	8.79	4.69	14.83	13.73	14.29	8.94
TPSS-QIDH	3.31	7.00	3.97	10.19	9.08	9.65	6.50
revPBE0-DH	3.95	9.27	3.38	22.57	19.80	21.21	11.56
MP2	4.07	10.27	9.46	6.11	8.44	7.25	7.03
SCS-MP2	2.93	8.20	7.87	7.31	6.00	6.67	5.90
SOS-MP2	3.28	10.74	9.94	12.37	8.45	10.45	8.18

SI.7 Analysis of normalised mean deviations and error ranges

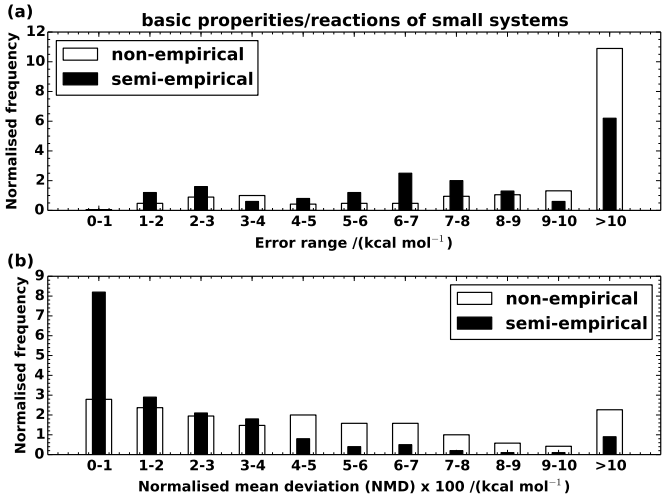

Figure S12: Histograms (1 kcal/mol bins) showing the error ranges (a) and normalised mean deviations (b) for semi- and non-empirical classes of DHDFs.

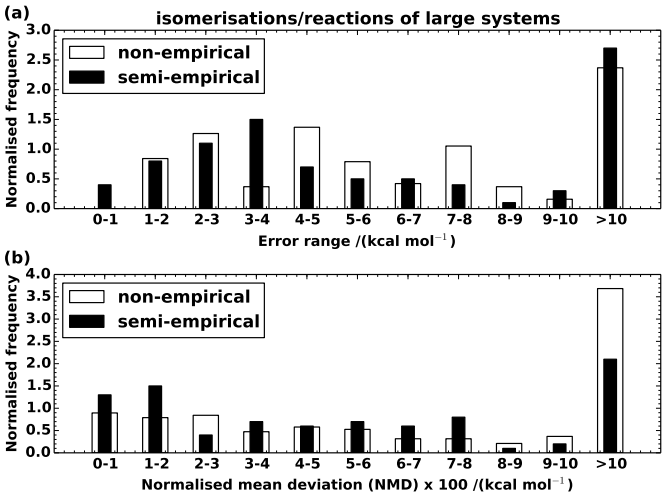

Figure S13: Histograms (1 kcal/mol bins) showing the error ranges (a) and normalised mean deviations (b) for semi- and non-empirical classes of DHDFs.

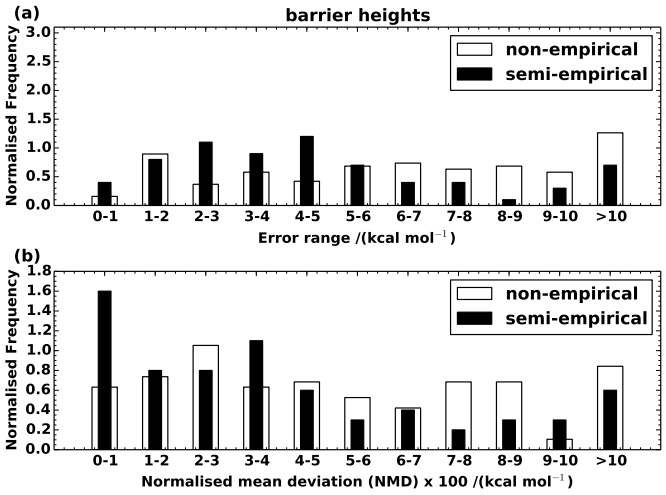

Figure S14: Histograms (1 kcal/mol bins) showing the error ranges (a) and normalised mean deviations (b) for semi- and non-empirical classes of DHDFs.

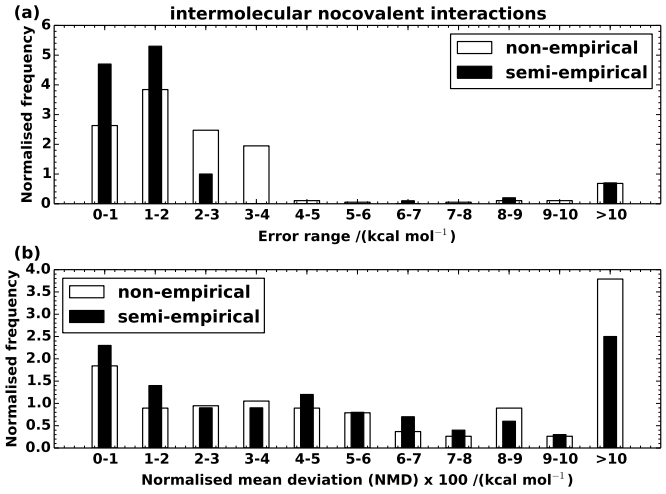

Figure S15: Histograms (1 kcal/mol bins) showing the error ranges (a) and normalised mean deviations (b) for semi- and non-empirical classes of DHDFs.

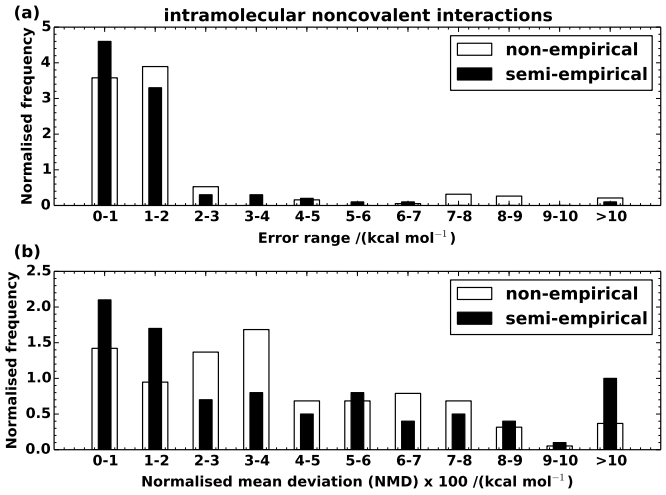

Figure S16: Histograms (1 kcal/mol bins) showing the error ranges (a) and normalised mean deviations (b) for semi- and non-empirical classes of DHDFs.

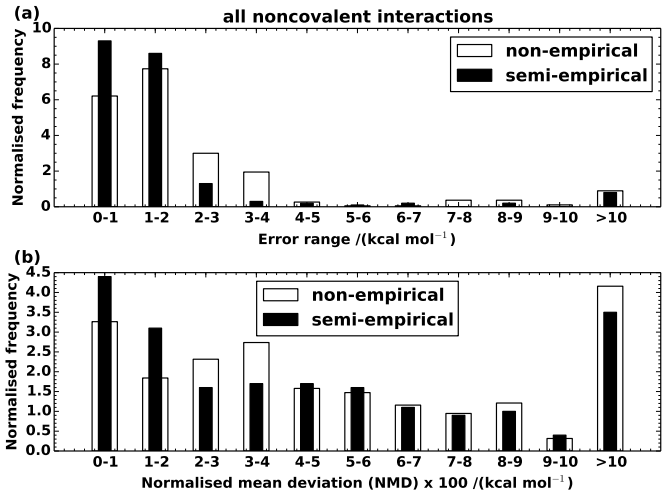

Figure S17: Histograms (1 kcal/mol bins) showing the error ranges (a) and normalised mean deviations (b) for semi- and non-empirical classes of DHDFs.

SI.8 Statistical results for all test sets and DFAs

SI.8.1 Results for dispersion-corrected functionals with DFT-D3(BJ)

Table S22: Statistical analysis of the ωB97X-2-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	4.79	4.79	5.59	0.14	10.47	11.56	1.08
ACONF	0.00	0.02	0.02	0.01	0.07	0.05	−0.02
ADIM6	0.28	0.28	0.29	0.08	0.18	0.36	0.19
AHB21	−0.38	0.42	0.47	0.02	1.56	0.41	−1.16
AL2X6	−0.97	0.97	1.11	0.03	1.68	−0.47	−2.14
ALK8	1.70	2.43	4.14	0.04	12.93	10.69	−2.25
ALKBDE10	−0.16	2.52	3.10	0.03	11.10	4.53	−6.57
AMINO20x4	−0.05	0.10	0.14	0.04	0.56	0.23	−0.34
BH76	−0.04	1.24	2.06	0.07	13.54	8.15	−5.38
BHDIV10	−0.55	0.76	1.07	0.02	3.75	1.05	−2.70
BHPERI	−2.25	2.28	2.39	0.11	3.67	0.38	−3.29
BHROT27	0.07	0.15	0.20	0.02	0.72	0.42	−0.31
BSR36	−1.18	1.18	1.38	0.07	2.98	−0.43	−3.41
BUT14DIOL	0.00	0.03	0.03	0.01	0.17	0.08	−0.09
C60ISO	−1.43	3.44	4.21	0.04	11.70	3.18	−8.52
CARBHB12	0.60	0.60	0.66	0.10	0.95	1.21	0.26
CDIE20	0.23	0.27	0.41	0.07	1.12	0.97	−0.15
CHB6	−0.83	0.83	0.90	0.03	0.98	−0.52	−1.50
DARC	−0.60	0.88	0.98	0.03	2.44	0.55	−1.89
DC13	−0.31	2.71	4.03	0.05	17.80	7.91	−9.89
DIPCS10	−5.11	5.11	5.66	0.01	8.09	−0.94	−9.03
FH51	−0.23	0.75	1.06	0.02	6.20	1.58	−4.62
G21EA	−1.04	1.60	1.91	0.05	8.00	4.47	−3.53
G21IP	−1.12	2.09	2.65	0.01	13.67	5.52	−8.15
G2RC	−0.46	1.23	1.53	0.02	6.38	2.61	−3.76
HAL59	0.40	0.41	0.51	0.09	1.63	1.37	−0.26
HEAVY28	0.31	0.31	0.33	0.25	0.51	0.63	0.13
HEAVYSB11	−1.41	1.41	1.63	0.02	2.90	−0.01	−2.92
ICONF	0.02	0.13	0.17	0.04	0.67	0.37	−0.30
IDISP	−0.43	0.65	1.17	0.05	3.20	0.40	−2.80
IL16	0.54	0.54	0.57	0.00	0.52	0.81	0.29
INV24	0.06	0.95	1.75	0.03	9.66	7.26	−2.40
ISO34	−0.06	0.56	0.85	0.04	4.26	2.63	−1.63
ISOL24	0.01	1.02	1.46	0.05	7.43	3.16	−4.28
MB16-43	−16.03	16.87	19.18	0.04	51.58	13.28	−38.29
MCONF	0.25	0.27	0.30	0.05	0.58	0.47	−0.11
NBPRC	0.08	0.97	1.18	0.04	4.07	2.90	−1.17
PA26	0.26	0.98	1.38	0.01	5.49	3.81	−1.68
PArel	0.04	0.36	0.58	0.08	2.77	0.77	−2.00
PCONF21	0.04	0.19	0.24	0.12	0.84	0.47	−0.37
PNICO23	0.31	0.31	0.32	0.07	0.35	0.45	0.10
PX13	−2.05	2.05	2.18	0.06	2.75	−0.59	−3.34
RC21	−1.08	1.57	1.94	0.04	5.93	1.76	−4.17
RG18	0.18	0.18	0.21	0.31	0.41	0.46	0.05
RSE43	0.49	0.60	1.26	0.08	5.80	5.44	−0.36
S22	0.27	0.34	0.38	0.05	1.03	0.64	−0.39
S66	0.33	0.33	0.35	0.06	0.54	0.57	0.03
SCONF	−0.01	0.05	0.07	0.01	0.27	0.06	−0.21
TAUT15	0.16	0.45	0.58	0.15	1.92	1.08	−0.83
UPU23	0.26	0.40	0.51	0.07	1.81	1.13	−0.68
W4-11	−3.50	3.64	4.48	0.01	20.60	2.32	−18.28
WATER27	3.64	3.77	5.17	0.05	14.61	12.83	−1.77
WCPT18	−1.24	1.67	2.01	0.05	7.17	3.75	−3.42
YBDE18	−0.08	1.40	1.71	0.03	6.86	4.47	−2.39
BH76RC	−0.14	0.71	0.97	0.03	4.68	2.23	−2.45

Table S23: Statistical analysis of the B2PPW91-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	18.25	18.25	20.26	0.54	31.85	36.21	4.36
ACONF	−0.20	0.20	0.23	0.11	0.41	−0.01	−0.43
ADIM6	0.16	0.26	0.32	0.08	0.77	0.53	−0.24
AHB21	0.09	0.56	0.77	0.02	3.79	2.24	−1.55
AL2X6	−2.27	2.27	3.36	0.06	6.78	−0.32	−7.10
ALK8	0.11	1.94	2.46	0.03	7.70	5.13	−2.57
ALKBDE10	4.08	5.69	8.92	0.06	26.61	23.86	−2.75
AMINO20x4	−0.02	0.36	0.45	0.15	2.26	1.19	−1.07
BH76	−6.75	6.79	7.54	0.36	23.95	1.51	−22.44
BHDIV10	−6.42	6.81	7.67	0.15	14.10	1.92	−12.18
BHPERI	−5.95	5.95	6.17	0.29	6.11	−2.90	−9.02
BHROT27	0.27	0.34	0.49	0.05	1.61	1.35	−0.26
BSR36	−1.82	1.82	1.99	0.11	3.52	−0.68	−4.20
BUT14DIOL	0.22	0.27	0.36	0.10	1.38	1.00	−0.38
C60ISO	−11.89	11.89	14.58	0.12	24.65	−1.16	−25.81
CARBHB12	0.85	0.89	1.32	0.15	3.17	3.00	−0.17
CDIE20	1.35	1.35	1.44	0.33	2.00	2.32	0.32
CHB6	1.70	1.70	2.13	0.06	3.87	4.26	0.38
DARC	1.58	2.21	2.58	0.07	6.40	4.29	−2.11
DC13	1.84	7.40	9.62	0.13	36.24	12.50	−23.74
DIPCS10	−1.49	3.02	3.68	0.00	10.75	3.55	−7.20
FH51	0.90	2.19	3.17	0.07	16.62	8.92	−7.70
G21EA	1.83	2.44	2.89	0.07	8.66	5.61	−3.06
G21IP	0.21	3.23	3.95	0.01	16.38	8.89	−7.48
G2RC	1.29	4.97	6.19	0.10	26.57	16.68	−9.88
HAL59	−0.39	0.94	1.16	0.20	5.64	4.15	−1.49
HEAVY28	−0.78	0.78	0.85	0.63	1.25	−0.17	−1.42
HEAVYSB11	−0.76	2.41	2.82	0.04	8.76	3.01	−5.75
ICONF	0.20	0.33	0.46	0.10	1.54	1.27	−0.27
IDISP	2.33	2.33	2.83	0.16	4.25	4.60	0.35
IL16	0.17	0.34	0.44	0.00	1.51	1.03	−0.48
INV24	−1.40	1.81	2.57	0.06	9.02	2.01	−7.01
ISO34	−0.41	1.13	1.50	0.08	8.34	3.87	−4.47
ISOL24	−0.86	3.11	4.51	0.14	21.14	13.16	−7.98
MB16-43	−7.08	18.82	24.83	0.05	116.53	47.51	−69.02
MCONF	0.31	0.52	0.65	0.10	2.27	1.43	−0.84
NBPRC	−0.11	1.63	1.95	0.06	6.60	2.52	−4.08
PA26	3.53	3.53	4.06	0.02	8.09	9.04	0.95
PArel	0.22	1.54	2.12	0.33	10.59	4.84	−5.75
PCONF21	−0.52	0.92	1.13	0.57	3.16	0.80	−2.36
PNICO23	0.03	0.62	0.96	0.15	4.75	3.78	−0.98
PX13	−9.60	9.60	9.80	0.29	7.12	−6.98	−14.10
RC21	3.62	4.47	5.00	0.13	12.25	9.36	−2.88
RG18	−0.71	0.71	0.94	1.22	2.24	−0.19	−2.44
RSE43	−1.86	1.86	2.10	0.24	4.73	−0.42	−5.15
S22	0.06	0.24	0.32	0.03	1.32	0.91	−0.41
S66	−0.01	0.22	0.26	0.04	1.07	0.53	−0.54
SCONF	0.36	0.97	1.18	0.21	4.22	1.19	−3.03
TAUT15	−0.05	1.56	1.82	0.51	5.93	3.67	−2.26
UPU23	0.20	0.44	0.56	0.08	2.29	1.42	−0.87
W4-11	8.10	9.35	11.79	0.03	46.27	35.33	−10.93
WATER27	0.88	1.61	2.43	0.02	13.98	4.73	−9.25
WCPT18	−7.31	7.31	7.68	0.21	9.21	−4.29	−13.50
YBDE18	−1.00	3.46	4.06	0.07	15.48	7.56	−7.92
BH76RC	0.49	2.73	4.35	0.13	22.92	17.77	−5.15

Table S24: Statistical analysis of the B2NC-PLYP-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	2.67	2.67	3.01	0.08	4.82	5.51	0.69
ACONF	−0.16	0.17	0.20	0.09	0.43	0.05	−0.38
ADIM6	−0.43	0.43	0.48	0.13	0.59	−0.16	−0.75
AHB21	−0.51	0.53	0.73	0.02	1.94	0.13	−1.81
AL2X6	−1.96	1.96	2.01	0.05	1.24	−1.50	−2.74
ALK8	−0.30	1.46	1.84	0.02	6.38	3.42	−2.96
ALKBDE10	−0.90	2.82	3.46	0.03	11.00	4.72	−6.27
AMINO20x4	−0.02	0.13	0.17	0.05	1.08	0.50	−0.58
BH76	1.20	1.80	2.88	0.10	18.98	15.48	−3.50
BHDIV10	0.27	1.08	1.48	0.02	5.35	3.28	−2.07
BHPERI	−2.04	2.07	2.25	0.10	4.20	0.43	−3.76
BHROT27	0.30	0.31	0.41	0.05	1.02	0.96	−0.06
BSR36	−1.67	1.67	1.79	0.10	2.18	−0.83	−3.01
BUT14DIOL	0.02	0.07	0.09	0.03	0.35	0.22	−0.14
C60ISO	−7.69	7.78	10.33	0.08	20.36	0.22	−20.14
CARBHB12	0.42	0.42	0.55	0.07	1.26	1.26	0.00
CDIE20	0.55	0.55	0.61	0.14	0.96	0.96	0.01
CHB6	−1.13	1.13	1.23	0.04	1.45	−0.36	−1.81
DARC	0.20	0.87	1.10	0.03	3.34	1.84	−1.50
DC13	1.65	3.28	4.20	0.06	15.13	8.45	−6.67
DIPCS10	−3.13	3.28	4.17	0.01	8.93	0.70	−8.22
FH51	−0.87	1.37	1.98	0.04	10.21	2.86	−7.35
G21EA	−1.42	2.13	2.55	0.06	11.23	7.19	−4.04
G21IP	−0.67	1.82	2.60	0.01	16.52	7.80	−8.72
G2RC	−1.58	1.92	2.53	0.04	8.49	2.28	−6.21
HAL59	0.06	0.23	0.34	0.05	1.94	1.31	−0.63
HEAVY28	−0.15	0.16	0.19	0.13	0.42	0.05	−0.37
HEAVYSB11	−0.79	0.98	1.33	0.02	3.15	0.53	−2.62
ICONF	0.08	0.20	0.27	0.06	0.98	0.49	−0.48
IDISP	0.38	0.57	0.67	0.04	1.38	0.95	−0.43
IL16	0.47	0.47	0.54	0.00	1.14	1.16	0.01
INV24	0.38	0.92	1.35	0.03	6.37	4.29	−2.07
ISO34	0.42	0.64	1.17	0.04	5.45	4.68	−0.77
ISOL24	0.32	1.54	2.26	0.07	10.43	5.97	−4.46
MB16-43	−5.79	10.54	13.15	0.03	55.96	21.14	−34.82
MCONF	0.26	0.30	0.34	0.06	0.89	0.64	−0.25
NBPRC	0.42	0.73	0.88	0.03	2.63	1.89	−0.74
PA26	0.32	0.96	1.48	0.01	7.44	3.85	−3.59
PArel	0.28	0.45	0.75	0.10	3.53	2.68	−0.86
PCONF21	0.13	0.25	0.29	0.15	0.87	0.56	−0.31
PNICO23	−0.09	0.13	0.16	0.03	0.45	0.14	−0.31
PX13	−1.08	1.11	1.25	0.03	2.07	0.15	−1.92
RC21	−2.15	2.83	3.25	0.08	7.72	2.58	−5.15
RG18	−0.09	0.18	0.24	0.31	1.04	0.54	−0.50
RSE43	1.15	1.15	2.22	0.15	9.30	9.26	−0.03
S22	0.14	0.18	0.23	0.02	0.79	0.59	−0.21
S66	0.01	0.21	0.26	0.04	1.24	0.72	−0.52
SCONF	0.02	0.06	0.10	0.01	0.44	0.34	−0.10
TAUT15	0.29	0.44	0.62	0.14	1.95	1.08	−0.87
UPU23	0.27	0.40	0.50	0.07	1.51	1.06	−0.45
W4-11	−3.14	3.46	4.98	0.01	27.25	3.97	−23.28
WATER27	1.41	1.58	2.04	0.02	8.57	6.30	−2.27
WCPT18	−0.48	0.90	1.14	0.03	4.08	2.22	−1.86
YBDE18	1.19	1.62	2.14	0.03	7.21	5.62	−1.59
BH76RC	−0.43	1.15	1.62	0.05	7.84	3.59	−4.26

Table S25: Statistical analysis of the mPW2NC-PLYP-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.52	7.52	8.39	0.22	12.51	13.69	1.18
ACONF	−0.13	0.14	0.16	0.08	0.35	0.04	−0.31
ADIM6	−0.38	0.38	0.45	0.11	0.70	−0.07	−0.77
AHB21	−0.40	0.44	0.56	0.02	1.90	0.25	−1.65
AL2X6	−0.81	0.81	0.98	0.02	1.58	−0.14	−1.72
ALK8	1.50	2.11	2.97	0.03	8.56	7.01	−1.55
ALKBDE10	4.69	5.19	6.89	0.05	13.79	12.45	−1.34
AMINO20x4	−0.04	0.13	0.18	0.05	1.13	0.65	−0.48
BH76	−1.61	2.27	2.80	0.12	15.39	8.14	−7.25
BHDIV10	−1.77	2.68	3.13	0.06	9.15	3.32	−5.83
BHPERI	−3.89	3.89	4.05	0.19	5.24	−1.93	−7.17
BHROT27	0.17	0.19	0.30	0.03	1.07	0.94	−0.13
BSR36	−1.46	1.46	1.59	0.09	2.31	−0.70	−3.01
BUT14DIOL	0.11	0.11	0.14	0.04	0.31	0.31	−0.01
C60ISO	−13.28	13.28	16.41	0.14	27.44	−2.34	−29.78
CARBHB12	0.66	0.66	0.80	0.11	1.42	1.58	0.16
CDIE20	0.73	0.73	0.77	0.18	0.98	1.23	0.25
CHB6	−1.57	1.57	1.66	0.06	1.73	−0.94	−2.67
DARC	3.45	3.45	3.65	0.11	3.47	5.29	1.82
DC13	2.81	5.19	6.54	0.09	22.21	9.24	−12.97
DIPCS10	−2.33	3.07	3.40	0.00	7.84	2.68	−5.15
FH51	0.42	1.44	1.98	0.05	12.13	6.50	−5.63
G21EA	0.61	1.48	1.98	0.04	6.53	4.41	−2.12
G21IP	−0.17	1.91	2.45	0.01	13.24	6.07	−7.18
G2RC	0.48	1.62	2.36	0.03	11.42	7.25	−4.17
HAL59	0.57	0.57	0.79	0.12	2.68	2.57	−0.11
HEAVY28	0.19	0.21	0.24	0.17	0.66	0.51	−0.15
HEAVYSB11	2.39	2.40	3.02	0.04	6.22	6.14	−0.07
ICONF	0.02	0.18	0.22	0.06	0.74	0.37	−0.37
IDISP	1.25	1.25	1.84	0.09	3.68	3.80	0.12
IL16	0.26	0.29	0.35	0.00	0.80	0.65	−0.14
INV24	−0.54	0.89	1.70	0.03	9.05	1.51	−7.53
ISO34	0.21	0.81	1.31	0.06	6.93	5.53	−1.40
ISOL24	−0.77	3.05	4.34	0.14	21.74	12.69	−9.04
MB16-43	1.39	4.91	6.15	0.01	28.82	18.61	−10.21
MCONF	0.19	0.25	0.30	0.05	1.02	0.64	−0.38
NBPRC	0.53	0.94	1.34	0.03	4.35	3.59	−0.75
PA26	−0.42	1.16	1.44	0.01	6.89	3.41	−3.48
PArel	0.03	0.68	1.20	0.15	6.52	2.72	−3.80
PCONF21	−0.02	0.36	0.41	0.22	1.39	0.65	−0.74
PNICO23	0.29	0.29	0.44	0.07	1.53	1.48	−0.06
PX13	−3.66	3.66	3.74	0.11	3.31	−2.16	−5.48
RC21	−1.54	2.13	2.65	0.06	7.29	1.85	−5.45
RG18	0.13	0.16	0.26	0.28	1.03	0.87	−0.16
RSE43	0.20	0.67	1.29	0.09	7.19	5.58	−1.62
S22	0.12	0.17	0.23	0.02	0.74	0.56	−0.18
S66	0.05	0.21	0.26	0.04	0.94	0.48	−0.46
SCONF	0.09	0.17	0.21	0.04	0.89	0.27	−0.62
TAUT15	0.20	0.63	0.79	0.21	2.47	1.74	−0.73
UPU23	0.22	0.37	0.46	0.06	1.62	1.09	−0.54
W4-11	5.66	6.03	7.54	0.02	24.84	20.06	−4.78
WATER27	2.49	2.81	3.84	0.03	15.64	11.23	−4.41
WCPT18	−2.96	2.96	3.14	0.08	3.49	−1.83	−5.32
YBDE18	1.79	2.02	3.08	0.04	9.55	8.20	−1.35
BH76RC	−0.01	1.50	2.22	0.08	12.07	9.38	−2.69

Table S26: Statistical analysis of the PBE0-DH-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.57	7.57	8.56	0.22	15.16	17.56	2.41
ACONF	−0.11	0.11	0.13	0.06	0.25	0.02	−0.23
ADIM6	−0.24	0.24	0.27	0.07	0.39	−0.07	−0.46
AHB21	−1.38	1.38	1.74	0.06	3.88	−0.08	−3.96
AL2X6	3.16	3.16	3.35	0.09	3.59	4.47	0.88
ALK8	7.21	7.27	9.46	0.12	17.15	16.92	−0.23
ALKBDE10	−5.76	5.88	7.12	0.06	15.28	0.58	−14.71
AMINO20x4	0.08	0.25	0.32	0.10	1.52	0.90	−0.62
BH76	−1.47	2.00	2.23	0.11	9.57	3.61	−5.96
BHDIV10	−2.43	2.95	3.38	0.07	7.60	2.41	−5.19
BHPERI	−2.31	2.90	3.23	0.14	9.90	4.11	−5.79
BHROT27	0.59	0.59	0.78	0.09	1.56	1.51	−0.05
BSR36	−2.30	2.30	2.51	0.14	3.64	−1.20	−4.84
BUT14DIOL	0.10	0.11	0.14	0.04	0.47	0.42	−0.06
C60ISO	2.09	3.12	3.33	0.03	8.00	5.44	−2.57
CARBHB12	1.18	1.18	1.42	0.20	2.28	2.65	0.37
CDIE20	0.91	0.94	1.16	0.23	2.73	2.46	−0.27
CHB6	−2.20	2.20	2.46	0.08	3.16	−0.91	−4.07
DARC	−7.44	7.44	7.67	0.23	6.53	−5.07	−11.61
DC13	−2.90	9.58	11.51	0.17	40.80	20.44	−20.37
DIPCS10	−0.65	2.60	3.35	0.00	11.18	4.07	−7.11
FH51	−2.44	3.12	3.93	0.10	15.43	5.57	−9.86
G21EA	−1.48	3.01	3.46	0.09	9.44	3.21	−6.23
G21IP	0.05	3.08	3.75	0.01	15.64	7.76	−7.88
G2RC	−4.15	6.63	8.42	0.13	35.20	11.95	−23.25
HAL59	0.57	0.64	0.96	0.14	3.40	3.07	−0.33
HEAVY28	0.36	0.37	0.46	0.30	1.09	0.95	−0.15
HEAVYSB11	1.17	1.36	1.72	0.02	3.97	3.27	−0.70
ICONF	0.21	0.29	0.42	0.09	1.42	1.13	−0.29
IDISP	−0.43	1.64	2.37	0.12	6.68	1.23	−5.45
IL16	−0.34	0.38	0.47	0.00	1.06	0.21	−0.84
INV24	0.51	1.56	2.29	0.05	10.42	6.22	−4.21
ISO34	0.07	1.44	1.98	0.10	7.99	4.19	−3.80
ISOL24	1.68	2.75	3.85	0.13	13.60	8.69	−4.92
MB16-43	19.03	20.86	25.48	0.05	90.02	76.51	−13.51
MCONF	0.19	0.25	0.32	0.05	1.11	0.69	−0.41
NBPRC	−1.59	3.51	4.07	0.13	12.92	5.82	−7.10
PA26	2.65	2.77	3.22	0.01	8.94	7.30	−1.64
PArel	0.88	1.21	1.59	0.26	6.04	4.13	−1.91
PCONF21	−0.05	0.58	0.70	0.36	2.28	0.93	−1.35
PNICO23	0.95	0.95	1.32	0.22	4.15	4.19	0.03
PX13	−3.90	3.90	4.10	0.12	4.58	−1.87	−6.44
RC21	4.28	4.79	5.97	0.13	18.36	16.08	−2.28
RG18	0.02	0.09	0.15	0.16	0.70	0.52	−0.18
RSE43	−0.48	0.50	0.64	0.07	2.19	0.40	−1.80
S22	0.33	0.41	0.59	0.06	2.21	1.87	−0.34
S66	0.21	0.32	0.49	0.06	2.34	1.94	−0.40
SCONF	0.06	0.20	0.26	0.04	1.09	0.29	−0.81
TAUT15	0.42	0.84	1.24	0.28	3.85	2.67	−1.18
UPU23	0.47	0.53	0.67	0.09	1.62	1.39	−0.23
W4-11	−4.75	5.38	7.14	0.02	37.14	10.02	−27.13
WATER27	3.75	4.11	5.06	0.05	18.27	13.43	−4.84
WCPT18	−2.35	2.56	3.26	0.07	8.22	1.91	−6.31
YBDE18	1.23	2.17	2.76	0.04	8.49	4.60	−3.89
BH76RC	−0.51	2.19	2.77	0.12	11.22	5.59	−5.63

Table S27: Statistical analysis of the LS1-DH-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	2.47	2.47	2.80	0.07	4.66	5.43	0.77
ACONF	−0.13	0.15	0.18	0.08	0.42	0.08	−0.34
ADIM6	−0.56	0.56	0.61	0.17	0.68	−0.24	−0.91
AHB21	−1.08	1.09	1.47	0.05	3.61	0.08	−3.53
AL2X6	0.69	0.88	0.91	0.02	1.81	1.25	−0.55
ALK8	3.15	3.15	4.79	0.05	11.25	11.53	0.28
ALKBDE10	−3.37	4.00	4.67	0.04	10.20	2.85	−7.35
AMINO20x4	0.05	0.19	0.24	0.08	1.18	0.67	−0.50
BH76	0.86	1.52	2.45	0.08	15.59	12.94	−2.65
BHDIV10	−1.20	1.88	2.11	0.04	5.39	2.07	−3.32
BHPERI	−2.83	3.17	3.45	0.15	8.58	2.94	−5.64
BHROT27	0.47	0.48	0.61	0.08	1.28	1.23	−0.05
BSR36	−1.38	1.38	1.43	0.09	1.36	−0.80	−2.16
BUT14DIOL	0.01	0.06	0.07	0.02	0.31	0.17	−0.14
C60ISO	−4.20	5.04	6.78	0.05	16.19	2.03	−14.16
CARBHB12	0.78	0.78	0.99	0.13	1.86	2.01	0.15
CDIE20	0.72	0.74	0.88	0.18	1.83	1.73	−0.10
CHB6	−1.33	1.33	1.50	0.05	1.95	−0.46	−2.40
DARC	−6.83	6.83	6.99	0.21	4.27	−4.63	−8.90
DC13	−1.29	8.20	10.21	0.15	35.45	18.87	−16.58
DIPCS10	−1.16	2.84	3.30	0.00	9.09	2.82	−6.27
FH51	−2.42	2.75	3.40	0.09	12.85	5.40	−7.45
G21EA	−1.42	2.43	2.89	0.07	10.32	5.05	−5.27
G21IP	0.13	2.28	2.87	0.01	14.11	5.56	−8.55
G2RC	−3.74	4.88	6.17	0.10	23.00	6.13	−16.87
HAL59	0.17	0.34	0.54	0.07	2.54	2.08	−0.45
HEAVY28	−0.01	0.13	0.16	0.10	0.59	0.32	−0.27
HEAVYSB11	0.39	0.73	0.89	0.01	2.81	1.77	−1.04
ICONF	0.20	0.21	0.34	0.06	0.89	0.82	−0.07
IDISP	−0.91	1.83	3.19	0.13	8.63	0.99	−7.64
IL16	−0.09	0.26	0.30	0.00	0.98	0.44	−0.54
INV24	0.59	1.24	1.78	0.04	9.16	5.89	−3.26
ISO34	0.30	1.30	1.88	0.09	7.54	3.99	−3.55
ISOL24	1.89	2.88	3.93	0.13	12.85	9.85	−3.00
MB16-43	12.46	14.23	17.19	0.03	59.33	41.96	−17.37
MCONF	0.31	0.34	0.40	0.07	0.95	0.72	−0.23
NBPRC	−1.09	2.39	2.87	0.09	9.73	4.20	−5.52
PA26	1.35	1.71	2.05	0.01	7.30	4.90	−2.39
PArel	0.75	0.96	1.26	0.21	4.73	3.02	−1.71
PCONF21	0.02	0.42	0.51	0.26	1.79	0.79	−1.01
PNICO23	0.34	0.37	0.60	0.09	2.26	2.13	−0.13
PX13	−2.63	2.63	2.81	0.08	3.62	−0.85	−4.48
RC21	1.35	2.54	3.85	0.07	15.82	11.84	−3.99
RG18	−0.12	0.18	0.24	0.31	0.98	0.48	−0.50
RSE43	0.70	0.76	1.44	0.10	6.10	5.79	−0.31
S22	0.18	0.25	0.35	0.03	1.53	1.18	−0.35
S66	0.01	0.28	0.37	0.05	1.94	1.28	−0.66
SCONF	0.14	0.17	0.18	0.04	0.50	0.28	−0.23
TAUT15	0.50	0.72	0.98	0.24	2.72	2.03	−0.69
UPU23	0.32	0.43	0.53	0.08	1.38	1.09	−0.29
W4-11	−3.53	4.04	5.68	0.01	27.62	5.60	−22.02
WATER27	1.74	2.04	2.43	0.03	10.99	6.99	−4.00
WCPT18	−1.69	1.93	2.35	0.06	6.40	2.16	−4.24
YBDE18	2.59	3.11	3.91	0.06	8.90	6.94	−1.97
BH76RC	−0.50	1.85	2.41	0.10	10.85	5.81	−5.04

Table S28: Statistical analysis of the PBE0-2-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	1.81	1.81	2.05	0.05	3.18	3.63	0.44
ACONF	−0.21	0.21	0.24	0.11	0.42	0.03	−0.39
ADIM6	−0.37	0.37	0.40	0.11	0.42	−0.16	−0.58
AHB21	−1.04	1.04	1.39	0.05	3.53	0.01	−3.52
AL2X6	1.14	1.25	1.34	0.03	2.18	1.85	−0.33
ALK8	4.00	4.01	6.69	0.06	17.08	17.05	−0.03
ALKBDE10	−2.13	3.58	3.88	0.04	10.57	3.85	−6.72
AMINO20x4	0.04	0.18	0.23	0.07	1.01	0.62	−0.40
BH76	1.22	1.86	2.93	0.10	18.07	15.01	−3.07
BHDIV10	−1.23	1.94	2.18	0.04	5.89	2.01	−3.88
BHPERI	−3.56	3.78	4.11	0.18	9.07	2.47	−6.60
BHROT27	0.43	0.44	0.56	0.07	1.22	1.17	−0.05
BSR36	−0.55	0.62	0.67	0.04	1.86	0.69	−1.18
BUT14DIOL	0.02	0.06	0.07	0.02	0.29	0.18	−0.11
C60ISO	−6.23	6.71	8.88	0.07	19.05	1.18	−17.87
CARBHB12	0.79	0.79	0.99	0.13	1.78	1.96	0.18
CDIE20	0.68	0.69	0.81	0.17	1.58	1.52	−0.07
CHB6	−1.49	1.49	1.64	0.06	1.96	−0.56	−2.52
DARC	−6.76	6.76	6.89	0.21	3.99	−4.58	−8.57
DC13	−0.97	8.18	10.26	0.15	36.05	20.19	−15.86
DIPCS10	−1.51	2.90	3.42	0.00	9.55	2.52	−7.03
FH51	−2.32	2.63	3.30	0.08	13.99	5.62	−8.37
G21EA	−1.27	2.23	2.77	0.07	11.57	6.58	−4.99
G21IP	0.09	2.11	2.83	0.01	16.27	7.43	−8.84
G2RC	−3.42	4.44	5.62	0.09	20.43	4.97	−15.46
HAL59	0.38	0.45	0.67	0.10	2.84	2.50	−0.33
HEAVY28	0.26	0.28	0.35	0.23	1.02	0.77	−0.26
HEAVYSB11	1.51	1.65	1.96	0.03	4.43	3.98	−0.46
ICONF	0.16	0.22	0.32	0.07	1.10	0.72	−0.38
IDISP	−1.00	2.89	4.06	0.20	11.90	2.76	−9.14
IL16	−0.23	0.30	0.35	0.00	0.84	0.17	−0.67
INV24	0.59	1.13	1.64	0.04	8.13	5.35	−2.78
ISO34	0.36	1.24	1.81	0.09	7.48	4.11	−3.37
ISOL24	2.00	2.96	4.06	0.14	14.13	11.25	−2.88
MB16-43	17.09	17.75	20.24	0.04	52.15	41.41	−10.74
MCONF	0.48	0.52	0.59	0.10	1.30	1.01	−0.29
NBPRC	−1.16	2.23	2.75	0.08	9.06	3.56	−5.50
PA26	0.96	1.48	1.80	0.01	7.46	4.19	−3.27
PArel	0.67	0.87	1.17	0.19	4.81	3.11	−1.70
PCONF21	0.06	0.31	0.41	0.19	1.54	0.69	−0.86
PNICO23	0.45	0.46	0.66	0.11	2.25	2.20	−0.05
PX13	−2.62	2.62	2.78	0.08	3.52	−0.89	−4.41
RC21	0.44	2.60	3.61	0.07	14.67	10.01	−4.66
RG18	−0.08	0.17	0.25	0.29	1.19	0.67	−0.52
RSE43	1.03	1.05	2.02	0.14	8.49	8.30	−0.18
S22	0.39	0.40	0.50	0.05	1.20	1.09	−0.11
S66	0.16	0.28	0.35	0.05	1.62	1.19	−0.43
SCONF	0.17	0.21	0.23	0.05	0.63	0.33	−0.31
TAUT15	0.52	0.71	0.94	0.23	2.48	1.91	−0.58
UPU23	0.20	0.36	0.46	0.06	1.69	0.95	−0.74
W4-11	−2.41	3.49	5.01	0.01	28.32	6.34	−21.99
WATER27	1.67	1.96	2.34	0.02	10.69	6.77	−3.91
WCPT18	−1.82	2.02	2.37	0.06	6.02	1.78	−4.24
YBDE18	3.33	3.58	4.50	0.07	8.97	7.70	−1.27
BH76RC	−0.42	1.94	2.44	0.10	10.97	6.55	−4.42

Table S29: Statistical analysis of the revPBE0-DH-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.36	7.36	8.52	0.22	16.22	18.20	1.98
ACONF	−0.11	0.12	0.13	0.07	0.25	0.02	−0.23
ADIM6	−0.21	0.21	0.24	0.06	0.33	−0.07	−0.41
AHB21	−0.50	0.69	1.01	0.03	3.37	0.51	−2.86
AL2X6	0.95	1.59	1.68	0.04	4.39	2.48	−1.91
ALK8	4.77	4.77	6.06	0.08	9.26	10.04	0.78
ALKBDE10	−8.45	8.45	9.37	0.08	15.23	−2.43	−17.66
AMINO20x4	0.00	0.26	0.33	0.11	1.58	0.86	−0.72
BH76	−0.73	1.35	1.65	0.07	8.85	5.07	−3.78
BHDIV10	−1.76	2.19	2.47	0.05	5.95	2.12	−3.83
BHPERI	−1.75	2.41	2.79	0.12	9.13	4.00	−5.13
BHROT27	0.50	0.50	0.66	0.08	1.28	1.24	−0.04
BSR36	−2.13	2.13	2.25	0.13	3.03	−1.11	−4.14
BUT14DIOL	−0.16	0.20	0.23	0.07	0.68	0.28	−0.40
C60ISO	2.15	3.14	3.36	0.03	7.88	5.37	−2.51
CARBHB12	0.74	0.74	0.98	0.12	1.92	2.03	0.11
CDIE20	0.85	0.87	1.10	0.21	2.59	2.39	−0.20
CHB6	−1.07	1.26	1.51	0.05	3.37	0.57	−2.80
DARC	−6.23	6.23	6.42	0.19	5.48	−4.16	−9.64
DC13	−1.56	7.56	9.56	0.14	34.45	19.43	−15.02
DIPCS10	−1.27	3.06	3.92	0.00	13.01	4.14	−8.87
FH51	−1.86	2.47	3.06	0.08	10.99	4.02	−6.97
G21EA	−2.10	3.36	3.99	0.10	10.55	2.97	−7.59
G21IP	−0.44	3.35	3.95	0.01	15.24	7.54	−7.70
G2RC	−3.08	5.34	6.93	0.10	31.00	11.02	−19.98
HAL59	0.03	0.38	0.52	0.08	2.76	1.90	−0.86
HEAVY28	0.10	0.15	0.21	0.12	0.74	0.45	−0.28
HEAVYSB11	−1.96	1.96	2.11	0.03	2.54	−0.74	−3.28
ICONF	0.17	0.24	0.40	0.07	1.39	1.15	−0.24
IDISP	−0.10	1.77	2.48	0.12	7.18	1.58	−5.60
IL16	0.45	0.51	0.60	0.00	1.16	0.97	−0.19
INV24	0.80	1.64	2.41	0.05	10.18	6.43	−3.75
ISO34	0.03	1.28	1.77	0.09	7.41	3.98	−3.43
ISOL24	1.40	2.36	3.22	0.11	11.49	7.36	−4.13
MB16-43	3.74	11.90	16.21	0.03	88.59	62.28	−26.31
MCONF	0.16	0.27	0.32	0.05	1.22	0.69	−0.54
NBPRC	−0.80	2.66	2.95	0.10	9.33	4.72	−4.61
PA26	4.05	4.05	4.46	0.02	8.46	9.09	0.63
PArel	0.77	0.95	1.34	0.21	4.43	3.67	−0.76
PCONF21	−0.08	0.42	0.54	0.26	2.07	0.77	−1.30
PNICO23	0.48	0.53	0.79	0.12	3.10	2.76	−0.34
PX13	−2.09	2.09	2.29	0.06	3.04	−0.59	−3.63
RC21	3.15	3.73	4.64	0.10	14.99	13.10	−1.89
RG18	−0.05	0.10	0.15	0.17	0.71	0.41	−0.30
RSE43	−0.19	0.31	0.41	0.04	1.97	0.69	−1.28
S22	0.13	0.20	0.27	0.03	1.14	0.74	−0.40
S66	0.02	0.18	0.22	0.03	1.20	0.82	−0.38
SCONF	−0.12	0.17	0.22	0.04	0.71	0.23	−0.48
TAUT15	0.14	0.66	0.78	0.22	2.65	1.54	−1.11
UPU23	0.44	0.54	0.65	0.09	1.58	1.31	−0.28
W4-11	−10.66	10.73	12.48	0.03	34.45	2.78	−31.67
WATER27	−1.65	2.09	3.25	0.03	10.70	1.25	−9.45
WCPT18	−1.17	1.62	2.09	0.05	6.89	2.26	−4.63
YBDE18	−0.70	2.65	3.01	0.05	9.96	3.14	−6.82
BH76RC	−0.48	2.01	2.57	0.09	9.35	3.54	−5.81

Table S30: Statistical analysis of the TPSS0-DH-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.69	7.69	9.67	0.23	19.63	20.92	1.29
ACONF	−0.11	0.12	0.13	0.07	0.25	0.02	−0.23
ADIM6	−0.47	0.47	0.52	0.14	0.63	−0.20	−0.82
AHB21	−0.98	1.02	1.39	0.05	3.62	0.32	−3.30
AL2X6	4.61	4.61	4.82	0.13	4.68	6.76	2.08
ALK8	10.04	10.10	14.24	0.16	32.12	31.90	−0.22
ALKBDE10	−7.01	7.01	8.18	0.07	15.46	−2.65	−18.11
AMINO20x4	0.06	0.23	0.29	0.09	1.53	1.00	−0.54
BH76	−1.99	2.60	3.37	0.14	14.08	4.83	−9.25
BHDIV10	−1.28	1.72	2.06	0.04	5.93	2.20	−3.73
BHPERI	−0.90	1.65	2.01	0.08	8.02	3.98	−4.04
BHROT27	0.55	0.57	0.77	0.09	1.67	1.56	−0.11
BSR36	−3.19	3.19	3.39	0.20	5.29	−1.46	−6.76
BUT14DIOL	−0.01	0.07	0.08	0.03	0.39	0.24	−0.15
C60ISO	3.12	3.53	4.00	0.04	8.01	7.09	−0.92
CARBHB12	0.97	0.97	1.19	0.16	2.00	2.30	0.30
CDIE20	0.94	0.95	1.16	0.23	2.51	2.40	−0.12
CHB6	−2.74	2.74	3.02	0.10	3.17	−1.08	−4.26
DARC	−4.02	4.02	4.30	0.12	5.55	−2.03	−7.57
DC13	−2.35	6.48	8.63	0.12	33.60	17.22	−16.38
DIPCS10	0.56	3.94	4.78	0.01	16.69	8.36	−8.33
FH51	−1.50	2.11	2.79	0.07	9.07	2.58	−6.49
G21EA	−1.75	3.38	4.11	0.10	12.19	4.53	−7.65
G21IP	0.49	3.25	4.12	0.01	16.22	9.99	−6.23
G2RC	−3.16	5.19	6.73	0.10	29.39	11.72	−17.67
HAL59	0.75	0.82	1.13	0.18	3.52	3.12	−0.40
HEAVY28	0.88	0.89	1.06	0.72	2.76	2.56	−0.19
HEAVYSB11	1.94	3.35	4.05	0.06	11.68	8.26	−3.42
ICONF	0.07	0.32	0.43	0.10	1.80	0.89	−0.90
IDISP	−0.15	1.45	1.68	0.10	4.55	1.59	−2.96
IL16	0.20	0.34	0.39	0.00	1.14	0.71	−0.43
INV24	0.72	1.59	2.67	0.05	11.98	8.04	−3.94
ISO34	−0.30	1.32	1.65	0.09	6.04	3.09	−2.95
ISOL24	0.55	1.73	2.30	0.08	9.75	5.24	−4.51
MB16-43	2.43	14.49	19.63	0.03	108.01	62.14	−45.87
MCONF	0.16	0.26	0.31	0.05	1.10	0.60	−0.51
NBPRC	−1.10	3.08	3.35	0.11	10.37	5.73	−4.64
PA26	4.09	4.09	4.53	0.02	8.17	9.52	1.36
PArel	0.76	0.98	1.33	0.21	4.71	3.66	−1.06
PCONF21	0.00	0.53	0.63	0.33	2.03	0.86	−1.17
PNICO23	1.02	1.05	1.30	0.25	3.63	3.37	−0.26
PX13	−1.72	1.73	1.94	0.05	2.97	0.07	−2.91
RC21	2.88	3.31	4.22	0.09	14.80	12.55	−2.24
RG18	0.05	0.12	0.21	0.21	0.95	0.74	−0.20
RSE43	−0.25	0.36	0.44	0.05	1.89	0.49	−1.41
S22	0.24	0.31	0.47	0.04	1.88	1.58	−0.30
S66	0.06	0.29	0.41	0.05	2.29	1.63	−0.67
SCONF	−0.03	0.16	0.26	0.03	1.06	0.21	−0.85
TAUT15	0.27	0.71	0.89	0.23	2.94	1.76	−1.18
UPU23	0.49	0.55	0.69	0.10	1.59	1.31	−0.28
W4-11	−7.99	11.13	13.98	0.04	60.63	22.67	−37.96
WATER27	0.95	1.44	1.90	0.02	9.50	5.24	−4.25
WCPT18	−0.37	1.61	1.87	0.05	7.02	4.17	−2.85
YBDE18	−1.75	3.48	4.02	0.07	12.10	3.24	−8.87
BH76RC	−1.25	3.59	4.38	0.19	17.37	8.21	−9.16

TPSS0-DH had convergence problems for the h2+_1.75 system in SIE4x4.

Table S31: Statistical analysis of the PBE-QIDH-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	3.45	3.45	3.93	0.10	6.95	8.04	1.09
ACONF	−0.07	0.12	0.14	0.07	0.45	0.15	−0.29
ADIM6	−0.73	0.73	0.80	0.22	0.91	−0.30	−1.21
AHB21	−1.14	1.15	1.55	0.05	3.78	0.14	−3.63
AL2X6	1.05	1.15	1.21	0.03	1.86	1.57	−0.29
ALK8	4.02	4.02	5.85	0.06	13.31	13.56	0.25
ALKBDE10	−4.42	4.82	5.56	0.05	11.85	1.99	−9.86
AMINO20x4	0.06	0.20	0.26	0.08	1.35	0.76	−0.59
BH76	0.38	1.22	1.99	0.07	12.97	10.43	−2.55
BHDIV10	−1.33	1.96	2.22	0.04	5.44	2.15	−3.29
BHPERI	−2.24	2.71	2.98	0.13	8.32	3.29	−5.02
BHROT27	0.51	0.51	0.67	0.08	1.37	1.32	−0.05
BSR36	−2.06	2.06	2.18	0.13	2.60	−1.12	−3.72
BUT14DIOL	0.00	0.06	0.08	0.02	0.36	0.20	−0.15
C60ISO	−2.16	3.47	4.87	0.04	13.29	2.87	−10.43
CARBHB12	0.82	0.82	1.04	0.14	1.94	2.12	0.18
CDIE20	0.77	0.81	0.97	0.20	2.11	1.97	−0.14
CHB6	−1.50	1.50	1.70	0.06	2.30	−0.51	−2.81
DARC	−6.90	6.90	7.07	0.21	4.87	−4.68	−9.55
DC13	−1.74	8.44	10.46	0.15	34.68	17.64	−17.04
DIPCS10	−0.86	2.74	3.26	0.00	9.28	3.18	−6.10
FH51	−2.46	2.84	3.50	0.09	13.62	5.17	−8.44
G21EA	−1.53	2.64	3.07	0.08	9.11	3.62	−5.49
G21IP	0.15	2.47	3.04	0.01	14.42	6.06	−8.35
G2RC	−3.98	5.33	6.78	0.10	26.25	7.79	−18.45
HAL59	0.19	0.38	0.59	0.08	2.59	2.10	−0.50
HEAVY28	0.11	0.17	0.22	0.14	0.73	0.48	−0.25
HEAVYSB11	0.20	0.68	0.87	0.01	3.06	1.85	−1.21
ICONF	0.20	0.23	0.36	0.07	1.01	0.92	−0.09
IDISP	−0.81	1.49	2.69	0.10	7.32	0.85	−6.47
IL16	−0.01	0.28	0.32	0.00	1.14	0.63	−0.50
INV24	0.58	1.35	1.97	0.04	9.93	6.24	−3.69
ISO34	0.23	1.35	1.94	0.09	7.80	4.10	−3.70
ISOL24	1.77	2.80	3.82	0.13	11.67	8.55	−3.13
MB16-43	12.12	14.67	17.86	0.04	67.12	48.86	−18.27
MCONF	0.18	0.22	0.27	0.04	0.81	0.53	−0.28
NBPRC	−1.09	2.62	3.08	0.09	10.39	4.79	−5.60
PA26	1.75	1.97	2.36	0.01	7.10	5.62	−1.48
PArel	0.81	1.05	1.36	0.23	4.71	2.97	−1.74
PCONF21	0.00	0.56	0.69	0.35	2.28	1.14	−1.14
PNICO23	0.40	0.43	0.68	0.10	2.51	2.41	−0.11
PX13	−2.76	2.76	2.94	0.08	3.78	−0.92	−4.70
RC21	2.22	3.01	4.30	0.08	16.65	13.37	−3.27
RG18	−0.11	0.16	0.22	0.28	0.92	0.44	−0.48
RSE43	0.35	0.50	0.90	0.07	4.45	3.80	−0.64
S22	0.04	0.28	0.42	0.04	2.05	1.29	−0.75
S66	−0.10	0.36	0.45	0.07	2.24	1.36	−0.88
SCONF	0.10	0.13	0.14	0.03	0.45	0.24	−0.20
TAUT15	0.48	0.73	1.03	0.24	2.97	2.18	−0.80
UPU23	0.44	0.52	0.65	0.09	1.58	1.35	−0.23
W4-11	−4.38	4.72	6.42	0.02	27.50	5.15	−22.35
WATER27	1.76	2.07	2.47	0.03	11.25	7.06	−4.18
WCPT18	−1.66	1.94	2.41	0.06	6.80	2.39	−4.41
YBDE18	1.95	2.76	3.45	0.06	9.14	6.24	−2.90
BH76RC	−0.55	1.91	2.46	0.10	10.66	5.29	−5.37

Table S32: Statistical analysis of the TPSS-QIDH-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	4.36	4.36	5.99	0.13	12.75	13.04	0.29
ACONF	−0.14	0.14	0.17	0.08	0.34	0.03	−0.32
ADIM6	−0.49	0.49	0.55	0.15	0.67	−0.19	−0.86
AHB21	−0.97	0.99	1.36	0.04	3.57	0.21	−3.36
AL2X6	1.48	1.48	1.63	0.04	2.22	2.37	0.15
ALK8	5.23	5.23	7.66	0.08	17.92	18.35	0.43
ALKBDE10	−4.22	4.22	5.18	0.04	11.14	−0.16	−11.29
AMINO20x4	0.03	0.19	0.24	0.08	1.18	0.70	−0.48
BH76	−0.44	2.02	2.74	0.11	17.75	12.29	−5.45
BHDIV10	−0.33	1.09	1.37	0.02	4.98	2.52	−2.46
BHPERI	−1.08	1.58	1.80	0.08	6.05	2.68	−3.37
BHROT27	0.47	0.48	0.65	0.08	1.42	1.35	−0.07
BSR36	−2.76	2.76	2.93	0.17	4.08	−1.46	−5.54
BUT14DIOL	−0.01	0.06	0.08	0.02	0.36	0.21	−0.15
C60ISO	−1.17	3.20	4.21	0.03	12.16	3.40	−8.76
CARBHB12	0.82	0.82	1.00	0.14	1.78	2.02	0.24
CDIE20	0.76	0.77	0.92	0.19	1.90	1.84	−0.05
CHB6	−1.77	1.77	1.91	0.07	2.02	−0.72	−2.74
DARC	−3.43	3.43	3.66	0.11	4.28	−1.58	−5.86
DC13	−1.03	5.05	6.62	0.09	25.59	11.76	−13.82
DIPCS10	2.24	3.81	4.69	0.01	13.24	8.51	−4.73
FH51	−1.84	2.09	2.57	0.07	8.30	1.74	−6.56
G21EA	−0.50	2.47	2.89	0.07	10.04	5.08	−4.97
G21IP	1.57	2.81	3.48	0.01	13.48	8.71	−4.77
G2RC	−3.77	4.65	5.77	0.09	19.72	6.42	−13.29
HAL59	0.36	0.46	0.66	0.10	2.60	2.17	−0.43
HEAVY28	0.41	0.43	0.51	0.35	1.39	1.15	−0.24
HEAVYSB11	0.12	1.75	1.98	0.03	6.67	3.34	−3.33
ICONF	0.10	0.25	0.34	0.08	1.24	0.78	−0.45
IDISP	−0.40	1.48	1.81	0.10	4.61	1.06	−3.55
IL16	0.19	0.30	0.36	0.00	1.07	0.72	−0.35
INV24	0.70	1.35	2.17	0.04	10.84	7.33	−3.51
ISO34	0.16	0.98	1.38	0.07	5.90	3.50	−2.40
ISOL24	0.85	1.56	2.17	0.07	8.87	5.28	−3.59
MB16-43	−4.04	14.50	19.04	0.03	95.76	34.15	−61.62
MCONF	0.24	0.29	0.33	0.06	0.90	0.60	−0.31
NBPRC	−0.28	2.70	3.06	0.10	8.50	5.59	−2.91
PA26	2.80	2.80	3.22	0.01	7.12	7.24	0.13
PArel	0.63	0.76	1.06	0.16	3.22	2.52	−0.70
PCONF21	0.05	0.44	0.52	0.27	1.74	0.84	−0.90
PNICO23	0.49	0.52	0.65	0.12	1.89	1.72	−0.17
PX13	−1.10	1.20	1.36	0.04	2.59	0.44	−2.15
RC21	1.18	1.99	3.02	0.06	13.26	9.97	−3.29
RG18	−0.01	0.13	0.22	0.22	1.08	0.76	−0.32
RSE43	0.43	0.52	0.95	0.07	4.45	3.86	−0.59
S22	0.22	0.28	0.40	0.04	1.50	1.30	−0.20
S66	0.05	0.28	0.37	0.05	2.00	1.36	−0.64
SCONF	−0.01	0.06	0.11	0.01	0.47	0.08	−0.39
TAUT15	0.31	0.56	0.77	0.18	2.53	1.49	−1.04
UPU23	0.43	0.50	0.63	0.09	1.42	1.20	−0.22
W4-11	−4.53	8.87	11.19	0.03	57.18	24.01	−33.17
WATER27	1.00	1.35	1.74	0.02	8.61	5.21	−3.41
WCPT18	−0.16	1.34	1.56	0.04	6.13	3.93	−2.20
YBDE18	−0.26	2.91	3.20	0.06	9.97	4.48	−5.49
BH76RC	−1.47	2.86	3.66	0.15	16.00	5.32	−10.68

TPSS-QIDH had convergence problems for the h2+_1.75 system in SIE4x4.

Table S33: Statistical analysis of the PBE-CIDH-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	6.41	6.41	7.27	0.19	12.86	15.03	2.17
ACONF	−0.06	0.08	0.10	0.04	0.31	0.08	−0.23
ADIM6	−0.40	0.40	0.44	0.12	0.52	−0.17	−0.70
AHB21	−1.28	1.29	1.68	0.06	3.96	0.08	−3.88
AL2X6	3.22	3.22	3.41	0.09	3.45	4.43	0.98
ALK8	8.39	8.54	12.34	0.14	29.18	28.57	−0.61
ALKBDE10	−5.62	5.76	6.85	0.06	14.54	0.71	−13.84
AMINO20x4	0.07	0.24	0.31	0.10	1.45	0.84	−0.61
BH76	−0.93	1.59	1.88	0.09	9.55	4.74	−4.81
BHDIV10	−2.02	2.58	2.94	0.06	7.06	2.38	−4.68
BHPERI	−1.91	2.52	2.84	0.12	8.98	3.86	−5.12
BHROT27	0.56	0.56	0.75	0.09	1.50	1.45	−0.05
BSR36	−2.49	2.49	2.67	0.15	3.68	−1.35	−5.03
BUT14DIOL	0.05	0.08	0.11	0.03	0.46	0.34	−0.12
C60ISO	1.12	2.84	3.13	0.03	8.52	4.20	−4.33
CARBHB12	1.09	1.09	1.32	0.18	2.12	2.48	0.36
CDIE20	0.90	0.94	1.15	0.23	2.66	2.39	−0.27
CHB6	−2.35	2.35	2.60	0.09	2.99	−0.89	−3.88
DARC	−6.99	6.99	7.21	0.22	6.17	−4.69	−10.86
DC13	−2.60	9.28	11.13	0.17	38.45	19.51	−18.94
DIPCS10	−0.60	2.65	3.28	0.00	10.88	4.04	−6.84
FH51	−2.38	2.97	3.70	0.10	14.31	4.97	−9.34
G21EA	−1.51	2.94	3.38	0.09	9.00	2.99	−6.01
G21IP	0.10	2.90	3.56	0.01	15.29	7.38	−7.90
G2RC	−4.08	6.31	7.95	0.12	33.07	11.49	−21.58
HAL59	0.68	0.73	1.02	0.16	3.29	2.95	−0.34
HEAVY28	0.82	0.83	0.98	0.67	2.39	2.26	−0.12
HEAVYSB11	1.82	2.14	2.89	0.04	7.63	6.65	−0.98
ICONF	0.17	0.29	0.41	0.09	1.49	1.07	−0.42
IDISP	−0.50	1.85	2.72	0.13	8.03	1.83	−6.20
IL16	−0.18	0.31	0.38	0.00	1.16	0.46	−0.71
INV24	0.50	1.51	2.22	0.05	10.10	5.94	−4.17
ISO34	0.08	1.42	1.96	0.10	7.85	4.05	−3.80
ISOL24	1.54	2.65	3.69	0.12	12.91	8.42	−4.49
MB16-43	19.53	21.01	25.30	0.05	84.61	72.61	−11.99
MCONF	0.22	0.27	0.33	0.05	1.02	0.66	−0.36
NBPRC	−1.35	3.21	3.69	0.12	11.68	5.58	−6.10
PA26	2.41	2.51	2.98	0.01	8.23	6.89	−1.34
PArel	0.87	1.17	1.54	0.25	5.66	3.83	−1.83
PCONF21	0.00	0.61	0.72	0.38	2.37	1.12	−1.25
PNICO23	0.92	0.93	1.22	0.22	3.78	3.69	−0.09
PX13	−3.47	3.47	3.66	0.10	4.34	−1.47	−5.81
RC21	3.69	4.23	5.39	0.12	17.46	15.25	−2.22
RG18	0.04	0.14	0.22	0.24	1.01	0.73	−0.28
RSE43	−0.32	0.38	0.52	0.05	2.43	0.93	−1.50
S22	0.27	0.34	0.50	0.05	1.97	1.61	−0.36
S66	0.12	0.30	0.43	0.05	2.25	1.71	−0.55
SCONF	0.03	0.10	0.15	0.02	0.69	0.20	−0.49
TAUT15	0.43	0.80	1.17	0.26	3.55	2.46	−1.09
UPU23	0.43	0.52	0.64	0.09	1.67	1.36	−0.32
W4-11	−4.95	5.43	7.17	0.02	33.97	8.24	−25.73
WATER27	2.59	2.95	3.54	0.04	14.77	10.01	−4.76
WCPT18	−2.03	2.27	2.88	0.06	7.64	2.13	−5.51
YBDE18	1.13	2.23	2.84	0.05	8.88	4.85	−4.04
BH76RC	−0.54	2.11	2.67	0.11	10.58	5.30	−5.28

Table S34: Statistical analysis of the TPSS-CIDH-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	6.79	6.79	8.70	0.20	17.94	18.91	0.97
ACONF	−0.12	0.12	0.14	0.07	0.27	0.02	−0.25
ADIM6	−0.50	0.50	0.55	0.15	0.66	−0.21	−0.86
AHB21	−0.99	1.03	1.40	0.05	3.67	0.29	−3.38
AL2X6	4.52	4.52	4.71	0.13	4.42	6.60	2.18
ALK8	10.00	10.13	14.46	0.16	33.63	33.09	−0.54
ALKBDE10	−6.36	6.36	7.46	0.06	14.27	−2.36	−16.63
AMINO20x4	0.05	0.22	0.28	0.09	1.45	0.93	−0.51
BH76	−1.62	2.41	3.15	0.13	14.89	6.61	−8.28
BHDIV10	−1.03	1.48	1.86	0.03	5.71	2.29	−3.42
BHPERI	−0.86	1.59	1.93	0.08	7.51	3.64	−3.87
BHROT27	0.54	0.55	0.74	0.09	1.63	1.54	−0.10
BSR36	−3.06	3.06	3.24	0.19	4.90	−1.45	−6.36
BUT14DIOL	−0.01	0.06	0.08	0.02	0.35	0.21	−0.14
C60ISO	2.24	3.29	3.55	0.03	8.54	5.98	−2.55
CARBHB12	0.94	0.94	1.15	0.16	1.96	2.25	0.28
CDIE20	0.90	0.91	1.10	0.22	2.39	2.28	−0.11
CHB6	−2.76	2.76	3.03	0.10	3.19	−1.10	−4.29
DARC	−3.98	3.98	4.24	0.12	5.24	−2.04	−7.28
DC13	−2.15	6.20	8.18	0.11	31.97	15.95	−16.02
DIPCS10	1.13	3.87	4.74	0.01	15.99	8.64	−7.35
FH51	−1.65	2.08	2.73	0.07	9.12	2.06	−7.06
G21EA	−1.44	3.15	3.79	0.09	11.28	4.55	−6.73
G21IP	0.81	3.08	3.91	0.01	15.15	9.74	−5.41
G2RC	−3.42	5.11	6.58	0.10	27.64	10.92	−16.73
HAL59	0.74	0.80	1.11	0.17	3.46	3.07	−0.38
HEAVY28	0.87	0.89	1.06	0.72	2.83	2.63	−0.20
HEAVYSB11	2.42	3.77	4.70	0.06	13.57	10.16	−3.42
ICONF	0.07	0.31	0.42	0.09	1.79	0.87	−0.92
IDISP	−0.29	1.54	1.83	0.11	4.82	1.49	−3.33
IL16	0.18	0.31	0.37	0.00	1.14	0.72	−0.42
INV24	0.74	1.55	2.59	0.05	11.93	8.04	−3.89
ISO34	−0.18	1.22	1.55	0.08	5.98	3.15	−2.82
ISOL24	0.65	1.69	2.28	0.08	9.32	5.32	−4.00
MB16-43	2.93	14.86	19.88	0.04	107.91	58.07	−49.84
MCONF	0.19	0.27	0.32	0.05	1.03	0.59	−0.45
NBPRC	−0.95	3.06	3.32	0.11	10.26	5.75	−4.52
PA26	3.78	3.78	4.22	0.02	7.83	9.01	1.18
PArel	0.74	0.94	1.28	0.20	4.34	3.40	−0.95
PCONF21	0.03	0.51	0.60	0.31	1.94	0.85	−1.09
PNICO23	0.96	0.99	1.23	0.23	3.40	3.15	−0.25
PX13	−1.49	1.53	1.72	0.05	2.84	0.23	−2.62
RC21	2.55	3.02	3.94	0.08	14.63	12.14	−2.49
RG18	0.05	0.13	0.23	0.22	1.03	0.78	−0.25
RSE43	−0.11	0.29	0.38	0.04	2.26	1.03	−1.23
S22	0.24	0.31	0.45	0.04	1.79	1.52	−0.27
S66	0.06	0.29	0.41	0.05	2.25	1.57	−0.68
SCONF	−0.03	0.12	0.21	0.03	0.88	0.16	−0.72
TAUT15	0.28	0.67	0.85	0.22	2.83	1.67	−1.16
UPU23	0.48	0.54	0.68	0.09	1.55	1.29	−0.26
W4-11	−7.18	10.71	13.41	0.03	60.52	23.37	−37.15
WATER27	0.87	1.38	1.81	0.02	8.95	4.94	−4.01
WCPT18	−0.26	1.55	1.78	0.04	6.84	4.20	−2.64
YBDE18	−1.36	3.36	3.83	0.07	11.97	3.74	−8.23
BH76RC	−1.33	3.45	4.23	0.19	17.34	7.53	−9.81

TPSS-CIDH had convergence problems for the h2+_1.75 system in SIE4x4.

Table S35: Statistical analysis of the SOS0-PBE0-2-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	1.22	1.85	2.30	0.05	6.77	5.20	−1.57
ACONF	−0.24	0.24	0.26	0.13	0.40	0.01	−0.39
ADIM6	−0.19	0.19	0.22	0.06	0.34	−0.04	−0.37
AHB21	−0.35	0.46	0.72	0.02	2.43	0.29	−2.14
AL2X6	−0.42	0.42	0.51	0.01	0.82	0.02	−0.80
ALK8	1.64	1.64	2.72	0.03	7.08	7.08	0.00
ALKBDE10	−5.11	5.11	5.49	0.05	6.40	−2.13	−8.53
AMINO20x4	−0.05	0.17	0.21	0.07	0.94	0.38	−0.56
BH76	2.30	2.43	3.63	0.13	18.81	17.16	−1.65
BHDIV10	0.77	0.90	1.13	0.02	2.12	1.76	−0.35
BHPERI	0.77	1.13	1.64	0.05	5.27	4.70	−0.58
BHROT27	0.23	0.26	0.33	0.04	0.85	0.71	−0.14
BSR36	−2.84	2.84	3.10	0.18	4.53	−1.46	−5.99
BUT14DIOL	−0.17	0.17	0.19	0.06	0.35	0.03	−0.32
C60ISO	−1.32	2.60	3.30	0.03	9.09	2.20	−6.89
CARBHB12	0.30	0.30	0.40	0.05	0.96	1.00	0.04
CDIE20	0.21	0.32	0.46	0.08	1.35	1.09	−0.26
CHB6	−1.00	1.05	1.24	0.04	2.01	0.12	−1.89
DARC	−4.99	4.99	5.02	0.15	1.83	−4.17	−6.00
DC13	−2.18	4.95	5.80	0.09	20.82	9.13	−11.69
DIPCS10	−1.96	3.10	3.60	0.00	10.50	2.89	−7.61
FH51	−1.55	1.82	2.37	0.06	11.19	2.37	−8.81
G21EA	−2.09	2.79	3.33	0.08	13.60	8.21	−5.39
G21IP	−0.12	1.95	2.81	0.01	16.37	10.82	−5.54
G2RC	−2.21	3.06	3.75	0.06	13.01	4.21	−8.81
HAL59	−0.24	0.32	0.39	0.07	1.60	0.69	−0.92
HEAVY28	−0.22	0.24	0.27	0.19	0.66	0.21	−0.44
HEAVYSB11	−0.46	1.01	1.35	0.02	4.03	1.13	−2.90
ICONF	0.05	0.18	0.23	0.06	0.82	0.52	−0.30
IDISP	−0.50	1.68	2.79	0.12	8.15	1.63	−6.52
IL16	0.54	0.55	0.63	0.01	1.25	1.20	−0.05
INV24	1.10	1.45	2.36	0.05	11.67	9.04	−2.63
ISO34	−0.02	0.83	1.19	0.06	5.54	3.11	−2.42
ISOL24	1.11	1.80	2.65	0.08	11.02	6.17	−4.85
MB16-43	−9.64	12.65	16.77	0.03	71.94	18.75	−53.19
MCONF	0.27	0.30	0.34	0.06	0.73	0.58	−0.15
NBPRC	−0.24	1.60	1.91	0.06	6.63	3.58	−3.05
PA26	3.13	3.13	3.36	0.02	5.05	6.10	1.05
PArel	0.35	0.43	0.64	0.09	2.18	1.56	−0.61
PCONF21	0.07	0.22	0.29	0.14	1.09	0.52	−0.58
PNICO23	−0.06	0.15	0.19	0.04	0.78	0.39	−0.39
PX13	0.02	0.52	0.60	0.02	1.81	1.10	−0.71
RC21	−0.47	1.96	2.67	0.05	11.11	6.36	−4.75
RG18	−0.10	0.17	0.23	0.29	1.03	0.54	−0.49
RSE43	1.55	1.55	2.45	0.20	10.33	10.58	0.25
S22	0.07	0.19	0.23	0.03	0.81	0.40	−0.41
S66	0.01	0.13	0.16	0.02	0.78	0.48	−0.31
SCONF	−0.14	0.17	0.20	0.04	0.42	0.11	−0.30
TAUT15	−0.32	0.54	0.58	0.18	1.41	0.55	−0.86
UPU23	0.43	0.50	0.62	0.09	1.49	1.16	−0.33
W4-11	−3.16	4.62	6.87	0.02	39.42	7.92	−31.50
WATER27	−0.47	0.89	1.39	0.01	5.29	0.91	−4.38
WCPT18	0.93	1.04	1.37	0.03	3.83	3.52	−0.31
YBDE18	1.91	2.04	2.40	0.04	5.27	4.24	−1.03
BH76RC	−0.44	1.45	1.93	0.08	7.86	2.95	−4.91

Table S36: Statistical analysis of the SOS0-PBE0-DH-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.35	7.35	8.48	0.22	15.68	18.07	2.39
ACONF	−0.08	0.09	0.10	0.05	0.25	0.03	−0.22
ADIM6	−0.29	0.29	0.33	0.09	0.45	−0.11	−0.56
AHB21	−1.11	1.13	1.49	0.05	3.53	0.14	−3.40
AL2X6	4.07	4.07	4.27	0.11	3.97	5.70	1.73
ALK8	9.78	10.03	13.90	0.16	31.70	30.67	−1.03
ALKBDE10	−6.64	6.64	7.72	0.07	14.75	−1.29	−16.05
AMINO20x4	0.05	0.23	0.30	0.09	1.52	0.89	−0.63
BH76	−1.04	1.64	1.89	0.09	7.60	3.84	−3.76
BHDIV10	−1.76	2.21	2.64	0.05	6.52	2.00	−4.51
BHPERI	−0.75	1.73	2.14	0.08	9.07	5.30	−3.76
BHROT27	0.51	0.51	0.69	0.08	1.38	1.34	−0.05
BSR36	−3.02	3.02	3.29	0.19	5.00	−1.63	−6.63
BUT14DIOL	0.01	0.08	0.11	0.03	0.49	0.34	−0.15
C60ISO	3.59	3.59	4.18	0.04	7.31	7.75	0.45
CARBHB12	1.03	1.03	1.24	0.17	1.96	2.33	0.37
CDIE20	0.79	0.85	1.09	0.21	2.76	2.37	−0.39
CHB6	−2.78	2.78	3.07	0.10	3.32	−1.10	−4.43
DARC	−6.47	6.47	6.68	0.20	6.07	−4.52	−10.59
DC13	−3.23	8.81	10.48	0.16	35.55	18.60	−16.95
DIPCS10	−0.78	2.73	3.52	0.00	12.24	4.53	−7.71
FH51	−2.07	2.76	3.57	0.09	13.74	3.95	−9.79
G21EA	−1.76	2.72	3.37	0.08	9.49	2.43	−7.06
G21IP	−0.05	2.90	3.53	0.01	14.41	7.56	−6.85
G2RC	−3.65	6.25	7.89	0.12	33.39	12.71	−20.68
HAL59	0.75	0.79	1.08	0.17	3.21	2.89	−0.32
HEAVY28	0.88	0.89	1.05	0.72	2.64	2.51	−0.13
HEAVYSB11	2.53	2.89	3.90	0.05	9.82	8.42	−1.40
ICONF	0.10	0.29	0.41	0.09	1.56	1.01	−0.55
IDISP	−0.28	1.89	2.55	0.13	7.81	2.38	−5.43
IL16	−0.02	0.31	0.35	0.00	1.16	0.58	−0.58
INV24	0.61	1.59	2.48	0.05	11.39	7.17	−4.21
ISO34	−0.08	1.32	1.78	0.09	7.25	3.76	−3.49
ISOL24	1.22	2.35	3.26	0.11	12.20	7.55	−4.64
MB16-43	18.08	20.31	24.89	0.05	87.37	74.15	−13.22
MCONF	0.16	0.22	0.28	0.04	1.00	0.60	−0.40
NBPRC	−1.21	3.25	3.66	0.12	11.27	5.69	−5.57
PA26	3.16	3.22	3.64	0.02	8.55	7.69	−0.87
PArel	0.78	1.05	1.44	0.23	5.73	4.15	−1.57
PCONF21	0.01	0.55	0.66	0.34	2.21	1.02	−1.19
PNICO23	1.02	1.04	1.32	0.24	3.88	3.75	−0.13
PX13	−3.00	3.00	3.19	0.09	3.95	−1.12	−5.07
RC21	3.65	4.19	5.18	0.12	16.00	14.18	−1.82
RG18	0.08	0.13	0.22	0.22	0.95	0.78	−0.17
RSE43	−0.35	0.38	0.51	0.05	1.92	0.29	−1.63
S22	0.25	0.32	0.45	0.04	1.83	1.48	−0.35
S66	0.14	0.27	0.38	0.05	2.01	1.57	−0.44
SCONF	−0.09	0.13	0.18	0.03	0.66	0.13	−0.54
TAUT15	0.17	0.73	0.97	0.24	3.09	1.90	−1.18
UPU23	0.50	0.58	0.71	0.10	1.78	1.46	−0.32
W4-11	−5.24	5.89	7.91	0.02	39.20	11.27	−27.93
WATER27	2.07	2.38	2.85	0.03	12.33	8.15	−4.18
WCPT18	−1.40	1.89	2.40	0.05	7.02	2.30	−4.72
YBDE18	0.70	1.87	2.38	0.04	8.07	3.82	−4.25
BH76RC	−0.51	2.10	2.61	0.11	8.40	3.48	−4.92

Table S37: Statistical analysis of the SOS1-PBE-QIDH-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	3.00	3.06	3.97	0.09	9.70	9.23	−0.47
ACONF	−0.10	0.12	0.14	0.07	0.37	0.07	−0.30
ADIM6	−0.69	0.69	0.76	0.21	0.94	−0.25	−1.19
AHB21	−0.64	0.70	1.07	0.03	3.33	0.39	−2.94
AL2X6	1.30	1.30	1.39	0.04	1.47	1.84	0.36
ALK8	4.73	4.73	6.60	0.08	14.86	14.93	0.06
ALKBDE10	−6.37	6.37	6.93	0.06	10.64	−2.21	−12.86
AMINO20x4	−0.01	0.18	0.23	0.07	1.15	0.55	−0.60
BH76	1.20	1.53	2.37	0.08	14.03	12.06	−1.98
BHDIV10	0.03	1.11	1.16	0.02	3.19	1.38	−1.81
BHPERI	0.78	1.21	1.78	0.06	6.65	5.65	−1.00
BHROT27	0.36	0.37	0.49	0.06	1.14	1.06	−0.09
BSR36	−3.30	3.30	3.57	0.20	5.33	−1.76	−7.09
BUT14DIOL	−0.14	0.16	0.18	0.06	0.44	0.14	−0.30
C60ISO	1.41	2.45	2.83	0.02	7.99	5.47	−2.53
CARBHB12	0.49	0.49	0.64	0.08	1.44	1.46	0.02
CDIE20	0.45	0.54	0.73	0.13	2.01	1.68	−0.34
CHB6	−1.82	1.82	2.04	0.07	2.57	−0.64	−3.20
DARC	−5.69	5.69	5.77	0.18	3.63	−4.42	−8.06
DC13	−2.75	6.68	7.91	0.12	26.47	12.24	−14.24
DIPCS10	−1.18	2.88	3.51	0.00	11.94	4.32	−7.63
FH51	−1.85	2.23	2.85	0.07	12.29	2.74	−9.55
G21EA	−2.14	2.58	3.21	0.08	11.22	4.52	−6.71
G21IP	−0.03	2.04	2.71	0.01	11.67	5.65	−6.02
G2RC	−3.01	4.46	5.58	0.09	21.96	8.11	−13.85
HAL59	0.04	0.34	0.45	0.07	2.08	1.46	−0.62
HEAVY28	0.12	0.18	0.23	0.15	0.91	0.60	−0.32
HEAVYSB11	0.39	1.43	1.66	0.02	5.34	2.85	−2.49
ICONF	0.08	0.22	0.30	0.07	1.07	0.71	−0.36
IDISP	−0.55	1.76	2.49	0.12	7.52	2.02	−5.50
IL16	0.53	0.53	0.62	0.00	1.15	1.13	−0.02
INV24	0.95	1.51	2.56	0.05	12.49	8.90	−3.59
ISO34	−0.04	1.05	1.49	0.07	6.49	3.53	−2.96
ISOL24	1.14	2.03	2.90	0.09	11.60	6.77	−4.83
MB16-43	1.14	10.72	13.82	0.03	74.02	35.07	−38.95
MCONF	0.07	0.11	0.14	0.02	0.62	0.36	−0.26
NBPRC	−0.52	2.18	2.50	0.08	8.44	4.69	−3.75
PA26	3.16	3.16	3.50	0.02	6.09	6.81	0.72
PArel	0.58	0.73	1.02	0.16	3.66	2.70	−0.96
PCONF21	0.05	0.41	0.53	0.25	1.87	1.00	−0.87
PNICO23	0.26	0.31	0.44	0.07	1.60	1.34	−0.26
PX13	−0.82	1.00	1.12	0.03	2.29	0.59	−1.70
RC21	1.39	2.22	3.20	0.06	13.61	10.05	−3.56
RG18	−0.09	0.15	0.21	0.26	0.94	0.52	−0.42
RSE43	0.69	0.72	1.13	0.09	5.04	4.66	−0.38
S22	−0.14	0.27	0.38	0.04	1.70	0.72	−0.98
S66	−0.21	0.30	0.39	0.05	1.67	0.78	−0.89
SCONF	−0.16	0.21	0.24	0.05	0.72	0.35	−0.37
TAUT15	−0.10	0.46	0.51	0.15	1.42	0.52	−0.90
UPU23	0.58	0.61	0.77	0.11	1.61	1.47	−0.14
W4-11	−4.97	5.74	7.99	0.02	35.85	7.21	−28.64
WATER27	−0.45	1.20	1.81	0.01	7.11	1.38	−5.73
WCPT18	0.35	1.16	1.36	0.03	5.00	3.53	−1.47
YBDE18	1.17	1.92	2.39	0.04	7.92	4.48	−3.44
BH76RC	−0.54	1.71	2.24	0.09	8.85	3.21	−5.64

Table S38: Statistical analysis of the SOS0-PBE-CIDH-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	6.11	6.11	7.17	0.18	13.86	15.64	1.77
ACONF	−0.10	0.10	0.12	0.05	0.28	0.03	−0.24
ADIM6	−0.32	0.32	0.36	0.10	0.48	−0.12	−0.60
AHB21	−1.04	1.06	1.43	0.05	3.55	0.16	−3.39
AL2X6	3.84	3.84	4.03	0.11	3.67	5.42	1.75
ALK8	9.29	9.51	13.29	0.15	30.68	29.79	−0.89
ALKBDE10	−6.83	6.83	7.76	0.07	14.14	−1.71	−15.85
AMINO20x4	0.04	0.22	0.28	0.09	1.44	0.84	−0.60
BH76	−0.45	1.31	1.66	0.07	9.05	5.76	−3.29
BHDIV10	−1.29	1.82	2.19	0.04	5.66	1.80	−3.86
BHPERI	−0.35	1.61	2.01	0.08	8.85	5.66	−3.19
BHROT27	0.48	0.49	0.66	0.08	1.36	1.31	−0.05
BSR36	−2.98	2.98	3.22	0.18	4.81	−1.61	−6.42
BUT14DIOL	−0.02	0.08	0.10	0.03	0.44	0.26	−0.18
C60ISO	3.44	3.44	4.08	0.04	7.46	7.75	0.29
CARBHB12	0.93	0.93	1.12	0.15	1.83	2.15	0.32
CDIE20	0.70	0.77	1.00	0.19	2.61	2.23	−0.39
CHB6	−2.71	2.71	3.00	0.10	3.24	−1.06	−4.30
DARC	−6.63	6.63	6.80	0.20	5.46	−4.86	−10.32
DC13	−3.27	8.62	10.15	0.16	32.74	17.38	−15.36
DIPCS10	−0.77	2.78	3.53	0.00	12.53	4.81	−7.72
FH51	−2.12	2.70	3.49	0.09	14.03	3.87	−10.16
G21EA	−1.92	2.66	3.39	0.08	9.41	2.15	−7.27
G21IP	−0.03	2.70	3.32	0.01	13.76	7.16	−6.61
G2RC	−3.62	5.98	7.51	0.12	31.53	11.99	−19.54
HAL59	0.66	0.72	0.99	0.16	3.03	2.67	−0.36
HEAVY28	0.82	0.83	0.98	0.67	2.54	2.37	−0.16
HEAVYSB11	2.38	2.91	3.80	0.05	10.02	8.26	−1.76
ICONF	0.09	0.29	0.40	0.09	1.57	0.96	−0.61
IDISP	−0.42	1.94	2.76	0.14	8.28	2.12	−6.16
IL16	0.04	0.31	0.34	0.00	1.15	0.63	−0.52
INV24	0.76	1.62	2.59	0.05	11.78	7.57	−4.21
ISO34	−0.06	1.28	1.74	0.09	7.12	3.82	−3.30
ISOL24	1.31	2.38	3.36	0.11	12.25	7.67	−4.58
MB16-43	16.07	18.56	22.97	0.04	84.36	68.22	−16.14
MCONF	0.19	0.24	0.29	0.05	0.96	0.61	−0.36
NBPRC	−1.13	3.10	3.52	0.11	11.07	5.61	−5.46
PA26	3.20	3.24	3.65	0.02	8.10	7.54	−0.56
PArel	0.76	0.99	1.36	0.21	5.27	3.83	−1.43
PCONF21	0.04	0.49	0.58	0.30	2.01	0.93	−1.08
PNICO23	0.92	0.94	1.18	0.22	3.45	3.29	−0.16
PX13	−2.39	2.39	2.60	0.07	3.53	−0.63	−4.16
RC21	3.25	3.81	4.79	0.11	15.53	13.67	−1.86
RG18	0.06	0.13	0.22	0.22	1.02	0.79	−0.23
RSE43	−0.11	0.25	0.37	0.03	2.10	0.77	−1.33
S22	0.24	0.29	0.41	0.04	1.64	1.37	−0.26
S66	0.12	0.26	0.35	0.05	1.91	1.45	−0.46
SCONF	−0.11	0.14	0.17	0.03	0.50	0.10	−0.41
TAUT15	0.11	0.66	0.84	0.22	2.63	1.55	−1.08
UPU23	0.50	0.57	0.70	0.10	1.77	1.42	−0.35
W4-11	−5.56	6.19	8.30	0.02	37.82	10.62	−27.20
WATER27	1.56	1.83	2.21	0.02	9.99	6.33	−3.66
WCPT18	−0.91	1.65	2.04	0.05	6.59	2.78	−3.81
YBDE18	0.86	1.99	2.54	0.04	8.76	4.32	−4.44
BH76RC	−0.55	2.04	2.55	0.11	7.89	2.85	−5.04

Table S39: Statistical analysis of the SOS0-PBE0-2(b)-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	4.47	4.47	4.95	0.13	7.45	8.55	1.09
ACONF	−0.32	0.32	0.34	0.17	0.42	−0.02	−0.44
ADIM6	−0.31	0.31	0.33	0.09	0.36	−0.13	−0.48
AHB21	−0.48	0.52	0.67	0.02	2.31	0.23	−2.08
AL2X6	0.31	0.83	0.95	0.02	2.81	1.27	−1.55
ALK8	2.59	3.61	6.07	0.06	18.92	15.42	−3.49
ALKBDE10	9.86	9.86	11.33	0.10	14.87	18.69	3.82
AMINO20x4	−0.07	0.14	0.18	0.06	0.74	0.28	−0.46
BH76	0.54	2.14	3.87	0.11	25.51	21.03	−4.48
BHDIV10	−2.06	2.53	2.87	0.06	7.21	2.32	−4.90
BHPERI	−3.13	3.27	3.54	0.16	7.31	1.93	−5.39
BHROT27	−0.01	0.13	0.17	0.02	0.73	0.23	−0.50
BSR36	−1.29	1.29	1.35	0.08	1.48	−0.69	−2.18
BUT14DIOL	−0.02	0.06	0.07	0.02	0.29	0.18	−0.12
C60ISO	−13.13	13.13	15.47	0.13	23.79	−3.36	−27.16
CARBHB12	0.50	0.50	0.64	0.08	1.27	1.32	0.05
CDIE20	0.28	0.31	0.38	0.08	1.12	0.98	−0.14
CHB6	−0.98	0.98	1.16	0.04	1.88	−0.31	−2.19
DARC	−5.34	5.34	5.40	0.16	2.44	−4.01	−6.45
DC13	−2.62	9.29	12.07	0.17	48.20	21.17	−27.02
DIPCS10	6.79	6.79	7.24	0.01	7.48	10.69	3.21
FH51	−0.78	1.54	2.21	0.05	12.85	5.99	−6.85
G21EA	4.53	5.41	6.53	0.16	20.55	16.50	−4.05
G21IP	5.18	5.72	7.37	0.02	32.47	24.24	−8.23
G2RC	−0.60	2.49	3.36	0.05	15.92	7.12	−8.81
HAL59	0.14	0.31	0.47	0.07	2.32	1.87	−0.44
HEAVY28	−0.09	0.13	0.16	0.10	0.54	0.20	−0.34
HEAVYSB11	6.80	6.80	6.84	0.12	3.12	7.91	4.80
ICONF	−0.03	0.17	0.24	0.05	1.07	0.39	−0.68
IDISP	−0.64	3.49	4.37	0.25	13.71	5.03	−8.68
IL16	−0.28	0.35	0.41	0.00	1.23	0.35	−0.88
INV24	−0.13	0.80	1.60	0.03	8.84	1.63	−7.21
ISO34	−0.28	1.03	1.53	0.07	7.23	3.28	−3.95
ISOL24	1.11	2.24	3.12	0.10	12.40	8.78	−3.62
MB16-43	9.88	12.57	15.18	0.03	50.13	32.19	−17.94
MCONF	0.36	0.40	0.46	0.08	1.09	0.86	−0.24
NBPRC	−1.06	2.10	2.55	0.08	8.11	2.31	−5.79
PA26	1.71	1.76	1.88	0.01	3.70	3.12	−0.58
PArel	−0.02	0.72	1.14	0.16	4.86	1.86	−2.99
PCONF21	−0.14	0.36	0.44	0.22	1.53	0.58	−0.95
PNICO23	0.09	0.24	0.38	0.06	1.97	1.53	−0.44
PX13	−4.21	4.21	4.33	0.13	4.20	−2.18	−6.37
RC21	−0.89	2.65	3.30	0.07	13.17	8.11	−5.06
RG18	−0.15	0.19	0.26	0.33	0.92	0.33	−0.59
RSE43	1.54	1.58	3.47	0.21	16.06	15.64	−0.42
S22	0.00	0.25	0.36	0.03	1.61	0.67	−0.95
S66	−0.08	0.19	0.24	0.03	1.00	0.33	−0.67
SCONF	0.14	0.30	0.34	0.07	1.13	0.40	−0.73
TAUT15	−0.29	0.65	0.81	0.21	2.48	0.88	−1.60
UPU23	0.24	0.35	0.45	0.06	1.53	1.07	−0.46
W4-11	28.46	28.52	32.80	0.09	71.73	68.92	−2.82
WATER27	1.09	1.45	1.91	0.02	10.55	5.64	−4.91
WCPT18	−2.75	2.75	2.86	0.08	2.84	−1.82	−4.66
YBDE18	9.32	9.32	9.93	0.19	11.54	16.89	5.35
BH76RC	−0.24	1.85	2.51	0.10	12.15	8.88	−3.27

Table S40: Statistical analysis of the SOS0-PBE0-DH(b)-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	8.43	8.43	9.50	0.25	16.72	19.10	2.38
ACONF	−0.10	0.10	0.12	0.05	0.26	0.03	−0.23
ADIM6	−0.32	0.32	0.36	0.10	0.46	−0.13	−0.59
AHB21	−1.13	1.14	1.44	0.05	3.55	0.10	−3.45
AL2X6	3.52	3.52	3.76	0.10	3.96	4.81	0.85
ALK8	8.80	9.01	12.88	0.14	29.97	29.13	−0.84
ALKBDE10	−2.26	3.41	4.08	0.03	12.44	5.09	−7.35
AMINO20x4	0.05	0.21	0.28	0.09	1.37	0.81	−0.56
BH76	−1.87	2.30	2.54	0.12	10.53	3.54	−6.99
BHDIV10	−2.64	2.99	3.42	0.07	6.65	1.40	−5.25
BHPERI	−1.79	2.31	2.62	0.11	8.49	3.76	−4.73
BHROT27	0.42	0.44	0.58	0.07	1.16	1.10	−0.06
BSR36	−2.74	2.74	2.97	0.17	4.44	−1.48	−5.92
BUT14DIOL	0.07	0.09	0.12	0.03	0.47	0.38	−0.09
C60ISO	−0.46	2.39	2.94	0.02	8.57	2.47	−6.11
CARBHB12	1.08	1.08	1.30	0.18	2.09	2.47	0.38
CDIE20	0.80	0.84	1.05	0.21	2.47	2.16	−0.31
CHB6	−2.40	2.40	2.66	0.09	2.94	−0.93	−3.87
DARC	−6.27	6.27	6.47	0.19	5.64	−4.25	−9.89
DC13	−3.07	8.89	10.88	0.16	36.73	17.13	−19.60
DIPCS10	1.59	2.52	2.85	0.00	7.55	4.31	−3.24
FH51	−1.76	2.58	3.39	0.08	13.96	4.81	−9.14
G21EA	0.11	1.45	1.69	0.04	5.65	2.70	−2.95
G21IP	1.34	2.45	2.96	0.01	12.59	7.85	−4.74
G2RC	−2.99	5.74	7.27	0.11	29.74	10.79	−18.95
HAL59	0.76	0.79	1.09	0.17	3.33	3.05	−0.28
HEAVY28	0.80	0.81	0.95	0.65	2.32	2.21	−0.11
HEAVYSB11	3.65	3.65	4.16	0.06	5.97	7.35	1.39
ICONF	0.10	0.24	0.34	0.07	1.30	0.83	−0.48
IDISP	−0.24	1.62	2.46	0.11	7.27	1.67	−5.60
IL16	−0.28	0.34	0.42	0.00	1.15	0.28	−0.87
INV24	0.22	1.19	1.65	0.04	7.28	4.33	−2.94
ISO34	−0.17	1.20	1.72	0.08	7.19	3.69	−3.50
ISOL24	1.10	2.22	3.10	0.10	11.95	7.36	−4.58
MB16-43	19.18	20.37	24.90	0.05	79.21	68.48	−10.73
MCONF	0.20	0.26	0.33	0.05	1.12	0.72	−0.40
NBPRC	−1.37	3.30	3.69	0.12	11.07	4.93	−6.13
PA26	2.79	2.86	3.21	0.02	7.86	6.96	−0.89
PArel	0.66	1.00	1.37	0.22	6.09	3.92	−2.17
PCONF21	−0.07	0.61	0.71	0.38	2.25	0.94	−1.31
PNICO23	0.97	0.98	1.29	0.23	4.02	3.95	−0.07
PX13	−4.38	4.38	4.55	0.13	4.76	−2.23	−7.00
RC21	3.72	4.22	5.10	0.12	14.85	12.56	−2.29
RG18	0.06	0.12	0.20	0.21	0.88	0.70	−0.18
RSE43	−0.50	0.53	0.66	0.07	2.33	0.58	−1.76
S22	0.23	0.31	0.43	0.04	1.77	1.36	−0.40
S66	0.12	0.27	0.36	0.05	1.93	1.46	−0.47
SCONF	0.01	0.16	0.24	0.03	1.08	0.26	−0.82
TAUT15	0.13	0.86	1.07	0.28	3.42	2.07	−1.34
UPU23	0.44	0.52	0.64	0.09	1.71	1.42	−0.29
W4-11	3.93	5.63	7.27	0.02	37.87	22.86	−15.01
WATER27	2.71	3.11	3.77	0.04	15.95	10.61	−5.34
WCPT18	−2.65	2.68	3.24	0.08	6.44	0.20	−6.24
YBDE18	2.56	2.58	2.78	0.05	4.06	3.82	−0.24
BH76RC	−0.41	1.94	2.58	0.10	11.33	6.49	−4.84

Table S41: Statistical analysis of the SOS0-PBE-QIDH(b)-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	5.43	5.43	6.07	0.16	10.09	11.64	1.55
ACONF	−0.20	0.21	0.22	0.11	0.36	0.01	−0.35
ADIM6	−0.43	0.43	0.46	0.13	0.50	−0.19	−0.69
AHB21	−0.78	0.80	1.02	0.04	2.83	0.18	−2.64
AL2X6	1.05	1.34	1.39	0.04	2.86	1.98	−0.89
ALK8	3.79	3.79	6.21	0.06	14.78	14.92	0.14
ALKBDE10	4.05	4.05	5.42	0.04	11.32	11.34	0.02
AMINO20x4	−0.01	0.15	0.18	0.06	0.81	0.49	−0.32
BH76	−0.35	1.75	2.55	0.09	15.44	11.39	−4.06
BHDIV10	−2.01	2.31	2.70	0.05	6.13	1.51	−4.62
BHPERI	−2.11	2.49	2.68	0.12	6.90	2.77	−4.13
BHROT27	0.18	0.20	0.26	0.03	0.67	0.61	−0.06
BSR36	−2.08	2.08	2.20	0.13	2.72	−1.23	−3.95
BUT14DIOL	0.00	0.06	0.07	0.02	0.25	0.14	−0.12
C60ISO	−7.31	7.31	9.21	0.07	16.85	−0.64	−17.49
CARBHB12	0.71	0.71	0.87	0.12	1.54	1.71	0.17
CDIE20	0.47	0.51	0.62	0.13	1.44	1.26	−0.17
CHB6	−1.30	1.30	1.48	0.05	1.89	−0.42	−2.31
DARC	−6.12	6.12	6.19	0.19	2.72	−4.63	−7.34
DC13	−2.89	8.17	10.87	0.15	39.20	18.75	−20.45
DIPCS10	4.81	4.81	5.09	0.01	5.68	7.40	1.72
FH51	−1.34	1.96	2.67	0.06	14.21	5.55	−8.66
G21EA	2.43	2.74	3.32	0.08	10.93	8.92	−2.01
G21IP	3.55	3.78	4.55	0.01	16.71	13.70	−3.01
G2RC	−1.82	3.57	4.72	0.07	17.69	6.41	−11.28
HAL59	0.26	0.38	0.58	0.08	2.50	2.10	−0.40
HEAVY28	0.11	0.15	0.18	0.12	0.60	0.36	−0.24
HEAVYSB11	4.35	4.35	4.38	0.07	1.76	5.20	3.44
ICONF	0.05	0.13	0.18	0.04	0.64	0.33	−0.31
IDISP	−0.65	2.26	3.56	0.16	10.71	2.56	−8.15
IL16	−0.27	0.32	0.38	0.00	1.00	0.15	−0.85
INV24	0.14	0.78	1.06	0.02	5.27	3.48	−1.79
ISO34	−0.22	1.05	1.57	0.07	7.19	3.62	−3.56
ISOL24	1.18	2.28	3.21	0.10	12.14	8.12	−4.02
MB16-43	10.02	12.36	14.74	0.03	42.26	29.49	−12.77
MCONF	0.32	0.36	0.42	0.07	1.07	0.81	−0.25
NBPRC	−1.17	2.57	2.99	0.09	9.24	3.17	−6.06
PA26	2.28	2.28	2.48	0.01	4.35	4.90	0.55
PArel	0.31	0.77	1.02	0.17	4.50	1.94	−2.55
PCONF21	−0.09	0.43	0.50	0.27	1.57	0.49	−1.08
PNICO23	0.35	0.37	0.59	0.09	2.29	2.17	−0.13
PX13	−3.99	3.99	4.13	0.12	4.30	−1.92	−6.22
RC21	1.44	2.45	3.21	0.07	12.03	8.62	−3.41
RG18	−0.08	0.14	0.21	0.24	0.91	0.51	−0.40
RSE43	0.57	0.74	1.59	0.10	7.61	6.82	−0.79
S22	0.10	0.22	0.27	0.03	1.06	0.59	−0.46
S66	−0.02	0.20	0.25	0.04	1.22	0.69	−0.53
SCONF	0.09	0.22	0.26	0.05	0.99	0.32	−0.67
TAUT15	−0.13	0.70	0.77	0.23	2.06	0.88	−1.18
UPU23	0.30	0.41	0.50	0.07	1.49	1.17	−0.32
W4-11	17.20	17.35	20.50	0.06	55.43	48.22	−7.21
WATER27	1.50	1.86	2.29	0.02	11.52	6.60	−4.91
WCPT18	−2.47	2.47	2.73	0.07	3.86	−1.01	−4.87
YBDE18	6.26	6.26	6.48	0.13	5.87	9.33	3.45
BH76RC	−0.38	1.54	2.19	0.08	11.10	7.15	−3.95

Table S42: Statistical analysis of the SOS0-PBE-CIDH(b)-D3(BJ) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.49	7.49	8.44	0.22	14.80	16.97	2.17
ACONF	−0.12	0.12	0.13	0.07	0.28	0.03	−0.25
ADIM6	−0.38	0.38	0.42	0.11	0.51	−0.16	−0.66
AHB21	−1.07	1.08	1.37	0.05	3.43	0.13	−3.30
AL2X6	3.14	3.14	3.37	0.09	3.66	4.31	0.64
ALK8	8.09	8.25	12.12	0.13	28.87	28.20	−0.66
ALKBDE10	−1.21	2.81	3.34	0.03	10.70	5.86	−4.84
AMINO20x4	0.04	0.20	0.25	0.08	1.21	0.73	−0.48
BH76	−1.45	2.02	2.27	0.11	9.83	3.81	−6.02
BHDIV10	−2.39	2.73	3.14	0.06	5.99	1.05	−4.93
BHPERI	−1.69	2.22	2.51	0.11	8.12	3.70	−4.42
BHROT27	0.38	0.39	0.51	0.06	1.06	1.00	−0.06
BSR36	−2.65	2.65	2.86	0.16	4.13	−1.45	−5.59
BUT14DIOL	0.04	0.08	0.10	0.03	0.42	0.32	−0.10
C60ISO	−1.67	2.66	3.72	0.03	10.03	1.92	−8.11
CARBHB12	0.99	0.99	1.20	0.16	1.93	2.27	0.34
CDIE20	0.72	0.76	0.95	0.19	2.25	1.96	−0.29
CHB6	−2.23	2.23	2.48	0.08	2.76	−0.84	−3.59
DARC	−6.40	6.40	6.56	0.20	4.95	−4.53	−9.48
DC13	−3.09	8.78	10.83	0.16	34.31	15.77	−18.55
DIPCS10	2.35	2.73	3.15	0.00	6.78	4.85	−1.93
FH51	−1.73	2.47	3.26	0.08	14.36	5.00	−9.36
G21EA	0.51	1.21	1.46	0.04	5.63	3.69	−1.94
G21IP	1.81	2.36	2.92	0.01	11.46	7.55	−3.91
G2RC	−2.82	5.32	6.75	0.10	26.96	9.54	−17.42
HAL59	0.67	0.71	0.99	0.15	3.19	2.87	−0.31
HEAVY28	0.74	0.75	0.88	0.60	2.18	2.04	−0.14
HEAVYSB11	3.86	3.86	4.23	0.07	5.21	7.06	1.85
ICONF	0.08	0.22	0.31	0.07	1.19	0.73	−0.47
IDISP	−0.39	1.75	2.73	0.12	7.68	1.27	−6.40
IL16	−0.27	0.32	0.40	0.00	1.13	0.27	−0.86
INV24	0.25	1.11	1.53	0.03	7.11	4.46	−2.64
ISO34	−0.18	1.16	1.69	0.08	7.24	3.74	−3.50
ISOL24	1.16	2.25	3.18	0.10	11.34	7.48	−3.87
MB16-43	17.63	18.86	22.93	0.05	71.33	60.96	−10.37
MCONF	0.22	0.28	0.35	0.06	1.09	0.73	−0.36
NBPRC	−1.34	3.17	3.57	0.11	10.86	4.67	−6.19
PA26	2.73	2.77	3.09	0.01	7.16	6.58	−0.58
PArel	0.60	0.93	1.27	0.20	5.73	3.51	−2.23
PCONF21	−0.06	0.57	0.66	0.35	2.10	0.85	−1.25
PNICO23	0.85	0.86	1.14	0.20	3.62	3.53	−0.10
PX13	−4.15	4.15	4.31	0.12	4.59	−2.03	−6.63
RC21	3.29	3.83	4.66	0.11	13.98	11.65	−2.33
RG18	0.03	0.13	0.20	0.22	0.94	0.69	−0.25
RSE43	−0.28	0.43	0.56	0.06	2.79	1.32	−1.47
S22	0.19	0.28	0.38	0.04	1.59	1.21	−0.38
S66	0.07	0.26	0.34	0.05	1.83	1.31	−0.52
SCONF	0.02	0.15	0.23	0.03	0.99	0.25	−0.74
TAUT15	0.07	0.82	0.97	0.27	3.01	1.75	−1.27
UPU23	0.42	0.51	0.62	0.09	1.66	1.37	−0.29
W4-11	6.32	7.35	9.35	0.02	36.93	26.09	−10.84
WATER27	2.34	2.72	3.28	0.03	14.49	9.33	−5.16
WCPT18	−2.49	2.51	3.02	0.07	5.87	0.14	−5.74
YBDE18	3.29	3.29	3.42	0.07	3.67	4.60	0.93
BH76RC	−0.43	1.82	2.45	0.10	10.99	6.29	−4.69

SI.8.2 Results for dispersion-corrected functionals with non-local (NL) correction

Table S43: Statistical analysis of the PBE-QIDH-NL DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	3.49	3.49	3.95	0.10	6.82	8.05	1.22
ACONF	−0.12	0.14	0.16	0.08	0.39	0.08	−0.31
ADIM6	−0.59	0.59	0.63	0.18	0.66	−0.26	−0.92
AHB21	−1.29	1.29	1.66	0.06	3.70	−0.11	−3.81
AL2X6	0.97	0.97	1.07	0.03	1.48	1.52	0.03
ALK8	3.02	3.02	4.34	0.05	9.51	9.79	0.27
ALKBDE10	−4.38	4.79	5.53	0.05	11.84	2.08	−9.76
AMINO20x4	0.07	0.20	0.25	0.08	1.26	0.73	−0.53
BH76	0.26	1.24	1.99	0.07	12.99	10.19	−2.80
BHDIV10	−1.46	2.06	2.36	0.05	5.47	2.06	−3.42
BHPERI	−2.59	3.05	3.32	0.15	8.74	3.36	−5.38
BHROT27	0.52	0.52	0.67	0.08	1.36	1.32	−0.05
BSR36	−1.54	1.54	1.60	0.10	1.46	−0.88	−2.33
BUT14DIOL	0.09	0.09	0.11	0.03	0.24	0.21	−0.03
C60ISO	−2.57	3.72	5.14	0.04	13.61	2.61	−10.99
CARBHB12	0.86	0.86	1.07	0.14	2.04	2.18	0.14
CDIE20	0.75	0.78	0.93	0.19	2.01	1.90	−0.10
CHB6	−1.41	1.41	1.60	0.05	2.20	−0.50	−2.69
DARC	−7.81	7.81	7.94	0.24	4.41	−5.71	−10.12
DC13	−1.95	9.12	11.24	0.17	38.82	20.21	−18.61
DIPCS10	−2.23	3.07	3.85	0.00	9.08	1.77	−7.31
FH51	−2.62	3.00	3.72	0.10	15.24	6.07	−9.17
G21EA	−2.18	2.84	3.45	0.08	9.13	2.96	−6.17
G21IP	−0.55	2.55	3.09	0.01	14.41	5.32	−9.10
G2RC	−4.07	5.43	6.94	0.11	26.62	7.38	−19.23
HAL59	0.22	0.36	0.61	0.08	2.62	2.23	−0.39
HEAVY28	−0.05	0.17	0.19	0.14	0.60	0.31	−0.29
HEAVYSB11	0.14	0.58	0.69	0.01	2.18	1.26	−0.92
ICONF	0.22	0.26	0.37	0.08	1.02	0.90	−0.12
IDISP	−1.01	1.64	3.17	0.12	8.65	0.98	−7.67
IL16	−0.29	0.33	0.40	0.00	0.94	0.24	−0.71
INV24	0.58	1.34	1.96	0.04	9.95	6.29	−3.66
ISO34	0.23	1.37	2.01	0.09	8.23	4.35	−3.88
ISOL24	2.01	3.16	4.46	0.14	15.44	11.48	−3.96
MB16-43	12.08	14.79	18.02	0.04	66.59	48.17	−18.43
MCONF	0.30	0.33	0.39	0.07	0.90	0.69	−0.21
NBPRC	−1.28	2.77	3.32	0.10	11.31	4.92	−6.40
PA26	1.60	1.89	2.27	0.01	7.16	5.44	−1.73
PArel	0.84	1.09	1.41	0.24	4.90	3.02	−1.88
PCONF21	0.00	0.36	0.44	0.22	1.48	0.57	−0.91
PNICO23	0.38	0.44	0.69	0.10	2.66	2.42	−0.24
PX13	−2.99	2.99	3.18	0.09	3.98	−1.07	−5.05
RC21	2.49	3.28	4.57	0.09	17.27	13.91	−3.36
RG18	−0.05	0.13	0.19	0.22	0.91	0.52	−0.39
RSE43	0.38	0.51	0.90	0.07	4.39	3.84	−0.55
S22	0.19	0.29	0.46	0.04	1.87	1.59	−0.28
S66	0.02	0.35	0.45	0.06	2.34	1.67	−0.67
SCONF	0.18	0.26	0.29	0.06	0.89	0.37	−0.52
TAUT15	0.51	0.76	1.07	0.25	3.02	2.25	−0.77
UPU23	0.30	0.41	0.51	0.07	1.40	1.08	−0.33
W4-11	−4.02	4.46	6.16	0.01	26.74	5.26	−21.48
WATER27	2.96	3.27	3.95	0.04	14.85	10.70	−4.15
WCPT18	−1.87	2.14	2.65	0.06	7.10	2.34	−4.76
YBDE18	2.26	2.90	3.61	0.06	9.05	6.56	−2.49
BH76RC	−0.54	1.95	2.51	0.09	10.70	5.50	−5.20

Table S44: Statistical analysis of the SOS1-PBE-QIDH-NL DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	3.11	3.11	3.99	0.09	9.23	9.25	0.02
ACONF	−0.24	0.24	0.25	0.13	0.37	0.01	−0.36
ADIM6	0.03	0.07	0.08	0.02	0.19	0.13	−0.05
AHB21	−1.04	1.04	1.36	0.05	3.32	−0.10	−3.43
AL2X6	1.21	1.21	1.27	0.03	1.14	1.92	0.78
ALK8	2.57	2.57	3.53	0.04	7.74	7.97	0.24
ALKBDE10	−6.23	6.23	6.81	0.06	10.65	−1.91	−12.55
AMINO20x4	0.04	0.16	0.20	0.07	1.00	0.56	−0.44
BH76	0.86	1.38	2.22	0.07	13.98	11.35	−2.63
BHDIV10	−0.37	1.20	1.36	0.03	3.35	1.30	−2.06
BHPERI	−0.25	1.57	2.00	0.08	8.17	5.76	−2.41
BHROT27	0.38	0.39	0.51	0.06	1.08	1.04	−0.04
BSR36	−1.72	1.72	1.79	0.11	1.73	−0.98	−2.71
BUT14DIOL	0.06	0.07	0.09	0.03	0.36	0.28	−0.07
C60ISO	0.42	2.02	2.52	0.02	7.70	3.73	−3.97
CARBHB12	0.66	0.66	0.80	0.11	1.53	1.67	0.14
CDIE20	0.37	0.45	0.63	0.11	1.76	1.50	−0.26
CHB6	−1.54	1.54	1.72	0.06	2.26	−0.57	−2.83
DARC	−8.29	8.29	8.32	0.26	2.51	−7.27	−9.78
DC13	−3.46	8.73	10.19	0.16	35.76	17.27	−18.49
DIPCS10	−4.01	4.28	5.13	0.01	11.30	1.36	−9.94
FH51	−2.32	2.74	3.58	0.09	17.18	5.36	−11.82
G21EA	−3.43	3.69	4.21	0.11	11.40	3.20	−8.20
G21IP	−1.47	2.44	3.09	0.01	11.93	4.32	−7.62
G2RC	−3.34	4.85	6.09	0.09	23.35	7.09	−16.27
HAL59	0.24	0.34	0.52	0.07	2.16	1.86	−0.30
HEAVY28	0.00	0.13	0.17	0.10	0.78	0.48	−0.29
HEAVYSB11	0.20	0.85	1.03	0.01	3.07	1.38	−1.70
ICONF	0.11	0.21	0.29	0.06	1.05	0.70	−0.35
IDISP	−1.11	2.77	4.43	0.19	13.04	2.70	−10.34
IL16	−0.29	0.31	0.38	0.00	0.94	0.10	−0.84
INV24	1.00	1.52	2.55	0.05	12.46	8.90	−3.56
ISO34	−0.05	1.19	1.76	0.08	7.82	4.23	−3.60
ISOL24	1.86	3.28	4.94	0.15	22.40	13.96	−8.44
MB16-43	1.79	11.97	15.01	0.03	74.59	36.43	−38.16
MCONF	0.53	0.55	0.62	0.11	1.16	0.97	−0.19
NBPRC	−1.12	2.71	3.17	0.10	11.29	5.11	−6.18
PA26	2.74	2.74	3.11	0.01	6.04	6.26	0.22
PArel	0.68	0.85	1.16	0.18	4.26	2.83	−1.43
PCONF21	0.09	0.40	0.49	0.25	1.63	1.07	−0.56
PNICO23	0.34	0.35	0.51	0.08	1.48	1.37	−0.11
PX13	−1.54	1.56	1.78	0.05	2.91	0.10	−2.81
RC21	2.21	2.98	3.97	0.08	14.53	11.70	−2.83
RG18	0.06	0.12	0.22	0.21	1.05	0.81	−0.24
RSE43	0.76	0.77	1.16	0.10	4.83	4.71	−0.12
S22	0.45	0.46	0.57	0.06	1.51	1.47	−0.04
S66	0.30	0.31	0.41	0.06	1.63	1.58	−0.06
SCONF	0.03	0.18	0.23	0.04	0.85	0.22	−0.64
TAUT15	0.00	0.48	0.54	0.16	1.59	0.78	−0.81
UPU23	0.15	0.44	0.61	0.08	2.93	1.21	−1.72
W4-11	−3.77	5.05	7.17	0.02	35.77	8.74	−27.03
WATER27	2.67	2.85	3.51	0.04	11.59	9.15	−2.45
WCPT18	−0.29	1.53	1.69	0.04	5.79	3.36	−2.43
YBDE18	2.10	2.40	2.87	0.05	7.58	5.29	−2.29
BH76RC	−0.54	1.81	2.31	0.08	8.14	3.02	−5.12

SI.8.3 Results for dispersion-corrected functionals with DFT-D3(0)

Table S45: Statistical analysis of the ωB97X-2-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	4.77	4.77	5.58	0.14	10.46	11.54	1.08
ACONF	0.04	0.04	0.05	0.02	0.09	0.09	0.00
ADIM6	0.07	0.09	0.10	0.03	0.21	0.16	−0.06
AHB21	−0.33	0.37	0.44	0.02	1.54	0.41	−1.13
AL2X6	−1.16	1.16	1.28	0.03	1.70	−0.53	−2.23
ALK8	1.54	2.29	3.93	0.04	12.40	10.11	−2.30
ALKBDE10	−0.16	2.52	3.10	0.03	11.10	4.53	−6.57
AMINO20x4	−0.05	0.11	0.15	0.05	0.64	0.26	−0.38
BH76	−0.01	1.23	2.05	0.07	13.47	8.16	−5.31
BHDIV10	−0.53	0.75	1.06	0.02	3.77	1.08	−2.68
BHPERI	−2.08	2.11	2.21	0.10	3.41	0.38	−3.03
BHROT27	0.07	0.15	0.21	0.02	0.75	0.42	−0.32
BSR36	−1.42	1.42	1.62	0.09	3.16	−0.56	−3.71
BUT14DIOL	−0.02	0.04	0.05	0.01	0.23	0.11	−0.12
C60ISO	−1.40	3.41	4.19	0.03	11.66	3.19	−8.47
CARBHB12	0.55	0.55	0.61	0.09	0.97	1.16	0.19
CDIE20	0.25	0.29	0.42	0.07	1.15	0.98	−0.17
CHB6	−0.78	0.78	0.85	0.03	1.02	−0.41	−1.43
DARC	−0.34	0.79	0.87	0.02	2.57	0.86	−1.72
DC13	−0.25	2.84	4.12	0.05	18.18	8.29	−9.89
DIPCS10	−5.11	5.11	5.66	0.01	8.08	−0.95	−9.03
FH51	−0.17	0.76	1.05	0.02	6.27	1.65	−4.62
G21EA	−1.04	1.60	1.91	0.05	8.00	4.47	−3.53
G21IP	−1.12	2.09	2.65	0.01	13.67	5.52	−8.15
G2RC	−0.45	1.20	1.50	0.02	6.31	2.60	−3.70
HAL59	0.31	0.34	0.45	0.07	1.57	1.23	−0.34
HEAVY28	0.23	0.23	0.27	0.19	0.51	0.57	0.06
HEAVYSB11	−1.53	1.53	1.72	0.03	2.73	−0.19	−2.92
ICONF	0.02	0.14	0.18	0.04	0.67	0.33	−0.34
IDISP	−0.42	0.89	1.06	0.06	3.00	0.91	−2.08
IL16	0.71	0.71	0.74	0.01	0.56	1.03	0.47
INV24	0.01	0.96	1.76	0.03	9.68	7.28	−2.40
ISO34	−0.06	0.58	0.86	0.04	4.32	2.69	−1.63
ISOL24	−0.08	1.12	1.58	0.05	7.76	3.40	−4.35
MB16-43	−17.05	17.73	20.02	0.04	51.38	12.38	−38.99
MCONF	0.15	0.17	0.19	0.03	0.42	0.32	−0.10
NBPRC	0.21	0.95	1.18	0.03	4.02	2.99	−1.03
PA26	0.24	0.99	1.38	0.01	5.55	3.80	−1.75
PArel	0.04	0.35	0.57	0.08	2.75	0.79	−1.97
PCONF21	0.01	0.12	0.13	0.07	0.47	0.22	−0.25
PNICO23	0.24	0.24	0.26	0.06	0.43	0.38	−0.06
PX13	−2.04	2.04	2.17	0.06	2.78	−0.57	−3.34
RC21	−1.16	1.61	2.00	0.05	6.06	1.78	−4.28
RG18	0.15	0.15	0.17	0.26	0.39	0.41	0.02
RSE43	0.46	0.60	1.25	0.08	5.81	5.42	−0.39
S22	0.10	0.27	0.30	0.04	1.06	0.49	−0.57
S66	0.18	0.20	0.24	0.04	0.80	0.53	−0.27
SCONF	−0.03	0.06	0.07	0.01	0.30	0.14	−0.16
TAUT15	0.18	0.44	0.58	0.14	1.92	1.10	−0.82
UPU23	0.33	0.44	0.56	0.08	1.60	1.16	−0.44
W4-11	−3.52	3.65	4.50	0.01	20.56	2.28	−18.28
WATER27	3.19	3.33	4.51	0.04	13.47	11.55	−1.92
WCPT18	−1.20	1.63	1.96	0.05	7.08	3.76	−3.32
YBDE18	−0.21	1.38	1.69	0.03	6.76	4.30	−2.45
BH76RC	−0.14	0.70	0.96	0.03	4.68	2.22	−2.46

Table S46: Statistical analysis of the B2PPW91-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	18.11	18.11	20.19	0.54	32.27	36.24	3.97
ACONF	−0.08	0.09	0.10	0.05	0.23	0.03	−0.20
ADIM6	−0.14	0.16	0.20	0.05	0.39	0.06	−0.33
AHB21	0.18	0.56	0.77	0.02	3.77	2.45	−1.33
AL2X6	−1.90	1.90	3.04	0.05	6.52	−0.41	−6.93
ALK8	0.93	3.39	4.87	0.05	15.84	11.09	−4.74
ALKBDE10	3.90	5.65	8.80	0.06	26.48	23.63	−2.85
AMINO20x4	−0.01	0.31	0.41	0.13	2.37	1.33	−1.04
BH76	−6.49	6.53	7.30	0.35	23.24	1.51	−21.73
BHDIV10	−5.94	6.24	7.07	0.14	12.02	1.50	−10.52
BHPERI	−5.24	5.24	5.44	0.25	5.50	−2.67	−8.17
BHROT27	0.26	0.29	0.44	0.05	1.33	1.20	−0.13
BSR36	−1.97	1.97	2.21	0.12	4.15	−0.58	−4.73
BUT14DIOL	0.10	0.21	0.27	0.08	1.18	0.79	−0.40
C60ISO	−12.59	12.59	15.27	0.13	25.15	−1.49	−26.64
CARBHB12	0.86	0.90	1.37	0.15	3.31	3.18	−0.13
CDIE20	1.36	1.36	1.45	0.33	2.05	2.42	0.37
CHB6	0.87	0.87	0.97	0.03	1.22	1.81	0.59
DARC	2.95	2.97	3.42	0.09	5.32	5.20	−0.12
DC13	2.38	7.54	9.40	0.14	34.76	12.23	−22.53
DIPCS10	−1.59	3.01	3.72	0.00	10.72	3.46	−7.26
FH51	1.06	2.17	3.11	0.07	16.21	8.89	−7.32
G21EA	1.83	2.45	2.90	0.07	8.66	5.61	−3.06
G21IP	0.20	3.23	3.95	0.01	16.38	8.89	−7.48
G2RC	1.75	4.66	5.93	0.09	25.95	16.61	−9.34
HAL59	0.15	0.67	1.06	0.15	5.53	4.33	−1.20
HEAVY28	−0.04	0.21	0.25	0.17	0.90	0.45	−0.45
HEAVYSB11	−0.45	2.06	2.40	0.04	6.98	2.62	−4.37
ICONF	0.16	0.35	0.44	0.11	1.42	0.96	−0.46
IDISP	2.63	2.63	3.13	0.18	4.97	5.52	0.54
IL16	−0.22	0.37	0.50	0.00	1.68	0.66	−1.02
INV24	−1.56	1.78	2.65	0.06	9.19	1.74	−7.46
ISO34	−0.55	1.16	1.56	0.08	8.73	3.98	−4.75
ISOL24	−1.55	3.48	4.81	0.16	21.06	12.52	−8.54
MB16-43	−2.46	18.00	23.46	0.04	116.90	53.74	−63.16
MCONF	0.41	0.55	0.66	0.11	2.02	1.34	−0.69
NBPRC	0.01	1.26	1.56	0.05	5.81	2.33	−3.48
PA26	3.06	3.06	3.66	0.02	7.79	8.48	0.69
PArel	0.21	1.46	2.12	0.32	11.35	5.14	−6.21
PCONF21	−0.26	0.74	0.82	0.46	2.73	1.31	−1.42
PNICO23	−0.05	0.54	0.82	0.13	3.95	3.22	−0.73
PX13	−8.76	8.76	8.92	0.26	6.60	−6.14	−12.74
RC21	3.07	4.03	4.56	0.11	12.00	9.02	−2.98
RG18	−0.50	0.50	0.73	0.86	2.22	−0.05	−2.26
RSE43	−1.75	1.75	2.00	0.23	4.60	−0.48	−5.08
S22	0.03	0.26	0.32	0.04	1.13	0.58	−0.55
S66	−0.06	0.19	0.23	0.03	1.00	0.39	−0.61
SCONF	0.23	0.81	1.05	0.18	3.87	1.04	−2.84
TAUT15	−0.25	1.48	1.68	0.49	5.52	3.14	−2.38
UPU23	0.06	0.38	0.47	0.07	1.93	1.04	−0.89
W4-11	7.13	8.59	11.10	0.03	47.03	34.59	−12.44
WATER27	−0.59	1.19	2.01	0.01	10.17	2.05	−8.12
WCPT18	−6.77	6.77	7.11	0.19	8.61	−3.92	−12.53
YBDE18	−1.51	3.52	4.16	0.07	15.15	7.26	−7.89
BH76RC	0.53	2.64	4.26	0.12	22.44	17.54	−4.89

Table S47: Statistical analysis of the B2NC-PLYP-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	2.67	2.67	3.02	0.08	4.82	5.52	0.70
ACONF	−0.17	0.17	0.20	0.09	0.42	0.04	−0.38
ADIM6	−0.38	0.38	0.43	0.11	0.54	−0.13	−0.67
AHB21	−0.50	0.52	0.72	0.02	1.86	0.10	−1.76
AL2X6	−2.07	2.07	2.12	0.06	1.30	−1.56	−2.86
ALK8	−0.44	1.58	1.99	0.03	6.76	3.47	−3.29
ALKBDE10	−0.93	2.83	3.47	0.03	11.05	4.69	−6.36
AMINO20x4	−0.02	0.14	0.18	0.06	1.11	0.52	−0.59
BH76	1.21	1.81	2.88	0.10	18.98	15.48	−3.51
BHDIV10	0.31	1.07	1.46	0.02	5.20	3.27	−1.93
BHPERI	−1.98	2.01	2.19	0.10	4.10	0.45	−3.65
BHROT27	0.30	0.31	0.40	0.05	1.02	0.96	−0.06
BSR36	−1.75	1.75	1.88	0.11	2.56	−0.84	−3.40
BUT14DIOL	0.03	0.07	0.09	0.03	0.36	0.23	−0.13
C60ISO	−7.74	7.81	10.37	0.08	20.36	0.17	−20.19
CARBHB12	0.42	0.42	0.55	0.07	1.24	1.26	0.02
CDIE20	0.55	0.55	0.61	0.14	0.96	0.97	0.01
CHB6	−1.11	1.11	1.21	0.04	1.42	−0.36	−1.79
DARC	0.32	0.83	1.12	0.03	3.35	1.94	−1.41
DC13	1.70	3.29	4.26	0.06	15.77	9.29	−6.48
DIPCS10	−3.14	3.28	4.17	0.01	8.92	0.70	−8.23
FH51	−0.87	1.37	1.98	0.04	10.24	2.85	−7.38
G21EA	−1.42	2.13	2.55	0.06	11.23	7.19	−4.04
G21IP	−0.67	1.82	2.60	0.01	16.52	7.80	−8.72
G2RC	−1.57	1.90	2.51	0.04	8.49	2.26	−6.23
HAL59	0.03	0.22	0.33	0.05	1.88	1.24	−0.64
HEAVY28	−0.17	0.17	0.20	0.14	0.41	0.02	−0.38
HEAVYSB11	−0.91	1.01	1.38	0.02	3.04	0.37	−2.67
ICONF	0.08	0.21	0.27	0.06	1.01	0.51	−0.50
IDISP	0.42	0.48	0.59	0.03	1.09	0.91	−0.18
IL16	0.45	0.45	0.52	0.00	1.21	1.18	−0.03
INV24	0.35	0.91	1.32	0.03	6.34	4.26	−2.08
ISO34	0.41	0.63	1.17	0.04	5.47	4.69	−0.78
ISOL24	0.27	1.56	2.27	0.07	10.57	6.06	−4.51
MB16-43	−6.45	11.03	13.62	0.03	56.45	20.64	−35.81
MCONF	0.27	0.30	0.34	0.06	0.85	0.64	−0.21
NBPRC	0.45	0.77	0.93	0.03	2.83	2.01	−0.83
PA26	0.31	0.96	1.48	0.01	7.45	3.84	−3.61
PArel	0.28	0.46	0.76	0.10	3.57	2.67	−0.90
PCONF21	0.14	0.23	0.27	0.14	0.74	0.50	−0.24
PNICO23	−0.15	0.17	0.21	0.04	0.52	0.10	−0.43
PX13	−1.04	1.07	1.22	0.03	2.08	0.19	−1.89
RC21	−2.18	2.82	3.25	0.08	7.74	2.57	−5.17
RG18	−0.08	0.16	0.23	0.28	1.02	0.49	−0.53
RSE43	1.16	1.16	2.22	0.15	9.27	9.26	−0.01
S22	0.13	0.17	0.22	0.02	0.80	0.58	−0.22
S66	0.03	0.20	0.25	0.04	1.18	0.71	−0.47
SCONF	0.03	0.06	0.10	0.01	0.41	0.33	−0.09
TAUT15	0.28	0.43	0.60	0.14	1.92	1.03	−0.88
UPU23	0.26	0.40	0.50	0.07	1.59	1.09	−0.50
W4-11	−3.21	3.51	5.02	0.01	27.19	3.88	−23.31
WATER27	1.77	1.92	2.53	0.02	9.46	7.35	−2.11
WCPT18	−0.46	0.90	1.13	0.03	4.04	2.23	−1.82
YBDE18	1.13	1.59	2.11	0.03	7.26	5.61	−1.65
BH76RC	−0.43	1.15	1.62	0.05	7.83	3.57	−4.26

Table S48: Statistical analysis of the mPW2NC-PLYP-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.55	7.55	8.41	0.22	12.55	13.72	1.17
ACONF	−0.13	0.13	0.16	0.07	0.35	0.04	−0.31
ADIM6	−0.30	0.30	0.36	0.09	0.59	−0.03	−0.62
AHB21	−0.42	0.44	0.54	0.02	1.73	0.24	−1.49
AL2X6	−2.60	2.60	2.66	0.07	1.69	−1.68	−3.36
ALK8	−0.60	1.73	2.22	0.03	7.52	4.04	−3.48
ALKBDE10	4.39	5.24	6.78	0.05	15.12	12.37	−2.76
AMINO20x4	−0.03	0.13	0.18	0.05	1.06	0.62	−0.44
BH76	−1.63	2.29	2.82	0.12	15.38	8.13	−7.25
BHDIV10	−1.66	2.57	3.06	0.06	8.86	3.22	−5.64
BHPERI	−3.84	3.84	4.00	0.18	5.28	−1.95	−7.23
BHROT27	0.17	0.19	0.29	0.03	1.02	0.92	−0.10
BSR36	−1.75	1.75	2.00	0.11	3.91	−0.69	−4.60
BUT14DIOL	0.11	0.11	0.13	0.04	0.27	0.27	0.00
C60ISO	−13.39	13.39	16.50	0.14	27.46	−2.42	−29.87
CARBHB12	0.66	0.66	0.80	0.11	1.39	1.55	0.16
CDIE20	0.71	0.71	0.74	0.17	0.91	1.15	0.25
CHB6	−1.01	1.01	1.11	0.04	1.38	−0.23	−1.61
DARC	3.59	3.59	3.78	0.11	3.49	5.37	1.87
DC13	3.11	5.33	6.64	0.10	21.00	9.30	−11.70
DIPCS10	−2.34	3.08	3.41	0.00	7.84	2.68	−5.16
FH51	0.36	1.33	1.87	0.04	11.47	5.85	−5.62
G21EA	0.61	1.48	1.99	0.04	6.54	4.41	−2.13
G21IP	−0.17	1.91	2.45	0.01	13.24	6.07	−7.18
G2RC	0.39	1.74	2.43	0.03	11.39	7.23	−4.16
HAL59	0.38	0.40	0.61	0.09	2.44	2.30	−0.15
HEAVY28	0.06	0.10	0.12	0.08	0.38	0.23	−0.15
HEAVYSB11	0.25	0.72	0.96	0.01	3.30	2.24	−1.06
ICONF	0.04	0.20	0.23	0.06	0.79	0.34	−0.45
IDISP	1.38	1.38	2.14	0.10	4.81	4.88	0.07
IL16	0.17	0.22	0.27	0.00	0.89	0.65	−0.24
INV24	−0.57	0.87	1.70	0.03	8.78	1.24	−7.54
ISO34	0.20	0.81	1.30	0.06	6.88	5.47	−1.41
ISOL24	−0.81	3.08	4.40	0.14	22.14	12.97	−9.17
MB16-43	−6.95	7.93	9.51	0.02	33.72	15.08	−18.64
MCONF	0.17	0.22	0.27	0.04	0.87	0.58	−0.30
NBPRC	0.57	1.06	1.44	0.04	4.41	3.62	−0.79
PA26	−0.38	1.18	1.45	0.01	6.84	3.42	−3.42
PArel	0.02	0.67	1.21	0.14	6.66	2.65	−4.01
PCONF21	−0.04	0.32	0.36	0.20	1.25	0.62	−0.63
PNICO23	0.07	0.16	0.23	0.04	0.95	0.75	−0.20
PX13	−3.64	3.64	3.72	0.11	3.20	−2.18	−5.38
RC21	−1.52	2.11	2.60	0.06	7.30	1.90	−5.41
RG18	0.12	0.18	0.26	0.31	0.97	0.75	−0.22
RSE43	0.25	0.65	1.30	0.09	7.16	5.62	−1.54
S22	0.11	0.15	0.20	0.02	0.63	0.41	−0.22
S66	0.07	0.21	0.26	0.04	0.88	0.54	−0.34
SCONF	0.12	0.22	0.26	0.05	1.01	0.32	−0.69
TAUT15	0.15	0.62	0.76	0.20	2.44	1.67	−0.78
UPU23	0.21	0.36	0.46	0.06	1.62	0.99	−0.63
W4-11	5.45	5.86	7.39	0.02	24.64	19.85	−4.79
WATER27	3.11	3.42	4.68	0.04	17.11	12.98	−4.12
WCPT18	−2.99	2.99	3.17	0.09	3.64	−1.78	−5.42
YBDE18	1.58	1.92	3.04	0.04	9.62	8.28	−1.35
BH76RC	−0.02	1.49	2.22	0.07	12.05	9.37	−2.69

Table S49: Statistical analysis of the mSD-PBEPBE-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	9.50	9.50	11.01	0.28	20.07	21.06	0.99
ACONF	−0.34	0.34	0.35	0.19	0.46	−0.02	−0.48
ADIM6	−0.23	0.23	0.27	0.07	0.44	−0.03	−0.47
AHB21	1.39	1.45	2.17	0.06	7.58	7.19	−0.39
AL2X6	−3.88	3.88	4.25	0.11	4.95	−2.76	−7.71
ALK8	−1.30	5.83	7.74	0.09	27.88	14.79	−13.08
ALKBDE10	17.82	17.82	21.99	0.18	37.55	45.17	7.62
AMINO20x4	−0.24	0.37	0.47	0.15	1.82	0.69	−1.13
BH76	−1.78	4.04	5.65	0.22	41.48	27.55	−13.93
BHDIV10	−4.36	4.70	5.87	0.10	12.51	1.73	−10.78
BHPERI	−7.36	7.36	7.70	0.35	11.26	−2.85	−14.12
BHROT27	−0.52	0.54	0.83	0.09	2.16	0.08	−2.08
BSR36	0.55	0.69	1.02	0.04	3.81	3.03	−0.78
BUT14DIOL	−0.17	0.19	0.21	0.07	0.58	0.21	−0.37
C60ISO	−32.03	32.03	36.90	0.33	52.17	−8.24	−60.41
CARBHB12	0.07	0.21	0.31	0.03	1.17	0.84	−0.33
CDIE20	0.45	0.58	0.70	0.14	2.61	1.93	−0.68
CHB6	0.45	0.73	1.05	0.03	3.00	2.24	−0.76
DARC	7.41	7.41	7.53	0.23	4.92	10.61	5.68
DC13	5.11	11.52	15.52	0.21	48.18	33.56	−14.63
DIPCS10	−3.67	5.94	7.75	0.01	22.01	3.38	−18.63
FH51	3.44	3.97	5.15	0.13	18.16	15.24	−2.92
G21EA	4.32	6.27	8.11	0.19	24.13	17.08	−7.05
G21IP	0.46	4.97	7.42	0.02	44.36	25.23	−19.13
G2RC	6.29	7.71	9.38	0.15	31.10	21.50	−9.61
HAL59	0.37	0.47	0.72	0.10	3.43	2.64	−0.79
HEAVY28	0.15	0.19	0.23	0.15	0.72	0.49	−0.23
HEAVYSB11	6.02	6.02	6.40	0.10	7.06	8.68	1.62
ICONF	−0.26	0.51	0.67	0.16	2.54	1.28	−1.26
IDISP	1.85	3.46	5.08	0.24	14.75	11.60	−3.15
IL16	0.76	0.77	0.95	0.01	1.72	1.66	−0.06
INV24	−2.05	3.01	5.93	0.09	32.52	7.84	−24.68
ISO34	−0.51	1.37	1.97	0.09	9.96	3.12	−6.84
ISOL24	−2.32	4.57	6.11	0.21	27.77	14.48	−13.29
MB16-43	−6.93	24.69	29.82	0.06	116.88	55.85	−61.03
MCONF	0.38	0.54	0.63	0.11	1.87	1.19	−0.67
NBPRC	1.27	2.78	3.42	0.10	13.29	6.32	−6.97
PA26	−1.32	2.15	2.45	0.01	9.33	4.63	−4.69
PArel	−1.00	1.32	2.96	0.29	12.86	0.90	−11.96
PCONF21	−0.22	0.60	0.72	0.37	2.55	1.23	−1.32
PNICO23	−0.25	0.36	0.53	0.08	2.37	0.75	−1.62
PX13	−6.46	6.46	6.55	0.19	3.54	−5.18	−8.72
RC21	−6.79	7.17	8.83	0.20	22.37	2.78	−19.59
RG18	0.00	0.14	0.22	0.24	1.02	0.71	−0.31
RSE43	1.80	2.06	4.56	0.27	21.05	19.84	−1.20
S22	−0.24	0.94	1.36	0.13	5.38	2.35	−3.03
S66	−0.26	0.51	0.82	0.09	4.03	1.31	−2.73
SCONF	0.05	0.23	0.32	0.05	1.31	0.43	−0.88
TAUT15	−0.62	1.14	1.31	0.37	3.40	1.27	−2.14
UPU23	−0.18	0.42	0.53	0.07	2.00	0.94	−1.06
W4-11	28.15	28.15	33.99	0.09	87.95	88.53	0.58
WATER27	−4.45	4.46	6.01	0.05	15.20	0.12	−15.08
WCPT18	−6.25	6.25	6.60	0.18	8.59	−2.79	−11.38
YBDE18	6.77	7.35	10.08	0.15	30.69	26.97	−3.72
BH76RC	1.44	4.56	6.57	0.21	29.94	21.40	−8.54

Table S50: Statistical analysis of the PBE0-DH-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.58	7.58	8.57	0.22	15.24	17.59	2.34
ACONF	0.00	0.08	0.09	0.04	0.34	0.16	−0.18
ADIM6	0.24	0.24	0.25	0.07	0.25	0.40	0.15
AHB21	−1.21	1.21	1.58	0.05	3.51	−0.08	−3.59
AL2X6	0.28	1.03	1.04	0.03	2.30	1.17	−1.14
ALK8	2.70	2.70	3.56	0.04	5.53	5.87	0.34
ALKBDE10	−6.30	6.38	7.57	0.06	15.63	0.37	−15.26
AMINO20x4	0.04	0.27	0.34	0.11	1.56	0.76	−0.80
BH76	−1.31	1.87	2.11	0.10	9.35	3.72	−5.63
BHDIV10	−1.84	2.37	2.73	0.05	7.23	2.34	−4.88
BHPERI	−1.31	1.96	2.24	0.09	7.43	3.96	−3.48
BHROT27	0.54	0.55	0.73	0.09	1.47	1.42	−0.06
BSR36	−3.93	3.93	4.47	0.24	8.10	−1.79	−9.89
BUT14DIOL	0.03	0.10	0.12	0.04	0.55	0.39	−0.16
C60ISO	1.83	2.96	3.17	0.03	7.96	5.18	−2.78
CARBHB12	1.04	1.04	1.25	0.17	2.19	2.38	0.19
CDIE20	0.98	1.03	1.25	0.25	2.86	2.53	−0.32
CHB6	−1.06	1.21	1.50	0.05	3.36	0.45	−2.91
DARC	−5.87	5.87	6.20	0.18	6.73	−3.55	−10.28
DC13	−2.23	8.17	10.35	0.15	37.58	20.32	−17.27
DIPCS10	−0.67	2.61	3.36	0.00	11.18	4.04	−7.14
FH51	−2.28	2.98	3.73	0.10	14.06	4.10	−9.96
G21EA	−1.47	3.01	3.46	0.09	9.44	3.21	−6.23
G21IP	0.05	3.08	3.75	0.01	15.64	7.76	−7.88
G2RC	−4.11	6.23	7.89	0.12	31.48	10.72	−20.76
HAL59	−0.11	0.46	0.58	0.10	2.68	1.97	−0.71
HEAVY28	−0.25	0.30	0.35	0.24	0.80	0.19	−0.61
HEAVYSB11	−1.86	1.86	1.96	0.03	1.94	−0.97	−2.91
ICONF	0.25	0.33	0.50	0.10	1.56	1.18	−0.38
IDISP	0.23	2.06	2.42	0.14	7.29	3.73	−3.56
IL16	−0.05	0.32	0.42	0.00	1.71	0.89	−0.82
INV24	0.20	1.45	2.15	0.05	10.43	6.16	−4.27
ISO34	0.01	1.46	2.00	0.10	7.85	4.00	−3.85
ISOL24	0.96	2.34	3.23	0.11	13.65	8.07	−5.57
MB16-43	1.20	12.91	16.41	0.03	76.89	47.66	−29.23
MCONF	0.21	0.22	0.27	0.04	0.65	0.50	−0.14
NBPRC	−0.87	2.95	3.34	0.11	10.78	5.69	−5.08
PA26	2.56	2.64	3.14	0.01	8.29	7.22	−1.07
PArel	0.83	1.16	1.54	0.25	5.94	4.18	−1.76
PCONF21	−0.06	0.53	0.64	0.33	2.21	0.95	−1.26
PNICO23	0.27	0.44	0.66	0.10	3.11	2.45	−0.66
PX13	−3.79	3.79	4.02	0.11	4.92	−1.59	−6.51
RC21	3.87	4.37	5.42	0.12	16.84	14.57	−2.27
RG18	0.03	0.14	0.18	0.24	0.73	0.37	−0.36
RSE43	−0.43	0.46	0.62	0.06	2.28	0.52	−1.75
S22	0.23	0.43	0.58	0.06	2.47	1.69	−0.78
S66	0.26	0.34	0.47	0.06	2.17	1.77	−0.40
SCONF	−0.09	0.16	0.23	0.03	0.92	0.22	−0.70
TAUT15	0.30	0.78	1.08	0.26	3.53	2.27	−1.26
UPU23	0.41	0.55	0.65	0.10	1.87	1.35	−0.52
W4-11	−5.52	5.77	7.51	0.02	30.59	3.07	−27.52
WATER27	4.33	4.65	5.88	0.06	18.62	14.28	−4.33
WCPT18	−2.11	2.33	2.95	0.07	7.86	1.95	−5.91
YBDE18	−0.01	1.69	2.06	0.03	6.86	2.63	−4.23
BH76RC	−0.54	2.15	2.73	0.10	10.62	5.39	−5.23

Table S51: Statistical analysis of the LS1-DH-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	2.48	2.48	2.81	0.07	4.67	5.44	0.77
ACONF	−0.15	0.16	0.19	0.09	0.41	0.06	−0.35
ADIM6	−0.40	0.40	0.44	0.12	0.54	−0.15	−0.69
AHB21	−1.06	1.07	1.44	0.05	3.45	0.03	−3.42
AL2X6	0.35	0.64	0.68	0.02	1.64	0.89	−0.75
ALK8	2.59	2.59	4.07	0.04	9.89	10.01	0.12
ALKBDE10	−3.42	4.04	4.71	0.04	10.21	2.82	−7.39
AMINO20x4	0.04	0.20	0.24	0.08	1.15	0.63	−0.52
BH76	0.88	1.53	2.45	0.08	15.55	12.93	−2.62
BHDIV10	−1.13	1.82	2.02	0.04	5.12	2.08	−3.04
BHPERI	−2.71	3.05	3.31	0.15	8.26	2.95	−5.31
BHROT27	0.46	0.47	0.60	0.07	1.23	1.19	−0.05
BSR36	−1.54	1.54	1.61	0.10	1.57	−0.87	−2.44
BUT14DIOL	0.03	0.06	0.07	0.02	0.30	0.19	−0.11
C60ISO	−4.24	5.05	6.81	0.05	16.17	1.98	−14.19
CARBHB12	0.76	0.76	0.96	0.13	1.88	1.97	0.10
CDIE20	0.73	0.75	0.90	0.18	1.84	1.74	−0.10
CHB6	−1.18	1.18	1.37	0.04	1.90	−0.37	−2.26
DARC	−6.64	6.64	6.80	0.20	4.36	−4.42	−8.78
DC13	−1.23	7.87	9.95	0.14	34.63	18.07	−16.55
DIPCS10	−1.16	2.84	3.30	0.00	9.09	2.82	−6.27
FH51	−2.41	2.73	3.38	0.09	12.75	5.26	−7.50
G21EA	−1.42	2.43	2.89	0.07	10.32	5.05	−5.27
G21IP	0.13	2.28	2.87	0.01	14.11	5.56	−8.55
G2RC	−3.73	4.85	6.11	0.09	22.54	5.99	−16.55
HAL59	0.03	0.32	0.49	0.07	2.30	1.77	−0.53
HEAVY28	−0.17	0.21	0.24	0.17	0.55	0.16	−0.39
HEAVYSB11	0.03	0.57	0.71	0.01	2.37	1.18	−1.18
ICONF	0.20	0.23	0.34	0.07	0.95	0.80	−0.15
IDISP	−0.82	1.57	2.83	0.11	7.80	1.02	−6.78
IL16	−0.06	0.25	0.30	0.00	1.00	0.55	−0.45
INV24	0.53	1.21	1.74	0.04	9.13	5.87	−3.26
ISO34	0.30	1.30	1.88	0.09	7.55	3.99	−3.56
ISOL24	1.78	2.79	3.80	0.13	12.12	9.15	−2.97
MB16-43	10.20	12.74	15.55	0.03	57.05	37.00	−20.05
MCONF	0.33	0.36	0.41	0.07	0.91	0.73	−0.18
NBPRC	−0.99	2.32	2.77	0.08	9.48	4.18	−5.30
PA26	1.34	1.71	2.05	0.01	7.32	4.90	−2.43
PArel	0.74	0.96	1.25	0.21	4.71	3.01	−1.70
PCONF21	0.03	0.38	0.46	0.23	1.62	0.73	−0.88
PNICO23	0.20	0.35	0.52	0.08	2.37	1.93	−0.44
PX13	−2.61	2.61	2.80	0.08	3.76	−0.79	−4.54
RC21	1.30	2.52	3.79	0.07	15.63	11.64	−3.98
RG18	−0.11	0.16	0.23	0.28	0.90	0.36	−0.53
RSE43	0.71	0.77	1.45	0.10	6.13	5.82	−0.31
S22	0.17	0.24	0.34	0.03	1.45	1.16	−0.29
S66	0.05	0.26	0.34	0.05	1.77	1.25	−0.53
SCONF	0.11	0.15	0.16	0.03	0.51	0.25	−0.26
TAUT15	0.48	0.71	0.95	0.23	2.65	1.96	−0.69
UPU23	0.31	0.44	0.53	0.08	1.43	1.08	−0.35
W4-11	−3.63	4.08	5.72	0.01	27.45	5.39	−22.06
WATER27	2.34	2.62	3.17	0.03	12.48	8.67	−3.81
WCPT18	−1.66	1.91	2.31	0.05	6.40	2.18	−4.22
YBDE18	2.44	2.99	3.75	0.06	8.77	6.68	−2.08
BH76RC	−0.50	1.85	2.41	0.09	10.82	5.78	−5.04

Table S52: Statistical analysis of the PBE0-2-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	1.79	1.79	2.05	0.05	3.22	3.66	0.45
ACONF	−0.16	0.17	0.20	0.09	0.43	0.06	−0.38
ADIM6	−0.40	0.40	0.45	0.12	0.56	−0.16	−0.72
AHB21	−0.94	0.95	1.32	0.04	3.45	0.13	−3.32
AL2X6	0.07	0.52	0.56	0.01	1.50	0.53	−0.97
ALK8	2.22	2.23	3.86	0.04	9.94	9.89	−0.04
ALKBDE10	−2.22	3.63	3.96	0.04	10.61	3.81	−6.80
AMINO20x4	0.01	0.20	0.24	0.08	1.12	0.61	−0.52
BH76	1.30	1.89	2.94	0.10	18.03	15.00	−3.03
BHDIV10	−1.08	1.83	2.01	0.04	5.49	2.09	−3.40
BHPERI	−3.15	3.37	3.65	0.16	8.21	2.47	−5.74
BHROT27	0.42	0.42	0.55	0.07	1.17	1.12	−0.05
BSR36	−1.10	1.10	1.14	0.07	1.31	−0.50	−1.80
BUT14DIOL	0.02	0.06	0.08	0.02	0.35	0.22	−0.12
C60ISO	−6.21	6.67	8.84	0.07	18.94	1.15	−17.79
CARBHB12	0.67	0.67	0.86	0.11	1.76	1.82	0.06
CDIE20	0.72	0.74	0.86	0.18	1.64	1.56	−0.08
CHB6	−1.06	1.06	1.23	0.04	1.61	−0.31	−1.92
DARC	−6.12	6.12	6.29	0.19	4.31	−3.85	−8.16
DC13	−0.77	7.43	9.64	0.14	34.28	18.50	−15.78
DIPCS10	−1.52	2.90	3.42	0.00	9.55	2.52	−7.03
FH51	−2.21	2.54	3.17	0.08	13.42	5.04	−8.38
G21EA	−1.27	2.23	2.77	0.07	11.57	6.58	−4.99
G21IP	0.10	2.11	2.83	0.01	16.27	7.43	−8.84
G2RC	−3.40	4.35	5.49	0.08	19.35	4.46	−14.89
HAL59	−0.02	0.34	0.48	0.07	2.32	1.73	−0.59
HEAVY28	−0.25	0.26	0.30	0.21	0.58	0.08	−0.50
HEAVYSB11	0.43	0.70	0.84	0.01	2.46	1.69	−0.78
ICONF	0.18	0.21	0.30	0.06	0.82	0.65	−0.17
IDISP	−0.84	1.89	3.05	0.13	8.45	1.24	−7.21
IL16	0.08	0.28	0.34	0.00	1.15	0.79	−0.35
INV24	0.44	1.06	1.55	0.03	8.12	5.35	−2.77
ISO34	0.35	1.25	1.81	0.09	7.63	4.27	−3.37
ISOL24	1.73	2.70	3.62	0.12	11.75	8.71	−3.05
MB16-43	10.82	12.40	15.02	0.03	47.78	30.90	−16.88
MCONF	0.38	0.41	0.46	0.08	1.04	0.83	−0.21
NBPRC	−0.82	2.00	2.40	0.07	8.15	3.52	−4.64
PA26	0.92	1.46	1.80	0.01	7.58	4.17	−3.41
PArel	0.66	0.87	1.17	0.19	4.84	3.11	−1.73
PCONF21	0.02	0.37	0.44	0.23	1.61	0.77	−0.84
PNICO23	0.14	0.32	0.47	0.07	2.24	1.77	−0.47
PX13	−2.59	2.59	2.77	0.08	3.75	−0.78	−4.54
RC21	0.25	2.59	3.54	0.07	14.39	9.73	−4.65
RG18	−0.14	0.18	0.25	0.31	0.95	0.36	−0.58
RSE43	1.01	1.03	2.01	0.14	8.45	8.25	−0.20
S22	0.14	0.21	0.28	0.03	1.17	0.87	−0.30
S66	0.01	0.24	0.30	0.04	1.58	0.97	−0.61
SCONF	0.11	0.13	0.15	0.03	0.44	0.29	−0.15
TAUT15	0.54	0.73	0.95	0.24	2.48	1.93	−0.55
UPU23	0.26	0.41	0.50	0.07	1.58	1.07	−0.51
W4-11	−2.58	3.51	5.04	0.01	27.99	5.95	−22.05
WATER27	1.72	2.02	2.45	0.02	10.99	7.02	−3.97
WCPT18	−1.72	1.92	2.24	0.05	5.84	1.80	−4.04
YBDE18	2.90	3.23	4.07	0.07	8.70	7.03	−1.67
BH76RC	−0.43	1.94	2.43	0.09	10.93	6.50	−4.43

Table S53: Statistical analysis of the revPBE0-DH-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.38	7.38	8.53	0.22	16.17	18.36	2.19
ACONF	−0.06	0.08	0.09	0.04	0.24	0.06	−0.18
ADIM6	−0.41	0.41	0.46	0.12	0.56	−0.09	−0.64
AHB21	−0.50	0.61	0.90	0.03	2.80	0.49	−2.32
AL2X6	−0.24	0.76	1.18	0.02	3.59	0.93	−2.66
ALK8	2.91	2.96	4.80	0.05	12.09	11.90	−0.19
ALKBDE10	−8.93	8.93	9.81	0.09	15.52	−2.77	−18.28
AMINO20x4	0.04	0.23	0.30	0.09	1.47	0.86	−0.60
BH76	−0.65	1.27	1.57	0.07	9.10	5.19	−3.91
BHDIV10	−1.24	1.56	1.86	0.03	5.21	1.61	−3.60
BHPERI	−1.17	1.93	2.22	0.09	7.07	3.56	−3.50
BHROT27	0.52	0.52	0.63	0.08	1.27	1.19	−0.08
BSR36	−3.38	3.38	3.91	0.21	7.91	−1.45	−9.36
BUT14DIOL	−0.24	0.26	0.29	0.09	0.73	0.22	−0.51
C60ISO	1.15	2.63	2.86	0.03	7.63	3.94	−3.69
CARBHB12	0.74	0.74	1.02	0.12	2.08	2.12	0.04
CDIE20	0.79	0.79	1.04	0.19	2.28	2.31	0.03
CHB6	−1.03	1.03	1.34	0.04	2.38	−0.16	−2.55
DARC	−4.87	4.87	5.03	0.15	4.36	−3.11	−7.47
DC13	−0.78	7.44	9.03	0.14	31.53	18.98	−12.55
DIPCS10	−1.34	3.06	3.91	0.00	12.79	3.89	−8.91
FH51	−1.93	2.47	3.09	0.08	11.39	2.87	−8.53
G21EA	−2.10	3.37	4.00	0.10	10.72	3.14	−7.59
G21IP	−0.44	3.35	3.95	0.01	15.24	7.54	−7.70
G2RC	−2.88	5.07	6.40	0.10	28.26	11.97	−16.29
HAL59	0.01	0.41	0.55	0.09	2.77	1.69	−1.08
HEAVY28	0.31	0.33	0.41	0.27	1.21	0.98	−0.23
HEAVYSB11	−3.01	3.01	3.10	0.05	2.70	−1.79	−4.49
ICONF	0.24	0.35	0.56	0.11	1.84	1.47	−0.36
IDISP	0.38	1.14	1.37	0.08	4.10	2.58	−1.53
IL16	0.01	0.36	0.41	0.00	1.27	0.66	−0.61
INV24	0.63	1.49	2.21	0.05	10.23	6.32	−3.91
ISO34	−0.11	1.23	1.69	0.08	6.95	3.46	−3.49
ISOL24	0.85	2.06	2.82	0.09	11.51	7.40	−4.11
MB16-43	−3.20	11.30	14.60	0.03	76.51	43.23	−33.29
MCONF	0.13	0.22	0.27	0.04	1.07	0.61	−0.47
NBPRC	−0.62	2.29	2.51	0.08	7.79	4.47	−3.32
PA26	3.87	3.87	4.29	0.02	8.32	8.86	0.54
PArel	0.78	0.94	1.42	0.20	4.48	3.86	−0.61
PCONF21	0.10	0.32	0.40	0.20	1.48	0.65	−0.82
PNICO23	0.04	0.25	0.38	0.06	1.82	1.30	−0.53
PX13	−1.60	1.60	1.79	0.05	2.35	−0.14	−2.48
RC21	2.83	3.43	4.11	0.10	12.70	10.57	−2.13
RG18	0.05	0.16	0.21	0.28	0.78	0.57	−0.21
RSE43	0.01	0.26	0.33	0.03	1.66	0.80	−0.86
S22	0.12	0.25	0.35	0.03	1.44	0.96	−0.48
S66	0.01	0.25	0.32	0.05	1.56	0.96	−0.59
SCONF	−0.09	0.17	0.24	0.04	0.79	0.17	−0.62
TAUT15	−0.17	0.57	0.69	0.19	2.39	0.94	−1.45
UPU23	0.40	0.55	0.72	0.10	2.33	1.83	−0.49
W4-11	−11.66	11.69	13.31	0.04	33.35	1.18	−32.17
WATER27	−1.16	1.69	2.81	0.02	9.00	1.12	−7.88
WCPT18	−0.96	1.48	1.94	0.04	6.65	2.26	−4.38
YBDE18	−1.49	2.73	3.18	0.06	9.08	2.22	−6.86
BH76RC	−0.51	1.97	2.54	0.09	9.41	3.56	−5.85

Table S54: Statistical analysis of the TPSS0-DH-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.76	7.76	9.71	0.23	19.51	21.01	1.51
ACONF	−0.06	0.07	0.09	0.04	0.26	0.06	−0.20
ADIM6	−0.10	0.10	0.12	0.03	0.17	0.01	−0.16
AHB21	−0.93	0.94	1.30	0.04	3.43	0.14	−3.29
AL2X6	0.51	0.82	0.91	0.02	2.20	1.32	−0.88
ALK8	2.69	2.69	3.61	0.04	8.21	8.40	0.19
ALKBDE10	−7.71	7.71	8.80	0.08	15.69	−3.11	−18.80
AMINO20x4	0.07	0.20	0.25	0.08	1.13	0.69	−0.44
BH76	−1.96	2.58	3.35	0.14	14.18	4.83	−9.35
BHDIV10	−0.79	1.18	1.49	0.03	5.25	1.95	−3.30
BHPERI	−0.29	1.21	1.51	0.06	5.74	3.74	−2.00
BHROT27	0.55	0.56	0.74	0.09	1.43	1.39	−0.04
BSR36	−4.55	4.55	5.13	0.28	9.10	−2.14	−11.24
BUT14DIOL	−0.04	0.07	0.09	0.03	0.39	0.22	−0.17
C60ISO	2.60	3.25	3.60	0.03	7.91	6.45	−1.46
CARBHB12	0.91	0.91	1.10	0.15	2.01	2.18	0.18
CDIE20	0.89	0.89	1.11	0.22	2.30	2.30	0.00
CHB6	−1.13	1.20	1.47	0.04	2.95	0.21	−2.74
DARC	−2.84	2.84	3.20	0.09	5.08	−1.05	−6.13
DC13	−1.43	6.83	8.44	0.12	29.65	17.18	−12.47
DIPCS10	0.53	3.92	4.76	0.01	16.59	8.24	−8.35
FH51	−1.58	2.23	2.94	0.07	10.82	2.73	−8.09
G21EA	−1.75	3.39	4.12	0.10	12.37	4.72	−7.65
G21IP	0.49	3.25	4.12	0.01	16.24	9.99	−6.25
G2RC	−3.25	5.00	6.26	0.10	25.19	10.05	−15.13
HAL59	−0.02	0.37	0.52	0.08	2.76	1.97	−0.79
HEAVY28	0.01	0.12	0.15	0.10	0.58	0.29	−0.29
HEAVYSB11	−2.73	2.73	2.92	0.05	3.04	−1.43	−4.47
ICONF	0.19	0.30	0.45	0.09	1.41	1.05	−0.37
IDISP	0.35	1.91	2.36	0.13	7.23	2.54	−4.69
IL16	0.08	0.40	0.48	0.00	1.70	0.97	−0.73
INV24	0.51	1.50	2.52	0.05	11.51	7.46	−4.05
ISO34	−0.38	1.33	1.66	0.09	6.18	3.12	−3.07
ISOL24	0.10	1.76	2.29	0.08	10.43	5.52	−4.91
MB16-43	−20.42	23.63	29.25	0.06	99.31	27.20	−72.11
MCONF	0.16	0.20	0.25	0.04	0.88	0.55	−0.33
NBPRC	−0.68	2.93	3.21	0.11	9.72	5.69	−4.04
PA26	4.08	4.08	4.52	0.02	8.31	9.51	1.20
PArel	0.74	0.92	1.30	0.20	4.53	3.73	−0.80
PCONF21	−0.03	0.40	0.47	0.25	1.53	0.58	−0.95
PNICO23	0.16	0.34	0.46	0.08	1.64	1.21	−0.44
PX13	−1.53	1.56	1.77	0.05	2.85	0.19	−2.66
RC21	2.70	3.14	3.91	0.09	13.22	10.97	−2.25
RG18	0.03	0.11	0.14	0.19	0.59	0.32	−0.27
RSE43	−0.10	0.28	0.35	0.04	1.57	0.63	−0.94
S22	0.20	0.44	0.58	0.06	2.40	1.69	−0.70
S66	0.16	0.32	0.45	0.06	2.31	1.74	−0.57
SCONF	0.01	0.20	0.29	0.04	1.09	0.25	−0.83
TAUT15	0.10	0.67	0.78	0.22	2.75	1.41	−1.34
UPU23	0.34	0.46	0.59	0.08	1.59	1.24	−0.35
W4-11	−8.78	11.52	14.36	0.04	59.79	21.55	−38.25
WATER27	2.90	3.14	3.91	0.04	13.88	10.68	−3.20
WCPT18	−0.29	1.61	1.85	0.05	6.80	4.16	−2.64
YBDE18	−2.76	3.28	4.17	0.07	10.15	1.34	−8.82
BH76RC	−1.29	3.63	4.41	0.17	17.40	8.22	−9.19

TPSS0-DH had convergence problems for the h2+_1.75 system in SIE4x4.

Table S55: Statistical analysis of the PBE-QIDH-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	3.50	3.50	3.95	0.10	6.83	8.06	1.23
ACONF	−0.12	0.13	0.16	0.07	0.38	0.07	−0.31
ADIM6	−0.33	0.33	0.38	0.10	0.57	−0.07	−0.64
AHB21	−1.17	1.17	1.54	0.05	3.52	−0.10	−3.63
AL2X6	0.54	0.77	0.85	0.02	1.77	1.19	−0.59
ALK8	2.78	2.78	4.01	0.04	8.92	9.17	0.25
ALKBDE10	−4.52	4.91	5.65	0.05	11.91	1.95	−9.96
AMINO20x4	0.05	0.21	0.26	0.09	1.18	0.63	−0.56
BH76	0.36	1.22	1.99	0.07	13.01	10.41	−2.60
BHDIV10	−1.22	1.85	2.09	0.04	5.28	2.06	−3.22
BHPERI	−2.27	2.74	2.99	0.13	8.17	3.31	−4.86
BHROT27	0.50	0.50	0.65	0.08	1.31	1.26	−0.05
BSR36	−2.22	2.22	2.42	0.14	3.45	−1.15	−4.59
BUT14DIOL	0.02	0.06	0.07	0.02	0.31	0.18	−0.12
C60ISO	−2.24	3.51	4.91	0.04	13.31	2.80	−10.51
CARBHB12	0.85	0.85	1.05	0.14	1.98	2.11	0.13
CDIE20	0.77	0.80	0.96	0.20	2.08	1.96	−0.12
CHB6	−1.23	1.23	1.46	0.05	2.21	−0.35	−2.56
DARC	−6.95	6.95	7.11	0.21	4.69	−4.76	−9.46
DC13	−1.71	8.20	10.27	0.15	34.30	17.17	−17.14
DIPCS10	−0.87	2.74	3.26	0.00	9.26	3.15	−6.10
FH51	−2.53	2.90	3.58	0.09	14.34	5.32	−9.03
G21EA	−1.53	2.64	3.07	0.08	9.11	3.62	−5.49
G21IP	0.15	2.47	3.04	0.01	14.42	6.06	−8.35
G2RC	−4.00	5.30	6.73	0.10	25.65	7.50	−18.14
HAL59	0.02	0.34	0.50	0.07	2.44	1.89	−0.56
HEAVY28	−0.15	0.22	0.23	0.18	0.56	0.20	−0.35
HEAVYSB11	−0.46	0.61	0.79	0.01	1.98	0.45	−1.53
ICONF	0.23	0.25	0.38	0.08	1.09	0.96	−0.13
IDISP	−0.63	1.48	2.40	0.10	6.62	0.93	−5.69
IL16	−0.14	0.27	0.32	0.00	1.06	0.44	−0.62
INV24	0.55	1.33	1.94	0.04	9.84	6.14	−3.70
ISO34	0.24	1.34	1.95	0.09	7.94	4.21	−3.74
ISOL24	1.72	2.79	3.86	0.13	11.86	8.63	−3.23
MB16-43	8.56	12.77	15.74	0.03	64.75	41.96	−22.79
MCONF	0.29	0.30	0.35	0.06	0.78	0.60	−0.18
NBPRC	−1.08	2.63	3.09	0.09	10.59	4.80	−5.79
PA26	1.76	1.98	2.37	0.01	7.04	5.64	−1.40
PArel	0.79	1.03	1.34	0.22	4.59	2.91	−1.68
PCONF21	0.03	0.39	0.48	0.24	1.70	0.72	−0.97
PNICO23	0.25	0.39	0.58	0.09	2.52	2.07	−0.45
PX13	−2.80	2.80	3.00	0.08	3.94	−0.90	−4.83
RC21	2.27	3.05	4.29	0.09	16.39	13.15	−3.24
RG18	−0.07	0.14	0.20	0.24	0.86	0.42	−0.44
RSE43	0.40	0.52	0.92	0.07	4.41	3.88	−0.53
S22	0.19	0.28	0.42	0.04	1.73	1.45	−0.28
S66	0.09	0.29	0.39	0.05	1.93	1.50	−0.43
SCONF	0.07	0.14	0.17	0.03	0.67	0.21	−0.46
TAUT15	0.41	0.68	0.94	0.22	2.74	1.93	−0.81
UPU23	0.40	0.49	0.62	0.09	1.47	1.21	−0.26
W4-11	−4.51	4.79	6.48	0.02	27.21	4.83	−22.38
WATER27	2.82	3.10	3.70	0.04	13.17	9.45	−3.73
WCPT18	−1.68	1.96	2.44	0.06	6.94	2.40	−4.54
YBDE18	1.87	2.69	3.34	0.05	8.85	6.02	−2.84
BH76RC	−0.56	1.91	2.47	0.09	10.63	5.26	−5.38

Table S56: Statistical analysis of the TPSS-QIDH-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	4.40	4.40	6.00	0.13	12.68	13.07	0.40
ACONF	−0.11	0.12	0.15	0.07	0.36	0.06	−0.30
ADIM6	−0.06	0.10	0.13	0.03	0.33	0.06	−0.27
AHB21	−0.91	0.91	1.29	0.04	3.32	−0.01	−3.33
AL2X6	−0.26	0.58	0.67	0.02	1.55	0.51	−1.04
ALK8	1.83	1.85	2.67	0.03	6.68	6.61	−0.07
ALKBDE10	−4.50	4.50	5.47	0.04	11.32	−0.26	−11.58
AMINO20x4	0.01	0.20	0.25	0.08	1.16	0.63	−0.52
BH76	−0.40	1.99	2.73	0.11	17.69	12.29	−5.40
BHDIV10	−0.02	0.79	1.16	0.02	4.68	2.40	−2.28
BHPERI	−0.70	1.27	1.44	0.06	5.03	2.74	−2.29
BHROT27	0.46	0.46	0.62	0.07	1.27	1.20	−0.07
BSR36	−3.64	3.64	4.03	0.22	6.43	−1.74	−8.17
BUT14DIOL	−0.04	0.07	0.09	0.03	0.34	0.16	−0.18
C60ISO	−1.29	3.19	4.24	0.03	12.09	3.26	−8.83
CARBHB12	0.76	0.76	0.93	0.13	1.78	1.90	0.12
CDIE20	0.78	0.79	0.95	0.19	1.92	1.86	−0.05
CHB6	−0.98	1.04	1.23	0.04	2.23	0.18	−2.04
DARC	−2.83	2.83	3.12	0.09	4.26	−1.02	−5.29
DC13	−0.73	5.01	6.49	0.09	23.90	11.72	−12.19
DIPCS10	2.23	3.81	4.68	0.01	13.22	8.48	−4.75
FH51	−1.83	2.11	2.62	0.07	9.39	2.25	−7.14
G21EA	−0.50	2.48	2.89	0.07	10.04	5.08	−4.97
G21IP	1.58	2.82	3.48	0.01	13.48	8.71	−4.77
G2RC	−3.79	4.58	5.61	0.09	17.73	5.60	−12.13
HAL59	−0.16	0.35	0.43	0.08	2.02	1.30	−0.72
HEAVY28	−0.23	0.25	0.29	0.20	0.60	0.14	−0.46
HEAVYSB11	−1.80	1.80	2.10	0.03	3.44	−0.33	−3.77
ICONF	0.15	0.26	0.37	0.08	1.40	0.84	−0.56
IDISP	0.01	1.63	1.91	0.11	5.76	2.10	−3.67
IL16	0.24	0.37	0.44	0.00	1.55	1.10	−0.46
INV24	0.56	1.29	2.09	0.04	10.62	7.08	−3.54
ISO34	0.14	0.99	1.40	0.07	6.08	3.63	−2.44
ISOL24	0.54	1.44	2.06	0.07	9.28	5.43	−3.86
MB16-43	−15.05	19.11	25.04	0.05	97.45	23.57	−73.89
MCONF	0.28	0.29	0.32	0.06	0.69	0.55	−0.14
NBPRC	0.02	2.52	3.05	0.09	9.18	6.38	−2.79
PA26	2.78	2.78	3.21	0.01	7.11	7.23	0.12
PArel	0.60	0.73	1.03	0.16	3.05	2.45	−0.60
PCONF21	0.03	0.34	0.40	0.21	1.37	0.59	−0.78
PNICO23	0.03	0.24	0.31	0.06	1.25	0.73	−0.52
PX13	−1.10	1.23	1.41	0.04	2.80	0.54	−2.27
RC21	1.07	1.93	2.87	0.05	12.56	9.25	−3.31
RG18	−0.02	0.12	0.19	0.21	0.92	0.57	−0.35
RSE43	0.50	0.55	0.97	0.07	4.44	3.97	−0.47
S22	0.22	0.27	0.40	0.04	1.57	1.34	−0.24
S66	0.16	0.24	0.35	0.04	1.60	1.39	−0.21
SCONF	−0.08	0.11	0.15	0.02	0.59	0.10	−0.49
TAUT15	0.22	0.51	0.66	0.17	2.23	1.17	−1.06
UPU23	0.40	0.50	0.61	0.09	1.53	1.17	−0.36
W4-11	−4.91	9.02	11.32	0.03	56.74	23.44	−33.30
WATER27	2.02	2.23	2.67	0.03	10.23	7.37	−2.86
WCPT18	−0.09	1.28	1.51	0.04	5.92	3.95	−1.97
YBDE18	−0.80	2.74	3.05	0.06	9.06	3.51	−5.55
BH76RC	−1.50	2.89	3.68	0.14	16.02	5.33	−10.69

TPSS-QIDH had convergence problems for the h2+_1.75 system in SIE4x4.

Table S57: Statistical analysis of the PBE-CIDH-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	6.47	6.47	7.32	0.19	12.94	15.08	2.14
ACONF	−0.05	0.08	0.10	0.04	0.32	0.09	−0.22
ADIM6	−0.11	0.12	0.14	0.04	0.24	0.01	−0.23
AHB21	−1.26	1.26	1.62	0.06	3.61	−0.09	−3.69
AL2X6	0.55	0.95	1.00	0.03	2.13	1.25	−0.88
ALK8	3.17	3.17	4.29	0.05	8.15	8.60	0.44
ALKBDE10	−6.00	6.12	7.19	0.06	14.79	0.59	−14.20
AMINO20x4	0.07	0.22	0.29	0.09	1.34	0.76	−0.58
BH76	−0.93	1.59	1.87	0.09	9.44	4.72	−4.72
BHDIV10	−1.76	2.29	2.62	0.05	6.66	2.24	−4.42
BHPERI	−1.63	2.25	2.52	0.11	7.78	3.76	−4.02
BHROT27	0.56	0.56	0.73	0.09	1.45	1.41	−0.04
BSR36	−3.19	3.19	3.61	0.20	6.56	−1.47	−8.04
BUT14DIOL	0.06	0.08	0.10	0.03	0.43	0.33	−0.10
C60ISO	0.87	2.70	3.05	0.03	8.50	3.95	−4.55
CARBHB12	1.03	1.03	1.25	0.17	2.19	2.40	0.21
CDIE20	0.87	0.90	1.12	0.22	2.53	2.33	−0.20
CHB6	−1.27	1.27	1.58	0.05	2.86	−0.11	−2.97
DARC	−6.42	6.42	6.67	0.20	6.00	−4.17	−10.17
DC13	−2.09	8.36	10.37	0.15	36.43	19.58	−16.85
DIPCS10	−0.62	2.64	3.28	0.00	10.83	3.97	−6.85
FH51	−2.46	3.04	3.79	0.10	15.03	4.68	−10.35
G21EA	−1.51	2.94	3.39	0.09	9.00	2.99	−6.01
G21IP	0.10	2.90	3.56	0.01	15.29	7.38	−7.90
G2RC	−4.15	6.10	7.70	0.12	30.60	10.28	−20.33
HAL59	0.07	0.36	0.56	0.08	2.77	2.19	−0.58
HEAVY28	−0.05	0.16	0.18	0.13	0.61	0.33	−0.28
HEAVYSB11	−1.35	1.35	1.45	0.02	1.60	−0.65	−2.25
ICONF	0.26	0.30	0.46	0.09	1.26	1.14	−0.12
IDISP	−0.21	1.84	2.10	0.13	5.56	2.11	−3.44
IL16	−0.24	0.32	0.41	0.00	1.21	0.36	−0.85
INV24	0.38	1.42	2.13	0.04	9.98	5.76	−4.21
ISO34	0.05	1.39	1.96	0.10	7.82	3.95	−3.87
ISOL24	1.33	2.44	3.47	0.11	12.94	8.22	−4.73
MB16-43	4.94	12.72	16.38	0.03	73.15	49.85	−23.30
MCONF	0.20	0.23	0.29	0.05	0.85	0.61	−0.25
NBPRC	−1.11	3.06	3.42	0.11	11.25	5.57	−5.68
PA26	2.41	2.50	2.99	0.01	8.08	6.90	−1.18
PArel	0.87	1.14	1.51	0.25	5.56	3.86	−1.70
PCONF21	−0.04	0.51	0.60	0.31	1.97	0.84	−1.13
PNICO23	0.33	0.45	0.68	0.11	2.94	2.47	−0.48
PX13	−3.39	3.39	3.59	0.10	4.31	−1.41	−5.72
RC21	3.63	4.18	5.25	0.12	16.81	14.47	−2.34
RG18	−0.01	0.12	0.16	0.21	0.64	0.28	−0.36
RSE43	−0.23	0.31	0.46	0.04	2.36	1.03	−1.33
S22	0.22	0.45	0.59	0.06	2.46	1.69	−0.77
S66	0.19	0.34	0.48	0.06	2.32	1.78	−0.54
SCONF	0.05	0.14	0.19	0.03	0.77	0.21	−0.56
TAUT15	0.33	0.77	1.08	0.25	3.41	2.26	−1.15
UPU23	0.36	0.48	0.59	0.08	1.53	1.16	−0.37
W4-11	−5.36	5.62	7.35	0.02	29.49	3.53	−25.95
WATER27	4.25	4.55	5.79	0.06	18.76	14.70	−4.06
WCPT18	−2.01	2.26	2.88	0.06	7.75	2.14	−5.61
YBDE18	0.61	1.93	2.37	0.04	7.74	3.79	−3.95
BH76RC	−0.56	2.11	2.67	0.10	10.50	5.23	−5.27

Table S58: Statistical analysis of the TPSS-CIDH-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	6.86	6.86	8.74	0.20	17.81	18.99	1.18
ACONF	−0.07	0.09	0.11	0.05	0.28	0.06	−0.22
ADIM6	−0.14	0.14	0.16	0.04	0.23	−0.01	−0.23
AHB21	−0.95	0.96	1.32	0.04	3.47	0.10	−3.37
AL2X6	0.37	0.72	0.79	0.02	2.01	1.16	−0.85
ALK8	2.60	2.60	3.55	0.04	8.26	8.42	0.16
ALKBDE10	−7.03	7.03	8.07	0.07	14.70	−2.56	−17.26
AMINO20x4	0.06	0.19	0.25	0.08	1.14	0.69	−0.46
BH76	−1.60	2.39	3.14	0.13	14.97	6.61	−8.36
BHDIV10	−0.58	1.00	1.36	0.02	5.10	2.07	−3.04
BHPERI	−0.32	1.20	1.47	0.06	5.45	3.44	−2.01
BHROT27	0.54	0.54	0.72	0.09	1.41	1.37	−0.05
BSR36	−4.28	4.28	4.81	0.26	8.28	−2.09	−10.37
BUT14DIOL	−0.04	0.07	0.08	0.03	0.35	0.19	−0.16
C60ISO	1.79	3.04	3.28	0.03	8.44	5.45	−2.99
CARBHB12	0.88	0.88	1.07	0.15	1.96	2.13	0.17
CDIE20	0.85	0.85	1.06	0.21	2.20	2.19	−0.01
CHB6	−1.13	1.18	1.44	0.04	2.75	0.13	−2.62
DARC	−2.95	2.95	3.28	0.09	4.84	−1.19	−6.03
DC13	−1.29	6.30	7.92	0.11	28.34	15.93	−12.41
DIPCS10	1.11	3.85	4.72	0.01	15.90	8.53	−7.37
FH51	−1.73	2.21	2.88	0.07	10.97	2.43	−8.54
G21EA	−1.43	3.16	3.80	0.09	11.45	4.71	−6.73
G21IP	0.81	3.09	3.92	0.01	15.17	9.74	−5.43
G2RC	−3.52	4.94	6.18	0.10	23.62	9.10	−14.52
HAL59	−0.03	0.35	0.49	0.08	2.59	1.84	−0.74
HEAVY28	−0.02	0.12	0.15	0.10	0.54	0.26	−0.28
HEAVYSB11	−2.50	2.50	2.72	0.04	3.21	−1.17	−4.38
ICONF	0.18	0.28	0.43	0.09	1.39	1.01	−0.38
IDISP	0.16	1.75	2.31	0.12	7.09	2.30	−4.79
IL16	0.09	0.36	0.45	0.00	1.57	0.89	−0.68
INV24	0.56	1.46	2.45	0.05	11.47	7.48	−3.99
ISO34	−0.25	1.22	1.56	0.08	6.13	3.20	−2.93
ISOL24	0.26	1.66	2.22	0.08	9.93	5.57	−4.36
MB16-43	−19.62	22.98	28.91	0.06	100.34	24.93	−75.41
MCONF	0.18	0.21	0.26	0.04	0.82	0.54	−0.28
NBPRC	−0.56	2.91	3.23	0.11	9.65	5.72	−3.92
PA26	3.79	3.79	4.23	0.02	7.96	9.01	1.05
PArel	0.73	0.88	1.25	0.19	4.18	3.45	−0.73
PCONF21	−0.01	0.39	0.45	0.24	1.48	0.59	−0.89
PNICO23	0.14	0.32	0.42	0.07	1.54	1.10	−0.43
PX13	−1.34	1.41	1.59	0.04	2.74	0.33	−2.41
RC21	2.41	2.88	3.67	0.08	13.25	10.77	−2.49
RG18	0.01	0.11	0.15	0.19	0.67	0.36	−0.30
RSE43	0.02	0.24	0.33	0.03	1.98	1.16	−0.82
S22	0.20	0.41	0.55	0.06	2.27	1.63	−0.63
S66	0.15	0.31	0.44	0.06	2.20	1.69	−0.51
SCONF	0.00	0.17	0.24	0.04	0.93	0.21	−0.72
TAUT15	0.13	0.64	0.75	0.21	2.66	1.36	−1.30
UPU23	0.35	0.47	0.59	0.08	1.57	1.24	−0.33
W4-11	−7.89	11.05	13.73	0.04	59.86	22.53	−37.32
WATER27	2.75	2.97	3.69	0.04	13.23	10.18	−3.05
WCPT18	−0.20	1.55	1.77	0.04	6.66	4.20	−2.46
YBDE18	−2.28	3.11	3.85	0.06	10.12	1.96	−8.16
BH76RC	−1.37	3.49	4.27	0.16	17.37	7.54	−9.83

TPSS-CIDH had convergence problems for the h2+_1.75 system in SIE4x4.

Table S59: Statistical analysis of the SOS0-PBE0-2-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	1.18	1.89	2.32	0.06	6.93	5.21	−1.71
ACONF	−0.11	0.13	0.16	0.07	0.42	0.08	−0.34
ADIM6	−0.56	0.56	0.63	0.17	0.83	−0.18	−1.01
AHB21	−0.18	0.50	0.68	0.02	2.65	0.56	−2.09
AL2X6	−1.33	1.33	1.39	0.04	1.16	−0.96	−2.12
ALK8	0.71	1.13	2.08	0.02	6.78	5.70	−1.07
ALKBDE10	−5.23	5.23	5.61	0.05	6.46	−2.22	−8.68
AMINO20x4	−0.07	0.19	0.24	0.08	1.11	0.45	−0.66
BH76	2.43	2.54	3.71	0.14	18.72	17.21	−1.50
BHDIV10	0.99	1.03	1.26	0.02	2.38	2.21	−0.17
BHPERI	1.51	1.51	1.90	0.07	4.45	4.63	0.18
BHROT27	0.22	0.26	0.33	0.04	0.85	0.65	−0.19
BSR36	−3.96	3.96	4.36	0.24	6.86	−2.03	−8.89
BUT14DIOL	−0.25	0.26	0.28	0.09	0.51	0.08	−0.43
C60ISO	−1.42	2.64	3.33	0.03	8.97	2.04	−6.93
CARBHB12	0.11	0.20	0.29	0.03	1.12	0.82	−0.30
CDIE20	0.26	0.37	0.51	0.09	1.53	1.23	−0.30
CHB6	−0.83	0.93	1.10	0.03	2.04	0.29	−1.75
DARC	−3.74	3.74	3.81	0.12	2.33	−2.81	−5.14
DC13	−1.87	4.15	5.11	0.08	18.25	7.15	−11.09
DIPCS10	−1.97	3.10	3.60	0.00	10.51	2.88	−7.63
FH51	−1.35	1.68	2.20	0.05	10.57	2.30	−8.27
G21EA	−2.09	2.79	3.33	0.08	13.60	8.21	−5.39
G21IP	−0.11	1.96	2.81	0.01	16.37	10.82	−5.54
G2RC	−2.11	2.93	3.52	0.06	12.24	4.23	−8.01
HAL59	−0.60	0.61	0.67	0.13	1.68	0.27	−1.41
HEAVY28	−0.45	0.45	0.48	0.36	0.64	−0.07	−0.72
HEAVYSB11	−1.11	1.18	1.54	0.02	3.50	0.35	−3.15
ICONF	0.05	0.18	0.24	0.06	0.90	0.50	−0.40
IDISP	−0.24	1.51	1.87	0.11	5.55	2.10	−3.46
IL16	1.04	1.04	1.12	0.01	1.53	1.93	0.41
INV24	0.89	1.29	2.27	0.04	11.59	8.95	−2.64
ISO34	−0.08	0.86	1.19	0.06	5.31	2.91	−2.40
ISOL24	0.68	1.45	1.98	0.07	8.23	4.84	−3.39
MB16-43	−15.25	16.35	20.58	0.04	72.99	14.56	−58.43
MCONF	−0.01	0.07	0.08	0.01	0.40	0.25	−0.15
NBPRC	0.25	1.27	1.70	0.05	5.03	3.47	−1.55
PA26	3.04	3.04	3.28	0.02	5.00	6.03	1.03
PArel	0.35	0.43	0.64	0.09	2.16	1.61	−0.56
PCONF21	0.00	0.41	0.49	0.25	1.73	1.01	−0.73
PNICO23	−0.40	0.41	0.46	0.10	1.19	0.07	−1.12
PX13	0.16	0.54	0.64	0.02	1.91	1.26	−0.65
RC21	−0.82	2.11	2.67	0.06	10.68	5.56	−5.12
RG18	−0.21	0.23	0.31	0.40	0.87	0.16	−0.71
RSE43	1.52	1.52	2.43	0.20	10.25	10.47	0.22
S22	−0.41	0.43	0.61	0.06	1.76	0.20	−1.57
S66	−0.36	0.38	0.47	0.07	1.49	0.11	−1.39
SCONF	−0.21	0.32	0.36	0.07	1.05	0.53	−0.52
TAUT15	−0.32	0.53	0.57	0.17	1.34	0.53	−0.81
UPU23	0.56	0.62	0.77	0.11	1.85	1.53	−0.32
W4-11	−3.48	4.70	6.97	0.02	38.52	6.87	−31.65
WATER27	−1.31	1.53	2.51	0.02	8.19	0.66	−7.52
WCPT18	1.12	1.12	1.41	0.03	3.50	3.53	0.03
YBDE18	1.23	1.54	1.80	0.03	4.79	3.43	−1.36
BH76RC	−0.45	1.45	1.94	0.07	7.89	2.96	−4.93

Table S60: Statistical analysis of the SOS0-PBE0-DH-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.41	7.41	8.52	0.22	15.81	18.15	2.34
ACONF	−0.04	0.06	0.08	0.03	0.25	0.06	−0.19
ADIM6	0.02	0.03	0.04	0.01	0.12	0.09	−0.03
AHB21	−1.08	1.08	1.42	0.05	3.35	0.01	−3.34
AL2X6	0.27	0.81	0.82	0.02	1.98	0.95	−1.03
ALK8	2.94	2.94	3.93	0.05	7.44	7.87	0.43
ALKBDE10	−7.26	7.26	8.27	0.07	15.16	−1.48	−16.64
AMINO20x4	0.06	0.20	0.26	0.08	1.24	0.69	−0.55
BH76	−1.02	1.63	1.88	0.09	7.66	3.81	−3.85
BHDIV10	−1.36	1.76	2.17	0.04	5.90	1.77	−4.13
BHPERI	−0.27	1.40	1.77	0.07	7.16	5.10	−2.06
BHROT27	0.51	0.52	0.67	0.08	1.30	1.26	−0.03
BSR36	−4.15	4.15	4.79	0.26	9.60	−1.73	−11.34
BUT14DIOL	−0.01	0.08	0.10	0.03	0.49	0.33	−0.16
C60ISO	3.14	3.15	3.79	0.03	7.27	7.23	−0.04
CARBHB12	0.97	0.97	1.17	0.16	2.01	2.23	0.23
CDIE20	0.74	0.79	1.04	0.19	2.56	2.28	−0.28
CHB6	−1.30	1.30	1.63	0.05	3.01	−0.07	−3.08
DARC	−5.52	5.52	5.75	0.17	5.73	−3.64	−9.37
DC13	−2.42	7.63	9.34	0.14	32.47	18.61	−13.86
DIPCS10	−0.81	2.73	3.51	0.00	12.15	4.42	−7.73
FH51	−2.17	2.90	3.71	0.09	14.64	3.33	−11.31
G21EA	−1.75	2.73	3.38	0.08	9.49	2.43	−7.06
G21IP	−0.04	2.90	3.53	0.01	14.41	7.56	−6.85
G2RC	−3.74	5.94	7.47	0.12	29.66	11.10	−18.56
HAL59	0.04	0.33	0.50	0.07	2.66	1.99	−0.67
HEAVY28	0.05	0.12	0.16	0.10	0.58	0.36	−0.22
HEAVYSB11	−1.95	1.95	2.02	0.03	1.44	−1.28	−2.72
ICONF	0.22	0.30	0.46	0.09	1.36	1.15	−0.21
IDISP	0.15	1.99	2.25	0.14	6.66	3.01	−3.66
IL16	−0.15	0.34	0.44	0.00	1.38	0.49	−0.89
INV24	0.44	1.47	2.35	0.05	11.44	7.13	−4.31
ISO34	−0.14	1.30	1.77	0.09	7.06	3.52	−3.54
ISOL24	0.89	2.14	2.96	0.10	12.48	7.50	−4.98
MB16-43	−2.60	13.24	16.80	0.03	76.72	43.10	−33.62
MCONF	0.14	0.19	0.23	0.04	0.80	0.55	−0.24
NBPRC	−0.88	3.02	3.29	0.11	10.44	5.67	−4.77
PA26	3.16	3.19	3.64	0.02	8.13	7.68	−0.45
PArel	0.77	1.00	1.41	0.22	5.56	4.21	−1.35
PCONF21	−0.03	0.44	0.52	0.27	1.68	0.67	−1.01
PNICO23	0.25	0.38	0.54	0.09	2.30	1.85	−0.45
PX13	−2.83	2.83	3.03	0.08	3.72	−1.04	−4.76
RC21	3.53	4.08	4.92	0.11	14.82	12.85	−1.97
RG18	0.04	0.12	0.15	0.21	0.61	0.33	−0.28
RSE43	−0.21	0.27	0.40	0.04	1.71	0.42	−1.29
S22	0.20	0.47	0.59	0.06	2.47	1.60	−0.88
S66	0.22	0.34	0.47	0.06	2.30	1.69	−0.61
SCONF	−0.04	0.11	0.16	0.02	0.69	0.16	−0.53
TAUT15	0.02	0.72	0.89	0.24	2.90	1.61	−1.29
UPU23	0.38	0.49	0.62	0.09	1.60	1.22	−0.39
W4-11	−5.91	6.25	8.22	0.02	31.81	3.51	−28.30
WATER27	3.97	4.21	5.37	0.05	16.87	13.64	−3.24
WCPT18	−1.35	1.88	2.39	0.05	7.10	2.29	−4.80
YBDE18	−0.13	1.49	1.82	0.03	6.36	2.20	−4.16
BH76RC	−0.54	2.09	2.62	0.10	8.46	3.36	−5.09

Table S61: Statistical analysis of the SOS1-PBE-QIDH-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	3.20	3.20	4.08	0.09	9.39	9.35	−0.05
ACONF	−0.21	0.22	0.23	0.12	0.35	0.01	−0.34
ADIM6	0.04	0.04	0.05	0.01	0.09	0.09	0.00
AHB21	−0.78	0.79	1.15	0.04	3.14	0.05	−3.09
AL2X6	−0.26	0.43	0.64	0.01	1.63	0.28	−1.35
ALK8	1.76	1.76	2.34	0.03	4.87	5.00	0.14
ALKBDE10	−6.62	6.62	7.20	0.07	10.88	−2.22	−13.10
AMINO20x4	0.01	0.18	0.22	0.07	1.05	0.48	−0.57
BH76	0.97	1.46	2.31	0.08	14.15	11.92	−2.23
BHDIV10	−0.01	1.09	1.17	0.02	3.17	1.18	−1.99
BHPERI	0.41	1.30	1.75	0.06	6.81	5.67	−1.14
BHROT27	0.37	0.38	0.51	0.06	1.14	1.09	−0.04
BSR36	−3.64	3.64	4.13	0.22	7.68	−1.64	−9.32
BUT14DIOL	−0.06	0.09	0.11	0.03	0.38	0.16	−0.23
C60ISO	1.39	2.43	2.82	0.02	8.01	5.48	−2.53
CARBHB12	0.57	0.57	0.70	0.09	1.45	1.53	0.07
CDIE20	0.40	0.48	0.68	0.12	1.85	1.62	−0.23
CHB6	−0.85	1.12	1.26	0.04	2.95	0.81	−2.15
DARC	−6.23	6.23	6.31	0.19	3.52	−4.93	−8.44
DC13	−2.73	6.28	7.60	0.11	26.51	12.22	−14.29
DIPCS10	−1.14	2.87	3.50	0.00	11.93	4.31	−7.62
FH51	−2.14	2.57	3.35	0.08	15.98	3.80	−12.18
G21EA	−2.13	2.58	3.21	0.08	11.22	4.52	−6.71
G21IP	−0.02	2.04	2.71	0.01	11.67	5.65	−6.02
G2RC	−3.32	4.70	5.81	0.09	21.58	7.24	−14.34
HAL59	−0.40	0.48	0.53	0.10	2.05	1.00	−1.05
HEAVY28	−0.38	0.39	0.43	0.31	0.85	0.14	−0.71
HEAVYSB11	−1.47	1.47	1.70	0.03	2.93	−0.22	−3.14
ICONF	0.15	0.22	0.33	0.07	1.00	0.83	−0.17
IDISP	−0.42	1.91	2.47	0.13	7.46	2.37	−5.09
IL16	0.28	0.45	0.55	0.00	1.76	1.23	−0.53
INV24	0.99	1.52	2.55	0.05	12.36	8.75	−3.62
ISO34	0.00	1.08	1.55	0.07	6.85	3.81	−3.04
ISOL24	1.42	2.21	3.23	0.10	12.23	7.81	−4.41
MB16-43	−9.32	14.54	18.67	0.04	76.74	23.03	−53.72
MCONF	0.17	0.19	0.24	0.04	0.79	0.57	−0.22
NBPRC	−0.64	2.59	2.98	0.09	10.30	5.09	−5.22
PA26	3.49	3.49	3.81	0.02	6.15	7.18	1.03
PArel	0.59	0.76	1.01	0.16	3.71	2.54	−1.17
PCONF21	−0.05	0.34	0.44	0.21	1.62	0.59	−1.03
PNICO23	−0.01	0.27	0.36	0.06	1.63	0.68	−0.95
PX13	−1.30	1.33	1.53	0.04	2.46	0.09	−2.37
RC21	1.78	2.56	3.47	0.07	13.56	10.41	−3.15
RG18	−0.12	0.16	0.24	0.28	0.88	0.25	−0.63
RSE43	0.76	0.77	1.16	0.10	4.74	4.66	−0.07
S22	0.12	0.33	0.42	0.05	1.87	1.21	−0.66
S66	0.12	0.26	0.34	0.05	1.79	1.26	−0.53
SCONF	−0.03	0.10	0.13	0.02	0.57	0.20	−0.37
TAUT15	−0.17	0.51	0.56	0.17	1.38	0.47	−0.91
UPU23	0.52	0.57	0.71	0.10	1.45	1.31	−0.13
W4-11	−4.81	5.78	7.99	0.02	36.42	7.98	−28.44
WATER27	2.70	2.84	3.50	0.04	10.58	8.66	−1.93
WCPT18	−0.05	1.46	1.61	0.04	5.64	3.44	−2.20
YBDE18	1.19	1.78	2.14	0.04	6.88	3.86	−3.02
BH76RC	−0.61	1.82	2.36	0.09	8.87	3.23	−5.65

Table S62: Statistical analysis of the SOS0-PBE-CIDH-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	6.18	6.18	7.21	0.18	13.73	15.72	1.99
ACONF	−0.06	0.08	0.10	0.04	0.27	0.05	−0.22
ADIM6	−0.03	0.05	0.06	0.01	0.13	0.06	−0.08
AHB21	−1.01	1.02	1.37	0.05	3.43	0.04	−3.39
AL2X6	0.26	0.71	0.72	0.02	1.74	0.91	−0.84
ALK8	2.82	2.82	3.80	0.05	7.68	8.05	0.37
ALKBDE10	−7.42	7.42	8.30	0.07	14.51	−1.89	−16.41
AMINO20x4	0.05	0.20	0.25	0.08	1.18	0.64	−0.54
BH76	−0.44	1.29	1.65	0.07	9.13	5.76	−3.37
BHDIV10	−0.91	1.40	1.76	0.03	5.08	1.57	−3.50
BHPERI	0.09	1.33	1.74	0.06	7.16	5.47	−1.69
BHROT27	0.49	0.49	0.64	0.08	1.26	1.22	−0.04
BSR36	−4.03	4.03	4.63	0.25	9.03	−1.76	−10.79
BUT14DIOL	−0.04	0.08	0.10	0.03	0.44	0.25	−0.19
C60ISO	3.01	3.05	3.71	0.03	7.43	7.25	−0.17
CARBHB12	0.88	0.88	1.05	0.15	1.87	2.06	0.20
CDIE20	0.65	0.71	0.95	0.17	2.42	2.14	−0.28
CHB6	−1.32	1.32	1.63	0.05	2.88	−0.16	−3.03
DARC	−5.75	5.75	5.93	0.18	5.14	−4.05	−9.19
DC13	−2.51	7.29	9.01	0.13	31.70	17.39	−14.31
DIPCS10	−0.79	2.78	3.53	0.00	12.44	4.70	−7.74
FH51	−2.21	2.84	3.64	0.09	14.92	3.32	−11.61
G21EA	−1.91	2.66	3.39	0.08	9.41	2.15	−7.27
G21IP	−0.02	2.70	3.33	0.01	13.76	7.16	−6.61
G2RC	−3.71	5.70	7.13	0.11	28.03	10.47	−17.56
HAL59	−0.01	0.32	0.45	0.07	2.48	1.80	−0.68
HEAVY28	0.02	0.11	0.14	0.09	0.52	0.30	−0.22
HEAVYSB11	−1.85	1.85	1.94	0.03	1.88	−0.98	−2.87
ICONF	0.20	0.29	0.44	0.09	1.29	1.09	−0.20
IDISP	−0.02	2.02	2.24	0.14	6.03	2.67	−3.35
IL16	−0.08	0.32	0.41	0.00	1.40	0.58	−0.82
INV24	0.60	1.50	2.46	0.05	11.54	7.24	−4.30
ISO34	−0.12	1.25	1.73	0.09	7.00	3.60	−3.40
ISOL24	1.00	2.20	3.05	0.10	11.58	7.33	−4.25
MB16-43	−3.38	13.14	16.67	0.03	77.55	38.95	−38.60
MCONF	0.17	0.19	0.24	0.04	0.77	0.56	−0.21
NBPRC	−0.83	2.91	3.18	0.11	10.32	5.59	−4.73
PA26	3.21	3.22	3.65	0.02	7.72	7.54	−0.17
PArel	0.75	0.95	1.34	0.21	5.10	3.89	−1.22
PCONF21	0.00	0.39	0.45	0.24	1.52	0.61	−0.92
PNICO23	0.19	0.33	0.46	0.08	1.96	1.51	−0.45
PX13	−2.24	2.24	2.45	0.07	3.32	−0.55	−3.86
RC21	3.14	3.71	4.55	0.10	14.28	12.42	−1.86
RG18	0.02	0.11	0.15	0.19	0.67	0.35	−0.32
RSE43	0.01	0.23	0.32	0.03	1.81	0.89	−0.92
S22	0.19	0.42	0.53	0.06	2.25	1.49	−0.76
S66	0.19	0.31	0.43	0.06	2.13	1.58	−0.56
SCONF	−0.06	0.10	0.14	0.02	0.53	0.12	−0.41
TAUT15	−0.03	0.66	0.77	0.22	2.44	1.26	−1.18
UPU23	0.38	0.49	0.62	0.09	1.55	1.19	−0.36
W4-11	−6.18	6.52	8.60	0.02	31.52	3.97	−27.55
WATER27	3.38	3.59	4.52	0.04	14.37	11.61	−2.76
WCPT18	−0.87	1.66	2.04	0.05	6.67	2.77	−3.90
YBDE18	0.09	1.59	1.96	0.03	7.16	2.82	−4.35
BH76RC	−0.58	2.04	2.56	0.10	8.01	2.86	−5.15

Table S63: Statistical analysis of the SOS0-PBE0-2(b)-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	4.45	4.45	4.94	0.13	7.46	8.53	1.07
ACONF	−0.32	0.32	0.33	0.17	0.41	−0.02	−0.43
ADIM6	−0.46	0.46	0.51	0.14	0.55	−0.20	−0.75
AHB21	−0.47	0.52	0.67	0.02	2.32	0.24	−2.07
AL2X6	0.29	0.80	0.92	0.02	2.73	1.17	−1.56
ALK8	2.60	3.63	6.12	0.06	19.07	15.56	−3.51
ALKBDE10	9.86	9.86	11.33	0.10	14.87	18.69	3.82
AMINO20x4	−0.06	0.14	0.18	0.06	0.73	0.30	−0.43
BH76	0.56	2.13	3.87	0.11	25.47	21.03	−4.44
BHDIV10	−2.05	2.51	2.85	0.06	7.16	2.33	−4.84
BHPERI	−3.05	3.20	3.46	0.15	7.22	1.93	−5.29
BHROT27	−0.01	0.13	0.17	0.02	0.75	0.24	−0.52
BSR36	−1.29	1.29	1.34	0.08	1.38	−0.82	−2.20
BUT14DIOL	−0.01	0.05	0.07	0.02	0.30	0.20	−0.10
C60ISO	−13.18	13.18	15.53	0.13	23.88	−3.36	−27.25
CARBHB12	0.49	0.49	0.64	0.08	1.26	1.31	0.05
CDIE20	0.29	0.32	0.40	0.08	1.15	1.00	−0.14
CHB6	−0.99	0.99	1.17	0.04	1.90	−0.30	−2.20
DARC	−5.26	5.26	5.32	0.16	2.36	−3.95	−6.31
DC13	−2.57	9.26	12.03	0.17	47.97	20.91	−27.06
DIPCS10	6.79	6.79	7.24	0.01	7.48	10.69	3.21
FH51	−0.75	1.51	2.16	0.05	12.40	5.83	−6.57
G21EA	4.53	5.41	6.53	0.16	20.55	16.50	−4.05
G21IP	5.18	5.72	7.37	0.02	32.47	24.24	−8.23
G2RC	−0.58	2.47	3.34	0.05	15.86	7.12	−8.74
HAL59	0.15	0.32	0.48	0.07	2.35	1.91	−0.44
HEAVY28	−0.09	0.12	0.16	0.10	0.50	0.18	−0.33
HEAVYSB11	6.78	6.78	6.83	0.12	3.03	7.85	4.82
ICONF	−0.03	0.17	0.23	0.05	1.05	0.40	−0.64
IDISP	−0.72	3.37	4.33	0.24	13.71	4.99	−8.72
IL16	−0.29	0.35	0.42	0.00	1.18	0.28	−0.89
INV24	−0.14	0.80	1.60	0.03	8.84	1.62	−7.21
ISO34	−0.28	1.03	1.52	0.07	7.13	3.22	−3.91
ISOL24	1.06	2.19	3.03	0.10	11.86	8.23	−3.62
MB16-43	10.07	12.68	15.32	0.03	50.21	32.68	−17.52
MCONF	0.39	0.43	0.50	0.09	1.14	0.88	−0.25
NBPRC	−1.02	2.08	2.48	0.08	7.80	2.25	−5.56
PA26	1.69	1.74	1.86	0.01	3.71	3.08	−0.63
PArel	−0.02	0.72	1.13	0.16	4.85	1.85	−3.00
PCONF21	−0.11	0.36	0.43	0.22	1.53	0.66	−0.87
PNICO23	0.09	0.23	0.38	0.05	1.84	1.52	−0.32
PX13	−4.17	4.17	4.29	0.13	4.22	−2.13	−6.35
RC21	−0.94	2.67	3.30	0.07	13.04	7.99	−5.06
RG18	−0.13	0.18	0.25	0.31	0.94	0.36	−0.57
RSE43	1.54	1.58	3.47	0.21	16.07	15.63	−0.44
S22	−0.01	0.27	0.37	0.04	1.72	0.72	−0.99
S66	−0.10	0.21	0.27	0.04	1.05	0.39	−0.65
SCONF	0.14	0.30	0.33	0.07	1.11	0.40	−0.71
TAUT15	−0.29	0.64	0.80	0.21	2.47	0.88	−1.59
UPU23	0.22	0.35	0.44	0.06	1.46	1.05	−0.41
W4-11	28.43	28.49	32.76	0.09	71.51	68.68	−2.83
WATER27	0.87	1.25	1.69	0.02	9.65	4.76	−4.90
WCPT18	−2.72	2.72	2.83	0.08	2.77	−1.81	−4.59
YBDE18	9.28	9.28	9.89	0.19	11.58	16.87	5.29
BH76RC	−0.24	1.84	2.51	0.09	12.13	8.87	−3.26

Table S64: Statistical analysis of the SOS0-PBE0-DH(b)-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	8.48	8.48	9.55	0.25	16.82	19.16	2.34
ACONF	−0.08	0.08	0.10	0.04	0.25	0.04	−0.21
ADIM6	−0.04	0.06	0.06	0.02	0.14	0.05	−0.09
AHB21	−1.10	1.10	1.37	0.05	2.86	−0.09	−2.95
AL2X6	0.49	1.05	1.10	0.03	2.65	1.35	−1.30
ALK8	3.20	3.20	4.47	0.05	9.17	9.50	0.33
ALKBDE10	−2.74	3.65	4.36	0.04	12.04	4.24	−7.80
AMINO20x4	0.06	0.19	0.25	0.08	1.14	0.68	−0.46
BH76	−1.85	2.29	2.52	0.12	10.38	3.52	−6.87
BHDIV10	−2.32	2.63	3.04	0.06	6.43	1.23	−5.20
BHPERI	−1.41	1.94	2.19	0.09	6.98	3.62	−3.36
BHROT27	0.43	0.43	0.56	0.07	1.10	1.07	−0.03
BSR36	−3.62	3.62	4.15	0.22	7.98	−1.63	−9.61
BUT14DIOL	0.06	0.08	0.11	0.03	0.47	0.37	−0.10
C60ISO	−0.79	2.38	3.03	0.02	8.55	2.14	−6.42
CARBHB12	1.02	1.02	1.23	0.17	2.08	2.30	0.22
CDIE20	0.77	0.80	1.01	0.20	2.35	2.12	−0.23
CHB6	−1.21	1.21	1.50	0.05	2.74	−0.07	−2.81
DARC	−5.51	5.51	5.74	0.17	5.36	−3.61	−8.97
DC13	−2.44	7.70	9.50	0.14	34.20	17.18	−17.02
DIPCS10	1.57	2.50	2.83	0.00	7.49	4.23	−3.26
FH51	−1.84	2.66	3.48	0.09	14.65	4.35	−10.30
G21EA	0.11	1.46	1.69	0.04	5.65	2.70	−2.95
G21IP	1.34	2.45	2.96	0.01	12.59	7.85	−4.74
G2RC	−3.06	5.49	6.94	0.11	26.80	9.48	−17.32
HAL59	0.13	0.37	0.60	0.08	2.81	2.25	−0.56
HEAVY28	0.02	0.12	0.16	0.10	0.56	0.34	−0.21
HEAVYSB11	0.10	0.63	0.69	0.01	1.94	1.00	−0.93
ICONF	0.19	0.23	0.37	0.07	1.02	0.92	−0.10
IDISP	0.09	1.66	1.79	0.12	4.72	2.14	−2.58
IL16	−0.35	0.39	0.47	0.00	1.06	0.19	−0.87
INV24	0.08	1.08	1.54	0.03	6.96	3.95	−3.01
ISO34	−0.22	1.18	1.71	0.08	7.06	3.51	−3.55
ISOL24	0.82	2.07	2.85	0.09	11.67	6.80	−4.87
MB16-43	2.64	11.27	14.50	0.03	63.38	42.99	−20.38
MCONF	0.18	0.23	0.29	0.05	0.93	0.66	−0.27
NBPRC	−1.09	3.04	3.36	0.11	10.42	4.92	−5.51
PA26	2.80	2.84	3.21	0.02	7.60	6.96	−0.63
PArel	0.65	0.96	1.34	0.21	5.96	3.96	−2.00
PCONF21	−0.11	0.52	0.60	0.32	1.79	0.64	−1.16
PNICO23	0.33	0.44	0.67	0.10	2.93	2.47	−0.46
PX13	−4.25	4.25	4.42	0.13	4.65	−2.15	−6.80
RC21	3.63	4.13	4.91	0.12	13.96	11.54	−2.42
RG18	0.02	0.12	0.15	0.21	0.58	0.27	−0.31
RSE43	−0.40	0.44	0.57	0.06	2.34	0.69	−1.65
S22	0.17	0.42	0.53	0.06	2.32	1.44	−0.88
S66	0.18	0.32	0.43	0.06	2.14	1.54	−0.60
SCONF	0.05	0.20	0.27	0.04	1.12	0.30	−0.83
TAUT15	0.02	0.85	1.01	0.28	3.26	1.84	−1.42
UPU23	0.34	0.46	0.57	0.08	1.58	1.17	−0.41
W4-11	3.41	5.27	6.84	0.02	35.10	19.81	−15.30
WATER27	4.40	4.74	6.08	0.06	20.04	15.48	−4.57
WCPT18	−2.62	2.64	3.22	0.08	6.51	0.19	−6.31
YBDE18	1.90	1.96	2.10	0.04	3.69	3.30	−0.39
BH76RC	−0.43	1.94	2.57	0.09	11.18	6.39	−4.79

Table S65: Statistical analysis of the SOS0-PBE-QIDH(b)-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	5.45	5.45	6.09	0.16	10.11	11.64	1.53
ACONF	−0.21	0.21	0.22	0.11	0.35	0.00	−0.35
ADIM6	−0.19	0.19	0.22	0.06	0.35	−0.05	−0.40
AHB21	−0.75	0.76	0.98	0.03	2.55	0.08	−2.47
AL2X6	0.45	0.90	0.98	0.03	2.56	1.35	−1.22
ALK8	2.76	2.82	4.85	0.05	12.17	11.94	−0.23
ALKBDE10	3.96	4.02	5.36	0.04	11.49	11.18	−0.31
AMINO20x4	−0.02	0.15	0.18	0.06	0.84	0.44	−0.40
BH76	−0.33	1.74	2.54	0.09	15.39	11.37	−4.02
BHDIV10	−1.88	2.20	2.54	0.05	6.08	1.57	−4.52
BHPERI	−1.92	2.31	2.46	0.11	6.40	2.78	−3.62
BHROT27	0.17	0.19	0.25	0.03	0.69	0.59	−0.10
BSR36	−2.40	2.40	2.60	0.15	3.70	−1.30	−5.00
BUT14DIOL	0.01	0.05	0.07	0.02	0.26	0.16	−0.09
C60ISO	−7.37	7.37	9.26	0.08	16.81	−0.72	−17.53
CARBHB12	0.67	0.67	0.83	0.11	1.56	1.65	0.09
CDIE20	0.48	0.53	0.64	0.13	1.47	1.30	−0.18
CHB6	−1.04	1.04	1.24	0.04	1.74	−0.26	−2.00
DARC	−5.82	5.82	5.90	0.18	2.89	−4.28	−7.17
DC13	−2.79	7.66	10.20	0.14	34.96	17.58	−17.38
DIPCS10	4.80	4.80	5.08	0.01	5.68	7.39	1.72
FH51	−1.33	1.93	2.65	0.06	14.11	5.34	−8.77
G21EA	2.43	2.74	3.32	0.08	10.93	8.92	−2.01
G21IP	3.55	3.79	4.55	0.01	16.71	13.70	−3.01
G2RC	−1.81	3.50	4.62	0.07	17.15	6.38	−10.77
HAL59	0.02	0.32	0.48	0.07	2.17	1.64	−0.53
HEAVY28	−0.18	0.20	0.24	0.16	0.55	0.18	−0.37
HEAVYSB11	3.72	3.72	3.78	0.06	2.15	4.53	2.38
ICONF	0.07	0.13	0.17	0.04	0.52	0.31	−0.21
IDISP	−0.47	1.81	2.92	0.13	8.45	1.71	−6.75
IL16	−0.23	0.32	0.36	0.00	1.15	0.43	−0.72
INV24	0.05	0.73	1.02	0.02	5.22	3.42	−1.80
ISO34	−0.22	1.05	1.57	0.07	7.23	3.63	−3.59
ISOL24	1.01	2.18	3.04	0.10	10.84	6.95	−3.90
MB16-43	6.09	9.94	11.97	0.02	42.14	24.79	−17.35
MCONF	0.34	0.36	0.42	0.07	0.94	0.77	−0.17
NBPRC	−1.02	2.45	2.86	0.09	8.88	3.15	−5.74
PA26	2.26	2.26	2.46	0.01	4.33	4.89	0.56
PArel	0.30	0.77	1.01	0.17	4.48	1.95	−2.52
PCONF21	−0.08	0.37	0.44	0.23	1.40	0.48	−0.92
PNICO23	0.13	0.30	0.46	0.07	2.30	1.82	−0.48
PX13	−3.98	3.98	4.14	0.12	4.50	−1.83	−6.33
RC21	1.38	2.42	3.14	0.07	11.85	8.46	−3.38
RG18	−0.07	0.13	0.20	0.22	0.84	0.36	−0.48
RSE43	0.59	0.74	1.59	0.10	7.55	6.81	−0.74
S22	0.07	0.20	0.24	0.03	1.00	0.58	−0.42
S66	0.03	0.18	0.22	0.03	1.13	0.67	−0.46
SCONF	0.05	0.18	0.25	0.04	1.09	0.35	−0.74
TAUT15	−0.16	0.69	0.76	0.23	1.94	0.75	−1.19
UPU23	0.29	0.43	0.52	0.08	1.60	1.16	−0.44
W4-11	17.03	17.19	20.33	0.06	55.15	47.88	−7.27
WATER27	2.28	2.62	3.21	0.03	13.19	8.56	−4.63
WCPT18	−2.42	2.42	2.68	0.07	3.85	−0.99	−4.84
YBDE18	6.01	6.01	6.27	0.12	6.26	9.31	3.04
BH76RC	−0.39	1.54	2.18	0.07	11.03	7.10	−3.92

Table S66: Statistical analysis of the SOS0-PBE-CIDH(b)-D3(0) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.54	7.54	8.49	0.22	14.88	17.02	2.14
ACONF	−0.11	0.11	0.13	0.06	0.27	0.03	−0.25
ADIM6	−0.09	0.10	0.11	0.03	0.19	0.02	−0.17
AHB21	−1.05	1.05	1.31	0.05	2.80	−0.09	−2.88
AL2X6	0.57	1.02	1.10	0.03	2.62	1.46	−1.16
ALK8	3.19	3.19	4.57	0.05	9.93	10.21	0.28
ALKBDE10	−1.60	3.00	3.50	0.03	10.37	5.17	−5.21
AMINO20x4	0.04	0.18	0.23	0.07	1.03	0.62	−0.40
BH76	−1.45	2.02	2.27	0.11	9.71	3.79	−5.92
BHDIV10	−2.14	2.44	2.82	0.05	5.81	0.90	−4.91
BHPERI	−1.41	1.93	2.19	0.09	6.92	3.59	−3.34
BHROT27	0.38	0.38	0.50	0.06	1.01	0.98	−0.03
BSR36	−3.33	3.33	3.78	0.21	6.98	−1.55	−8.54
BUT14DIOL	0.05	0.07	0.09	0.03	0.40	0.31	−0.09
C60ISO	−1.94	2.71	3.86	0.03	10.00	1.64	−8.36
CARBHB12	0.95	0.95	1.14	0.16	1.97	2.16	0.19
CDIE20	0.69	0.73	0.91	0.18	2.15	1.93	−0.22
CHB6	−1.21	1.21	1.48	0.05	2.52	−0.18	−2.70
DARC	−5.83	5.83	6.00	0.18	4.70	−4.05	−8.75
DC13	−2.59	7.67	9.52	0.14	32.09	15.66	−16.43
DIPCS10	2.33	2.72	3.13	0.00	6.78	4.84	−1.94
FH51	−1.81	2.55	3.36	0.08	15.10	4.69	−10.41
G21EA	0.51	1.21	1.47	0.04	5.63	3.69	−1.94
G21IP	1.81	2.36	2.92	0.01	11.46	7.55	−3.91
G2RC	−2.89	5.12	6.49	0.10	24.53	8.41	−16.12
HAL59	0.11	0.35	0.56	0.08	2.67	2.13	−0.54
HEAVY28	−0.01	0.13	0.15	0.10	0.49	0.29	−0.20
HEAVYSB11	0.83	0.86	1.04	0.01	1.83	1.68	−0.15
ICONF	0.17	0.20	0.32	0.06	0.91	0.81	−0.10
IDISP	−0.11	1.61	1.91	0.11	5.34	1.52	−3.82
IL16	−0.36	0.38	0.45	0.00	1.00	0.18	−0.82
INV24	0.14	1.01	1.43	0.03	6.86	4.16	−2.69
ISO34	−0.21	1.15	1.68	0.08	7.19	3.62	−3.56
ISOL24	0.95	2.13	2.97	0.10	10.83	6.75	−4.09
MB16-43	3.63	10.77	13.71	0.03	58.98	39.10	−19.88
MCONF	0.22	0.25	0.32	0.05	0.95	0.70	−0.25
NBPRC	−1.12	2.97	3.32	0.11	10.43	4.66	−5.77
PA26	2.73	2.76	3.10	0.01	6.99	6.58	−0.41
PArel	0.60	0.90	1.25	0.19	5.63	3.54	−2.09
PCONF21	−0.10	0.47	0.55	0.29	1.63	0.54	−1.09
PNICO23	0.29	0.41	0.62	0.10	2.76	2.31	−0.45
PX13	−4.06	4.06	4.22	0.12	4.52	−1.97	−6.49
RC21	3.24	3.78	4.52	0.11	13.29	10.84	−2.45
RG18	−0.01	0.12	0.16	0.21	0.64	0.28	−0.36
RSE43	−0.19	0.36	0.50	0.05	2.67	1.41	−1.26
S22	0.16	0.36	0.46	0.05	2.03	1.30	−0.73
S66	0.15	0.29	0.38	0.05	1.91	1.40	−0.51
SCONF	0.06	0.20	0.27	0.04	1.08	0.29	−0.79
TAUT15	−0.02	0.81	0.93	0.27	2.88	1.55	−1.33
UPU23	0.33	0.45	0.55	0.08	1.54	1.15	−0.39
W4-11	5.90	7.00	8.97	0.02	36.71	25.64	−11.07
WATER27	3.96	4.30	5.47	0.05	18.49	14.03	−4.46
WCPT18	−2.47	2.49	3.01	0.07	5.97	0.14	−5.84
YBDE18	2.78	2.78	2.90	0.06	3.48	4.18	0.70
BH76RC	−0.45	1.82	2.45	0.09	10.90	6.22	−4.68

SI.8.4 Results for dispersion-corrected functionals with DFT-D3(CSO)

Table S67: Statistical analysis of the B2PLYP-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	10.08	10.08	11.39	0.30	19.47	21.40	1.94
ACONF	−0.13	0.13	0.15	0.07	0.28	0.02	−0.26
ADIM6	−0.24	0.24	0.27	0.07	0.35	−0.10	−0.45
AHB21	−0.20	0.26	0.37	0.01	1.21	0.24	−0.96
AL2X6	−1.36	1.36	1.43	0.04	1.16	−0.74	−1.90
ALK8	−1.60	2.26	3.36	0.04	10.17	1.74	−8.42
ALKBDE10	0.46	3.13	3.75	0.03	11.51	6.92	−4.60
AMINO20x4	−0.03	0.15	0.20	0.06	1.32	0.85	−0.47
BH76	−2.54	2.61	2.95	0.14	7.13	0.98	−6.15
BHDIV10	−1.21	2.15	2.64	0.05	8.47	4.05	−4.42
BHPERI	−1.77	1.80	2.02	0.09	5.15	0.34	−4.81
BHROT27	0.25	0.27	0.38	0.04	1.17	1.02	−0.16
BSR36	−2.14	2.14	2.35	0.13	3.63	−1.07	−4.70
BUT14DIOL	0.13	0.13	0.16	0.05	0.39	0.33	−0.06
C60ISO	−6.66	6.69	9.06	0.07	17.88	0.15	−17.73
CARBHB12	0.57	0.57	0.72	0.09	1.34	1.46	0.12
CDIE20	0.74	0.74	0.79	0.18	1.01	1.32	0.31
CHB6	−1.44	1.44	1.58	0.05	1.85	−0.30	−2.15
DARC	4.78	4.78	4.91	0.15	3.99	6.28	2.29
DC13	2.44	6.62	7.77	0.12	25.29	12.55	−12.74
DIPCS10	−3.11	3.96	4.55	0.01	13.37	4.23	−9.13
FH51	0.68	1.49	2.04	0.05	11.82	6.79	−5.03
G21EA	−0.53	1.29	1.64	0.04	6.89	4.04	−2.85
G21IP	−0.91	2.28	2.78	0.01	13.01	5.78	−7.23
G2RC	0.51	1.47	1.98	0.03	10.19	6.22	−3.97
HAL59	0.40	0.46	0.67	0.10	2.45	2.19	−0.25
HEAVY28	0.09	0.14	0.17	0.11	0.64	0.38	−0.26
HEAVYSB11	−0.51	1.31	1.53	0.02	4.50	2.19	−2.31
ICONF	0.01	0.18	0.24	0.06	1.05	0.51	−0.54
IDISP	1.86	1.86	2.71	0.13	5.54	6.19	0.64
IL16	0.56	0.56	0.63	0.01	1.05	1.03	−0.02
INV24	−0.17	0.70	1.00	0.02	5.06	2.51	−2.54
ISO34	0.14	1.09	1.69	0.07	9.42	7.27	−2.14
ISOL24	−1.17	3.75	5.29	0.17	25.91	14.06	−11.85
MB16-43	−12.03	13.30	15.92	0.03	50.94	11.56	−39.38
MCONF	0.17	0.26	0.30	0.05	0.93	0.54	−0.38
NBPRC	0.87	1.21	1.96	0.04	6.12	5.31	−0.82
PA26	1.19	1.29	1.98	0.01	7.01	6.19	−0.82
PArel	0.07	0.74	1.14	0.16	5.73	3.01	−2.72
PCONF21	0.02	0.32	0.37	0.20	1.23	0.59	−0.64
PNICO23	0.25	0.26	0.39	0.06	1.30	1.17	−0.13
PX13	−2.75	2.75	2.84	0.08	2.78	−1.39	−4.17
RC21	−0.26	1.15	1.42	0.03	5.10	1.94	−3.16
RG18	−0.08	0.15	0.19	0.26	0.70	0.26	−0.44
RSE43	−0.52	0.58	0.74	0.08	3.31	1.21	−2.10
S22	0.16	0.18	0.23	0.02	0.68	0.55	−0.13
S66	0.08	0.18	0.24	0.03	1.01	0.73	−0.28
SCONF	−0.03	0.12	0.24	0.03	1.12	0.19	−0.93
TAUT15	−0.01	0.72	0.86	0.24	2.92	1.72	−1.20
UPU23	0.30	0.45	0.56	0.08	2.01	1.20	−0.80
W4-11	−0.26	1.96	2.69	0.01	20.84	9.18	−11.65
WATER27	1.77	2.03	2.82	0.03	11.52	8.09	−3.43
WCPT18	−1.73	1.73	2.12	0.05	4.07	−0.08	−4.15
YBDE18	−1.09	1.93	2.37	0.04	8.57	3.80	−4.77
BH76RC	−0.20	1.12	1.53	0.05	7.74	4.92	−2.82

Table S68: Statistical analysis of the mPW2PLYP-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	9.23	9.23	10.47	0.27	18.41	20.08	1.66
ACONF	−0.08	0.09	0.11	0.05	0.28	0.04	−0.24
ADIM6	−0.07	0.15	0.18	0.04	0.49	0.11	−0.37
AHB21	−0.68	0.68	0.77	0.03	1.19	−0.14	−1.33
AL2X6	−1.53	1.53	1.62	0.04	1.51	−1.03	−2.54
ALK8	−1.12	1.99	2.59	0.03	7.34	2.22	−5.13
ALKBDE10	−0.95	3.13	3.41	0.03	10.17	4.11	−6.06
AMINO20x4	0.00	0.13	0.19	0.05	1.21	0.74	−0.47
BH76	−2.23	2.37	2.68	0.13	7.40	1.48	−5.92
BHDIV10	−0.56	1.75	2.17	0.04	7.64	4.32	−3.32
BHPERI	−0.92	1.05	1.22	0.05	4.54	1.14	−3.40
BHROT27	0.31	0.33	0.45	0.05	1.26	1.10	−0.16
BSR36	−3.33	3.33	3.87	0.21	7.81	−1.33	−9.14
BUT14DIOL	0.16	0.16	0.18	0.06	0.33	0.35	0.02
C60ISO	−4.07	4.59	6.34	0.05	14.34	1.23	−13.11
CARBHB12	0.79	0.79	0.90	0.13	1.37	1.68	0.31
CDIE20	0.69	0.69	0.75	0.17	1.13	1.35	0.22
CHB6	−1.92	1.92	2.07	0.07	2.09	−0.53	−2.62
DARC	4.15	4.15	4.35	0.13	4.46	5.59	1.12
DC13	1.75	6.01	7.54	0.11	26.13	12.34	−13.80
DIPCS10	−2.37	3.49	4.50	0.01	15.12	5.57	−9.55
FH51	0.15	1.14	1.65	0.04	10.07	5.10	−4.97
G21EA	−0.72	1.48	1.80	0.04	7.33	4.44	−2.90
G21IP	−0.66	2.34	2.82	0.01	12.84	6.10	−6.73
G2RC	−0.52	1.63	1.99	0.03	8.71	4.47	−4.24
HAL59	0.49	0.50	0.70	0.11	2.30	2.21	−0.09
HEAVY28	0.17	0.19	0.23	0.15	0.63	0.52	−0.11
HEAVYSB11	−1.30	1.60	1.84	0.03	4.03	1.00	−3.03
ICONF	0.05	0.20	0.28	0.06	1.22	0.60	−0.62
IDISP	1.75	2.41	3.46	0.17	9.55	7.58	−1.98
IL16	0.36	0.40	0.48	0.00	1.22	0.91	−0.31
INV24	0.09	0.72	1.00	0.02	4.15	2.38	−1.77
ISO34	0.19	1.04	1.65	0.07	9.31	7.44	−1.87
ISOL24	−1.10	3.64	5.20	0.17	26.16	13.46	−12.70
MB16-43	−15.91	17.52	22.72	0.04	87.71	13.25	−74.46
MCONF	0.04	0.10	0.13	0.02	0.53	0.29	−0.24
NBPRC	0.75	1.42	1.91	0.05	5.87	4.56	−1.31
PA26	1.00	1.18	1.89	0.01	7.20	6.03	−1.17
PArel	0.16	0.67	1.05	0.14	4.80	3.20	−1.60
PCONF21	0.02	0.36	0.41	0.22	1.50	0.77	−0.73
PNICO23	0.37	0.37	0.43	0.09	0.94	0.90	−0.04
PX13	−2.19	2.19	2.31	0.07	2.59	−0.86	−3.45
RC21	0.19	1.08	1.38	0.03	4.68	2.35	−2.33
RG18	0.27	0.27	0.40	0.47	1.10	1.07	−0.03
RSE43	−0.43	0.49	0.64	0.06	2.95	1.07	−1.89
S22	0.24	0.31	0.42	0.04	1.55	1.20	−0.35
S66	0.25	0.30	0.44	0.05	1.58	1.34	−0.24
SCONF	−0.06	0.10	0.18	0.02	0.77	0.12	−0.65
TAUT15	0.04	0.66	0.85	0.22	3.04	1.72	−1.32
UPU23	0.41	0.50	0.63	0.09	1.63	1.28	−0.35
W4-11	−1.79	2.49	3.54	0.01	21.82	5.02	−16.80
WATER27	4.80	5.01	6.93	0.06	20.71	17.91	−2.80
WCPT18	−1.16	1.51	1.88	0.04	4.79	1.31	−3.48
YBDE18	−1.23	1.72	2.10	0.03	7.05	2.34	−4.71
BH76RC	−0.39	1.24	1.56	0.06	6.65	3.04	−3.61

Table S69: Statistical analysis of the B2GPPLYP-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	6.70	6.70	7.59	0.20	13.09	14.52	1.42
ACONF	−0.15	0.16	0.18	0.09	0.33	0.02	−0.31
ADIM6	−0.34	0.34	0.38	0.10	0.49	−0.13	−0.62
AHB21	−0.41	0.43	0.56	0.02	1.53	0.14	−1.39
AL2X6	−1.25	1.25	1.30	0.03	1.04	−0.84	−1.89
ALK8	−1.00	1.74	2.49	0.03	7.47	1.69	−5.78
ALKBDE10	−0.93	2.96	3.31	0.03	10.01	4.30	−5.71
AMINO20x4	−0.02	0.13	0.18	0.05	1.11	0.67	−0.44
BH76	−0.88	1.40	1.82	0.08	11.68	6.88	−4.80
BHDIV10	−0.34	1.47	1.86	0.03	6.62	3.83	−2.79
BHPERI	−1.51	1.52	1.69	0.07	3.53	0.18	−3.35
BHROT27	0.32	0.33	0.48	0.05	1.74	1.64	−0.10
BSR36	−2.05	2.05	2.24	0.13	3.42	−0.98	−4.41
BUT14DIOL	0.09	0.10	0.12	0.04	0.26	0.26	−0.01
C60ISO	−5.78	6.03	8.22	0.06	17.27	0.76	−16.51
CARBHB12	0.53	0.53	0.66	0.09	1.28	1.37	0.08
CDIE20	0.63	0.63	0.69	0.16	1.02	1.18	0.16
CHB6	−1.51	1.51	1.63	0.06	1.74	−0.51	−2.25
DARC	2.47	2.47	2.69	0.08	3.48	3.97	0.49
DC13	1.79	4.17	5.31	0.08	17.99	9.87	−8.12
DIPCS10	−2.94	3.40	4.20	0.01	11.44	2.27	−9.16
FH51	−0.14	1.02	1.52	0.03	8.27	4.08	−4.18
G21EA	−1.14	1.71	1.92	0.05	6.57	3.22	−3.36
G21IP	−0.81	2.01	2.52	0.01	12.84	5.08	−7.76
G2RC	−0.63	1.37	1.79	0.03	8.11	3.92	−4.19
HAL59	0.29	0.36	0.53	0.08	2.03	1.75	−0.27
HEAVY28	0.02	0.11	0.14	0.09	0.55	0.27	−0.28
HEAVYSB11	−0.63	1.26	1.54	0.02	4.41	1.52	−2.89
ICONF	0.05	0.19	0.25	0.06	1.02	0.55	−0.47
IDISP	1.16	1.16	1.53	0.08	2.88	3.36	0.48
IL16	0.48	0.49	0.55	0.00	1.09	1.00	−0.08
INV24	0.20	0.80	1.12	0.03	4.52	3.08	−1.44
ISO34	0.28	0.73	1.33	0.05	7.19	6.22	−0.97
ISOL24	−0.43	2.61	3.67	0.12	17.35	10.10	−7.26
MB16-43	−9.23	11.39	14.12	0.03	55.29	15.94	−39.35
MCONF	0.20	0.26	0.29	0.05	0.84	0.54	−0.30
NBPRC	0.60	0.94	1.36	0.03	4.33	3.52	−0.81
PA26	0.99	1.15	1.75	0.01	6.55	5.50	−1.05
PArel	0.20	0.54	0.89	0.12	3.79	2.36	−1.43
PCONF21	0.09	0.26	0.31	0.16	0.93	0.53	−0.40
PNICO23	0.16	0.19	0.25	0.04	0.84	0.68	−0.16
PX13	−1.71	1.71	1.83	0.05	2.31	−0.44	−2.75
RC21	−0.63	1.36	1.60	0.04	5.66	2.43	−3.23
RG18	−0.07	0.15	0.20	0.26	0.82	0.43	−0.39
RSE43	0.12	0.41	0.76	0.05	4.45	3.28	−1.17
S22	0.17	0.18	0.25	0.02	0.86	0.75	−0.11
S66	0.07	0.21	0.27	0.04	1.24	0.89	−0.36
SCONF	−0.02	0.07	0.15	0.02	0.67	0.11	−0.56
TAUT15	0.11	0.54	0.68	0.18	2.53	1.40	−1.14
UPU23	0.33	0.45	0.56	0.08	1.70	1.14	−0.56
W4-11	−2.46	2.76	3.87	0.01	19.13	2.76	−16.37
WATER27	1.94	2.13	2.84	0.03	10.65	8.05	−2.60
WCPT18	−0.87	1.18	1.47	0.03	4.27	1.52	−2.75
YBDE18	−0.25	0.92	1.28	0.02	5.77	3.26	−2.51
BH76RC	−0.38	0.97	1.34	0.05	6.55	3.26	−3.30

Table S70: Statistical analysis of the ωB97X-2-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	4.77	4.77	5.58	0.14	10.48	11.55	1.08
ACONF	0.05	0.05	0.06	0.03	0.10	0.10	0.00
ADIM6	0.10	0.10	0.11	0.03	0.15	0.17	0.01
AHB21	−0.33	0.37	0.43	0.02	1.50	0.42	−1.08
AL2X6	−1.38	1.38	1.48	0.04	1.70	−0.69	−2.39
ALK8	1.53	2.31	3.91	0.04	12.45	10.15	−2.29
ALKBDE10	−0.22	2.54	3.13	0.03	11.29	4.50	−6.79
AMINO20x4	−0.05	0.11	0.14	0.05	0.62	0.25	−0.38
BH76	−0.01	1.23	2.06	0.07	13.50	8.17	−5.33
BHDIV10	−0.48	0.73	1.04	0.02	3.73	1.06	−2.68
BHPERI	−2.02	2.05	2.15	0.10	3.39	0.40	−2.98
BHROT27	0.07	0.15	0.21	0.02	0.73	0.42	−0.31
BSR36	−1.53	1.53	1.78	0.09	3.68	−0.56	−4.24
BUT14DIOL	−0.03	0.05	0.05	0.02	0.23	0.10	−0.13
C60ISO	−1.46	3.43	4.21	0.03	11.67	3.13	−8.53
CARBHB12	0.54	0.54	0.60	0.09	0.96	1.15	0.19
CDIE20	0.24	0.28	0.42	0.07	1.14	0.98	−0.17
CHB6	−0.72	0.72	0.79	0.03	0.98	−0.33	−1.32
DARC	−0.22	0.74	0.83	0.02	2.59	0.96	−1.63
DC13	−0.17	2.91	4.21	0.05	18.72	9.17	−9.55
DIPCS10	−5.11	5.11	5.66	0.01	8.09	−0.94	−9.03
FH51	−0.17	0.78	1.06	0.03	6.30	1.67	−4.63
G21EA	−1.04	1.60	1.91	0.05	8.00	4.47	−3.53
G21IP	−1.12	2.09	2.65	0.01	13.67	5.52	−8.15
G2RC	−0.45	1.19	1.48	0.02	6.27	2.59	−3.67
HAL59	0.28	0.32	0.42	0.07	1.48	1.14	−0.34
HEAVY28	0.23	0.23	0.25	0.19	0.49	0.55	0.07
HEAVYSB11	−1.75	1.75	1.90	0.03	2.70	−0.39	−3.09
ICONF	0.03	0.15	0.19	0.05	0.74	0.38	−0.36
IDISP	−0.36	0.88	1.00	0.06	2.76	1.03	−1.73
IL16	0.70	0.70	0.72	0.01	0.58	1.02	0.44
INV24	−0.01	0.97	1.75	0.03	9.63	7.23	−2.40
ISO34	−0.07	0.58	0.87	0.04	4.32	2.67	−1.65
ISOL24	−0.12	1.15	1.61	0.05	7.88	3.47	−4.41
MB16-43	−18.16	18.71	21.09	0.05	52.41	11.82	−40.60
MCONF	0.13	0.15	0.16	0.03	0.36	0.29	−0.07
NBPRC	0.24	0.98	1.23	0.04	4.11	3.13	−0.98
PA26	0.24	0.99	1.38	0.01	5.55	3.79	−1.76
PArel	0.04	0.36	0.58	0.08	2.79	0.77	−2.02
PCONF21	0.01	0.11	0.12	0.07	0.45	0.20	−0.24
PNICO23	0.19	0.20	0.23	0.05	0.52	0.39	−0.13
PX13	−2.01	2.01	2.15	0.06	2.72	−0.55	−3.28
RC21	−1.19	1.61	2.01	0.05	6.05	1.76	−4.29
RG18	0.15	0.15	0.17	0.26	0.39	0.42	0.03
RSE43	0.48	0.60	1.26	0.08	5.80	5.43	−0.37
S22	0.08	0.26	0.29	0.04	1.05	0.50	−0.56
S66	0.18	0.20	0.24	0.04	0.86	0.54	−0.32
SCONF	−0.03	0.05	0.07	0.01	0.28	0.12	−0.15
TAUT15	0.16	0.45	0.58	0.15	1.92	1.08	−0.84
UPU23	0.33	0.43	0.56	0.08	1.61	1.14	−0.47
W4-11	−3.60	3.72	4.56	0.01	20.46	2.13	−18.33
WATER27	3.30	3.43	4.69	0.04	13.82	11.97	−1.84
WCPT18	−1.18	1.62	1.95	0.05	7.07	3.76	−3.31
YBDE18	−0.29	1.38	1.70	0.03	6.83	4.29	−2.54
BH76RC	−0.15	0.70	0.96	0.03	4.72	2.21	−2.52

Table S71: Statistical analysis of the DSD-BLYP-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	5.88	5.88	6.65	0.17	11.22	12.48	1.26
ACONF	−0.18	0.18	0.20	0.10	0.35	0.02	−0.33
ADIM6	−0.36	0.36	0.40	0.11	0.51	−0.14	−0.64
AHB21	−0.17	0.24	0.34	0.01	1.33	0.26	−1.07
AL2X6	−1.47	1.47	1.51	0.04	1.01	−0.98	−1.99
ALK8	−1.09	2.06	2.77	0.03	8.15	1.89	−6.26
ALKBDE10	−0.35	2.84	3.28	0.03	10.44	5.16	−5.28
AMINO20x4	−0.05	0.14	0.18	0.06	1.13	0.62	−0.51
BH76	−0.35	1.20	1.93	0.06	13.66	8.96	−4.69
BHDIV10	−0.32	1.35	1.74	0.03	6.12	3.31	−2.81
BHPERI	−1.72	1.72	1.90	0.08	3.76	−0.11	−3.87
BHROT27	0.22	0.23	0.31	0.04	0.90	0.83	−0.06
BSR36	−1.72	1.72	1.84	0.11	2.30	−0.90	−3.19
BUT14DIOL	0.03	0.06	0.08	0.02	0.31	0.24	−0.07
C60ISO	−7.68	7.68	10.17	0.08	19.58	−0.10	−19.68
CARBHB12	0.40	0.40	0.53	0.07	1.15	1.14	−0.01
CDIE20	0.57	0.57	0.61	0.14	0.87	1.01	0.14
CHB6	−1.31	1.31	1.42	0.05	1.65	−0.49	−2.14
DARC	2.63	2.63	2.77	0.08	2.72	4.01	1.29
DC13	2.09	4.12	4.97	0.07	15.83	8.90	−6.93
DIPCS10	−3.81	3.99	4.61	0.01	10.01	0.91	−9.10
FH51	0.10	1.11	1.57	0.04	8.90	4.49	−4.41
G21EA	−1.27	1.76	1.99	0.05	7.36	3.85	−3.52
G21IP	−1.12	2.02	2.52	0.01	12.60	4.75	−7.85
G2RC	−0.08	1.00	1.45	0.02	7.18	4.21	−2.97
HAL59	0.23	0.32	0.46	0.07	2.02	1.62	−0.40
HEAVY28	0.01	0.10	0.13	0.08	0.55	0.24	−0.31
HEAVYSB11	−0.34	1.26	1.48	0.02	4.44	1.85	−2.58
ICONF	0.01	0.18	0.23	0.06	0.86	0.40	−0.46
IDISP	1.08	1.08	1.32	0.08	2.27	2.45	0.19
IL16	0.62	0.62	0.66	0.01	1.00	1.14	0.14
INV24	0.16	0.77	1.12	0.02	5.59	2.80	−2.80
ISO34	0.24	0.71	1.20	0.05	6.42	5.42	−1.00
ISOL24	−0.48	2.45	3.43	0.11	16.46	9.43	−7.03
MB16-43	−11.98	12.96	21.08	0.03	123.81	14.14	−109.67
MCONF	0.24	0.31	0.34	0.06	0.95	0.62	−0.33
NBPRC	0.72	0.91	1.43	0.03	4.42	3.82	−0.60
PA26	0.87	1.03	1.60	0.01	6.17	4.96	−1.21
PArel	0.10	0.53	0.84	0.11	4.15	2.08	−2.07
PCONF21	0.10	0.24	0.30	0.15	0.94	0.64	−0.30
PNICO23	0.07	0.11	0.16	0.03	0.60	0.43	−0.17
PX13	−1.56	1.56	1.66	0.05	2.04	−0.40	−2.44
RC21	−1.55	1.86	2.28	0.05	5.07	1.19	−3.88
RG18	−0.08	0.16	0.22	0.28	0.89	0.46	−0.42
RSE43	0.48	0.59	1.22	0.08	5.72	4.99	−0.74
S22	0.11	0.18	0.24	0.02	0.86	0.58	−0.28
S66	0.01	0.17	0.21	0.03	0.83	0.44	−0.40
SCONF	−0.04	0.06	0.12	0.01	0.50	0.05	−0.44
TAUT15	0.03	0.45	0.53	0.15	1.94	0.95	−0.99
UPU23	0.29	0.42	0.52	0.07	1.71	1.11	−0.60
W4-11	−2.10	2.58	3.54	0.01	21.75	4.47	−17.29
WATER27	0.60	0.87	1.20	0.01	6.30	3.81	−2.48
WCPT18	−0.87	1.01	1.26	0.03	3.40	0.91	−2.49
YBDE18	0.13	0.92	1.45	0.02	6.26	4.42	−1.84
BH76RC	−0.22	0.82	1.22	0.04	7.04	4.07	−2.97

Table S72: Statistical analysis of the PWPB95-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	10.07	10.07	11.47	0.30	20.84	22.65	1.81
ACONF	−0.34	0.34	0.37	0.19	0.58	−0.03	−0.60
ADIM6	0.25	0.26	0.37	0.08	0.70	0.68	−0.01
AHB21	0.16	0.25	0.30	0.01	0.99	0.65	−0.34
AL2X6	1.12	1.27	1.37	0.04	2.56	2.12	−0.44
ALK8	1.90	2.56	3.98	0.04	11.22	9.39	−1.83
ALKBDE10	−2.12	3.06	3.43	0.03	8.85	3.80	−5.05
AMINO20x4	0.00	0.24	0.33	0.10	1.63	1.06	−0.56
BH76	−1.79	1.86	2.17	0.10	5.96	0.92	−5.04
BHDIV10	−1.77	1.77	1.98	0.04	3.03	0.01	−3.03
BHPERI	−0.47	0.92	1.08	0.04	3.39	1.08	−2.32
BHROT27	0.29	0.33	0.43	0.05	1.02	0.79	−0.23
BSR36	−1.79	1.79	1.89	0.11	2.73	−0.47	−3.20
BUT14DIOL	−0.31	0.31	0.33	0.11	0.60	−0.07	−0.67
C60ISO	−2.94	3.43	4.73	0.03	10.95	1.12	−9.83
CARBHB12	0.14	0.22	0.29	0.04	1.01	0.74	−0.28
CDIE20	0.64	0.65	0.80	0.16	1.70	1.62	−0.08
CHB6	−1.59	1.59	1.91	0.06	2.81	−0.40	−3.21
DARC	0.83	1.18	1.37	0.04	3.30	2.43	−0.88
DC13	−0.48	3.78	5.06	0.07	20.84	10.51	−10.33
DIPCS10	−4.04	4.08	4.62	0.01	8.87	0.21	−8.65
FH51	0.32	1.05	1.39	0.03	6.46	3.06	−3.40
G21EA	−1.64	1.71	1.84	0.05	3.27	0.53	−2.73
G21IP	−1.29	1.94	2.37	0.01	10.15	3.61	−6.54
G2RC	0.19	2.27	2.92	0.04	10.60	6.13	−4.47
HAL59	0.06	0.31	0.43	0.07	2.14	1.37	−0.77
HEAVY28	−0.02	0.13	0.16	0.10	0.58	0.33	−0.26
HEAVYSB11	0.72	1.06	1.25	0.02	3.70	2.38	−1.33
ICONF	−0.02	0.21	0.32	0.06	1.42	0.51	−0.91
IDISP	0.98	1.85	2.05	0.13	5.33	3.25	−2.08
IL16	0.99	0.99	1.04	0.01	1.05	1.65	0.59
INV24	−0.17	0.75	1.00	0.02	4.73	2.48	−2.25
ISO34	−0.48	0.75	0.90	0.05	3.37	1.21	−2.16
ISOL24	−0.76	1.69	2.20	0.08	8.89	4.77	−4.12
MB16-43	2.62	7.05	8.79	0.02	36.25	22.88	−13.37
MCONF	0.35	0.43	0.53	0.09	1.54	1.11	−0.43
NBPRC	−0.23	0.82	0.91	0.03	2.81	0.99	−1.82
PA26	1.95	1.97	2.37	0.01	6.31	6.09	−0.22
PArel	0.36	0.68	1.06	0.15	4.87	3.60	−1.27
PCONF21	0.28	0.57	0.69	0.35	2.02	1.34	−0.67
PNICO23	0.15	0.23	0.35	0.05	1.59	1.29	−0.29
PX13	−1.36	1.36	1.45	0.04	1.93	−0.24	−2.17
RC21	0.94	1.50	1.74	0.04	5.63	3.35	−2.27
RG18	0.09	0.22	0.34	0.38	1.35	1.02	−0.32
RSE43	−0.94	0.94	1.11	0.12	2.55	−0.27	−2.82
S22	−0.15	0.39	0.55	0.05	1.99	0.62	−1.37
S66	−0.07	0.21	0.28	0.04	1.43	0.47	−0.95
SCONF	−0.02	0.24	0.37	0.05	1.64	0.42	−1.22
TAUT15	−0.08	0.63	0.74	0.21	2.38	1.37	−1.02
UPU23	0.40	0.52	0.63	0.09	1.53	1.14	−0.39
W4-11	0.13	1.92	3.08	0.01	30.77	17.43	−13.34
WATER27	−2.61	2.69	4.31	0.03	12.14	0.57	−11.57
WCPT18	−1.25	1.25	1.48	0.04	2.90	−0.27	−3.17
YBDE18	−0.12	1.74	1.97	0.04	6.24	2.98	−3.25
BH76RC	0.15	1.16	1.62	0.05	8.90	5.23	−3.67

Table S73: Statistical analysis of the DSD-PBEP86-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	5.00	5.00	5.76	0.15	10.37	11.45	1.08
ACONF	−0.20	0.20	0.22	0.11	0.36	0.01	−0.35
ADIM6	−0.27	0.27	0.32	0.08	0.50	−0.05	−0.55
AHB21	−0.34	0.36	0.47	0.02	1.54	0.09	−1.45
AL2X6	−0.70	0.70	0.79	0.02	1.06	−0.42	−1.48
ALK8	0.13	1.64	2.65	0.03	10.22	6.11	−4.11
ALKBDE10	−2.18	3.08	3.57	0.03	8.33	2.21	−6.12
AMINO20x4	−0.04	0.13	0.16	0.05	0.76	0.42	−0.35
BH76	−0.17	1.24	2.06	0.07	14.11	9.83	−4.27
BHDIV10	−1.01	1.56	1.72	0.03	4.83	1.89	−2.94
BHPERI	−2.01	2.06	2.18	0.10	4.01	0.58	−3.44
BHROT27	0.19	0.20	0.26	0.03	0.67	0.63	−0.04
BSR36	−1.84	1.84	2.01	0.11	3.11	−0.88	−4.00
BUT14DIOL	0.00	0.06	0.07	0.02	0.25	0.14	−0.10
C60ISO	−7.69	7.69	9.94	0.08	18.61	−0.43	−19.04
CARBHB12	0.53	0.53	0.65	0.09	1.25	1.38	0.13
CDIE20	0.50	0.50	0.58	0.12	1.05	1.06	0.01
CHB6	−0.99	0.99	1.08	0.04	1.28	−0.55	−1.83
DARC	−0.64	1.01	1.10	0.03	2.66	0.74	−1.92
DC13	0.88	2.62	3.41	0.05	13.05	4.33	−8.73
DIPCS10	−3.91	3.91	4.36	0.01	6.73	−0.96	−7.69
FH51	−0.43	0.80	1.13	0.03	5.95	2.61	−3.34
G21EA	−1.01	1.51	1.80	0.04	7.69	4.72	−2.98
G21IP	−1.06	2.10	2.62	0.01	13.63	6.57	−7.05
G2RC	−0.58	1.70	2.27	0.03	10.46	5.06	−5.40
HAL59	0.30	0.36	0.51	0.08	1.96	1.71	−0.25
HEAVY28	0.11	0.15	0.18	0.12	0.61	0.38	−0.23
HEAVYSB11	−0.58	0.98	1.14	0.02	3.02	1.08	−1.94
ICONF	0.03	0.14	0.18	0.04	0.67	0.35	−0.32
IDISP	0.42	0.69	0.89	0.05	2.39	1.71	−0.68
IL16	0.34	0.39	0.42	0.00	1.13	0.74	−0.39
INV24	0.15	0.69	1.06	0.02	5.86	3.61	−2.25
ISO34	0.04	0.43	0.66	0.03	3.29	2.32	−0.97
ISOL24	0.04	1.20	1.65	0.05	6.79	4.33	−2.47
MB16-43	−5.91	7.46	8.64	0.02	26.75	10.86	−15.89
MCONF	0.25	0.29	0.33	0.06	0.85	0.60	−0.26
NBPRC	0.11	0.66	0.87	0.02	3.04	1.89	−1.15
PA26	0.96	1.05	1.45	0.01	5.15	4.35	−0.80
PArel	0.20	0.49	0.76	0.11	3.42	1.75	−1.67
PCONF21	0.03	0.23	0.29	0.14	1.04	0.52	−0.52
PNICO23	0.26	0.27	0.37	0.06	1.17	1.13	−0.04
PX13	−2.41	2.41	2.49	0.07	2.55	−1.15	−3.70
RC21	−0.96	1.77	2.09	0.05	6.03	2.45	−3.59
RG18	−0.05	0.15	0.22	0.26	1.04	0.61	−0.43
RSE43	0.88	0.89	1.71	0.12	7.18	7.15	−0.03
S22	0.11	0.19	0.23	0.03	0.81	0.47	−0.34
S66	0.04	0.15	0.19	0.03	0.78	0.45	−0.33
SCONF	−0.01	0.06	0.11	0.01	0.53	0.12	−0.41
TAUT15	0.01	0.45	0.50	0.15	1.43	0.78	−0.65
UPU23	0.31	0.42	0.52	0.07	1.50	1.13	−0.37
W4-11	−2.81	2.94	3.94	0.01	21.70	2.25	−19.45
WATER27	1.29	1.52	1.93	0.02	8.76	5.77	−2.99
WCPT18	−1.63	1.64	1.92	0.05	3.76	0.10	−3.66
YBDE18	0.72	1.38	1.74	0.03	6.62	4.91	−1.71
BH76RC	−0.22	0.85	1.39	0.04	8.36	5.00	−3.35

Table S74: Statistical analysis of the B2PPW91-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	18.05	18.05	20.12	0.54	31.96	36.07	4.12
ACONF	−0.18	0.18	0.20	0.10	0.33	−0.02	−0.35
ADIM6	−0.63	0.63	0.63	0.19	0.21	−0.53	−0.73
AHB21	0.09	0.76	0.99	0.03	4.70	2.38	−2.32
AL2X6	2.27	3.58	3.66	0.10	8.57	4.63	−3.94
ALK8	1.32	5.03	7.43	0.08	24.90	11.50	−13.40
ALKBDE10	4.96	6.11	9.51	0.06	28.04	25.53	−2.50
AMINO20x4	−0.02	0.36	0.46	0.15	2.23	1.20	−1.03
BH76	−6.62	6.66	7.42	0.36	23.79	1.54	−22.25
BHDIV10	−6.85	7.23	8.03	0.16	15.11	1.92	−13.19
BHPERI	−6.37	6.37	6.67	0.31	7.37	−3.06	−10.44
BHROT27	0.28	0.34	0.49	0.05	1.52	1.32	−0.20
BSR36	0.36	1.11	1.44	0.07	6.39	4.93	−1.45
BUT14DIOL	0.19	0.22	0.29	0.08	1.14	0.82	−0.32
C60ISO	−11.47	11.47	14.20	0.12	24.49	−0.91	−25.41
CARBHB12	0.93	0.94	1.43	0.16	3.43	3.34	−0.10
CDIE20	1.39	1.39	1.49	0.34	2.13	2.30	0.18
CHB6	−0.49	0.52	0.78	0.02	1.57	0.06	−1.51
DARC	0.74	1.80	2.07	0.06	6.30	3.41	−2.88
DC13	1.00	7.85	10.61	0.14	41.59	12.72	−28.87
DIPCS10	−1.53	3.02	3.70	0.00	10.87	3.53	−7.34
FH51	1.08	2.49	3.47	0.08	17.31	9.07	−8.24
G21EA	1.81	2.43	2.87	0.07	8.66	5.61	−3.06
G21IP	0.20	3.23	3.96	0.01	16.38	8.89	−7.48
G2RC	1.69	5.31	6.57	0.10	28.38	16.85	−11.53
HAL59	0.77	0.98	1.48	0.21	5.25	4.54	−0.72
HEAVY28	0.25	0.32	0.41	0.26	1.47	1.04	−0.43
HEAVYSB11	3.81	3.81	4.57	0.07	8.64	8.92	0.29
ICONF	0.07	0.29	0.34	0.09	1.13	0.66	−0.46
IDISP	1.68	1.92	3.31	0.14	8.29	7.89	−0.40
IL16	0.02	0.34	0.42	0.00	1.59	0.94	−0.65
INV24	−1.19	1.85	2.57	0.06	9.70	3.26	−6.44
ISO34	−0.39	1.07	1.48	0.07	8.45	3.84	−4.61
ISOL24	−0.61	2.83	4.11	0.13	19.16	11.53	−7.63
MB16-43	18.61	28.65	35.75	0.07	130.86	83.49	−47.37
MCONF	0.40	0.61	0.74	0.12	2.41	1.46	−0.95
NBPRC	−0.67	2.10	2.66	0.08	9.44	4.08	−5.36
PA26	3.18	3.18	3.76	0.02	8.64	8.62	−0.02
PArel	0.24	1.52	2.09	0.33	10.08	4.97	−5.11
PCONF21	−0.36	0.83	1.01	0.51	3.09	1.14	−1.95
PNICO23	0.91	1.07	1.74	0.25	6.98	6.26	−0.73
PX13	−9.16	9.16	9.34	0.27	7.03	−6.49	−13.52
RC21	3.57	4.46	4.96	0.12	11.97	9.28	−2.69
RG18	−0.61	0.61	0.88	1.05	2.64	−0.18	−2.83
RSE43	−1.98	1.98	2.22	0.26	4.68	−0.42	−5.10
S22	0.04	0.43	0.56	0.06	2.14	1.49	−0.65
S66	−0.21	0.36	0.43	0.07	1.63	0.81	−0.82
SCONF	0.27	0.80	1.00	0.17	3.68	0.97	−2.70
TAUT15	0.11	1.52	1.82	0.50	6.00	3.92	−2.08
UPU23	0.17	0.43	0.55	0.08	2.54	1.41	−1.13
W4-11	8.60	9.59	12.28	0.03	44.23	35.16	−9.07
WATER27	−2.62	3.05	4.53	0.04	13.97	2.02	−11.95
WCPT18	−7.02	7.02	7.36	0.20	8.51	−4.27	−12.78
YBDE18	0.22	2.76	3.35	0.06	12.56	7.42	−5.14
BH76RC	0.60	2.75	4.38	0.13	22.89	17.87	−5.02

Table S75: Statistical analysis of the DSD-PBEB95-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	5.94	5.94	6.80	0.18	12.47	13.62	1.14
ACONF	−0.31	0.31	0.33	0.17	0.44	−0.01	−0.45
ADIM6	0.12	0.13	0.19	0.04	0.40	0.36	−0.04
AHB21	0.15	0.23	0.28	0.01	1.02	0.57	−0.45
AL2X6	0.28	0.52	0.58	0.01	1.67	0.97	−0.70
ALK8	1.69	2.21	3.96	0.04	11.76	10.17	−1.59
ALKBDE10	−2.93	3.02	3.66	0.03	6.62	0.43	−6.19
AMINO20x4	−0.03	0.21	0.27	0.09	1.38	0.91	−0.48
BH76	0.16	1.03	1.64	0.06	11.04	7.57	−3.48
BHDIV10	−0.65	0.74	0.93	0.02	1.79	0.26	−1.53
BHPERI	−0.15	0.78	0.96	0.04	3.34	1.63	−1.71
BHROT27	0.24	0.25	0.33	0.04	0.71	0.66	−0.04
BSR36	−1.74	1.74	1.83	0.11	2.33	−0.64	−2.97
BUT14DIOL	−0.31	0.31	0.32	0.11	0.44	−0.05	−0.48
C60ISO	−4.56	4.79	6.44	0.05	13.65	0.70	−12.95
CARBHB12	0.08	0.20	0.26	0.03	1.02	0.61	−0.41
CDIE20	0.50	0.53	0.65	0.13	1.46	1.34	−0.12
CHB6	−1.44	1.44	1.69	0.05	2.26	−0.41	−2.67
DARC	−0.45	0.89	0.93	0.03	2.31	0.93	−1.38
DC13	−0.20	3.25	4.46	0.06	16.73	8.89	−7.84
DIPCS10	−6.03	6.03	6.33	0.01	7.78	−2.50	−10.28
FH51	−0.17	0.72	0.98	0.02	4.47	2.03	−2.44
G21EA	−2.76	2.89	2.97	0.09	5.92	1.59	−4.34
G21IP	−2.18	2.58	2.89	0.01	10.89	3.22	−7.68
G2RC	−0.22	1.64	2.24	0.03	9.17	4.53	−4.64
HAL59	−0.01	0.25	0.33	0.05	1.61	0.88	−0.73
HEAVY28	−0.03	0.12	0.15	0.10	0.56	0.26	−0.30
HEAVYSB11	0.50	0.74	0.90	0.01	2.40	1.60	−0.79
ICONF	−0.01	0.17	0.26	0.05	1.17	0.43	−0.74
IDISP	0.49	1.21	1.41	0.09	3.88	2.38	−1.50
IL16	0.99	0.99	1.02	0.01	0.88	1.55	0.66
INV24	0.22	0.74	1.16	0.02	6.08	4.50	−1.58
ISO34	−0.31	0.58	0.78	0.04	3.21	1.29	−1.92
ISOL24	−0.36	1.12	1.49	0.05	6.80	3.23	−3.57
MB16-43	−0.45	4.39	5.46	0.01	21.25	12.58	−8.67
MCONF	0.35	0.40	0.47	0.08	1.22	0.91	−0.31
NBPRC	−0.06	0.63	0.73	0.02	2.47	1.27	−1.19
PA26	1.47	1.48	1.82	0.01	4.99	4.83	−0.16
PArel	0.31	0.55	0.78	0.12	3.41	2.34	−1.07
PCONF21	0.29	0.48	0.61	0.30	1.48	1.11	−0.37
PNICO23	0.01	0.10	0.15	0.02	0.79	0.52	−0.27
PX13	−0.11	0.45	0.53	0.01	1.78	0.94	−0.85
RC21	−0.35	1.15	1.42	0.03	6.39	2.69	−3.69
RG18	0.05	0.17	0.26	0.29	1.07	0.70	−0.37
RSE43	0.02	0.45	0.73	0.06	3.91	2.58	−1.33
S22	−0.11	0.39	0.53	0.05	1.90	0.62	−1.28
S66	−0.06	0.19	0.26	0.03	1.26	0.35	−0.91
SCONF	−0.10	0.15	0.19	0.03	0.63	0.13	−0.50
TAUT15	−0.15	0.41	0.48	0.13	1.45	0.51	−0.94
UPU23	0.32	0.46	0.56	0.08	1.60	1.11	−0.49
W4-11	−1.70	2.42	3.53	0.01	29.62	12.29	−17.33
WATER27	−2.47	2.50	3.96	0.03	10.81	0.23	−10.58
WCPT18	−0.38	0.55	0.70	0.02	2.21	0.69	−1.52
YBDE18	0.89	1.01	1.23	0.02	3.08	2.20	−0.88
BH76RC	0.14	1.12	1.52	0.05	8.36	4.78	−3.58

Table S76: Statistical analysis of the B2NC-PLYP-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	2.68	2.68	3.02	0.08	4.76	5.51	0.75
ACONF	−0.19	0.20	0.22	0.11	0.42	0.03	−0.39
ADIM6	−0.53	0.53	0.59	0.16	0.75	−0.17	−0.93
AHB21	−0.55	0.56	0.76	0.02	2.00	0.08	−1.92
AL2X6	−1.40	1.40	1.44	0.04	0.95	−1.07	−2.02
ALK8	−0.26	1.72	2.14	0.03	6.95	4.12	−2.83
ALKBDE10	−0.78	2.77	3.37	0.03	10.60	4.79	−5.81
AMINO20x4	−0.01	0.13	0.16	0.05	0.99	0.47	−0.52
BH76	1.16	1.79	2.87	0.10	19.02	15.47	−3.55
BHDIV10	0.17	1.09	1.50	0.02	5.64	3.27	−2.37
BHPERI	−2.27	2.30	2.48	0.11	4.50	0.40	−4.09
BHROT27	0.31	0.31	0.41	0.05	1.02	0.96	−0.06
BSR36	−1.43	1.43	1.51	0.09	1.73	−0.72	−2.45
BUT14DIOL	0.04	0.07	0.09	0.03	0.34	0.25	−0.10
C60ISO	−7.65	7.76	10.30	0.08	20.38	0.27	−20.11
CARBHB12	0.45	0.45	0.59	0.07	1.32	1.32	−0.01
CDIE20	0.54	0.54	0.59	0.13	0.90	0.93	0.02
CHB6	−1.35	1.35	1.44	0.05	1.45	−0.73	−2.18
DARC	−0.15	0.96	1.07	0.03	3.25	1.49	−1.76
DC13	1.49	3.25	4.11	0.06	14.35	7.03	−7.32
DIPCS10	−3.13	3.27	4.17	0.00	8.92	0.71	−8.22
FH51	−0.92	1.37	1.99	0.04	10.22	2.86	−7.36
G21EA	−1.42	2.13	2.55	0.06	11.23	7.19	−4.03
G21IP	−0.67	1.82	2.60	0.01	16.52	7.80	−8.72
G2RC	−1.59	1.92	2.57	0.04	8.41	2.31	−6.11
HAL59	0.16	0.27	0.41	0.06	2.14	1.59	−0.54
HEAVY28	−0.06	0.11	0.14	0.09	0.48	0.15	−0.33
HEAVYSB11	−0.27	1.07	1.27	0.02	3.65	1.21	−2.44
ICONF	0.07	0.20	0.26	0.06	0.85	0.49	−0.36
IDISP	0.28	0.74	0.82	0.05	2.01	1.41	−0.60
IL16	0.39	0.39	0.45	0.00	0.97	0.95	−0.02
INV24	0.43	0.96	1.38	0.03	6.44	4.37	−2.07
ISO34	0.44	0.63	1.16	0.04	5.32	4.59	−0.73
ISOL24	0.45	1.50	2.21	0.07	10.02	5.73	−4.29
MB16-43	−2.83	9.03	11.69	0.02	55.39	24.02	−31.38
MCONF	0.26	0.30	0.34	0.06	0.91	0.63	−0.28
NBPRC	0.26	0.57	0.70	0.02	2.05	1.40	−0.65
PA26	0.34	0.96	1.48	0.01	7.39	3.86	−3.52
PArel	0.29	0.44	0.75	0.10	3.48	2.70	−0.78
PCONF21	0.16	0.25	0.28	0.15	0.82	0.53	−0.29
PNICO23	0.02	0.12	0.15	0.03	0.51	0.28	−0.23
PX13	−1.13	1.14	1.28	0.03	2.06	0.10	−1.97
RC21	−2.05	2.80	3.20	0.08	7.62	2.60	−5.02
RG18	−0.08	0.17	0.23	0.29	1.06	0.58	−0.48
RSE43	1.16	1.16	2.22	0.15	9.32	9.29	−0.03
S22	0.14	0.19	0.25	0.03	0.90	0.67	−0.23
S66	0.00	0.23	0.29	0.04	1.38	0.77	−0.62
SCONF	0.05	0.08	0.10	0.02	0.39	0.22	−0.18
TAUT15	0.30	0.44	0.62	0.14	1.95	1.09	−0.86
UPU23	0.32	0.43	0.53	0.08	1.37	1.05	−0.31
W4-11	−2.99	3.35	4.90	0.01	27.52	4.28	−23.24
WATER27	1.51	1.67	2.10	0.02	8.49	6.27	−2.22
WCPT18	−0.53	0.96	1.20	0.03	4.21	2.22	−2.00
YBDE18	1.46	1.79	2.35	0.04	7.19	5.81	−1.37
BH76RC	−0.42	1.16	1.61	0.05	7.87	3.62	−4.25

Table S77: Statistical analysis of the mPW2NC-PLYP-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.54	7.54	8.40	0.22	12.50	13.69	1.19
ACONF	−0.15	0.16	0.18	0.09	0.34	0.02	−0.32
ADIM6	−0.37	0.37	0.45	0.11	0.73	−0.05	−0.78
AHB21	−0.42	0.45	0.56	0.02	1.85	0.24	−1.61
AL2X6	−2.03	2.03	2.10	0.06	1.42	−1.25	−2.67
ALK8	−0.59	1.76	2.35	0.03	7.81	3.86	−3.95
ALKBDE10	4.53	5.26	6.86	0.05	14.64	12.46	−2.18
AMINO20x4	−0.03	0.13	0.18	0.05	1.06	0.61	−0.45
BH76	−1.64	2.30	2.82	0.12	15.44	8.13	−7.31
BHDIV10	−1.78	2.69	3.16	0.06	9.24	3.29	−5.94
BHPERI	−3.99	3.99	4.15	0.19	5.04	−2.11	−7.15
BHROT27	0.17	0.19	0.30	0.03	1.07	0.93	−0.14
BSR36	−1.46	1.46	1.61	0.09	2.63	−0.65	−3.28
BUT14DIOL	0.13	0.13	0.15	0.05	0.27	0.28	0.02
C60ISO	−13.27	13.27	16.39	0.14	27.42	−2.34	−29.76
CARBHB12	0.67	0.67	0.81	0.11	1.39	1.57	0.18
CDIE20	0.72	0.72	0.75	0.18	0.94	1.20	0.25
CHB6	−1.17	1.17	1.27	0.04	1.47	−0.41	−1.88
DARC	3.29	3.29	3.50	0.10	3.49	5.13	1.65
DC13	2.91	5.19	6.52	0.09	21.72	9.14	−12.58
DIPCS10	−2.33	3.07	3.40	0.00	7.83	2.68	−5.15
FH51	0.37	1.37	1.91	0.04	11.80	6.20	−5.60
G21EA	0.61	1.48	1.98	0.04	6.53	4.41	−2.12
G21IP	−0.17	1.91	2.45	0.01	13.24	6.07	−7.18
G2RC	0.39	1.75	2.43	0.03	11.45	7.26	−4.18
HAL59	0.43	0.46	0.68	0.10	2.49	2.30	−0.20
HEAVY28	0.07	0.12	0.14	0.10	0.42	0.26	−0.16
HEAVYSB11	0.82	0.95	1.32	0.02	3.65	3.10	−0.55
ICONF	0.03	0.18	0.21	0.06	0.74	0.35	−0.39
IDISP	1.23	1.25	1.93	0.09	4.15	4.10	−0.06
IL16	0.23	0.27	0.32	0.00	0.85	0.69	−0.16
INV24	−0.53	0.88	1.70	0.03	9.08	1.55	−7.53
ISO34	0.23	0.79	1.29	0.05	6.79	5.44	−1.35
ISOL24	−0.70	3.02	4.30	0.14	21.44	12.75	−8.70
MB16-43	−4.23	6.09	7.54	0.01	34.34	16.44	−17.90
MCONF	0.18	0.24	0.29	0.05	0.97	0.61	−0.36
NBPRC	0.50	0.88	1.28	0.03	4.10	3.33	−0.77
PA26	−0.37	1.16	1.44	0.01	6.85	3.44	−3.40
PArel	0.02	0.68	1.20	0.15	6.56	2.70	−3.86
PCONF21	−0.03	0.35	0.40	0.22	1.35	0.64	−0.71
PNICO23	0.19	0.22	0.33	0.05	1.30	1.16	−0.14
PX13	−3.71	3.71	3.79	0.11	3.33	−2.22	−5.55
RC21	−1.46	2.10	2.60	0.06	7.32	1.93	−5.40
RG18	0.12	0.17	0.25	0.29	0.99	0.79	−0.21
RSE43	0.22	0.67	1.30	0.09	7.19	5.59	−1.60
S22	0.12	0.17	0.22	0.02	0.60	0.43	−0.17
S66	0.05	0.22	0.26	0.04	0.95	0.51	−0.43
SCONF	0.11	0.21	0.26	0.05	1.01	0.31	−0.70
TAUT15	0.20	0.63	0.79	0.21	2.48	1.76	−0.72
UPU23	0.24	0.38	0.48	0.07	1.62	1.09	−0.53
W4-11	5.65	6.03	7.55	0.02	24.89	20.15	−4.74
WATER27	2.83	3.15	4.27	0.04	16.36	12.03	−4.33
WCPT18	−3.02	3.02	3.20	0.09	3.62	−1.84	−5.45
YBDE18	1.80	2.03	3.11	0.04	9.48	8.29	−1.19
BH76RC	−0.01	1.50	2.22	0.07	12.10	9.41	−2.69

Table S78: Statistical analysis of the mSD-PBEPBE-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	9.48	9.48	11.00	0.28	20.06	21.06	1.00
ACONF	−0.33	0.33	0.35	0.18	0.44	−0.02	−0.47
ADIM6	−0.31	0.31	0.36	0.09	0.55	−0.08	−0.62
AHB21	1.41	1.47	2.19	0.07	7.58	7.19	−0.39
AL2X6	−3.65	3.65	4.05	0.10	4.98	−2.55	−7.53
ALK8	−1.38	5.87	7.76	0.09	27.45	14.34	−13.12
ALKBDE10	17.90	17.90	22.06	0.18	37.52	45.26	7.74
AMINO20x4	−0.24	0.37	0.46	0.15	1.76	0.64	−1.11
BH76	−1.77	4.03	5.64	0.22	41.50	27.55	−13.95
BHDIV10	−4.41	4.76	5.91	0.11	12.66	1.74	−10.92
BHPERI	−7.35	7.35	7.69	0.35	11.21	−2.86	−14.07
BHROT27	−0.52	0.54	0.83	0.09	2.12	0.07	−2.05
BSR36	0.64	0.80	1.19	0.05	4.58	3.75	−0.83
BUT14DIOL	−0.17	0.19	0.21	0.07	0.61	0.24	−0.37
C60ISO	−31.91	31.91	36.78	0.32	52.09	−8.19	−60.28
CARBHB12	0.05	0.21	0.30	0.03	1.14	0.80	−0.34
CDIE20	0.46	0.59	0.72	0.15	2.63	1.97	−0.66
CHB6	0.38	0.71	1.02	0.03	3.00	2.12	−0.88
DARC	7.37	7.37	7.49	0.23	4.74	10.42	5.68
DC13	5.02	11.63	15.60	0.21	48.63	33.49	−15.14
DIPCS10	−3.66	5.94	7.75	0.01	21.98	3.38	−18.60
FH51	3.48	4.02	5.22	0.13	18.53	15.63	−2.90
G21EA	4.32	6.27	8.11	0.19	24.13	17.08	−7.05
G21IP	0.46	4.98	7.42	0.02	44.36	25.23	−19.13
G2RC	6.29	7.72	9.36	0.15	31.10	21.52	−9.58
HAL59	0.37	0.47	0.71	0.10	3.24	2.60	−0.64
HEAVY28	0.12	0.16	0.21	0.13	0.72	0.48	−0.24
HEAVYSB11	6.29	6.29	6.65	0.11	7.10	9.11	2.02
ICONF	−0.27	0.53	0.69	0.16	2.57	1.31	−1.26
IDISP	1.79	3.33	5.06	0.23	14.70	11.64	−3.06
IL16	0.87	0.87	1.05	0.01	1.77	1.79	0.02
INV24	−2.05	3.01	5.93	0.09	32.53	7.86	−24.67
ISO34	−0.49	1.36	1.96	0.09	9.98	3.12	−6.86
ISOL24	−2.30	4.58	6.14	0.21	28.04	14.55	−13.49
MB16-43	−5.90	24.65	29.86	0.06	118.00	57.83	−60.17
MCONF	0.36	0.53	0.61	0.11	1.87	1.17	−0.70
NBPRC	1.30	2.80	3.46	0.10	13.32	6.35	−6.97
PA26	−1.33	2.15	2.45	0.01	9.30	4.60	−4.69
PArel	−1.00	1.34	2.95	0.29	12.76	0.90	−11.85
PCONF21	−0.23	0.57	0.68	0.35	2.36	1.16	−1.20
PNICO23	−0.21	0.37	0.55	0.09	2.57	0.96	−1.61
PX13	−6.49	6.49	6.58	0.19	3.67	−5.16	−8.83
RC21	−6.80	7.17	8.83	0.20	22.17	2.82	−19.35
RG18	−0.02	0.14	0.21	0.24	1.02	0.67	−0.35
RSE43	1.77	2.06	4.55	0.27	21.07	19.82	−1.25
S22	−0.30	0.96	1.38	0.13	5.43	2.33	−3.10
S66	−0.31	0.55	0.86	0.10	4.14	1.34	−2.81
SCONF	0.04	0.21	0.29	0.05	1.23	0.43	−0.81
TAUT15	−0.58	1.10	1.27	0.36	3.37	1.30	−2.07
UPU23	−0.15	0.43	0.53	0.08	1.97	0.93	−1.03
W4-11	28.25	28.25	34.08	0.09	87.97	88.58	0.61
WATER27	−4.79	4.80	6.50	0.06	16.48	0.10	−16.37
WCPT18	−6.24	6.24	6.59	0.18	8.59	−2.79	−11.38
YBDE18	6.82	7.38	10.10	0.15	30.54	26.93	−3.61
BH76RC	1.45	4.57	6.57	0.21	29.96	21.42	−8.54

Table S79: Statistical analysis of the PBE0-DH-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.58	7.58	8.56	0.22	15.15	17.56	2.41
ACONF	−0.10	0.10	0.12	0.05	0.25	0.02	−0.23
ADIM6	−0.32	0.32	0.36	0.10	0.51	−0.09	−0.60
AHB21	−1.37	1.37	1.73	0.06	3.85	−0.06	−3.91
AL2X6	2.59	2.59	2.78	0.07	3.33	3.80	0.47
ALK8	3.58	3.69	5.00	0.06	9.63	9.19	−0.43
ALKBDE10	−5.81	5.93	7.16	0.06	15.31	0.58	−14.73
AMINO20x4	0.08	0.25	0.32	0.10	1.47	0.84	−0.63
BH76	−1.47	2.00	2.24	0.11	9.60	3.62	−5.97
BHDIV10	−2.42	2.93	3.36	0.06	7.59	2.40	−5.19
BHPERI	−2.27	2.87	3.18	0.14	9.75	4.12	−5.63
BHROT27	0.59	0.60	0.78	0.10	1.57	1.52	−0.05
BSR36	−2.47	2.47	2.70	0.15	4.05	−1.27	−5.32
BUT14DIOL	0.10	0.11	0.14	0.04	0.48	0.42	−0.06
C60ISO	2.10	3.12	3.34	0.03	8.02	5.47	−2.55
CARBHB12	1.16	1.16	1.40	0.19	2.28	2.62	0.34
CDIE20	0.90	0.94	1.16	0.23	2.73	2.46	−0.27
CHB6	−1.97	1.97	2.25	0.07	3.09	−0.81	−3.89
DARC	−7.37	7.37	7.61	0.23	6.59	−4.99	−11.58
DC13	−2.83	9.41	11.41	0.17	40.64	20.47	−20.16
DIPCS10	−0.65	2.60	3.35	0.00	11.18	4.07	−7.11
FH51	−2.44	3.12	3.93	0.10	15.46	5.54	−9.92
G21EA	−1.48	3.01	3.46	0.09	9.44	3.21	−6.23
G21IP	0.05	3.08	3.75	0.01	15.64	7.76	−7.88
G2RC	−4.18	6.60	8.40	0.13	34.86	11.64	−23.22
HAL59	0.45	0.56	0.87	0.12	3.36	2.97	−0.38
HEAVY28	0.22	0.24	0.33	0.19	0.83	0.67	−0.16
HEAVYSB11	0.43	0.77	0.92	0.01	2.65	1.91	−0.74
ICONF	0.22	0.28	0.42	0.09	1.35	1.13	−0.22
IDISP	−0.42	1.60	2.20	0.11	6.32	1.39	−4.93
IL16	−0.29	0.34	0.44	0.00	1.08	0.28	−0.80
INV24	0.50	1.55	2.28	0.05	10.43	6.22	−4.21
ISO34	0.07	1.45	1.99	0.10	8.01	4.20	−3.81
ISOL24	1.67	2.72	3.80	0.12	13.63	8.65	−4.98
MB16-43	14.42	18.04	22.74	0.04	95.82	73.26	−22.56
MCONF	0.14	0.23	0.29	0.05	1.04	0.62	−0.42
NBPRC	−1.54	3.48	4.02	0.13	12.88	5.84	−7.05
PA26	2.66	2.78	3.23	0.01	8.87	7.31	−1.57
PArel	0.88	1.21	1.59	0.26	6.04	4.12	−1.92
PCONF21	−0.06	0.62	0.75	0.38	2.43	1.03	−1.40
PNICO23	0.85	0.85	1.22	0.20	3.97	4.00	0.03
PX13	−3.92	3.92	4.12	0.12	4.58	−1.89	−6.46
RC21	4.28	4.79	5.97	0.13	18.37	16.08	−2.28
RG18	0.00	0.09	0.14	0.16	0.66	0.44	−0.23
RSE43	−0.47	0.49	0.64	0.06	2.19	0.40	−1.80
S22	0.26	0.42	0.59	0.06	2.47	1.86	−0.61
S66	0.16	0.35	0.49	0.06	2.43	1.93	−0.50
SCONF	0.07	0.20	0.26	0.04	1.08	0.30	−0.78
TAUT15	0.42	0.84	1.24	0.28	3.85	2.68	−1.18
UPU23	0.49	0.56	0.70	0.10	1.67	1.42	−0.25
W4-11	−4.76	5.37	7.13	0.02	36.20	9.08	−27.12
WATER27	3.77	4.12	5.06	0.05	18.20	13.37	−4.83
WCPT18	−2.36	2.57	3.26	0.07	8.23	1.91	−6.32
YBDE18	1.17	2.11	2.68	0.04	8.31	4.44	−3.88
BH76RC	−0.51	2.19	2.77	0.10	11.23	5.59	−5.64

Table S80: Statistical analysis of the LS1-DH-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	2.49	2.49	2.81	0.07	4.61	5.44	0.83
ACONF	−0.18	0.19	0.22	0.10	0.38	0.02	−0.36
ADIM6	−0.65	0.65	0.72	0.19	0.86	−0.23	−1.09
AHB21	−1.14	1.14	1.51	0.05	3.65	0.00	−3.65
AL2X6	1.24	1.27	1.40	0.04	2.11	2.00	−0.11
ALK8	2.92	2.92	4.63	0.05	11.15	11.33	0.17
ALKBDE10	−3.25	3.94	4.56	0.04	10.16	2.89	−7.26
AMINO20x4	0.05	0.19	0.24	0.08	1.10	0.63	−0.46
BH76	0.81	1.51	2.44	0.08	15.69	12.92	−2.77
BHDIV10	−1.34	2.01	2.27	0.04	5.75	2.04	−3.70
BHPERI	−3.13	3.48	3.77	0.17	9.12	2.90	−6.21
BHROT27	0.48	0.48	0.62	0.08	1.31	1.26	−0.05
BSR36	−1.10	1.10	1.14	0.07	1.19	−0.63	−1.82
BUT14DIOL	0.04	0.06	0.08	0.02	0.26	0.17	−0.09
C60ISO	−4.16	5.01	6.76	0.05	16.21	2.08	−14.13
CARBHB12	0.81	0.81	1.03	0.13	1.92	2.07	0.16
CDIE20	0.70	0.72	0.86	0.18	1.79	1.71	−0.08
CHB6	−1.52	1.52	1.70	0.06	2.04	−0.59	−2.63
DARC	−7.29	7.29	7.43	0.22	4.20	−5.07	−9.28
DC13	−1.48	8.62	10.61	0.16	36.92	19.91	−17.01
DIPCS10	−1.16	2.84	3.30	0.00	9.09	2.82	−6.27
FH51	−2.49	2.82	3.49	0.09	13.55	5.90	−7.66
G21EA	−1.42	2.42	2.89	0.07	10.32	5.05	−5.27
G21IP	0.13	2.28	2.87	0.01	14.11	5.56	−8.55
G2RC	−3.76	4.96	6.30	0.10	23.83	6.34	−17.49
HAL59	0.24	0.38	0.62	0.08	2.71	2.30	−0.41
HEAVY28	0.01	0.14	0.18	0.11	0.65	0.35	−0.30
HEAVYSB11	0.88	1.11	1.26	0.02	2.95	2.27	−0.67
ICONF	0.19	0.26	0.36	0.08	1.09	0.83	−0.25
IDISP	−1.02	2.07	3.23	0.15	8.98	1.38	−7.60
IL16	−0.19	0.29	0.34	0.00	0.96	0.29	−0.67
INV24	0.65	1.27	1.83	0.04	9.23	5.99	−3.24
ISO34	0.33	1.29	1.90	0.09	7.71	4.13	−3.58
ISOL24	2.06	3.04	4.15	0.14	13.84	10.81	−3.04
MB16-43	15.21	16.39	19.43	0.04	61.01	46.73	−14.28
MCONF	0.29	0.33	0.38	0.07	0.95	0.68	−0.27
NBPRC	−1.28	2.54	3.12	0.09	10.44	4.25	−6.19
PA26	1.39	1.74	2.07	0.01	7.23	4.93	−2.30
PArel	0.75	0.97	1.27	0.21	4.77	3.04	−1.74
PCONF21	0.05	0.38	0.48	0.23	1.70	0.71	−0.99
PNICO23	0.43	0.46	0.72	0.11	2.63	2.49	−0.14
PX13	−2.70	2.70	2.87	0.08	3.63	−0.93	−4.56
RC21	1.49	2.60	3.98	0.07	16.27	12.20	−4.06
RG18	−0.11	0.18	0.23	0.31	0.94	0.46	−0.48
RSE43	0.71	0.77	1.45	0.10	6.09	5.79	−0.30
S22	0.18	0.24	0.37	0.03	1.61	1.30	−0.31
S66	0.00	0.30	0.40	0.05	2.09	1.35	−0.74
SCONF	0.17	0.23	0.25	0.05	0.72	0.32	−0.40
TAUT15	0.50	0.72	0.98	0.24	2.71	2.03	−0.67
UPU23	0.40	0.49	0.61	0.09	1.59	1.31	−0.28
W4-11	−3.34	3.97	5.62	0.01	27.90	5.93	−21.97
WATER27	2.03	2.32	2.71	0.03	11.36	7.45	−3.91
WCPT18	−1.77	2.01	2.46	0.06	6.58	2.16	−4.43
YBDE18	2.92	3.36	4.19	0.07	9.07	7.42	−1.66
BH76RC	−0.49	1.86	2.43	0.09	10.89	5.86	−5.03

Table S81: Statistical analysis of the PBE0-2-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	1.81	1.81	2.05	0.05	3.17	3.61	0.44
ACONF	−0.20	0.20	0.23	0.11	0.41	0.02	−0.39
ADIM6	−0.69	0.69	0.76	0.21	0.89	−0.25	−1.14
AHB21	−1.03	1.04	1.38	0.05	3.55	0.04	−3.50
AL2X6	0.75	0.91	0.95	0.03	1.87	1.38	−0.49
ALK8	2.63	2.63	4.56	0.04	11.59	11.71	0.12
ALKBDE10	−2.12	3.58	3.88	0.04	10.57	3.86	−6.72
AMINO20x4	0.04	0.17	0.22	0.07	0.97	0.54	−0.42
BH76	1.21	1.85	2.93	0.10	18.11	15.00	−3.12
BHDIV10	−1.25	1.96	2.21	0.04	5.99	1.99	−4.00
BHPERI	−3.57	3.79	4.11	0.18	9.02	2.46	−6.55
BHROT27	0.44	0.44	0.56	0.07	1.22	1.18	−0.05
BSR36	−0.76	0.77	0.83	0.05	1.43	0.05	−1.39
BUT14DIOL	0.03	0.06	0.07	0.02	0.29	0.19	−0.10
C60ISO	−6.24	6.70	8.88	0.07	19.03	1.17	−17.87
CARBHB12	0.73	0.73	0.94	0.12	1.82	1.94	0.12
CDIE20	0.67	0.68	0.81	0.17	1.58	1.51	−0.07
CHB6	−1.36	1.36	1.51	0.05	1.79	−0.52	−2.31
DARC	−6.71	6.71	6.86	0.21	4.11	−4.49	−8.61
DC13	−0.92	8.08	10.25	0.15	36.55	20.51	−16.04
DIPCS10	−1.51	2.90	3.42	0.00	9.55	2.52	−7.03
FH51	−2.33	2.63	3.30	0.08	14.00	5.67	−8.34
G21EA	−1.27	2.23	2.77	0.07	11.57	6.58	−4.99
G21IP	0.09	2.11	2.83	0.01	16.26	7.43	−8.84
G2RC	−3.44	4.44	5.64	0.09	20.33	4.79	−15.54
HAL59	0.21	0.35	0.57	0.08	2.65	2.24	−0.41
HEAVY28	−0.02	0.14	0.17	0.11	0.61	0.29	−0.32
HEAVYSB11	1.05	1.19	1.38	0.02	2.89	2.41	−0.48
ICONF	0.18	0.24	0.33	0.07	1.04	0.73	−0.31
IDISP	−1.01	2.34	3.36	0.16	9.77	2.14	−7.63
IL16	−0.13	0.26	0.31	0.00	0.93	0.32	−0.62
INV24	0.56	1.11	1.62	0.03	8.16	5.39	−2.77
ISO34	0.37	1.25	1.83	0.09	7.55	4.16	−3.40
ISOL24	2.00	2.93	3.96	0.13	13.22	10.38	−2.84
MB16-43	14.99	15.91	18.45	0.04	51.52	38.73	−12.79
MCONF	0.32	0.36	0.41	0.07	1.01	0.75	−0.27
NBPRC	−1.13	2.21	2.73	0.08	9.12	3.58	−5.54
PA26	0.97	1.49	1.81	0.01	7.45	4.20	−3.25
PArel	0.67	0.88	1.18	0.19	4.82	3.11	−1.72
PCONF21	0.05	0.35	0.43	0.22	1.58	0.66	−0.92
PNICO23	0.32	0.38	0.60	0.09	2.33	2.15	−0.18
PX13	−2.65	2.65	2.81	0.08	3.50	−0.93	−4.43
RC21	0.44	2.61	3.63	0.07	14.76	10.08	−4.68
RG18	−0.13	0.19	0.26	0.33	0.99	0.47	−0.52
RSE43	1.04	1.06	2.02	0.14	8.50	8.32	−0.18
S22	0.15	0.22	0.31	0.03	1.40	1.07	−0.33
S66	−0.04	0.29	0.37	0.05	1.91	1.13	−0.79
SCONF	0.18	0.23	0.25	0.05	0.68	0.35	−0.33
TAUT15	0.52	0.71	0.94	0.23	2.50	1.92	−0.57
UPU23	0.36	0.46	0.57	0.08	1.58	1.25	−0.33
W4-11	−2.38	3.48	5.00	0.01	28.30	6.33	−21.97
WATER27	1.59	1.88	2.21	0.02	10.08	6.20	−3.88
WCPT18	−1.84	2.03	2.39	0.06	6.06	1.77	−4.28
YBDE18	3.33	3.57	4.48	0.07	8.89	7.68	−1.21
BH76RC	−0.42	1.95	2.45	0.09	10.98	6.56	−4.42

Table S82: Statistical analysis of the revPBE0-DH-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.30	7.30	8.48	0.22	16.44	18.18	1.73
ACONF	−0.08	0.09	0.11	0.05	0.26	0.03	−0.22
ADIM6	−0.29	0.29	0.32	0.09	0.40	−0.13	−0.53
AHB21	−0.43	0.70	0.99	0.03	3.57	0.66	−2.92
AL2X6	2.46	2.70	2.93	0.08	4.89	4.18	−0.71
ALK8	2.57	4.18	5.12	0.07	15.53	9.10	−6.44
ALKBDE10	−8.14	8.14	9.11	0.08	14.98	−2.42	−17.41
AMINO20x4	−0.01	0.27	0.34	0.11	1.63	0.87	−0.76
BH76	−0.63	1.28	1.58	0.07	8.84	5.13	−3.71
BHDIV10	−1.76	2.19	2.46	0.05	5.89	2.14	−3.75
BHPERI	−1.53	2.24	2.62	0.11	9.08	3.99	−5.09
BHROT27	0.50	0.50	0.65	0.08	1.28	1.24	−0.04
BSR36	−1.83	1.83	1.91	0.11	2.31	−0.86	−3.17
BUT14DIOL	−0.19	0.22	0.25	0.08	0.65	0.22	−0.43
C60ISO	2.23	3.18	3.41	0.03	7.91	5.49	−2.42
CARBHB12	0.70	0.70	0.95	0.12	1.91	1.99	0.08
CDIE20	0.90	0.92	1.16	0.23	2.71	2.45	−0.25
CHB6	−1.77	1.77	2.11	0.07	3.17	−0.50	−3.68
DARC	−5.94	5.94	6.14	0.18	5.59	−3.86	−9.45
DC13	−1.81	7.79	9.71	0.14	35.98	19.50	−16.48
DIPCS10	−1.29	3.06	3.92	0.00	13.02	4.14	−8.88
FH51	−1.72	2.37	2.90	0.08	9.97	3.52	−6.44
G21EA	−2.11	3.35	3.99	0.10	10.55	2.97	−7.59
G21IP	−0.44	3.35	3.95	0.01	15.24	7.54	−7.70
G2RC	−2.88	5.34	6.93	0.10	32.16	12.42	−19.74
HAL59	0.20	0.46	0.65	0.10	2.94	2.17	−0.77
HEAVY28	0.20	0.24	0.30	0.19	0.98	0.69	−0.29
HEAVYSB11	−0.14	1.36	1.59	0.02	5.09	2.17	−2.92
ICONF	0.16	0.25	0.39	0.08	1.40	1.08	−0.32
IDISP	−0.15	1.91	2.75	0.13	8.17	1.99	−6.17
IL16	0.56	0.56	0.69	0.01	1.12	1.10	−0.02
INV24	0.78	1.65	2.42	0.05	10.13	6.42	−3.71
ISO34	0.01	1.27	1.74	0.09	7.22	3.83	−3.39
ISOL24	1.26	2.26	3.06	0.10	11.36	7.13	−4.22
MB16-43	9.05	15.58	19.45	0.04	96.35	69.30	−27.05
MCONF	0.21	0.32	0.36	0.06	1.21	0.70	−0.51
NBPRC	−0.74	2.73	2.98	0.10	8.92	4.54	−4.38
PA26	3.90	3.91	4.34	0.02	9.02	8.95	−0.07
PArel	0.78	0.95	1.35	0.21	4.39	3.73	−0.67
PCONF21	−0.04	0.43	0.54	0.27	2.04	0.85	−1.19
PNICO23	0.60	0.68	1.00	0.16	3.90	3.43	−0.47
PX13	−1.90	1.90	2.10	0.06	2.97	−0.44	−3.41
RC21	2.98	3.54	4.47	0.10	14.92	12.91	−2.01
RG18	−0.05	0.10	0.17	0.17	0.86	0.47	−0.40
RSE43	−0.24	0.34	0.45	0.04	2.11	0.65	−1.46
S22	0.12	0.25	0.29	0.03	0.97	0.53	−0.43
S66	−0.01	0.20	0.24	0.04	1.12	0.61	−0.51
SCONF	−0.19	0.24	0.28	0.05	0.70	0.23	−0.48
TAUT15	0.18	0.64	0.77	0.21	2.65	1.53	−1.12
UPU23	0.36	0.51	0.61	0.09	2.00	1.27	−0.73
W4-11	−10.65	10.81	12.55	0.04	37.38	5.53	−31.86
WATER27	−2.76	3.05	4.78	0.04	14.66	1.07	−13.59
WCPT18	−1.00	1.46	1.90	0.04	6.53	2.30	−4.23
YBDE18	−0.62	2.83	3.21	0.06	10.71	3.67	−7.04
BH76RC	−0.46	1.97	2.53	0.09	9.36	3.54	−5.81

Table S83: Statistical analysis of the TPSS0-DH-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.73	7.73	9.69	0.23	19.48	20.92	1.45
ACONF	−0.13	0.13	0.14	0.07	0.25	0.01	−0.24
ADIM6	−0.39	0.39	0.43	0.12	0.56	−0.14	−0.70
AHB21	−1.01	1.04	1.40	0.05	3.57	0.23	−3.34
AL2X6	3.07	3.07	3.22	0.09	2.99	3.97	0.97
ALK8	2.98	3.63	4.40	0.06	11.42	8.81	−2.61
ALKBDE10	−7.09	7.09	8.23	0.07	15.32	−2.80	−18.12
AMINO20x4	0.06	0.23	0.29	0.09	1.39	0.88	−0.52
BH76	−2.06	2.64	3.41	0.14	14.07	4.79	−9.29
BHDIV10	−1.32	1.75	2.06	0.04	5.92	2.15	−3.77
BHPERI	−1.08	1.76	2.12	0.08	8.08	4.03	−4.05
BHROT27	0.57	0.58	0.78	0.09	1.66	1.52	−0.13
BSR36	−3.24	3.24	3.48	0.20	5.46	−1.54	−7.00
BUT14DIOL	0.00	0.07	0.08	0.03	0.39	0.25	−0.14
C60ISO	3.23	3.58	4.08	0.04	8.11	7.31	−0.80
CARBHB12	0.96	0.96	1.16	0.16	2.00	2.29	0.29
CDIE20	0.90	0.91	1.12	0.22	2.45	2.35	−0.09
CHB6	−1.96	1.96	2.23	0.07	3.00	−0.79	−3.78
DARC	−4.30	4.30	4.58	0.13	5.55	−2.30	−7.85
DC13	−2.29	6.58	8.69	0.12	33.55	17.34	−16.20
DIPCS10	0.57	3.94	4.78	0.01	16.68	8.36	−8.32
FH51	−1.60	2.21	2.92	0.07	9.62	2.59	−7.03
G21EA	−1.76	3.38	4.10	0.10	12.09	4.44	−7.65
G21IP	0.49	3.25	4.12	0.01	16.21	9.99	−6.22
G2RC	−3.30	5.22	6.76	0.10	28.89	10.75	−18.14
HAL59	0.36	0.50	0.79	0.11	3.25	2.73	−0.52
HEAVY28	0.21	0.25	0.32	0.20	0.93	0.69	−0.24
HEAVYSB11	−0.18	1.45	1.72	0.02	5.45	2.01	−3.43
ICONF	0.13	0.26	0.37	0.08	1.26	0.92	−0.34
IDISP	−0.16	1.37	1.53	0.10	3.64	1.23	−2.41
IL16	0.16	0.35	0.39	0.00	1.15	0.67	−0.48
INV24	0.75	1.59	2.67	0.05	11.91	7.99	−3.92
ISO34	−0.27	1.32	1.67	0.09	6.22	3.25	−2.97
ISOL24	0.69	1.82	2.38	0.08	9.92	5.38	−4.54
MB16-43	−7.53	16.35	21.63	0.04	107.59	53.30	−54.29
MCONF	0.15	0.23	0.29	0.05	1.09	0.60	−0.49
NBPRC	−1.15	3.17	3.39	0.11	10.39	5.82	−4.57
PA26	4.16	4.16	4.60	0.02	8.21	9.60	1.38
PArel	0.75	0.98	1.33	0.21	4.73	3.62	−1.11
PCONF21	−0.05	0.53	0.63	0.33	2.01	0.77	−1.24
PNICO23	0.75	0.77	1.04	0.18	3.17	3.00	−0.17
PX13	−1.83	1.83	2.04	0.05	3.01	−0.06	−3.07
RC21	3.05	3.47	4.38	0.10	14.98	12.89	−2.09
RG18	−0.01	0.08	0.14	0.14	0.67	0.47	−0.19
RSE43	−0.22	0.35	0.42	0.05	1.82	0.52	−1.31
S22	0.23	0.35	0.52	0.05	2.12	1.71	−0.41
S66	0.09	0.30	0.43	0.05	2.31	1.74	−0.57
SCONF	0.02	0.21	0.30	0.05	1.18	0.26	−0.92
TAUT15	0.28	0.72	0.90	0.24	2.97	1.80	−1.16
UPU23	0.48	0.53	0.68	0.09	1.54	1.26	−0.28
W4-11	−7.90	11.10	13.93	0.04	60.79	22.93	−37.86
WATER27	1.69	2.00	2.46	0.02	11.42	7.27	−4.15
WCPT18	−0.48	1.70	1.97	0.05	7.18	4.15	−3.03
YBDE18	−1.67	3.33	3.86	0.07	11.74	3.07	−8.68
BH76RC	−1.25	3.59	4.38	0.17	17.36	8.20	−9.15

TPSS0-DH had convergence problems for the h2+_1.75 system in SIE4x4.

Table S84: Statistical analysis of the PBE-QIDH-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	3.49	3.49	3.95	0.10	6.78	8.04	1.26
ACONF	−0.17	0.17	0.20	0.09	0.35	0.02	−0.33
ADIM6	−0.60	0.60	0.67	0.18	0.83	−0.20	−1.03
AHB21	−1.24	1.24	1.62	0.06	3.74	−0.04	−3.78
AL2X6	1.75	1.75	1.90	0.05	2.42	2.66	0.24
ALK8	3.20	3.20	4.74	0.05	10.70	10.89	0.20
ALKBDE10	−4.26	4.67	5.42	0.05	11.74	2.06	−9.68
AMINO20x4	0.06	0.21	0.26	0.09	1.22	0.72	−0.50
BH76	0.28	1.23	2.00	0.07	13.08	10.39	−2.69
BHDIV10	−1.52	2.14	2.45	0.05	5.71	2.11	−3.60
BHPERI	−2.75	3.21	3.51	0.15	9.19	3.24	−5.95
BHROT27	0.52	0.52	0.67	0.08	1.40	1.35	−0.05
BSR36	−1.46	1.46	1.52	0.09	1.50	−0.81	−2.31
BUT14DIOL	0.06	0.07	0.09	0.03	0.27	0.20	−0.07
C60ISO	−2.08	3.44	4.83	0.04	13.33	2.96	−10.37
CARBHB12	0.91	0.91	1.12	0.15	2.02	2.23	0.20
CDIE20	0.74	0.76	0.93	0.19	2.03	1.93	−0.10
CHB6	−1.69	1.69	1.89	0.06	2.41	−0.66	−3.07
DARC	−7.71	7.71	7.86	0.24	4.67	−5.49	−10.16
DC13	−2.04	9.05	11.01	0.16	36.63	19.00	−17.63
DIPCS10	−0.85	2.73	3.26	0.00	9.26	3.18	−6.09
FH51	−2.60	2.97	3.67	0.10	14.93	6.04	−8.90
G21EA	−1.53	2.64	3.07	0.08	9.11	3.62	−5.49
G21IP	0.15	2.47	3.04	0.01	14.42	6.06	−8.35
G2RC	−4.03	5.47	6.98	0.11	27.42	7.97	−19.45
HAL59	0.30	0.42	0.68	0.09	2.79	2.39	−0.40
HEAVY28	0.07	0.16	0.21	0.13	0.71	0.44	−0.27
HEAVYSB11	0.74	1.03	1.17	0.02	2.92	2.10	−0.82
ICONF	0.21	0.27	0.38	0.08	1.12	0.94	−0.18
IDISP	−0.97	1.73	3.05	0.12	8.26	0.94	−7.33
IL16	−0.25	0.31	0.38	0.00	0.99	0.25	−0.74
INV24	0.69	1.41	2.04	0.04	10.03	6.36	−3.66
ISO34	0.28	1.33	1.96	0.09	8.03	4.32	−3.71
ISOL24	2.07	3.09	4.25	0.14	14.20	10.83	−3.37
MB16-43	15.41	17.17	20.59	0.04	69.71	55.26	−14.45
MCONF	0.27	0.31	0.36	0.06	0.98	0.67	−0.31
NBPRC	−1.42	2.89	3.48	0.10	11.63	4.87	−6.76
PA26	1.81	2.02	2.41	0.01	6.97	5.68	−1.29
PArel	0.81	1.06	1.37	0.23	4.76	2.99	−1.78
PCONF21	0.04	0.43	0.53	0.27	1.85	0.78	−1.07
PNICO23	0.56	0.57	0.86	0.13	2.99	2.90	−0.09
PX13	−2.87	2.87	3.06	0.09	3.81	−1.04	−4.86
RC21	2.47	3.26	4.56	0.09	17.36	13.96	−3.40
RG18	−0.09	0.16	0.21	0.28	0.88	0.47	−0.42
RSE43	0.37	0.51	0.91	0.07	4.37	3.81	−0.56
S22	0.22	0.28	0.44	0.04	1.80	1.52	−0.28
S66	0.04	0.32	0.43	0.06	2.22	1.56	−0.66
SCONF	0.15	0.23	0.25	0.05	0.81	0.31	−0.50
TAUT15	0.47	0.73	1.03	0.24	2.93	2.16	−0.78
UPU23	0.43	0.51	0.64	0.09	1.59	1.36	−0.23
W4-11	−4.12	4.60	6.31	0.01	28.96	6.76	−22.20
WATER27	2.50	2.80	3.28	0.03	12.82	8.80	−4.02
WCPT18	−1.80	2.08	2.60	0.06	7.12	2.38	−4.74
YBDE18	2.47	3.10	3.86	0.06	9.61	6.94	−2.67
BH76RC	−0.54	1.93	2.49	0.09	10.72	5.36	−5.36

Table S85: Statistical analysis of the TPSS-QIDH-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	4.39	4.39	6.00	0.13	12.62	13.05	0.43
ACONF	−0.17	0.18	0.20	0.10	0.34	0.01	−0.33
ADIM6	−0.51	0.51	0.57	0.15	0.75	−0.17	−0.91
AHB21	−1.02	1.03	1.39	0.05	3.51	0.12	−3.39
AL2X6	1.42	1.42	1.53	0.04	1.73	1.92	0.19
ALK8	2.47	2.90	3.97	0.05	11.01	9.32	−1.69
ALKBDE10	−4.13	4.13	5.10	0.04	11.08	−0.10	−11.18
AMINO20x4	0.04	0.18	0.23	0.07	1.10	0.64	−0.45
BH76	−0.51	2.02	2.76	0.11	17.76	12.26	−5.50
BHDIV10	−0.44	1.18	1.46	0.03	5.00	2.48	−2.51
BHPERI	−1.36	1.83	2.06	0.09	6.45	2.65	−3.80
BHROT27	0.49	0.50	0.66	0.08	1.43	1.36	−0.07
BSR36	−2.56	2.56	2.72	0.16	3.69	−1.43	−5.12
BUT14DIOL	0.01	0.05	0.07	0.02	0.29	0.18	−0.11
C60ISO	−1.08	3.19	4.17	0.03	12.12	3.47	−8.65
CARBHB12	0.84	0.84	1.02	0.14	1.84	2.06	0.23
CDIE20	0.74	0.74	0.89	0.18	1.84	1.81	−0.03
CHB6	−1.65	1.65	1.82	0.06	2.05	−0.72	−2.77
DARC	−3.87	3.87	4.08	0.12	4.24	−2.01	−6.24
DC13	−1.16	5.30	6.85	0.10	26.24	11.86	−14.38
DIPCS10	2.25	3.81	4.69	0.01	13.23	8.51	−4.72
FH51	−1.93	2.17	2.66	0.07	8.59	1.68	−6.90
G21EA	−0.51	2.47	2.88	0.07	10.04	5.08	−4.97
G21IP	1.57	2.81	3.48	0.01	13.48	8.71	−4.77
G2RC	−3.83	4.73	5.89	0.09	20.23	6.28	−13.95
HAL59	0.25	0.38	0.59	0.08	2.49	2.04	−0.44
HEAVY28	0.10	0.17	0.22	0.14	0.74	0.46	−0.28
HEAVYSB11	−0.11	1.34	1.57	0.02	5.17	2.01	−3.16
ICONF	0.13	0.24	0.34	0.07	1.15	0.80	−0.35
IDISP	−0.49	1.28	1.67	0.09	4.16	0.79	−3.37
IL16	0.10	0.26	0.31	0.00	0.98	0.58	−0.40
INV24	0.76	1.38	2.20	0.04	10.89	7.40	−3.49
ISO34	0.19	0.98	1.39	0.07	5.98	3.57	−2.41
ISOL24	1.04	1.70	2.32	0.08	8.81	5.45	−3.36
MB16-43	−5.31	14.95	19.70	0.04	96.35	34.26	−62.09
MCONF	0.22	0.27	0.31	0.05	0.86	0.56	−0.31
NBPRC	−0.44	2.85	3.16	0.10	8.84	5.57	−3.27
PA26	2.84	2.84	3.27	0.02	7.04	7.29	0.25
PArel	0.64	0.77	1.07	0.17	3.26	2.52	−0.74
PCONF21	0.04	0.41	0.49	0.25	1.65	0.73	−0.92
PNICO23	0.45	0.47	0.66	0.11	1.99	1.89	−0.10
PX13	−1.20	1.27	1.44	0.04	2.61	0.34	−2.27
RC21	1.35	2.08	3.15	0.06	13.53	10.38	−3.14
RG18	−0.04	0.13	0.19	0.22	0.94	0.61	−0.33
RSE43	0.45	0.53	0.96	0.07	4.39	3.87	−0.52
S22	0.22	0.28	0.42	0.04	1.67	1.42	−0.25
S66	0.05	0.29	0.40	0.05	2.07	1.46	−0.61
SCONF	0.03	0.12	0.16	0.03	0.66	0.14	−0.52
TAUT15	0.31	0.57	0.78	0.19	2.54	1.52	−1.02
UPU23	0.46	0.51	0.65	0.09	1.42	1.20	−0.22
W4-11	−4.35	8.83	11.16	0.03	57.47	24.42	−33.05
WATER27	1.51	1.75	2.14	0.02	9.72	6.41	−3.31
WCPT18	−0.26	1.43	1.65	0.04	6.33	3.92	−2.41
YBDE18	0.03	2.95	3.22	0.06	10.11	4.81	−5.31
BH76RC	−1.47	2.86	3.66	0.13	15.99	5.32	−10.67

TPSS-QIDH had convergence problems for the h2+_1.75 system in SIE4x4.

Table S86: Statistical analysis of the PBE-CIDH-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	6.46	6.46	7.30	0.19	12.84	15.04	2.20
ACONF	−0.11	0.12	0.14	0.07	0.27	0.02	−0.25
ADIM6	−0.39	0.39	0.44	0.12	0.59	−0.12	−0.71
AHB21	−1.35	1.35	1.73	0.06	3.84	−0.07	−3.91
AL2X6	2.44	2.44	2.62	0.07	3.11	3.60	0.49
ALK8	3.54	3.55	4.95	0.06	9.74	9.66	−0.08
ALKBDE10	−5.57	5.73	6.82	0.06	14.51	0.78	−13.73
AMINO20x4	0.08	0.24	0.30	0.10	1.39	0.82	−0.57
BH76	−1.03	1.67	1.95	0.09	9.68	4.69	−4.99
BHDIV10	−2.16	2.70	3.10	0.06	7.05	2.32	−4.74
BHPERI	−2.32	2.91	3.22	0.14	9.61	3.95	−5.66
BHROT27	0.58	0.58	0.76	0.09	1.53	1.49	−0.05
BSR36	−2.22	2.22	2.41	0.14	3.39	−1.16	−4.55
BUT14DIOL	0.09	0.10	0.13	0.04	0.43	0.37	−0.06
C60ISO	1.24	2.90	3.17	0.03	8.57	4.38	−4.19
CARBHB12	1.10	1.10	1.33	0.18	2.22	2.53	0.31
CDIE20	0.86	0.89	1.10	0.22	2.57	2.34	−0.24
CHB6	−1.92	1.92	2.18	0.07	2.96	−0.78	−3.74
DARC	−7.61	7.61	7.82	0.23	6.10	−5.29	−11.39
DC13	−2.70	9.41	11.35	0.17	39.05	19.63	−19.42
DIPCS10	−0.60	2.65	3.28	0.00	10.86	4.03	−6.83
FH51	−2.53	3.11	3.89	0.10	15.71	5.76	−9.95
G21EA	−1.52	2.94	3.38	0.09	9.00	2.99	−6.01
G21IP	0.10	2.90	3.56	0.01	15.29	7.38	−7.90
G2RC	−4.20	6.38	8.10	0.12	33.34	10.86	−22.48
HAL59	0.41	0.52	0.82	0.11	3.23	2.84	−0.39
HEAVY28	0.18	0.22	0.29	0.18	0.80	0.61	−0.19
HEAVYSB11	0.50	0.79	0.96	0.01	2.64	1.82	−0.82
ICONF	0.22	0.28	0.42	0.09	1.30	1.10	−0.20
IDISP	−0.59	1.55	2.42	0.11	6.61	0.93	−5.68
IL16	−0.29	0.34	0.43	0.00	1.06	0.26	−0.80
INV24	0.57	1.54	2.25	0.05	10.16	6.02	−4.14
ISO34	0.13	1.42	1.99	0.10	8.11	4.30	−3.81
ISOL24	1.81	2.86	3.97	0.13	13.42	9.07	−4.35
MB16-43	14.77	17.80	22.25	0.04	88.18	69.47	−18.71
MCONF	0.18	0.24	0.30	0.05	1.03	0.64	−0.39
NBPRC	−1.54	3.38	3.94	0.12	12.80	5.69	−7.12
PA26	2.49	2.60	3.06	0.01	8.46	6.97	−1.49
PArel	0.88	1.18	1.55	0.25	5.67	3.78	−1.89
PCONF21	−0.03	0.56	0.69	0.35	2.27	0.96	−1.31
PNICO23	0.79	0.79	1.14	0.19	3.74	3.74	0.00
PX13	−3.61	3.61	3.81	0.11	4.37	−1.63	−6.00
RC21	3.94	4.48	5.68	0.13	18.11	15.81	−2.29
RG18	−0.03	0.11	0.15	0.19	0.72	0.44	−0.28
RSE43	−0.29	0.35	0.50	0.05	2.34	0.95	−1.39
S22	0.26	0.39	0.56	0.05	2.30	1.81	−0.49
S66	0.13	0.35	0.48	0.06	2.39	1.86	−0.53
SCONF	0.09	0.21	0.25	0.05	1.01	0.30	−0.71
TAUT15	0.43	0.80	1.18	0.26	3.60	2.52	−1.08
UPU23	0.48	0.54	0.69	0.09	1.65	1.41	−0.25
W4-11	−4.73	5.30	7.05	0.02	34.21	8.62	−25.60
WATER27	3.48	3.82	4.64	0.05	16.85	12.26	−4.59
WCPT18	−2.18	2.42	3.08	0.07	7.99	2.12	−5.87
YBDE18	1.48	2.38	2.98	0.05	8.92	5.15	−3.77
BH76RC	−0.53	2.14	2.70	0.10	10.96	5.38	−5.58

Table S87: Statistical analysis of the TPSS-CIDH-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	6.83	6.83	8.72	0.20	17.77	18.92	1.14
ACONF	−0.14	0.14	0.16	0.08	0.27	0.01	−0.26
ADIM6	−0.42	0.42	0.47	0.13	0.60	−0.15	−0.75
AHB21	−1.03	1.05	1.42	0.05	3.61	0.20	−3.42
AL2X6	2.70	2.70	2.83	0.08	2.66	3.49	0.83
ALK8	2.90	3.48	4.31	0.06	11.28	8.95	−2.32
ALKBDE10	−6.47	6.47	7.54	0.06	14.33	−2.32	−16.64
AMINO20x4	0.06	0.22	0.27	0.09	1.31	0.82	−0.49
BH76	−1.70	2.45	3.19	0.13	14.88	6.57	−8.31
BHDIV10	−1.06	1.51	1.86	0.03	5.70	2.23	−3.47
BHPERI	−1.06	1.72	2.05	0.08	7.60	3.70	−3.90
BHROT27	0.55	0.56	0.75	0.09	1.61	1.49	−0.12
BSR36	−3.09	3.09	3.31	0.19	5.05	−1.53	−6.58
BUT14DIOL	0.00	0.06	0.08	0.02	0.35	0.22	−0.13
C60ISO	2.34	3.33	3.61	0.03	8.61	6.19	−2.42
CARBHB12	0.93	0.93	1.13	0.15	1.96	2.24	0.28
CDIE20	0.86	0.87	1.06	0.21	2.32	2.23	−0.09
CHB6	−1.90	1.90	2.15	0.07	2.80	−0.77	−3.57
DARC	−4.31	4.31	4.55	0.13	5.22	−2.36	−7.58
DC13	−2.07	6.11	8.22	0.11	31.84	16.07	−15.77
DIPCS10	1.14	3.88	4.74	0.01	15.98	8.64	−7.34
FH51	−1.76	2.20	2.87	0.07	9.46	1.81	−7.64
G21EA	−1.44	3.15	3.78	0.09	11.20	4.46	−6.73
G21IP	0.81	3.08	3.91	0.01	15.15	9.74	−5.41
G2RC	−3.57	5.14	6.62	0.10	26.99	9.75	−17.24
HAL59	0.33	0.47	0.74	0.10	3.06	2.55	−0.50
HEAVY28	0.18	0.23	0.29	0.19	0.89	0.64	−0.25
HEAVYSB11	−0.19	1.43	1.69	0.02	5.35	1.91	−3.44
ICONF	0.14	0.26	0.36	0.08	1.23	0.90	−0.33
IDISP	−0.31	1.43	1.67	0.10	3.81	1.06	−2.75
IL16	0.14	0.32	0.37	0.00	1.08	0.61	−0.47
INV24	0.78	1.55	2.59	0.05	11.87	7.99	−3.88
ISO34	−0.15	1.22	1.57	0.08	6.16	3.31	−2.85
ISOL24	0.80	1.79	2.38	0.08	9.49	5.48	−4.01
MB16-43	−7.82	16.49	21.82	0.04	108.02	48.76	−59.26
MCONF	0.17	0.24	0.29	0.05	1.02	0.58	−0.44
NBPRC	−1.00	3.16	3.37	0.11	10.18	5.85	−4.33
PA26	3.86	3.86	4.30	0.02	7.88	9.09	1.21
PArel	0.74	0.94	1.27	0.20	4.36	3.36	−1.00
PCONF21	−0.03	0.50	0.60	0.31	1.92	0.76	−1.16
PNICO23	0.68	0.70	0.95	0.16	2.88	2.73	−0.15
PX13	−1.61	1.63	1.83	0.05	2.88	0.09	−2.78
RC21	2.73	3.20	4.11	0.09	14.82	12.50	−2.32
RG18	−0.02	0.09	0.15	0.16	0.74	0.51	−0.23
RSE43	−0.08	0.28	0.37	0.04	2.19	1.06	−1.13
S22	0.23	0.33	0.50	0.05	2.02	1.66	−0.36
S66	0.08	0.30	0.43	0.05	2.28	1.70	−0.58
SCONF	0.02	0.18	0.26	0.04	1.03	0.22	−0.81
TAUT15	0.28	0.68	0.86	0.22	2.86	1.72	−1.14
UPU23	0.48	0.53	0.67	0.09	1.51	1.24	−0.27
W4-11	−7.09	10.67	13.37	0.03	60.80	23.79	−37.00
WATER27	1.66	1.95	2.39	0.02	10.96	7.07	−3.89
WCPT18	−0.38	1.64	1.89	0.05	7.03	4.19	−2.84
YBDE18	−1.27	3.20	3.67	0.06	11.57	3.55	−8.02
BH76RC	−1.33	3.46	4.24	0.16	17.33	7.53	−9.80

TPSS-CIDH had convergence problems for the h2+_1.75 system in SIE4x4.

Table S88: Statistical analysis of the SOS0-PBE0-2-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	1.18	1.86	2.29	0.06	6.85	5.17	−1.67
ACONF	−0.24	0.24	0.26	0.13	0.40	0.00	−0.40
ADIM6	−0.38	0.38	0.43	0.11	0.57	−0.12	−0.68
AHB21	−0.35	0.46	0.72	0.02	2.43	0.32	−2.10
AL2X6	0.93	0.93	1.08	0.03	1.42	1.59	0.17
ALK8	1.58	2.26	3.56	0.04	11.97	9.25	−2.72
ALKBDE10	−4.85	4.85	5.20	0.05	6.19	−2.08	−8.28
AMINO20x4	−0.05	0.18	0.22	0.07	0.97	0.42	−0.55
BH76	2.33	2.46	3.64	0.13	18.76	17.20	−1.57
BHDIV10	0.66	0.96	1.13	0.02	2.60	1.75	−0.84
BHPERI	0.67	1.20	1.66	0.06	5.68	4.73	−0.95
BHROT27	0.24	0.25	0.33	0.04	0.95	0.80	−0.15
BSR36	−2.34	2.34	2.48	0.14	3.17	−1.32	−4.50
BUT14DIOL	−0.18	0.18	0.20	0.06	0.37	0.04	−0.33
C60ISO	−1.24	2.58	3.27	0.03	9.11	2.29	−6.83
CARBHB12	0.32	0.32	0.43	0.05	1.07	1.04	−0.03
CDIE20	0.23	0.33	0.47	0.08	1.36	1.11	−0.25
CHB6	−1.64	1.64	1.84	0.06	2.27	−0.63	−2.90
DARC	−5.18	5.18	5.21	0.16	1.64	−4.37	−6.01
DC13	−2.45	5.58	6.49	0.10	21.80	9.86	−11.94
DIPCS10	−1.97	3.10	3.60	0.00	10.47	2.86	−7.61
FH51	−1.50	1.74	2.25	0.06	10.39	2.39	−8.00
G21EA	−2.09	2.79	3.33	0.08	13.60	8.21	−5.39
G21IP	−0.12	1.95	2.81	0.01	16.37	10.82	−5.54
G2RC	−2.09	3.04	3.78	0.06	14.19	5.03	−9.16
HAL59	0.04	0.31	0.40	0.07	1.86	1.10	−0.76
HEAVY28	0.04	0.14	0.17	0.11	0.77	0.40	−0.37
HEAVYSB11	0.95	1.81	1.97	0.03	5.01	2.73	−2.28
ICONF	0.02	0.19	0.27	0.06	1.21	0.50	−0.71
IDISP	−0.65	1.82	3.11	0.13	8.76	1.43	−7.33
IL16	0.51	0.51	0.57	0.00	0.88	0.90	0.02
INV24	1.15	1.49	2.40	0.05	11.78	9.18	−2.60
ISO34	−0.02	0.81	1.17	0.06	5.50	3.10	−2.41
ISOL24	1.16	1.87	2.75	0.09	11.68	6.39	−5.29
MB16-43	−2.75	9.49	12.65	0.02	63.76	20.50	−43.26
MCONF	0.30	0.35	0.38	0.07	0.83	0.60	−0.22
NBPRC	−0.38	1.62	1.93	0.06	6.58	3.48	−3.10
PA26	3.05	3.05	3.28	0.02	5.00	6.00	1.00
PArel	0.36	0.44	0.66	0.10	2.16	1.60	−0.57
PCONF21	0.12	0.25	0.32	0.15	1.12	0.63	−0.49
PNICO23	0.17	0.21	0.28	0.05	0.83	0.62	−0.21
PX13	0.12	0.52	0.62	0.02	1.85	1.20	−0.65
RC21	−0.48	1.98	2.70	0.06	11.15	6.36	−4.79
RG18	−0.07	0.16	0.24	0.28	1.09	0.67	−0.42
RSE43	1.52	1.52	2.43	0.20	10.40	10.57	0.18
S22	0.07	0.23	0.28	0.03	1.01	0.52	−0.49
S66	−0.03	0.15	0.20	0.03	0.86	0.39	−0.47
SCONF	−0.17	0.20	0.22	0.04	0.51	0.16	−0.35
TAUT15	−0.29	0.51	0.55	0.17	1.37	0.57	−0.80
UPU23	0.42	0.50	0.61	0.09	1.60	1.19	−0.41
W4-11	−3.03	4.53	6.81	0.01	40.68	9.14	−31.53
WATER27	−1.27	1.52	2.52	0.02	8.23	0.70	−7.53
WCPT18	0.99	1.05	1.39	0.03	3.80	3.55	−0.25
YBDE18	2.22	2.34	2.78	0.05	6.06	4.99	−1.07
BH76RC	−0.42	1.43	1.90	0.07	7.86	2.95	−4.91

Table S89: Statistical analysis of the SOS0-PBE0-DH-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.39	7.39	8.50	0.22	15.65	18.08	2.42
ACONF	−0.11	0.11	0.12	0.06	0.25	0.02	−0.23
ADIM6	−0.23	0.23	0.27	0.07	0.43	−0.05	−0.48
AHB21	−1.16	1.16	1.52	0.05	3.41	0.03	−3.38
AL2X6	2.62	2.62	2.79	0.07	3.18	3.85	0.67
ALK8	3.21	3.58	4.55	0.06	9.76	8.26	−1.50
ALKBDE10	−6.69	6.69	7.76	0.07	14.78	−1.24	−16.02
AMINO20x4	0.05	0.23	0.29	0.09	1.41	0.80	−0.61
BH76	−1.12	1.71	1.95	0.09	7.58	3.78	−3.80
BHDIV10	−1.84	2.25	2.71	0.05	6.51	1.95	−4.57
BHPERI	−1.02	1.87	2.31	0.09	9.33	5.37	−3.95
BHROT27	0.52	0.53	0.71	0.08	1.42	1.38	−0.05
BSR36	−2.97	2.97	3.27	0.18	5.12	−1.52	−6.65
BUT14DIOL	0.03	0.08	0.11	0.03	0.48	0.36	−0.12
C60ISO	3.70	3.70	4.29	0.04	7.42	7.97	0.55
CARBHB12	1.03	1.03	1.23	0.17	2.00	2.34	0.34
CDIE20	0.75	0.81	1.05	0.20	2.68	2.32	−0.36
CHB6	−2.06	2.06	2.35	0.08	3.18	−0.84	−4.03
DARC	−6.89	6.89	7.09	0.21	6.05	−4.92	−10.96
DC13	−3.21	8.66	10.42	0.16	35.69	18.73	−16.96
DIPCS10	−0.78	2.73	3.51	0.00	12.23	4.53	−7.70
FH51	−2.19	2.88	3.73	0.09	14.90	4.52	−10.38
G21EA	−1.76	2.72	3.37	0.08	9.49	2.43	−7.06
G21IP	−0.05	2.90	3.53	0.01	14.41	7.56	−6.85
G2RC	−3.79	6.26	7.95	0.12	33.06	11.73	−21.33
HAL59	0.39	0.49	0.77	0.11	3.11	2.68	−0.42
HEAVY28	0.24	0.26	0.33	0.21	0.87	0.69	−0.18
HEAVYSB11	0.37	0.88	1.02	0.02	3.21	1.85	−1.36
ICONF	0.17	0.25	0.38	0.08	1.28	1.04	−0.24
IDISP	−0.31	1.74	2.26	0.12	6.68	1.82	−4.86
IL16	−0.09	0.31	0.36	0.00	1.10	0.45	−0.66
INV24	0.66	1.60	2.49	0.05	11.38	7.19	−4.20
ISO34	−0.04	1.33	1.82	0.09	7.51	3.95	−3.56
ISOL24	1.42	2.46	3.42	0.11	12.48	7.86	−4.61
MB16-43	9.19	15.24	19.76	0.04	93.42	66.95	−26.48
MCONF	0.13	0.20	0.25	0.04	0.99	0.59	−0.40
NBPRC	−1.31	3.31	3.76	0.12	12.10	5.80	−6.31
PA26	3.24	3.30	3.71	0.02	8.61	7.77	−0.85
PArel	0.78	1.05	1.44	0.23	5.74	4.11	−1.63
PCONF21	−0.04	0.54	0.65	0.33	2.16	0.91	−1.25
PNICO23	0.79	0.79	1.10	0.19	3.51	3.48	−0.03
PX13	−3.12	3.12	3.32	0.09	3.97	−1.27	−5.24
RC21	3.86	4.38	5.40	0.12	16.48	14.61	−1.87
RG18	0.01	0.09	0.14	0.16	0.69	0.50	−0.19
RSE43	−0.31	0.35	0.48	0.05	1.84	0.32	−1.52
S22	0.24	0.37	0.51	0.05	2.19	1.64	−0.55
S66	0.16	0.29	0.42	0.05	2.11	1.70	−0.41
SCONF	−0.04	0.13	0.19	0.03	0.85	0.20	−0.66
TAUT15	0.18	0.74	0.99	0.24	3.13	1.96	−1.17
UPU23	0.52	0.57	0.71	0.10	1.64	1.43	−0.21
W4-11	−5.11	5.80	7.81	0.02	37.77	9.94	−27.83
WATER27	2.90	3.20	3.86	0.04	14.44	10.39	−4.05
WCPT18	−1.53	2.01	2.56	0.06	7.32	2.29	−5.03
YBDE18	0.88	1.82	2.33	0.04	7.82	3.79	−4.03
BH76RC	−0.51	2.12	2.64	0.10	8.72	3.54	−5.17

Table S90: Statistical analysis of the SOS1-PBE-QIDH-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	3.05	3.09	3.98	0.09	9.58	9.23	−0.35
ACONF	−0.19	0.19	0.21	0.10	0.35	0.01	−0.34
ADIM6	−0.35	0.35	0.39	0.10	0.53	−0.10	−0.64
AHB21	−0.75	0.77	1.12	0.03	3.16	0.18	−2.98
AL2X6	1.80	1.80	1.92	0.05	2.06	2.72	0.67
ALK8	2.40	2.84	3.79	0.05	10.69	8.93	−1.76
ALKBDE10	−6.24	6.24	6.80	0.06	10.56	−2.14	−12.70
AMINO20x4	0.00	0.18	0.23	0.07	1.10	0.60	−0.51
BH76	1.10	1.47	2.33	0.08	14.07	12.02	−2.05
BHDIV10	−0.14	1.15	1.26	0.03	3.24	1.30	−1.94
BHPERI	0.26	1.42	1.83	0.07	7.64	5.72	−1.92
BHROT27	0.37	0.38	0.50	0.06	1.13	1.07	−0.05
BSR36	−2.61	2.61	2.81	0.16	3.95	−1.42	−5.37
BUT14DIOL	−0.09	0.10	0.12	0.04	0.34	0.10	−0.23
C60ISO	1.53	2.51	2.89	0.03	8.02	5.59	−2.43
CARBHB12	0.60	0.60	0.74	0.10	1.40	1.57	0.17
CDIE20	0.41	0.50	0.69	0.12	1.94	1.63	−0.30
CHB6	−1.87	1.87	2.11	0.07	2.63	−0.73	−3.36
DARC	−6.55	6.55	6.62	0.20	3.37	−5.31	−8.68
DC13	−3.05	7.26	8.43	0.13	27.00	12.25	−14.75
DIPCS10	−1.17	2.88	3.51	0.00	11.93	4.31	−7.61
FH51	−2.01	2.36	3.04	0.08	13.72	3.63	−10.09
G21EA	−2.14	2.57	3.21	0.08	11.22	4.52	−6.71
G21IP	−0.03	2.04	2.71	0.01	11.67	5.65	−6.02
G2RC	−3.09	4.60	5.77	0.09	23.01	8.13	−14.88
HAL59	0.16	0.34	0.49	0.07	2.23	1.74	−0.49
HEAVY28	0.10	0.17	0.22	0.14	0.83	0.52	−0.30
HEAVYSB11	0.59	1.43	1.57	0.02	4.45	2.19	−2.25
ICONF	0.09	0.22	0.31	0.07	1.19	0.73	−0.46
IDISP	−0.70	1.66	2.95	0.12	8.11	1.04	−7.08
IL16	0.23	0.34	0.38	0.00	0.97	0.65	−0.32
INV24	1.08	1.59	2.63	0.05	12.55	8.99	−3.57
ISO34	0.00	1.04	1.51	0.07	6.74	3.74	−3.00
ISOL24	1.46	2.31	3.39	0.11	13.51	7.93	−5.59
MB16-43	2.62	11.25	14.55	0.03	76.87	40.69	−36.18
MCONF	0.24	0.28	0.31	0.06	0.85	0.60	−0.25
NBPRC	−0.86	2.46	2.85	0.09	9.74	4.78	−4.96
PA26	3.24	3.24	3.57	0.02	6.27	6.88	0.61
PArel	0.58	0.73	1.02	0.16	3.67	2.66	−1.00
PCONF21	0.07	0.29	0.36	0.18	1.28	0.49	−0.79
PNICO23	0.42	0.44	0.60	0.10	1.88	1.74	−0.14
PX13	−0.94	1.08	1.21	0.03	2.33	0.46	−1.87
RC21	1.67	2.47	3.45	0.07	13.93	10.66	−3.27
RG18	−0.05	0.13	0.20	0.22	0.95	0.60	−0.35
RSE43	0.72	0.74	1.14	0.10	5.02	4.73	−0.29
S22	0.15	0.20	0.28	0.03	1.18	0.97	−0.20
S66	0.04	0.20	0.26	0.04	1.45	1.03	−0.42
SCONF	−0.11	0.13	0.15	0.03	0.29	0.07	−0.22
TAUT15	−0.10	0.47	0.51	0.15	1.39	0.51	−0.88
UPU23	0.48	0.53	0.66	0.09	1.49	1.27	−0.23
W4-11	−4.71	5.65	7.89	0.02	37.67	9.13	−28.54
WATER27	0.50	0.94	1.16	0.01	4.28	2.15	−2.13
WCPT18	0.20	1.28	1.46	0.04	5.34	3.53	−1.81
YBDE18	1.68	2.25	2.76	0.05	8.31	5.13	−3.18
BH76RC	−0.54	1.73	2.25	0.08	8.83	3.20	−5.63

Table S91: Statistical analysis of the SOS0-PBE-CIDH-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	6.16	6.16	7.19	0.18	13.69	15.65	1.96
ACONF	−0.13	0.13	0.14	0.07	0.27	0.02	−0.26
ADIM6	−0.27	0.27	0.31	0.08	0.46	−0.07	−0.53
AHB21	−1.09	1.10	1.46	0.05	3.48	0.05	−3.43
AL2X6	2.50	2.50	2.66	0.07	2.89	3.67	0.78
ALK8	3.08	3.43	4.36	0.05	9.84	8.43	−1.41
ALKBDE10	−6.87	6.87	7.80	0.07	14.15	−1.66	−15.81
AMINO20x4	0.05	0.22	0.28	0.09	1.36	0.76	−0.60
BH76	−0.53	1.36	1.70	0.07	9.04	5.72	−3.32
BHDIV10	−1.37	1.87	2.25	0.04	5.66	1.74	−3.92
BHPERI	−0.63	1.71	2.15	0.08	9.14	5.73	−3.41
BHROT27	0.50	0.50	0.67	0.08	1.38	1.33	−0.05
BSR36	−2.90	2.90	3.18	0.18	4.87	−1.51	−6.37
BUT14DIOL	0.00	0.08	0.09	0.03	0.44	0.29	−0.15
C60ISO	3.54	3.54	4.18	0.04	7.57	7.96	0.39
CARBHB12	0.93	0.93	1.11	0.15	1.86	2.17	0.30
CDIE20	0.66	0.73	0.96	0.18	2.54	2.18	−0.36
CHB6	−2.04	2.04	2.32	0.08	3.12	−0.83	−3.94
DARC	−7.06	7.06	7.22	0.22	5.43	−5.27	−10.70
DC13	−3.27	8.51	10.11	0.15	33.25	17.50	−15.75
DIPCS10	−0.76	2.78	3.53	0.00	12.51	4.80	−7.71
FH51	−2.24	2.83	3.65	0.09	15.19	4.46	−10.73
G21EA	−1.92	2.65	3.38	0.08	9.41	2.15	−7.27
G21IP	−0.03	2.70	3.32	0.01	13.76	7.16	−6.61
G2RC	−3.76	5.99	7.58	0.12	31.28	11.08	−20.20
HAL59	0.33	0.44	0.69	0.10	2.91	2.46	−0.45
HEAVY28	0.20	0.23	0.30	0.19	0.86	0.65	−0.21
HEAVYSB11	0.36	0.99	1.13	0.02	3.38	1.67	−1.71
ICONF	0.16	0.26	0.37	0.08	1.22	0.98	−0.24
IDISP	−0.47	1.78	2.49	0.13	7.18	1.54	−5.64
IL16	−0.03	0.30	0.35	0.00	1.09	0.49	−0.60
INV24	0.81	1.63	2.61	0.05	11.76	7.57	−4.19
ISO34	−0.02	1.28	1.78	0.09	7.34	4.01	−3.32
ISOL24	1.50	2.52	3.54	0.11	12.29	8.00	−4.29
MB16-43	7.88	14.29	18.58	0.03	85.86	61.71	−24.15
MCONF	0.16	0.21	0.26	0.04	0.96	0.60	−0.36
NBPRC	−1.24	3.18	3.63	0.11	11.92	5.71	−6.20
PA26	3.28	3.32	3.73	0.02	8.17	7.62	−0.55
PArel	0.75	1.00	1.36	0.22	5.28	3.79	−1.49
PCONF21	−0.01	0.47	0.57	0.29	1.95	0.81	−1.14
PNICO23	0.71	0.71	0.99	0.17	3.13	3.07	−0.06
PX13	−2.52	2.52	2.72	0.08	3.55	−0.77	−4.33
RC21	3.45	4.01	5.01	0.11	16.04	14.10	−1.94
RG18	0.00	0.10	0.15	0.17	0.76	0.52	−0.24
RSE43	−0.08	0.25	0.36	0.03	2.02	0.80	−1.22
S22	0.23	0.33	0.47	0.05	1.97	1.53	−0.44
S66	0.13	0.28	0.39	0.05	2.00	1.59	−0.41
SCONF	−0.06	0.11	0.16	0.02	0.69	0.16	−0.53
TAUT15	0.12	0.67	0.85	0.22	2.65	1.59	−1.07
UPU23	0.51	0.56	0.71	0.10	1.57	1.39	−0.18
W4-11	−5.42	6.10	8.21	0.02	36.61	9.51	−27.10
WATER27	2.37	2.63	3.12	0.03	12.03	8.49	−3.53
WCPT18	−1.03	1.76	2.19	0.05	6.89	2.77	−4.12
YBDE18	1.05	2.00	2.53	0.04	8.55	4.33	−4.22
BH76RC	−0.55	2.07	2.57	0.10	7.87	2.85	−5.03

Table S92: Statistical analysis of the SOS0-PBE0-2(b)-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	4.46	4.46	4.94	0.13	7.45	8.53	1.09
ACONF	−0.34	0.34	0.36	0.19	0.42	−0.03	−0.45
ADIM6	−0.61	0.61	0.68	0.18	0.80	−0.22	−1.03
AHB21	−0.52	0.55	0.71	0.02	2.39	0.20	−2.19
AL2X6	0.77	1.17	1.21	0.03	2.99	1.78	−1.20
ALK8	2.78	3.75	6.40	0.06	19.69	16.25	−3.44
ALKBDE10	9.95	9.95	11.41	0.10	14.68	18.79	4.11
AMINO20x4	−0.06	0.14	0.18	0.06	0.75	0.29	−0.46
BH76	0.52	2.15	3.87	0.12	25.53	21.03	−4.50
BHDIV10	−2.15	2.61	2.98	0.06	7.46	2.31	−5.15
BHPERI	−3.31	3.46	3.75	0.17	7.58	1.90	−5.68
BHROT27	0.00	0.12	0.17	0.02	0.72	0.23	−0.49
BSR36	−1.10	1.10	1.15	0.07	1.54	−0.50	−2.04
BUT14DIOL	0.00	0.05	0.07	0.02	0.30	0.22	−0.09
C60ISO	−13.15	13.15	15.51	0.13	23.93	−3.32	−27.24
CARBHB12	0.52	0.52	0.67	0.09	1.29	1.37	0.08
CDIE20	0.27	0.30	0.38	0.07	1.09	0.97	−0.13
CHB6	−1.17	1.17	1.36	0.04	2.08	−0.42	−2.50
DARC	−5.64	5.64	5.70	0.17	2.33	−4.33	−6.66
DC13	−2.72	9.64	12.60	0.18	50.88	21.97	−28.91
DIPCS10	6.79	6.79	7.24	0.01	7.48	10.69	3.21
FH51	−0.80	1.57	2.22	0.05	12.91	6.22	−6.69
G21EA	4.53	5.41	6.53	0.16	20.55	16.50	−4.05
G21IP	5.18	5.72	7.37	0.02	32.47	24.24	−8.23
G2RC	−0.60	2.53	3.42	0.05	16.01	7.15	−8.87
HAL59	0.26	0.38	0.56	0.08	2.58	2.19	−0.39
HEAVY28	0.01	0.12	0.15	0.10	0.63	0.29	−0.34
HEAVYSB11	7.22	7.22	7.26	0.12	3.11	8.48	5.37
ICONF	−0.04	0.19	0.27	0.06	1.21	0.35	−0.86
IDISP	−0.82	3.59	4.60	0.25	14.69	5.63	−9.06
IL16	−0.36	0.40	0.48	0.00	1.32	0.30	−1.03
INV24	−0.08	0.82	1.62	0.03	8.89	1.68	−7.21
ISO34	−0.26	1.04	1.55	0.07	7.36	3.35	−4.02
ISOL24	1.20	2.32	3.21	0.11	12.89	8.97	−3.93
MB16-43	12.77	14.62	17.34	0.04	50.80	35.83	−14.97
MCONF	0.37	0.42	0.48	0.08	1.19	0.87	−0.31
NBPRC	−1.18	2.24	2.68	0.08	8.41	2.32	−6.09
PA26	1.71	1.76	1.88	0.01	3.66	3.10	−0.56
PArel	−0.02	0.73	1.14	0.16	4.86	1.92	−2.94
PCONF21	−0.08	0.34	0.42	0.21	1.62	0.71	−0.90
PNICO23	0.21	0.27	0.46	0.06	1.97	1.81	−0.16
PX13	−4.24	4.24	4.36	0.13	4.17	−2.21	−6.39
RC21	−0.83	2.68	3.31	0.08	13.37	8.26	−5.11
RG18	−0.12	0.18	0.25	0.31	0.99	0.47	−0.52
RSE43	1.54	1.58	3.47	0.21	16.10	15.67	−0.43
S22	0.00	0.26	0.38	0.04	1.77	0.77	−1.01
S66	−0.14	0.23	0.30	0.04	1.06	0.37	−0.69
SCONF	0.16	0.35	0.39	0.08	1.27	0.42	−0.85
TAUT15	−0.29	0.65	0.81	0.21	2.48	0.87	−1.61
UPU23	0.29	0.39	0.49	0.07	1.43	1.11	−0.32
W4-11	28.58	28.63	32.91	0.09	71.86	69.08	−2.78
WATER27	0.69	1.14	1.54	0.01	8.58	3.69	−4.89
WCPT18	−2.78	2.78	2.90	0.08	2.89	−1.82	−4.71
YBDE18	9.54	9.54	10.11	0.19	11.40	16.96	5.56
BH76RC	−0.23	1.85	2.52	0.09	12.20	8.91	−3.29

Table S93: Statistical analysis of the SOS0-PBE0-DH(b)-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	8.47	8.47	9.53	0.25	16.70	19.11	2.41
ACONF	−0.14	0.14	0.15	0.08	0.25	0.01	−0.24
ADIM6	−0.29	0.29	0.33	0.09	0.48	−0.08	−0.56
AHB21	−1.19	1.19	1.48	0.05	3.42	−0.02	−3.44
AL2X6	2.57	2.57	2.82	0.07	3.73	3.93	0.20
ALK8	3.53	3.65	5.14	0.06	10.74	10.29	−0.45
ALKBDE10	−2.25	3.44	4.08	0.03	12.38	5.10	−7.27
AMINO20x4	0.05	0.22	0.27	0.09	1.29	0.77	−0.53
BH76	−1.96	2.37	2.62	0.13	10.63	3.48	−7.15
BHDIV10	−2.75	3.08	3.55	0.07	6.87	1.35	−5.52
BHPERI	−2.13	2.65	2.94	0.13	8.95	3.84	−5.12
BHROT27	0.44	0.45	0.59	0.07	1.17	1.09	−0.08
BSR36	−2.57	2.57	2.82	0.16	4.31	−1.33	−5.65
BUT14DIOL	0.09	0.11	0.14	0.04	0.47	0.40	−0.07
C60ISO	−0.35	2.38	2.91	0.02	8.49	2.52	−5.97
CARBHB12	1.09	1.09	1.30	0.18	2.12	2.45	0.33
CDIE20	0.76	0.80	1.00	0.20	2.39	2.11	−0.28
CHB6	−1.89	1.89	2.15	0.07	2.88	−0.77	−3.65
DARC	−6.78	6.78	6.97	0.21	5.59	−4.75	−10.34
DC13	−3.13	8.93	10.91	0.16	37.16	17.25	−19.91
DIPCS10	1.59	2.52	2.85	0.00	7.54	4.31	−3.23
FH51	−1.89	2.70	3.56	0.09	15.19	5.48	−9.70
G21EA	0.11	1.45	1.68	0.04	5.65	2.70	−2.95
G21IP	1.34	2.45	2.96	0.01	12.59	7.85	−4.74
G2RC	−3.11	5.79	7.39	0.11	29.80	10.08	−19.72
HAL59	0.48	0.56	0.87	0.12	3.26	2.91	−0.35
HEAVY28	0.23	0.25	0.33	0.20	0.83	0.67	−0.16
HEAVYSB11	2.13	2.13	2.25	0.04	2.38	3.65	1.27
ICONF	0.15	0.21	0.32	0.06	1.10	0.86	−0.24
IDISP	−0.32	1.39	2.20	0.10	6.10	0.97	−5.13
IL16	−0.37	0.38	0.47	0.00	1.04	0.12	−0.92
INV24	0.28	1.20	1.67	0.04	7.30	4.38	−2.92
ISO34	−0.13	1.21	1.75	0.08	7.47	3.90	−3.57
ISOL24	1.32	2.31	3.31	0.11	12.27	7.78	−4.49
MB16-43	13.26	16.39	20.58	0.04	82.03	64.28	−17.75
MCONF	0.17	0.25	0.31	0.05	1.12	0.70	−0.42
NBPRC	−1.53	3.39	3.88	0.12	12.04	5.04	−7.00
PA26	2.87	2.95	3.29	0.02	8.01	7.04	−0.97
PArel	0.66	1.01	1.38	0.22	6.10	3.88	−2.22
PCONF21	−0.11	0.58	0.70	0.36	2.16	0.80	−1.36
PNICO23	0.81	0.81	1.17	0.19	3.86	3.89	0.03
PX13	−4.50	4.50	4.68	0.13	4.79	−2.38	−7.17
RC21	3.94	4.45	5.35	0.12	15.41	13.05	−2.36
RG18	0.00	0.09	0.14	0.16	0.66	0.44	−0.22
RSE43	−0.47	0.50	0.64	0.07	2.33	0.60	−1.73
S22	0.22	0.35	0.48	0.05	2.09	1.53	−0.56
S66	0.13	0.30	0.41	0.05	2.05	1.60	−0.45
SCONF	0.07	0.25	0.33	0.05	1.32	0.35	−0.98
TAUT15	0.14	0.87	1.09	0.29	3.47	2.13	−1.33
UPU23	0.47	0.53	0.66	0.09	1.63	1.41	−0.22
W4-11	4.11	5.76	7.45	0.02	37.56	22.67	−14.89
WATER27	3.52	3.91	4.79	0.05	17.92	12.72	−5.20
WCPT18	−2.79	2.81	3.42	0.08	6.75	0.18	−6.57
YBDE18	2.83	2.83	3.01	0.06	3.83	4.08	0.25
BH76RC	−0.40	1.95	2.61	0.09	11.67	6.56	−5.11

Table S94: Statistical analysis of the SOS0-PBE-QIDH(b)-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	5.46	5.46	6.08	0.16	10.07	11.64	1.57
ACONF	−0.27	0.27	0.28	0.15	0.35	−0.02	−0.37
ADIM6	−0.51	0.51	0.56	0.15	0.69	−0.18	−0.87
AHB21	−0.86	0.86	1.08	0.04	2.87	0.08	−2.79
AL2X6	1.70	1.81	1.97	0.05	3.22	2.89	−0.33
ALK8	3.23	3.23	5.65	0.05	13.81	13.83	0.02
ALKBDE10	4.21	4.21	5.54	0.04	11.05	11.63	0.57
AMINO20x4	0.00	0.15	0.18	0.06	0.88	0.52	−0.36
BH76	−0.43	1.80	2.58	0.10	15.53	11.37	−4.16
BHDIV10	−2.18	2.48	2.91	0.05	6.33	1.48	−4.86
BHPERI	−2.49	2.88	3.10	0.14	7.58	2.73	−4.86
BHROT27	0.20	0.20	0.27	0.03	0.67	0.61	−0.07
BSR36	−1.71	1.71	1.81	0.11	2.13	−0.99	−3.12
BUT14DIOL	0.04	0.06	0.08	0.02	0.28	0.21	−0.08
C60ISO	−7.24	7.24	9.15	0.07	16.84	−0.59	−17.43
CARBHB12	0.75	0.75	0.92	0.12	1.62	1.79	0.17
CDIE20	0.44	0.48	0.59	0.12	1.33	1.18	−0.14
CHB6	−1.52	1.52	1.72	0.06	2.11	−0.58	−2.69
DARC	−6.70	6.70	6.77	0.21	2.65	−5.18	−7.83
DC13	−3.13	8.60	11.49	0.16	42.09	20.01	−22.08
DIPCS10	4.81	4.81	5.09	0.01	5.68	7.40	1.72
FH51	−1.44	2.06	2.80	0.07	15.14	6.20	−8.95
G21EA	2.43	2.74	3.32	0.08	10.93	8.92	−2.01
G21IP	3.55	3.78	4.55	0.01	16.71	13.70	−3.01
G2RC	−1.85	3.70	4.89	0.07	18.55	6.46	−12.09
HAL59	0.33	0.42	0.66	0.09	2.69	2.34	−0.35
HEAVY28	0.08	0.16	0.20	0.13	0.73	0.45	−0.28
HEAVYSB11	4.93	4.93	4.97	0.08	2.05	6.01	3.96
ICONF	0.05	0.17	0.23	0.05	0.98	0.41	−0.57
IDISP	−0.78	2.58	3.66	0.18	11.22	3.11	−8.11
IL16	−0.41	0.42	0.48	0.00	1.11	0.10	−1.01
INV24	0.22	0.81	1.11	0.03	5.38	3.60	−1.78
ISO34	−0.18	1.05	1.60	0.07	7.43	3.81	−3.61
ISOL24	1.41	2.44	3.49	0.11	13.74	9.43	−4.31
MB16-43	13.15	14.54	17.17	0.04	44.45	35.03	−9.42
MCONF	0.30	0.34	0.40	0.07	1.11	0.81	−0.29
NBPRC	−1.41	2.82	3.31	0.10	10.18	3.24	−6.94
PA26	2.33	2.33	2.52	0.01	4.59	4.94	0.35
PArel	0.32	0.78	1.03	0.17	4.60	2.01	−2.59
PCONF21	−0.07	0.38	0.46	0.23	1.57	0.50	−1.07
PNICO23	0.45	0.47	0.74	0.11	2.70	2.62	−0.08
PX13	−4.08	4.08	4.22	0.12	4.30	−2.02	−6.33
RC21	1.63	2.54	3.40	0.07	12.58	9.07	−3.51
RG18	−0.08	0.15	0.20	0.26	0.88	0.45	−0.42
RSE43	0.59	0.74	1.59	0.10	7.65	6.87	−0.77
S22	0.10	0.21	0.27	0.03	1.18	0.74	−0.43
S66	−0.03	0.22	0.28	0.04	1.35	0.79	−0.55
SCONF	0.13	0.31	0.36	0.07	1.26	0.38	−0.88
TAUT15	−0.13	0.71	0.78	0.23	2.07	0.89	−1.18
UPU23	0.38	0.45	0.57	0.08	1.45	1.22	−0.22
W4-11	17.43	17.58	20.75	0.06	55.99	48.85	−7.14
WATER27	1.93	2.28	2.71	0.03	12.24	7.44	−4.80
WCPT18	−2.58	2.58	2.87	0.07	4.10	−1.02	−5.11
YBDE18	6.68	6.68	6.85	0.14	5.46	9.52	4.06
BH76RC	−0.38	1.55	2.20	0.07	11.30	7.21	−4.09

Table S95: Statistical analysis of the SOS0-PBE-CIDH(b)-D3(CSO) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.53	7.53	8.47	0.22	14.77	16.97	2.20
ACONF	−0.17	0.17	0.18	0.09	0.27	0.00	−0.27
ADIM6	−0.35	0.35	0.39	0.10	0.54	−0.11	−0.64
AHB21	−1.14	1.14	1.42	0.05	3.32	−0.01	−3.33
AL2X6	2.45	2.45	2.68	0.07	3.58	3.78	0.20
ALK8	3.51	3.51	5.19	0.06	11.08	11.07	−0.02
ALKBDE10	−1.16	2.82	3.33	0.03	10.67	5.94	−4.73
AMINO20x4	0.05	0.20	0.25	0.08	1.17	0.72	−0.45
BH76	−1.55	2.10	2.35	0.11	9.97	3.79	−6.18
BHDIV10	−2.53	2.84	3.29	0.06	6.21	0.99	−5.22
BHPERI	−2.08	2.61	2.88	0.13	8.72	3.78	−4.94
BHROT27	0.39	0.40	0.52	0.06	1.07	0.99	−0.08
BSR36	−2.39	2.39	2.61	0.15	3.84	−1.26	−5.10
BUT14DIOL	0.08	0.09	0.12	0.03	0.40	0.34	−0.06
C60ISO	−1.56	2.65	3.65	0.03	9.96	1.98	−7.98
CARBHB12	1.01	1.01	1.21	0.17	1.99	2.27	0.29
CDIE20	0.68	0.72	0.90	0.18	2.18	1.91	−0.26
CHB6	−1.84	1.84	2.08	0.07	2.73	−0.73	−3.46
DARC	−6.99	6.99	7.14	0.22	4.88	−5.10	−9.98
DC13	−3.20	8.92	10.95	0.16	35.58	16.58	−19.00
DIPCS10	2.35	2.73	3.15	0.00	6.77	4.85	−1.92
FH51	−1.87	2.60	3.44	0.08	15.67	5.74	−9.93
G21EA	0.50	1.20	1.46	0.04	5.63	3.69	−1.94
G21IP	1.80	2.36	2.92	0.01	11.46	7.55	−3.91
G2RC	−2.94	5.39	6.89	0.11	27.24	8.96	−18.27
HAL59	0.44	0.52	0.82	0.11	3.09	2.74	−0.36
HEAVY28	0.19	0.22	0.29	0.18	0.82	0.62	−0.19
HEAVYSB11	2.66	2.66	2.74	0.05	2.08	3.99	1.91
ICONF	0.13	0.20	0.30	0.06	0.99	0.75	−0.23
IDISP	−0.48	1.54	2.55	0.11	7.13	1.09	−6.05
IL16	−0.38	0.39	0.47	0.00	1.02	0.07	−0.94
INV24	0.33	1.13	1.56	0.04	7.15	4.54	−2.62
ISO34	−0.13	1.18	1.72	0.08	7.55	3.97	−3.58
ISOL24	1.41	2.36	3.44	0.11	11.86	8.13	−3.73
MB16-43	13.26	15.80	19.61	0.04	71.13	58.38	−12.75
MCONF	0.20	0.26	0.33	0.05	1.12	0.73	−0.38
NBPRC	−1.53	3.30	3.81	0.12	11.92	4.77	−7.15
PA26	2.80	2.86	3.17	0.02	7.36	6.65	−0.71
PArel	0.61	0.94	1.28	0.20	5.74	3.46	−2.28
PCONF21	−0.09	0.53	0.63	0.33	1.97	0.68	−1.29
PNICO23	0.73	0.73	1.07	0.17	3.57	3.58	0.01
PX13	−4.28	4.28	4.45	0.13	4.61	−2.18	−6.80
RC21	3.53	4.06	4.93	0.11	14.58	12.18	−2.40
RG18	−0.02	0.11	0.15	0.19	0.72	0.45	−0.27
RSE43	−0.25	0.41	0.55	0.05	2.71	1.34	−1.37
S22	0.20	0.31	0.43	0.04	1.85	1.40	−0.45
S66	0.09	0.28	0.38	0.05	1.92	1.46	−0.46
SCONF	0.08	0.26	0.33	0.06	1.28	0.35	−0.94
TAUT15	0.08	0.83	0.99	0.27	3.07	1.81	−1.26
UPU23	0.45	0.52	0.65	0.09	1.56	1.36	−0.20
W4-11	6.52	7.52	9.57	0.02	37.68	26.97	−10.71
WATER27	3.17	3.54	4.30	0.04	16.48	11.47	−5.01
WCPT18	−2.63	2.65	3.20	0.08	6.20	0.12	−6.08
YBDE18	3.63	3.63	3.73	0.07	3.43	4.88	1.45
BH76RC	−0.43	1.84	2.48	0.09	11.34	6.37	−4.97

SI.8.5 Results for dispersion-uncorrected functionals

Table S96: Statistical analysis of the ωB97X-2 DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	4.79	4.79	5.59	0.14	10.47	11.56	1.08
ACONF	0.00	0.02	0.02	0.01	0.07	0.05	−0.02
ADIM6	0.28	0.28	0.28	0.08	0.17	0.36	0.18
AHB21	−0.38	0.42	0.47	0.02	1.56	0.41	−1.16
AL2X6	−0.97	0.97	1.12	0.03	1.68	−0.47	−2.14
ALK8	1.69	2.43	4.14	0.04	12.93	10.69	−2.25
ALKBDE10	−0.16	2.52	3.10	0.03	11.10	4.53	−6.57
AMINO20x4	−0.05	0.10	0.14	0.04	0.56	0.23	−0.34
BH76	−0.04	1.24	2.06	0.07	13.54	8.15	−5.38
BHDIV10	−0.55	0.76	1.07	0.02	3.75	1.05	−2.70
BHPERI	−2.25	2.28	2.39	0.11	3.67	0.38	−3.29
BHROT27	0.07	0.15	0.20	0.02	0.72	0.42	−0.31
BSR36	−1.18	1.18	1.38	0.07	2.98	−0.43	−3.41
BUT14DIOL	0.00	0.03	0.03	0.01	0.17	0.08	−0.09
C60ISO	−1.43	3.44	4.21	0.04	11.70	3.18	−8.52
CARBHB12	0.60	0.60	0.66	0.10	0.95	1.21	0.26
CDIE20	0.23	0.27	0.41	0.07	1.12	0.97	−0.15
CHB6	−0.83	0.83	0.90	0.03	0.98	−0.52	−1.50
DARC	−0.60	0.88	0.98	0.03	2.44	0.55	−1.89
DC13	−0.31	2.71	4.03	0.05	17.80	7.91	−9.89
DIPCS10	−5.11	5.11	5.66	0.01	8.09	−0.94	−9.03
FH51	−0.23	0.75	1.06	0.02	6.20	1.58	−4.62
G21EA	−1.04	1.60	1.91	0.05	8.00	4.47	−3.53
G21IP	−1.12	2.09	2.65	0.01	13.67	5.52	−8.15
G2RC	−0.46	1.23	1.53	0.02	6.38	2.61	−3.76
HAL59	0.39	0.41	0.51	0.09	1.63	1.37	−0.26
HEAVY28	0.31	0.31	0.33	0.25	0.51	0.63	0.13
HEAVYSB11	−1.41	1.41	1.63	0.02	2.90	−0.02	−2.92
ICONF	0.02	0.13	0.17	0.04	0.67	0.37	−0.30
IDISP	−0.43	0.65	1.16	0.05	3.19	0.40	−2.79
IL16	0.55	0.55	0.57	0.01	0.52	0.81	0.29
INV24	0.06	0.95	1.75	0.03	9.66	7.26	−2.40
ISO34	−0.06	0.56	0.85	0.04	4.26	2.63	−1.63
ISOL24	−0.01	1.04	1.48	0.05	7.43	3.16	−4.28
MB16-43	−16.04	16.87	19.18	0.04	51.58	13.28	−38.29
MCONF	0.25	0.27	0.30	0.05	0.58	0.47	−0.11
NBPRC	0.09	0.97	1.18	0.04	4.07	2.90	−1.16
PA26	0.26	0.98	1.37	0.01	5.49	3.81	−1.68
PArel	0.04	0.36	0.58	0.08	2.77	0.77	−2.00
PCONF21	0.04	0.19	0.23	0.12	0.84	0.47	−0.37
PNICO23	0.31	0.31	0.32	0.07	0.35	0.45	0.10
PX13	−2.05	2.05	2.18	0.06	2.75	−0.59	−3.34
RC21	−1.08	1.57	1.94	0.04	5.93	1.76	−4.17
RG18	0.18	0.18	0.21	0.31	0.41	0.46	0.05
RSE43	0.49	0.60	1.26	0.08	5.80	5.44	−0.36
S22	0.27	0.34	0.38	0.05	1.03	0.63	−0.39
S66	0.33	0.33	0.35	0.06	0.54	0.57	0.03
SCONF	−0.01	0.05	0.07	0.01	0.27	0.06	−0.21
TAUT15	0.16	0.45	0.58	0.15	1.92	1.08	−0.83
UPU23	0.26	0.40	0.51	0.07	1.80	1.13	−0.68
W4-11	−3.50	3.64	4.48	0.01	20.60	2.32	−18.28
WATER27	3.63	3.76	5.16	0.05	14.58	12.80	−1.77
WCPT18	−1.24	1.67	2.01	0.05	7.17	3.75	−3.42
YBDE18	−0.08	1.40	1.71	0.03	6.86	4.47	−2.39
BH76RC	−0.14	0.71	0.97	0.03	4.68	2.23	−2.45

Table S97: Statistical analysis of the B2PPW91 DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	17.67	17.67	19.84	0.52	32.08	36.04	3.97
ACONF	0.75	0.75	0.84	0.41	1.22	1.45	0.22
ADIM6	−5.65	5.65	6.10	1.68	6.66	−2.46	−9.12
AHB21	1.03	1.15	1.40	0.05	3.92	3.02	−0.90
AL2X6	−5.79	5.79	6.26	0.16	6.96	−1.70	−8.66
ALK8	−1.90	3.12	4.32	0.05	13.57	3.16	−10.41
ALKBDE10	3.88	5.64	8.80	0.06	26.49	23.63	−2.86
AMINO20x4	−0.09	0.53	0.67	0.22	3.18	1.77	−1.40
BH76	−5.90	5.95	6.77	0.32	23.02	1.80	−21.23
BHDIV10	−5.59	6.01	6.79	0.13	11.98	2.07	−9.92
BHPERI	−1.98	2.18	3.00	0.10	7.73	1.04	−6.70
BHROT27	0.23	0.32	0.47	0.05	1.57	1.30	−0.28
BSR36	−7.25	7.25	8.02	0.45	16.65	−0.96	−17.60
BUT14DIOL	−0.26	0.39	0.45	0.14	1.58	0.72	−0.86
C60ISO	−12.20	12.20	14.75	0.12	24.34	−1.52	−25.86
CARBHB12	−0.36	0.82	0.92	0.14	2.93	1.38	−1.55
CDIE20	1.61	1.61	1.78	0.40	2.98	2.91	−0.07
CHB6	1.60	1.60	1.85	0.06	2.76	3.51	0.76
DARC	8.24	8.24	8.88	0.25	9.59	11.50	1.91
DC13	3.75	10.16	11.99	0.18	40.29	17.83	−22.46
DIPCS10	−1.54	3.03	3.71	0.00	10.90	3.53	−7.37
FH51	2.32	3.22	4.24	0.10	17.12	11.34	−5.78
G21EA	1.82	2.44	2.89	0.07	8.66	5.61	−3.06
G21IP	0.21	3.23	3.95	0.01	16.38	8.89	−7.48
G2RC	2.00	4.23	5.62	0.08	25.70	16.47	−9.23
HAL59	−1.58	1.81	2.23	0.39	10.60	3.06	−7.54
HEAVY28	−1.36	1.36	1.42	1.10	1.60	−0.63	−2.23
HEAVYSB11	−2.91	3.84	4.82	0.07	10.94	2.60	−8.34
ICONF	0.14	0.44	0.61	0.13	2.52	1.72	−0.80
IDISP	2.74	11.16	12.75	0.78	36.71	22.23	−14.48
IL16	3.20	3.20	3.31	0.03	3.36	5.13	1.77
INV24	−2.44	2.59	3.15	0.08	8.91	1.83	−7.08
ISO34	−0.70	1.55	2.20	0.11	12.15	4.82	−7.32
ISOL24	−3.15	6.35	10.04	0.29	55.39	22.02	−33.37
MB16-43	−21.53	26.31	32.36	0.06	123.70	40.18	−83.52
MCONF	−2.04	2.11	2.38	0.42	4.79	0.94	−3.85
NBPRC	2.36	2.98	4.07	0.11	11.46	9.11	−2.36
PA26	2.68	2.68	3.33	0.01	7.88	8.18	0.31
PArel	0.24	1.50	2.12	0.32	10.86	5.24	−5.62
PCONF21	−0.72	3.87	4.25	2.39	11.74	6.55	−5.18
PNICO23	−1.44	1.62	1.73	0.38	5.47	2.13	−3.34
PX13	−8.53	8.53	8.67	0.26	6.58	−5.81	−12.39
RC21	1.41	2.53	3.09	0.07	10.22	7.53	−2.69
RG18	−1.30	1.30	1.60	2.24	3.42	−0.35	−3.76
RSE43	−2.17	2.17	2.41	0.29	4.96	−0.62	−5.58
S22	−3.97	3.97	4.83	0.54	11.21	−0.92	−12.14
S66	−3.63	3.63	4.06	0.66	8.25	−0.86	−9.12
SCONF	0.06	0.32	0.37	0.07	1.32	0.66	−0.65
TAUT15	0.19	1.50	1.82	0.49	5.61	3.58	−2.03
UPU23	1.84	2.42	3.54	0.42	11.74	9.75	−1.99
W4-11	6.77	8.43	10.95	0.03	48.49	34.17	−14.32
WATER27	−10.91	11.03	16.99	0.14	46.67	1.23	−45.44
WCPT18	−5.98	5.98	6.23	0.17	6.84	−3.47	−10.31
YBDE18	−3.97	5.10	6.31	0.10	18.26	5.76	−12.50
BH76RC	0.57	2.65	4.27	0.12	22.18	17.40	−4.78

Table S98: Statistical analysis of the B2NC-PLYP DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	2.63	2.63	2.99	0.08	4.93	5.51	0.58
ACONF	−0.07	0.13	0.16	0.07	0.52	0.18	−0.34
ADIM6	−1.20	1.20	1.33	0.36	1.63	−0.42	−2.05
AHB21	−0.43	0.48	0.70	0.02	2.08	0.35	−1.73
AL2X6	−2.46	2.46	2.56	0.07	1.96	−1.69	−3.65
ALK8	−0.68	1.58	1.99	0.03	6.33	2.81	−3.52
ALKBDE10	−0.93	2.83	3.47	0.03	11.05	4.69	−6.36
AMINO20x4	−0.02	0.15	0.20	0.06	1.21	0.56	−0.65
BH76	1.26	1.84	2.90	0.10	18.92	15.50	−3.42
BHDIV10	0.33	1.08	1.47	0.02	5.22	3.28	−1.94
BHPERI	−1.69	1.72	1.90	0.08	3.59	0.46	−3.13
BHROT27	0.30	0.31	0.41	0.05	1.02	0.96	−0.06
BSR36	−2.43	2.43	2.64	0.15	3.61	−1.25	−4.86
BUT14DIOL	−0.02	0.09	0.11	0.03	0.45	0.23	−0.22
C60ISO	−7.75	7.80	10.36	0.08	20.31	0.13	−20.17
CARBHB12	0.28	0.33	0.47	0.05	1.42	1.18	−0.24
CDIE20	0.57	0.57	0.63	0.14	1.02	1.00	−0.02
CHB6	−1.07	1.07	1.18	0.04	1.45	−0.28	−1.73
DARC	0.84	1.11	1.44	0.03	3.64	2.56	−1.08
DC13	1.85	3.45	4.37	0.06	16.28	9.81	−6.48
DIPCS10	−3.14	3.28	4.17	0.01	8.93	0.70	−8.23
FH51	−0.75	1.39	1.99	0.04	10.32	2.99	−7.33
G21EA	−1.42	2.13	2.55	0.06	11.23	7.19	−4.04
G21IP	−0.67	1.82	2.60	0.01	16.52	7.80	−8.72
G2RC	−1.55	1.88	2.48	0.04	8.49	2.26	−6.23
HAL59	−0.15	0.31	0.39	0.07	1.79	1.02	−0.77
HEAVY28	−0.27	0.27	0.30	0.22	0.40	−0.08	−0.48
HEAVYSB11	−1.16	1.16	1.47	0.02	2.59	−0.08	−2.67
ICONF	0.08	0.21	0.28	0.06	0.99	0.49	−0.50
IDISP	0.41	1.06	1.29	0.07	3.70	2.40	−1.31
IL16	0.81	0.81	0.85	0.01	1.06	1.48	0.42
INV24	0.26	0.87	1.29	0.03	6.36	4.29	−2.07
ISO34	0.40	0.67	1.20	0.05	5.71	4.72	−1.00
ISOL24	0.12	1.88	2.71	0.09	12.63	7.12	−5.51
MB16-43	−8.12	12.00	14.48	0.03	55.77	18.69	−37.08
MCONF	−0.06	0.11	0.13	0.02	0.54	0.23	−0.31
NBPRC	0.66	0.93	1.15	0.03	3.41	2.67	−0.73
PA26	0.27	0.94	1.48	0.00	7.52	3.80	−3.72
PArel	0.29	0.46	0.76	0.10	3.54	2.69	−0.85
PCONF21	0.11	0.49	0.61	0.30	1.79	1.33	−0.46
PNICO23	−0.29	0.29	0.34	0.07	0.74	−0.03	−0.76
PX13	−1.04	1.07	1.21	0.03	2.00	0.19	−1.82
RC21	−2.33	2.93	3.39	0.08	7.98	2.60	−5.39
RG18	−0.17	0.22	0.28	0.38	0.93	0.32	−0.62
RSE43	1.13	1.13	2.20	0.15	9.29	9.23	−0.06
S22	−0.39	0.44	0.56	0.06	1.76	0.37	−1.38
S66	−0.45	0.53	0.66	0.10	1.88	0.45	−1.43
SCONF	0.02	0.09	0.15	0.02	0.69	0.53	−0.16
TAUT15	0.31	0.45	0.63	0.15	2.02	1.15	−0.88
UPU23	0.54	0.67	0.84	0.12	2.16	1.83	−0.33
W4-11	−3.23	3.53	5.04	0.01	27.19	3.87	−23.32
WATER27	0.40	0.81	1.07	0.01	5.19	2.81	−2.37
WCPT18	−0.39	0.82	1.04	0.02	3.87	2.22	−1.65
YBDE18	0.91	1.50	1.97	0.03	7.28	5.37	−1.91
BH76RC	−0.43	1.15	1.62	0.05	7.82	3.56	−4.26

Table S99: Statistical analysis of the mPW2NC-PLYP DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.49	7.49	8.36	0.22	12.52	13.69	1.17
ACONF	−0.02	0.11	0.13	0.06	0.43	0.16	−0.27
ADIM6	−1.08	1.08	1.22	0.32	1.66	−0.31	−1.97
AHB21	−0.29	0.38	0.47	0.02	1.74	0.32	−1.42
AL2X6	−3.16	3.16	3.26	0.09	2.54	−1.85	−4.40
ALK8	−1.04	1.67	2.14	0.03	6.08	2.48	−3.60
ALKBDE10	4.38	5.24	6.78	0.05	15.12	12.37	−2.76
AMINO20x4	−0.04	0.16	0.21	0.07	1.07	0.56	−0.51
BH76	−1.54	2.22	2.76	0.12	15.32	8.16	−7.17
BHDIV10	−1.60	2.55	3.03	0.06	8.80	3.31	−5.49
BHPERI	−3.38	3.38	3.57	0.16	5.93	−1.26	−7.19
BHROT27	0.16	0.18	0.29	0.03	1.08	0.94	−0.14
BSR36	−2.52	2.52	2.81	0.16	4.56	−1.13	−5.69
BUT14DIOL	0.07	0.09	0.12	0.03	0.39	0.33	−0.06
C60ISO	−13.37	13.37	16.47	0.14	27.40	−2.42	−29.83
CARBHB12	0.49	0.50	0.65	0.08	1.47	1.41	−0.06
CDIE20	0.75	0.75	0.79	0.18	1.04	1.25	0.21
CHB6	−0.88	0.91	1.02	0.03	1.47	0.07	−1.40
DARC	4.33	4.33	4.53	0.13	3.86	6.23	2.37
DC13	3.29	5.60	6.96	0.10	21.29	9.60	−11.69
DIPCS10	−2.33	3.07	3.40	0.00	7.84	2.68	−5.16
FH51	0.54	1.51	2.06	0.05	12.32	6.75	−5.56
G21EA	0.61	1.48	1.98	0.04	6.53	4.41	−2.12
G21IP	−0.17	1.91	2.45	0.01	13.24	6.07	−7.18
G2RC	0.43	1.75	2.43	0.03	11.35	7.21	−4.14
HAL59	0.11	0.36	0.54	0.08	2.86	2.09	−0.77
HEAVY28	−0.15	0.16	0.20	0.13	0.46	0.06	−0.41
HEAVYSB11	−0.12	0.75	0.97	0.01	3.64	1.86	−1.77
ICONF	0.04	0.21	0.25	0.06	0.96	0.41	−0.54
IDISP	1.37	2.01	3.19	0.14	9.18	7.27	−1.92
IL16	0.68	0.68	0.73	0.01	1.06	1.24	0.18
INV24	−0.70	0.91	1.72	0.03	8.25	0.71	−7.54
ISO34	0.18	0.86	1.35	0.06	6.98	5.58	−1.41
ISOL24	−1.05	3.49	5.10	0.16	27.13	14.21	−12.92
MB16-43	−9.82	10.41	11.90	0.03	35.21	12.53	−22.67
MCONF	−0.16	0.19	0.23	0.04	0.71	0.21	−0.50
NBPRC	0.92	1.29	1.85	0.05	5.45	4.67	−0.78
PA26	−0.44	1.21	1.48	0.01	6.99	3.38	−3.61
PArel	0.02	0.67	1.21	0.14	6.62	2.69	−3.93
PCONF21	−0.08	0.74	0.80	0.46	2.56	1.49	−1.07
PNICO23	−0.14	0.23	0.28	0.05	1.24	0.59	−0.65
PX13	−3.62	3.62	3.70	0.11	3.26	−2.13	−5.39
RC21	−1.76	2.23	2.79	0.06	7.12	1.68	−5.44
RG18	0.02	0.17	0.22	0.29	0.86	0.51	−0.35
RSE43	0.19	0.68	1.29	0.09	7.20	5.57	−1.63
S22	−0.44	0.50	0.65	0.07	2.08	0.31	−1.77
S66	−0.43	0.53	0.64	0.10	1.89	0.39	−1.49
SCONF	0.08	0.13	0.16	0.03	0.64	0.27	−0.37
TAUT15	0.21	0.63	0.80	0.21	2.45	1.71	−0.74
UPU23	0.48	0.66	0.84	0.12	2.36	1.82	−0.54
W4-11	5.40	5.82	7.34	0.02	24.60	19.78	−4.82
WATER27	1.66	2.00	2.67	0.02	12.86	8.37	−4.49
WCPT18	−2.87	2.87	3.03	0.08	3.28	−1.83	−5.11
YBDE18	1.22	1.91	2.95	0.04	10.31	8.06	−2.25
BH76RC	−0.02	1.51	2.22	0.07	12.03	9.34	−2.69

Table S100: Statistical analysis of the mSD-PBEPBE DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	9.45	9.45	10.97	0.28	20.05	21.04	0.99
ACONF	−0.26	0.26	0.28	0.14	0.38	0.00	−0.37
ADIM6	−0.70	0.70	0.79	0.21	1.04	−0.24	−1.28
AHB21	1.49	1.52	2.22	0.07	7.50	7.21	−0.29
AL2X6	−4.30	4.30	4.62	0.12	5.01	−2.90	−7.91
ALK8	−1.64	5.77	7.55	0.09	26.72	13.53	−13.19
ALKBDE10	17.82	17.82	21.98	0.18	37.55	45.17	7.62
AMINO20x4	−0.25	0.36	0.46	0.15	1.70	0.56	−1.13
BH76	−1.72	4.00	5.62	0.21	41.44	27.57	−13.86
BHDIV10	−4.31	4.66	5.82	0.10	12.40	1.75	−10.65
BHPERI	−6.99	6.99	7.35	0.33	11.21	−2.88	−14.09
BHROT27	−0.53	0.54	0.83	0.09	2.12	0.07	−2.05
BSR36	0.03	0.67	0.78	0.04	3.49	2.36	−1.13
BUT14DIOL	−0.20	0.23	0.25	0.08	0.65	0.27	−0.39
C60ISO	−31.97	31.97	36.83	0.33	52.08	−8.23	−60.31
CARBHB12	−0.06	0.23	0.29	0.04	1.06	0.64	−0.41
CDIE20	0.48	0.60	0.73	0.15	2.64	2.00	−0.63
CHB6	0.55	0.76	1.07	0.03	2.88	2.29	−0.59
DARC	7.98	7.98	8.08	0.25	4.66	10.81	6.16
DC13	5.24	11.62	15.56	0.21	48.27	33.66	−14.62
DIPCS10	−3.66	5.94	7.75	0.01	21.98	3.38	−18.60
FH51	3.58	4.10	5.33	0.13	18.85	15.94	−2.91
G21EA	4.32	6.27	8.11	0.19	24.13	17.08	−7.05
G21IP	0.46	4.97	7.42	0.02	44.36	25.23	−19.13
G2RC	6.32	7.76	9.43	0.15	31.23	21.48	−9.76
HAL59	0.18	0.38	0.60	0.08	3.41	2.48	−0.94
HEAVY28	−0.01	0.13	0.16	0.10	0.69	0.42	−0.28
HEAVYSB11	5.75	5.75	6.16	0.10	7.25	8.46	1.21
ICONF	−0.26	0.53	0.69	0.16	2.64	1.34	−1.30
IDISP	1.88	3.18	4.66	0.22	12.42	10.39	−2.04
IL16	1.13	1.13	1.27	0.01	1.77	2.06	0.28
INV24	−2.15	3.06	5.95	0.10	32.52	7.85	−24.67
ISO34	−0.52	1.35	1.97	0.09	10.06	3.13	−6.93
ISOL24	−2.50	4.86	6.58	0.22	31.28	15.39	−15.89
MB16-43	−9.15	25.49	30.46	0.06	117.77	54.32	−63.45
MCONF	0.16	0.35	0.41	0.07	1.49	0.82	−0.67
NBPRC	1.54	3.07	3.72	0.11	14.14	7.13	−7.01
PA26	−1.37	2.18	2.48	0.01	9.34	4.57	−4.77
PArel	−1.00	1.32	2.95	0.29	12.79	0.89	−11.89
PCONF21	−0.26	0.51	0.56	0.31	1.73	0.70	−1.03
PNICO23	−0.40	0.47	0.63	0.11	2.40	0.63	−1.77
PX13	−6.44	6.44	6.53	0.19	3.59	−5.15	−8.73
RC21	−6.97	7.32	9.00	0.21	22.52	2.82	−19.70
RG18	−0.08	0.15	0.22	0.26	0.93	0.51	−0.43
RSE43	1.75	2.05	4.54	0.27	21.05	19.78	−1.27
S22	−0.62	0.96	1.40	0.13	4.92	1.56	−3.36
S66	−0.59	0.69	0.95	0.13	3.86	0.86	−3.00
SCONF	0.02	0.17	0.22	0.04	0.99	0.38	−0.61
TAUT15	−0.57	1.10	1.26	0.36	3.35	1.29	−2.05
UPU23	−0.02	0.43	0.52	0.08	2.03	0.98	−1.05
W4-11	28.10	28.10	33.94	0.09	87.95	88.52	0.58
WATER27	−5.45	5.45	7.48	0.07	19.06	0.04	−19.02
WCPT18	−6.16	6.16	6.53	0.18	8.76	−2.61	−11.37
YBDE18	6.48	7.17	9.96	0.15	30.88	26.80	−4.08
BH76RC	1.44	4.57	6.57	0.21	29.92	21.38	−8.54

Table S101: Statistical analysis of the PBE0-DH DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.40	7.40	8.45	0.22	15.21	17.55	2.34
ACONF	0.33	0.34	0.41	0.19	0.82	0.77	−0.05
ADIM6	−2.71	2.71	2.99	0.81	3.65	−0.97	−4.62
AHB21	−0.94	1.06	1.49	0.05	4.44	0.87	−3.57
AL2X6	−1.09	1.35	1.47	0.04	2.92	0.79	−2.13
ALK8	2.11	2.26	3.12	0.04	6.35	5.76	−0.58
ALKBDE10	−6.31	6.38	7.57	0.06	15.63	0.37	−15.26
AMINO20x4	0.05	0.34	0.45	0.14	2.38	1.35	−1.03
BH76	−1.15	1.72	1.99	0.09	9.30	3.79	−5.51
BHDIV10	−1.85	2.39	2.73	0.05	7.19	2.46	−4.72
BHPERI	−0.26	1.29	1.60	0.06	7.16	3.97	−3.19
BHROT27	0.56	0.58	0.77	0.09	1.62	1.53	−0.09
BSR36	−5.98	5.98	6.70	0.37	12.35	−2.15	−14.51
BUT14DIOL	−0.11	0.18	0.22	0.06	0.74	0.35	−0.40
C60ISO	1.76	2.91	3.13	0.03	8.08	5.31	−2.77
CARBHB12	0.57	0.63	0.89	0.10	2.44	2.14	−0.30
CDIE20	1.00	1.07	1.29	0.26	2.96	2.58	−0.38
CHB6	−1.02	1.21	1.50	0.05	3.51	0.55	−2.96
DARC	−3.95	3.95	4.68	0.12	8.19	−1.21	−9.40
DC13	−1.57	8.42	10.36	0.15	38.01	20.75	−17.26
DIPCS10	−0.65	2.61	3.36	0.00	11.22	4.08	−7.14
FH51	−1.88	2.67	3.32	0.09	13.22	5.12	−8.10
G21EA	−1.47	3.01	3.46	0.09	9.44	3.21	−6.23
G21IP	0.05	3.08	3.75	0.01	15.64	7.76	−7.88
G2RC	−4.04	6.07	7.69	0.12	30.41	10.64	−19.77
HAL59	−0.62	0.86	1.11	0.19	5.81	1.80	−4.01
HEAVY28	−0.52	0.52	0.58	0.42	0.98	−0.02	−1.00
HEAVYSB11	−2.63	2.63	2.90	0.05	3.41	−1.06	−4.47
ICONF	0.26	0.34	0.49	0.10	1.39	1.09	−0.30
IDISP	0.03	5.49	5.95	0.39	15.45	7.23	−8.22
IL16	1.17	1.17	1.30	0.01	2.29	2.59	0.30
INV24	−0.07	1.45	2.16	0.05	10.46	6.20	−4.26
ISO34	−0.07	1.57	2.13	0.11	8.21	3.68	−4.53
ISOL24	0.51	2.44	3.42	0.11	14.93	8.46	−6.47
MB16-43	−3.81	13.33	16.71	0.03	75.51	41.35	−34.16
MCONF	−0.98	1.05	1.21	0.21	2.70	0.62	−2.08
NBPRC	−0.16	2.53	2.90	0.09	9.47	5.59	−3.89
PA26	2.42	2.53	3.04	0.01	8.41	7.10	−1.31
PArel	0.88	1.22	1.60	0.26	6.09	4.24	−1.84
PCONF21	−0.21	2.15	2.44	1.33	6.65	3.91	−2.74
PNICO23	−0.22	0.50	0.70	0.12	3.94	2.12	−1.82
PX13	−3.63	3.63	3.81	0.11	4.37	−1.58	−5.95
RC21	3.30	3.86	4.89	0.11	16.32	14.13	−2.19
RG18	−0.32	0.32	0.41	0.55	0.88	0.02	−0.85
RSE43	−0.56	0.58	0.72	0.08	2.22	0.32	−1.90
S22	−1.59	1.68	2.40	0.23	7.39	0.84	−6.55
S66	−1.44	1.49	1.90	0.27	5.46	0.81	−4.65
SCONF	−0.02	0.21	0.29	0.05	1.31	0.90	−0.41
TAUT15	0.45	0.86	1.28	0.28	3.98	2.78	−1.19
UPU23	1.28	1.52	2.15	0.27	5.84	5.19	−0.65
W4-11	−5.59	5.81	7.55	0.02	30.49	2.97	−27.52
WATER27	−0.13	1.83	2.60	0.02	10.04	2.61	−7.43
WCPT18	−1.88	2.10	2.62	0.06	7.12	1.92	−5.20
YBDE18	−0.74	1.69	2.14	0.03	6.83	1.81	−5.03
BH76RC	−0.52	2.12	2.69	0.10	10.49	5.37	−5.11

Table S102: Statistical analysis of the LS1-DH DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	2.43	2.43	2.77	0.07	4.78	5.43	0.66
ACONF	−0.03	0.12	0.16	0.07	0.57	0.27	−0.30
ADIM6	−1.50	1.50	1.65	0.45	1.98	−0.54	−2.53
AHB21	−0.99	1.02	1.42	0.05	3.75	0.33	−3.42
AL2X6	−0.08	0.35	0.47	0.01	1.51	0.64	−0.87
ALK8	2.39	2.40	3.91	0.04	9.75	9.72	−0.04
ALKBDE10	−3.42	4.04	4.71	0.04	10.21	2.82	−7.39
AMINO20x4	0.04	0.19	0.25	0.08	1.39	0.76	−0.63
BH76	0.92	1.55	2.46	0.08	15.55	12.96	−2.59
BHDIV10	−1.13	1.81	2.03	0.04	5.24	2.06	−3.17
BHPERI	−2.41	2.76	2.99	0.13	7.84	2.97	−4.86
BHROT27	0.47	0.48	0.61	0.08	1.28	1.24	−0.05
BSR36	−2.35	2.35	2.51	0.15	3.37	−1.14	−4.51
BUT14DIOL	−0.03	0.07	0.10	0.03	0.39	0.18	−0.21
C60ISO	−4.28	5.07	6.82	0.05	16.14	1.93	−14.21
CARBHB12	0.60	0.61	0.84	0.10	1.97	1.90	−0.06
CDIE20	0.74	0.76	0.90	0.19	1.89	1.76	−0.13
CHB6	−1.18	1.18	1.38	0.04	1.94	−0.36	−2.30
DARC	−6.06	6.06	6.27	0.19	4.65	−3.77	−8.43
DC13	−1.03	7.76	9.94	0.14	35.08	18.59	−16.50
DIPCS10	−1.16	2.84	3.30	0.00	9.09	2.82	−6.27
FH51	−2.29	2.65	3.25	0.09	12.08	4.74	−7.34
G21EA	−1.42	2.43	2.89	0.07	10.32	5.05	−5.27
G21IP	0.13	2.28	2.87	0.01	14.11	5.56	−8.55
G2RC	−3.71	4.83	6.06	0.09	22.24	5.98	−16.25
HAL59	−0.14	0.44	0.57	0.10	2.99	1.73	−1.26
HEAVY28	−0.25	0.26	0.30	0.21	0.56	0.10	−0.46
HEAVYSB11	−0.22	0.59	0.68	0.01	1.80	0.62	−1.18
ICONF	0.21	0.23	0.34	0.07	1.00	0.82	−0.19
IDISP	−0.85	1.38	1.85	0.10	5.01	1.15	−3.86
IL16	0.33	0.40	0.50	0.00	1.39	1.11	−0.28
INV24	0.45	1.17	1.71	0.04	9.15	5.88	−3.26
ISO34	0.28	1.34	1.90	0.09	7.64	4.06	−3.58
ISOL24	1.65	2.60	3.42	0.12	11.19	8.15	−3.04
MB16-43	8.67	11.86	14.58	0.03	56.61	35.29	−21.32
MCONF	−0.11	0.20	0.25	0.04	0.98	0.46	−0.52
NBPRC	−0.78	2.14	2.57	0.08	8.78	4.16	−4.62
PA26	1.30	1.68	2.03	0.01	7.39	4.86	−2.53
PArel	0.75	0.98	1.27	0.21	4.74	3.03	−1.71
PCONF21	0.00	0.87	1.00	0.54	2.94	1.74	−1.20
PNICO23	0.05	0.34	0.50	0.08	2.63	1.82	−0.81
PX13	−2.60	2.60	2.77	0.08	3.55	−0.82	−4.38
RC21	1.14	2.48	3.70	0.07	15.42	11.50	−3.92
RG18	−0.23	0.24	0.31	0.41	0.78	0.14	−0.64
RSE43	0.68	0.75	1.43	0.10	6.07	5.76	−0.31
S22	−0.48	0.59	0.83	0.08	3.11	0.93	−2.17
S66	−0.57	0.68	0.86	0.12	2.81	0.95	−1.85
SCONF	0.13	0.14	0.16	0.03	0.42	0.35	−0.07
TAUT15	0.51	0.74	0.99	0.24	2.79	2.10	−0.70
UPU23	0.68	0.82	1.07	0.14	2.84	2.48	−0.36
W4-11	−3.64	4.09	5.72	0.01	27.44	5.37	−22.06
WATER27	0.64	1.36	1.68	0.02	7.13	3.03	−4.09
WCPT18	−1.60	1.84	2.23	0.05	6.19	2.16	−4.02
YBDE18	2.23	2.85	3.58	0.06	8.79	6.48	−2.32
BH76RC	−0.50	1.84	2.40	0.09	10.82	5.78	−5.04

Table S103: Statistical analysis of the PBE0-2 DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	1.77	1.77	2.03	0.05	3.23	3.59	0.35
ACONF	−0.11	0.15	0.18	0.08	0.50	0.15	−0.35
ADIM6	−1.22	1.22	1.34	0.36	1.60	−0.44	−2.04
AHB21	−0.94	0.96	1.33	0.04	3.61	0.24	−3.37
AL2X6	−0.02	0.34	0.45	0.01	1.43	0.49	−0.94
ALK8	2.32	2.32	4.15	0.04	10.79	10.77	−0.02
ALKBDE10	−2.22	3.63	3.96	0.04	10.60	3.81	−6.79
AMINO20x4	0.03	0.17	0.22	0.07	1.16	0.65	−0.52
BH76	1.28	1.89	2.94	0.10	18.04	15.02	−3.01
BHDIV10	−1.14	1.85	2.07	0.04	5.70	2.00	−3.70
BHPERI	−3.15	3.37	3.64	0.16	8.27	2.51	−5.76
BHROT27	0.43	0.44	0.56	0.07	1.20	1.16	−0.05
BSR36	−1.52	1.52	1.58	0.09	1.79	−0.79	−2.58
BUT14DIOL	−0.01	0.06	0.08	0.02	0.33	0.16	−0.17
C60ISO	−6.31	6.73	8.93	0.07	19.00	1.08	−17.92
CARBHB12	0.61	0.61	0.83	0.10	1.86	1.84	−0.02
CDIE20	0.69	0.71	0.83	0.17	1.64	1.54	−0.10
CHB6	−1.17	1.17	1.32	0.04	1.62	−0.39	−2.00
DARC	−5.99	5.99	6.17	0.18	4.36	−3.74	−8.11
DC13	−0.66	7.54	9.84	0.14	35.50	19.74	−15.75
DIPCS10	−1.51	2.90	3.42	0.00	9.55	2.52	−7.03
FH51	−2.21	2.54	3.17	0.08	13.30	4.99	−8.31
G21EA	−1.27	2.23	2.77	0.07	11.57	6.58	−4.99
G21IP	0.10	2.11	2.83	0.01	16.27	7.43	−8.84
G2RC	−3.41	4.38	5.50	0.09	19.40	4.59	−14.82
HAL59	−0.02	0.36	0.51	0.08	2.46	1.82	−0.63
HEAVY28	−0.18	0.21	0.24	0.17	0.55	0.14	−0.41
HEAVYSB11	0.41	0.68	0.81	0.01	2.21	1.43	−0.77
ICONF	0.18	0.22	0.32	0.07	0.89	0.72	−0.16
IDISP	−0.92	1.26	2.25	0.09	6.04	0.64	−5.40
IL16	0.18	0.31	0.38	0.00	1.19	0.80	−0.39
INV24	0.44	1.05	1.54	0.03	8.11	5.32	−2.78
ISO34	0.34	1.26	1.83	0.09	7.62	4.22	−3.40
ISOL24	1.76	2.67	3.50	0.12	10.93	7.92	−3.01
MB16-43	11.19	12.73	15.36	0.03	48.97	32.09	−16.88
MCONF	0.09	0.17	0.21	0.03	0.79	0.50	−0.29
NBPRC	−0.84	1.96	2.40	0.07	8.15	3.53	−4.62
PA26	0.92	1.46	1.79	0.01	7.55	4.16	−3.39
PArel	0.67	0.88	1.18	0.19	4.80	3.10	−1.70
PCONF21	0.03	0.59	0.71	0.36	2.32	1.28	−1.04
PNICO23	0.10	0.32	0.47	0.07	2.34	1.76	−0.58
PX13	−2.59	2.59	2.75	0.08	3.46	−0.87	−4.33
RC21	0.24	2.59	3.53	0.07	14.27	9.67	−4.59
RG18	−0.20	0.23	0.30	0.40	0.90	0.28	−0.62
RSE43	1.02	1.04	2.01	0.14	8.46	8.27	−0.19
S22	−0.24	0.36	0.50	0.05	2.08	0.85	−1.23
S66	−0.39	0.51	0.64	0.09	2.31	0.89	−1.42
SCONF	0.16	0.18	0.20	0.04	0.40	0.30	−0.10
TAUT15	0.53	0.72	0.95	0.24	2.55	1.96	−0.59
UPU23	0.53	0.66	0.84	0.12	2.29	1.95	−0.33
W4-11	−2.55	3.50	5.03	0.01	28.03	6.01	−22.02
WATER27	0.74	1.27	1.59	0.02	7.39	3.40	−3.99
WCPT18	−1.74	1.93	2.26	0.06	5.82	1.78	−4.05
YBDE18	2.93	3.24	4.11	0.07	8.72	7.13	−1.60
BH76RC	−0.43	1.94	2.43	0.09	10.95	6.52	−4.43

Table S104: Statistical analysis of the revPBE0-DH DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.01	7.01	8.34	0.21	17.41	18.15	0.74
ACONF	0.62	0.62	0.73	0.34	1.24	1.31	0.07
ADIM6	−4.12	4.12	4.52	1.23	5.38	−1.55	−6.93
AHB21	0.29	0.87	1.07	0.04	4.16	2.19	−1.97
AL2X6	−3.63	3.63	3.88	0.10	3.95	−1.19	−5.14
ALK8	0.13	1.81	2.30	0.03	8.33	4.14	−4.19
ALKBDE10	−8.96	8.96	9.83	0.09	15.52	−2.77	−18.29
AMINO20x4	−0.07	0.50	0.62	0.20	2.97	1.68	−1.28
BH76	−0.09	1.08	1.44	0.06	8.90	5.32	−3.58
BHDIV10	−0.82	1.26	1.61	0.03	5.22	2.24	−2.98
BHPERI	1.79	2.18	2.41	0.10	6.77	4.08	−2.68
BHROT27	0.46	0.48	0.64	0.08	1.40	1.24	−0.16
BSR36	−7.59	7.59	8.41	0.47	16.21	−2.00	−18.21
BUT14DIOL	−0.53	0.58	0.64	0.21	1.35	0.37	−0.99
C60ISO	1.67	2.84	3.06	0.03	8.01	5.24	−2.77
CARBHB12	−0.27	0.53	0.66	0.09	2.28	1.06	−1.21
CDIE20	1.06	1.14	1.37	0.28	3.28	2.84	−0.44
CHB6	−0.19	0.96	1.20	0.04	3.87	1.75	−2.12
DARC	−0.28	2.27	2.58	0.07	8.17	2.34	−5.83
DC13	0.27	7.87	9.47	0.14	31.89	19.95	−11.94
DIPCS10	−1.30	3.06	3.94	0.00	13.09	4.16	−8.94
FH51	−0.78	1.81	2.35	0.06	12.32	7.54	−4.78
G21EA	−2.10	3.36	3.99	0.10	10.55	2.97	−7.59
G21IP	−0.44	3.34	3.94	0.01	15.24	7.54	−7.70
G2RC	−2.64	4.57	5.74	0.09	24.75	10.76	−13.99
HAL59	−1.57	1.58	1.84	0.34	5.98	0.22	−5.76
HEAVY28	−1.02	1.02	1.06	0.82	1.05	−0.56	−1.61
HEAVYSB11	−5.22	5.22	5.38	0.09	3.91	−3.65	−7.56
ICONF	0.21	0.42	0.56	0.13	2.17	1.40	−0.77
IDISP	0.64	8.94	9.60	0.63	23.60	11.64	−11.96
IL16	2.97	2.97	3.06	0.03	3.00	4.87	1.87
INV24	−0.17	1.47	2.23	0.05	10.19	6.38	−3.80
ISO34	−0.23	1.52	2.11	0.10	10.12	3.46	−6.66
ISOL24	−0.66	3.16	4.72	0.14	24.01	7.28	−16.73
MB16-43	−21.26	22.14	25.39	0.05	67.99	16.20	−51.79
MCONF	−1.63	1.69	1.92	0.34	3.53	0.56	−2.97
NBPRC	1.55	1.93	2.52	0.07	6.23	4.60	−1.63
PA26	3.52	3.52	3.99	0.02	8.26	8.61	0.36
PArel	0.79	0.97	1.38	0.21	5.04	3.93	−1.12
PCONF21	−0.30	2.91	3.35	1.80	9.86	5.57	−4.29
PNICO23	−1.17	1.20	1.27	0.28	3.00	0.32	−2.69
PX13	−1.43	1.44	1.61	0.04	2.63	0.08	−2.55
RC21	1.35	2.11	2.90	0.06	13.63	9.68	−3.96
RG18	−0.57	0.57	0.69	0.98	1.20	−0.09	−1.29
RSE43	−0.38	0.43	0.56	0.06	2.39	0.53	−1.86
S22	−2.92	2.92	3.73	0.40	9.17	−0.51	−9.67
S66	−2.61	2.61	3.00	0.48	6.58	−0.37	−6.95
SCONF	−0.34	0.79	0.94	0.17	3.36	2.03	−1.33
TAUT15	0.23	0.66	0.82	0.22	2.84	1.63	−1.21
UPU23	1.64	2.01	2.88	0.35	8.58	7.52	−1.06
W4-11	−12.03	12.05	13.69	0.04	33.65	0.96	−32.69
WATER27	−9.06	9.11	14.50	0.11	39.55	0.48	−39.07
WCPT18	−0.21	0.79	1.07	0.02	4.69	2.33	−2.36
YBDE18	−3.78	3.78	4.51	0.08	8.23	−0.57	−8.79
BH76RC	−0.48	1.87	2.45	0.09	9.42	3.56	−5.85

Table S105: Statistical analysis of the TPSS0-DH DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.50	7.50	9.60	0.22	20.23	20.91	0.68
ACONF	0.42	0.42	0.51	0.23	0.96	0.95	−0.01
ADIM6	−3.41	3.41	3.75	1.01	4.51	−1.26	−5.77
AHB21	−0.45	0.89	1.19	0.04	4.64	1.43	−3.21
AL2X6	−1.69	1.85	2.13	0.05	4.00	0.48	−3.52
ALK8	1.08	1.36	1.72	0.02	3.86	2.98	−0.88
ALKBDE10	−7.72	7.72	8.81	0.08	15.69	−3.11	−18.80
AMINO20x4	0.02	0.39	0.50	0.16	2.55	1.47	−1.08
BH76	−1.64	2.44	3.23	0.13	14.12	4.94	−9.18
BHDIV10	−0.58	1.03	1.39	0.02	5.40	2.23	−3.17
BHPERI	1.51	1.67	1.96	0.08	5.64	3.84	−1.80
BHROT27	0.53	0.58	0.77	0.09	1.73	1.54	−0.19
BSR36	−7.75	7.75	8.62	0.48	16.40	−2.43	−18.83
BUT14DIOL	−0.26	0.31	0.35	0.11	0.88	0.31	−0.57
C60ISO	2.79	3.31	3.75	0.03	8.40	7.10	−1.30
CARBHB12	0.20	0.45	0.64	0.07	2.35	1.67	−0.68
CDIE20	1.03	1.07	1.29	0.26	2.79	2.55	−0.24
CHB6	−0.73	1.13	1.37	0.04	3.71	1.15	−2.57
DARC	0.12	2.16	2.38	0.07	7.52	2.51	−5.02
DC13	−0.66	7.62	9.22	0.14	30.13	17.70	−12.43
DIPCS10	0.56	3.94	4.79	0.01	16.74	8.38	−8.37
FH51	−0.87	1.95	2.50	0.06	11.53	6.13	−5.40
G21EA	−1.75	3.39	4.11	0.10	12.28	4.63	−7.65
G21IP	0.49	3.25	4.12	0.01	16.22	9.99	−6.23
G2RC	−3.11	4.68	5.90	0.09	23.12	9.46	−13.66
HAL59	−1.02	1.11	1.40	0.24	6.07	1.20	−4.86
HEAVY28	−0.74	0.74	0.79	0.60	0.98	−0.29	−1.27
HEAVYSB11	−4.15	4.15	4.34	0.07	4.80	−2.22	−7.02
ICONF	0.18	0.33	0.45	0.10	1.58	0.93	−0.64
IDISP	0.44	7.80	8.51	0.55	22.40	11.03	−11.37
IL16	2.05	2.05	2.13	0.02	2.32	3.60	1.28
INV24	0.01	1.63	2.59	0.05	11.61	7.61	−3.99
ISO34	−0.46	1.55	1.96	0.11	9.21	3.67	−5.54
ISOL24	−0.81	3.25	4.64	0.15	23.02	7.20	−15.83
MB16-43	−31.17	32.26	37.49	0.08	91.68	11.90	−79.78
MCONF	−1.29	1.36	1.54	0.27	3.09	0.57	−2.51
NBPRC	0.65	2.35	2.94	0.08	8.89	5.67	−3.23
PA26	3.86	3.86	4.33	0.02	8.27	9.34	1.07
PArel	0.76	1.00	1.34	0.22	4.97	3.78	−1.19
PCONF21	−0.26	2.49	2.84	1.54	7.87	4.44	−3.43
PNICO23	−0.63	0.70	0.81	0.16	2.84	0.57	−2.26
PX13	−1.46	1.51	1.70	0.05	2.96	0.34	−2.62
RC21	1.77	2.32	3.29	0.06	13.98	10.37	−3.61
RG18	−0.41	0.41	0.52	0.71	1.04	−0.03	−1.07
RSE43	−0.33	0.41	0.51	0.05	2.04	0.42	−1.62
S22	−2.15	2.18	3.01	0.30	8.45	0.39	−8.06
S66	−1.96	1.96	2.42	0.36	6.12	0.31	−5.81
SCONF	−0.10	0.28	0.36	0.06	1.48	0.85	−0.63
TAUT15	0.32	0.74	0.94	0.24	3.08	1.84	−1.24
UPU23	1.48	1.75	2.52	0.31	7.24	6.44	−0.80
W4-11	−8.97	11.56	14.42	0.04	59.15	20.87	−38.28
WATER27	−3.27	3.81	6.37	0.05	20.63	1.51	−19.12
WCPT18	0.14	1.11	1.40	0.03	5.75	4.17	−1.58
YBDE18	−4.14	4.14	5.02	0.08	9.56	−0.09	−9.66
BH76RC	−1.27	3.58	4.38	0.17	17.41	8.22	−9.19

TPSS0-DH had convergence problems for the h2+_1.75 system in SIE4x4.

Table S106: Statistical analysis of the PBE-QIDH DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	3.40	3.40	3.90	0.10	7.09	8.03	0.94
ACONF	0.06	0.15	0.18	0.08	0.64	0.40	−0.24
ADIM6	−1.82	1.82	2.01	0.54	2.41	−0.66	−3.07
AHB21	−1.01	1.06	1.48	0.05	4.07	0.45	−3.61
AL2X6	−0.22	0.50	0.59	0.01	1.67	0.76	−0.91
ALK8	2.41	2.45	3.67	0.04	8.60	8.47	−0.12
ALKBDE10	−4.52	4.91	5.65	0.05	11.91	1.95	−9.96
AMINO20x4	0.05	0.21	0.30	0.09	1.63	0.88	−0.75
BH76	0.46	1.23	2.00	0.07	12.89	10.45	−2.44
BHDIV10	−1.21	1.85	2.08	0.04	5.29	2.15	−3.14
BHPERI	−1.68	2.19	2.40	0.10	7.28	3.34	−3.94
BHROT27	0.51	0.51	0.67	0.08	1.37	1.32	−0.05
BSR36	−3.31	3.31	3.60	0.20	5.69	−1.44	−7.12
BUT14DIOL	−0.05	0.10	0.12	0.04	0.47	0.22	−0.25
C60ISO	−2.26	3.48	4.90	0.04	13.22	2.74	−10.48
CARBHB12	0.59	0.61	0.85	0.10	2.09	1.97	−0.12
CDIE20	0.79	0.84	1.00	0.21	2.16	2.00	−0.16
CHB6	−1.18	1.18	1.43	0.04	2.32	−0.24	−2.57
DARC	−5.89	5.89	6.15	0.18	5.48	−3.51	−8.99
DC13	−1.37	7.91	10.10	0.14	33.85	16.99	−16.86
DIPCS10	−0.86	2.74	3.26	0.00	9.29	3.18	−6.11
FH51	−2.30	2.71	3.31	0.09	12.25	4.31	−7.94
G21EA	−1.53	2.64	3.07	0.08	9.11	3.62	−5.49
G21IP	0.15	2.47	3.04	0.01	14.42	6.06	−8.35
G2RC	−3.95	5.24	6.61	0.10	25.03	7.43	−17.60
HAL59	−0.27	0.55	0.69	0.12	3.75	1.75	−1.99
HEAVY28	−0.32	0.33	0.37	0.27	0.66	0.07	−0.59
HEAVYSB11	−0.90	0.90	1.11	0.02	1.93	−0.11	−2.04
ICONF	0.23	0.26	0.38	0.08	1.13	0.91	−0.21
IDISP	−0.71	2.03	2.27	0.14	5.84	2.70	−3.14
IL16	0.53	0.55	0.67	0.01	1.63	1.48	−0.15
INV24	0.39	1.27	1.90	0.04	9.90	6.20	−3.69
ISO34	0.20	1.40	1.98	0.10	7.78	4.03	−3.74
ISOL24	1.45	2.43	3.30	0.11	11.50	8.17	−3.33
MB16-43	5.62	11.23	14.21	0.03	63.11	38.08	−25.02
MCONF	−0.33	0.39	0.46	0.08	1.39	0.52	−0.87
NBPRC	−0.68	2.29	2.71	0.08	9.15	4.74	−4.41
PA26	1.68	1.94	2.33	0.01	7.22	5.57	−1.65
PArel	0.81	1.07	1.37	0.23	4.71	2.97	−1.73
PCONF21	−0.05	1.19	1.36	0.73	3.69	2.30	−1.39
PNICO23	−0.02	0.37	0.54	0.09	2.97	1.89	−1.08
PX13	−2.72	2.72	2.89	0.08	3.70	−0.88	−4.58
RC21	1.95	2.77	4.06	0.08	16.10	12.92	−3.18
RG18	−0.26	0.26	0.34	0.45	0.69	0.02	−0.67
RSE43	0.33	0.49	0.88	0.06	4.46	3.77	−0.69
S22	−0.76	0.87	1.24	0.12	4.28	0.98	−3.30
S66	−0.80	0.89	1.13	0.16	3.54	0.97	−2.57
SCONF	0.09	0.10	0.15	0.02	0.56	0.49	−0.07
TAUT15	0.49	0.75	1.05	0.25	3.07	2.26	−0.81
UPU23	0.84	1.01	1.35	0.18	3.50	3.07	−0.43
W4-11	−4.56	4.81	6.50	0.02	27.20	4.76	−22.44
WATER27	0.48	1.47	1.84	0.02	6.84	2.54	−4.30
WCPT18	−1.55	1.83	2.26	0.05	6.52	2.39	−4.14
YBDE18	1.44	2.46	3.03	0.05	8.63	5.53	−3.10
BH76RC	−0.55	1.90	2.44	0.09	10.62	5.24	−5.38

Table S107: Statistical analysis of the TPSS-QIDH DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	4.26	4.26	5.96	0.13	13.06	13.04	−0.02
ACONF	0.15	0.21	0.25	0.11	0.74	0.54	−0.20
ADIM6	−2.26	2.26	2.49	0.67	3.03	−0.81	−3.85
AHB21	−0.70	0.83	1.23	0.04	4.14	0.82	−3.32
AL2X6	−1.35	1.39	1.63	0.04	2.85	0.13	−2.72
ALK8	1.37	1.57	2.35	0.03	6.60	5.92	−0.68
ALKBDE10	−4.50	4.50	5.48	0.04	11.32	−0.26	−11.58
AMINO20x4	0.01	0.25	0.33	0.10	1.80	1.01	−0.79
BH76	−0.26	2.04	2.75	0.11	17.74	12.35	−5.39
BHDIV10	−0.01	0.79	1.16	0.02	4.69	2.53	−2.16
BHPERI	0.15	0.82	1.06	0.04	4.71	2.80	−1.91
BHROT27	0.47	0.48	0.65	0.08	1.38	1.31	−0.08
BSR36	−5.18	5.18	5.70	0.32	10.05	−1.95	−12.00
BUT14DIOL	−0.15	0.19	0.21	0.07	0.63	0.26	−0.37
C60ISO	−1.33	3.16	4.23	0.03	11.97	3.16	−8.81
CARBHB12	0.39	0.47	0.67	0.08	2.03	1.70	−0.33
CDIE20	0.81	0.84	0.99	0.21	2.04	1.91	−0.13
CHB6	−0.93	1.03	1.21	0.04	2.36	0.30	−2.06
DARC	−1.30	1.70	2.14	0.05	5.38	0.78	−4.59
DC13	−0.22	5.22	6.67	0.09	24.25	12.07	−12.18
DIPCS10	2.24	3.82	4.69	0.01	13.27	8.52	−4.75
FH51	−1.50	1.92	2.35	0.06	9.36	3.82	−5.55
G21EA	−0.50	2.47	2.89	0.07	10.04	5.08	−4.97
G21IP	1.58	2.81	3.48	0.01	13.48	8.71	−4.77
G2RC	−3.73	4.49	5.47	0.09	17.03	5.52	−11.51
HAL59	−0.56	0.68	0.83	0.15	3.81	1.13	−2.68
HEAVY28	−0.45	0.45	0.49	0.36	0.67	−0.09	−0.76
HEAVYSB11	−2.42	2.42	2.60	0.04	3.11	−0.83	−3.94
ICONF	0.15	0.27	0.37	0.08	1.29	0.77	−0.52
IDISP	−0.13	4.10	4.59	0.29	12.13	4.85	−7.28
IL16	1.20	1.20	1.29	0.01	1.75	2.31	0.56
INV24	0.33	1.27	2.07	0.04	10.71	7.18	−3.53
ISO34	0.08	1.09	1.51	0.07	7.07	3.82	−3.25
ISOL24	0.16	2.09	2.87	0.10	12.81	5.59	−7.23
MB16-43	−19.09	21.48	27.51	0.05	96.38	19.43	−76.95
MCONF	−0.61	0.66	0.75	0.13	1.66	0.43	−1.23
NBPRC	0.61	2.17	2.98	0.08	9.60	6.87	−2.73
PA26	2.67	2.69	3.13	0.01	7.39	7.14	−0.25
PArel	0.64	0.78	1.08	0.17	3.19	2.50	−0.69
PCONF21	−0.08	1.52	1.73	0.94	4.80	2.88	−1.92
PNICO23	−0.35	0.42	0.49	0.10	1.88	0.47	−1.40
PX13	−0.98	1.10	1.25	0.03	2.45	0.57	−1.88
RC21	0.61	1.97	2.75	0.06	12.95	8.91	−4.03
RG18	−0.28	0.28	0.36	0.48	0.72	0.01	−0.71
RSE43	0.38	0.50	0.92	0.07	4.51	3.81	−0.70
S22	−1.17	1.23	1.72	0.17	5.30	0.66	−4.64
S66	−1.14	1.17	1.48	0.21	4.13	0.63	−3.50
SCONF	−0.04	0.17	0.23	0.04	0.99	0.66	−0.33
TAUT15	0.34	0.58	0.81	0.19	2.73	1.66	−1.06
UPU23	1.04	1.20	1.66	0.21	4.32	3.80	−0.51
W4-11	−4.98	9.01	11.31	0.03	56.61	23.22	−33.39
WATER27	−1.37	2.09	3.45	0.03	12.52	1.58	−10.94
WCPT18	0.10	1.08	1.34	0.03	5.45	3.93	−1.51
YBDE18	−1.41	2.67	3.13	0.05	8.84	2.82	−6.02
BH76RC	−1.48	2.86	3.66	0.13	16.02	5.33	−10.69

TPSS-QIDH had convergence problems for the h2+_1.75 system in SIE4x4.

Table S108: Statistical analysis of the PBE-CIDH DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	6.31	6.31	7.21	0.19	13.01	15.02	2.01
ACONF	0.26	0.28	0.35	0.15	0.78	0.68	−0.10
ADIM6	−2.50	2.50	2.75	0.74	3.36	−0.89	−4.25
AHB21	−0.97	1.07	1.50	0.05	4.41	0.76	−3.65
AL2X6	−0.83	1.10	1.20	0.03	2.56	0.82	−1.74
ALK8	2.23	2.34	3.24	0.04	6.11	5.66	−0.45
ALKBDE10	−6.01	6.13	7.19	0.06	14.79	0.59	−14.20
AMINO20x4	0.05	0.31	0.41	0.13	2.19	1.22	−0.97
BH76	−0.74	1.44	1.77	0.08	9.37	4.80	−4.57
BHDIV10	−1.66	2.21	2.52	0.05	6.70	2.37	−4.32
BHPERI	−0.54	1.40	1.69	0.07	6.97	3.82	−3.14
BHROT27	0.55	0.57	0.75	0.09	1.56	1.50	−0.06
BSR36	−5.34	5.34	5.95	0.33	10.73	−1.99	−12.72
BUT14DIOL	−0.09	0.16	0.19	0.06	0.66	0.30	−0.36
C60ISO	0.93	2.71	3.05	0.03	8.62	4.24	−4.38
CARBHB12	0.57	0.62	0.88	0.10	2.35	2.10	−0.26
CDIE20	0.95	1.01	1.21	0.25	2.79	2.45	−0.33
CHB6	−1.07	1.19	1.49	0.04	3.26	0.35	−2.91
DARC	−4.58	4.58	5.13	0.14	7.52	−1.93	−9.45
DC13	−1.58	8.15	10.26	0.15	36.71	19.88	−16.83
DIPCS10	−0.60	2.65	3.29	0.00	10.91	4.04	−6.86
FH51	−2.02	2.70	3.32	0.09	12.82	4.48	−8.34
G21EA	−1.51	2.94	3.38	0.09	9.00	2.99	−6.01
G21IP	0.10	2.90	3.56	0.01	15.29	7.38	−7.90
G2RC	−4.07	5.91	7.48	0.12	29.38	9.97	−19.40
HAL59	−0.54	0.79	1.00	0.17	5.32	1.79	−3.54
HEAVY28	−0.47	0.47	0.53	0.38	0.90	0.00	−0.90
HEAVYSB11	−2.23	2.23	2.45	0.04	2.94	−0.85	−3.79
ICONF	0.25	0.33	0.46	0.10	1.34	1.05	−0.28
IDISP	−0.18	4.48	4.92	0.32	13.16	6.12	−7.05
IL16	1.00	1.00	1.13	0.01	2.13	2.31	0.18
INV24	0.07	1.42	2.12	0.04	9.97	5.79	−4.18
ISO34	0.00	1.53	2.09	0.11	7.76	3.83	−3.93
ISOL24	0.78	2.31	3.23	0.11	14.16	8.48	−5.67
MB16-43	−1.44	12.40	15.57	0.03	70.83	41.07	−29.76
MCONF	−0.81	0.89	1.02	0.18	2.38	0.60	−1.78
NBPRC	−0.31	2.47	2.86	0.09	9.27	5.46	−3.80
PA26	2.28	2.40	2.89	0.01	8.05	6.79	−1.26
PArel	0.88	1.20	1.55	0.26	5.71	3.89	−1.82
PCONF21	−0.17	1.92	2.17	1.19	5.89	3.52	−2.37
PNICO23	−0.17	0.46	0.66	0.11	3.71	2.07	−1.64
PX13	−3.36	3.36	3.54	0.10	4.19	−1.36	−5.54
RC21	3.06	3.66	4.73	0.10	16.29	14.08	−2.21
RG18	−0.31	0.31	0.39	0.53	0.83	0.02	−0.81
RSE43	−0.37	0.41	0.55	0.05	2.48	0.88	−1.60
S22	−1.39	1.48	2.12	0.20	6.67	0.90	−5.77
S66	−1.29	1.35	1.72	0.25	5.02	0.87	−4.14
SCONF	0.01	0.17	0.25	0.04	1.11	0.80	−0.31
TAUT15	0.46	0.83	1.22	0.27	3.74	2.65	−1.09
UPU23	1.18	1.39	1.96	0.24	5.23	4.63	−0.59
W4-11	−5.47	5.69	7.41	0.02	29.31	3.36	−25.95
WATER27	0.03	1.75	2.38	0.02	9.17	2.59	−6.58
WCPT18	−1.76	2.00	2.51	0.06	7.00	2.13	−4.87
YBDE18	−0.21	1.78	2.18	0.04	7.39	2.81	−4.58
BH76RC	−0.54	2.08	2.62	0.10	10.20	5.19	−5.01

Table S109: Statistical analysis of the TPSS-CIDH DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	6.62	6.62	8.64	0.20	18.45	18.90	0.45
ACONF	0.36	0.37	0.45	0.20	0.91	0.85	−0.06
ADIM6	−3.13	3.13	3.45	0.93	4.15	−1.15	−5.30
AHB21	−0.53	0.88	1.21	0.04	4.58	1.28	−3.30
AL2X6	−1.58	1.72	1.99	0.05	3.72	0.41	−3.30
ALK8	1.19	1.41	1.85	0.02	4.47	3.70	−0.77
ALKBDE10	−7.04	7.04	8.07	0.07	14.70	−2.56	−17.26
AMINO20x4	0.02	0.35	0.46	0.14	2.37	1.36	−1.02
BH76	−1.32	2.30	3.06	0.12	14.93	6.71	−8.22
BHDIV10	−0.40	0.86	1.29	0.02	5.23	2.31	−2.92
BHPERI	1.27	1.44	1.74	0.07	5.32	3.53	−1.78
BHROT27	0.52	0.55	0.75	0.09	1.65	1.50	−0.15
BSR36	−7.15	7.15	7.93	0.44	14.88	−2.33	−17.22
BUT14DIOL	−0.23	0.28	0.31	0.10	0.82	0.30	−0.52
C60ISO	1.94	3.10	3.37	0.03	8.68	6.01	−2.67
CARBHB12	0.25	0.44	0.64	0.07	2.27	1.68	−0.59
CDIE20	0.97	1.01	1.21	0.25	2.60	2.39	−0.22
CHB6	−0.79	1.10	1.33	0.04	3.42	0.94	−2.48
DARC	−0.33	1.93	2.22	0.06	7.00	1.96	−5.04
DC13	−0.60	6.99	8.53	0.13	28.77	16.39	−12.37
DIPCS10	1.13	3.88	4.74	0.01	16.03	8.65	−7.39
FH51	−1.10	1.89	2.44	0.06	11.13	5.58	−5.55
G21EA	−1.43	3.16	3.79	0.09	11.37	4.64	−6.73
G21IP	0.81	3.08	3.92	0.01	15.15	9.74	−5.41
G2RC	−3.40	4.67	5.87	0.09	21.80	8.58	−13.21
HAL59	−0.92	1.00	1.26	0.22	5.52	1.18	−4.34
HEAVY28	−0.67	0.67	0.72	0.54	0.90	−0.25	−1.15
HEAVYSB11	−3.75	3.75	3.93	0.06	4.41	−1.86	−6.27
ICONF	0.18	0.32	0.43	0.10	1.45	0.84	−0.61
IDISP	0.23	6.95	7.62	0.49	20.08	9.33	−10.75
IL16	1.83	1.83	1.92	0.02	2.19	3.29	1.10
INV24	0.12	1.54	2.49	0.05	11.60	7.65	−3.94
ISO34	−0.32	1.40	1.82	0.10	8.68	3.68	−5.00
ISOL24	−0.55	2.95	4.13	0.13	20.10	6.56	−13.53
MB16-43	−29.08	30.38	35.94	0.07	94.60	12.41	−82.19
MCONF	−1.13	1.19	1.35	0.24	2.75	0.54	−2.21
NBPRC	0.61	2.35	2.98	0.08	9.26	6.03	−3.23
PA26	3.59	3.59	4.06	0.02	7.92	8.85	0.93
PArel	0.74	0.96	1.29	0.21	4.57	3.50	−1.07
PCONF21	−0.21	2.26	2.57	1.40	7.12	4.07	−3.05
PNICO23	−0.57	0.63	0.73	0.15	2.60	0.54	−2.06
PX13	−1.28	1.35	1.53	0.04	2.82	0.45	−2.37
RC21	1.59	2.17	3.17	0.06	13.93	10.23	−3.70
RG18	−0.38	0.38	0.48	0.66	0.96	−0.02	−0.97
RSE43	−0.18	0.32	0.41	0.04	2.38	0.97	−1.41
S22	−1.91	1.95	2.70	0.27	7.72	0.48	−7.24
S66	−1.76	1.77	2.19	0.32	5.66	0.41	−5.25
SCONF	−0.09	0.26	0.34	0.06	1.38	0.82	−0.56
TAUT15	0.32	0.70	0.90	0.23	3.00	1.79	−1.21
UPU23	1.38	1.62	2.31	0.28	6.54	5.81	−0.73
W4-11	−8.06	11.06	13.77	0.04	59.47	21.94	−37.52
WATER27	−2.79	3.37	5.63	0.04	18.61	1.52	−17.09
WCPT18	0.18	1.11	1.39	0.03	5.68	4.20	−1.48
YBDE18	−3.50	3.61	4.50	0.07	9.48	0.58	−8.90
BH76RC	−1.35	3.45	4.24	0.16	17.38	7.54	−9.83

TPSS-CIDH had convergence problems for the h2+_1.75 system in SIE4x4.

Table S110: Statistical analysis of the SOS0-PBE0-2 DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	1.04	1.94	2.32	0.06	7.13	5.16	−1.96
ACONF	0.10	0.19	0.23	0.10	0.73	0.47	−0.26
ADIM6	−2.25	2.25	2.47	0.67	3.01	−0.81	−3.81
AHB21	0.00	0.56	0.72	0.02	3.09	1.07	−2.02
AL2X6	−2.03	2.03	2.12	0.06	1.73	−1.21	−2.94
ALK8	0.40	1.24	2.13	0.02	7.25	5.62	−1.63
ALKBDE10	−5.25	5.25	5.63	0.05	6.45	−2.25	−8.70
AMINO20x4	−0.08	0.26	0.33	0.11	1.60	0.83	−0.77
BH76	2.59	2.69	3.81	0.14	18.63	17.29	−1.34
BHDIV10	1.12	1.12	1.34	0.02	2.45	2.46	0.01
BHPERI	2.29	2.29	2.48	0.11	4.46	4.61	0.15
BHROT27	0.22	0.26	0.33	0.04	0.96	0.74	−0.22
BSR36	−5.14	5.14	5.65	0.32	9.78	−2.06	−11.84
BUT14DIOL	−0.34	0.36	0.38	0.13	0.73	0.14	−0.60
C60ISO	−1.51	2.67	3.37	0.03	8.91	1.92	−7.00
CARBHB12	−0.16	0.31	0.41	0.05	1.47	0.65	−0.82
CDIE20	0.31	0.42	0.56	0.10	1.70	1.32	−0.38
CHB6	−0.88	0.92	1.14	0.03	1.99	0.12	−1.87
DARC	−2.43	2.43	2.60	0.07	2.86	−1.38	−4.24
DC13	−1.43	4.05	4.98	0.07	17.87	7.09	−10.78
DIPCS10	−1.97	3.10	3.61	0.00	10.49	2.85	−7.64
FH51	−1.04	1.47	1.93	0.05	10.24	3.69	−6.56
G21EA	−2.09	2.79	3.33	0.08	13.60	8.21	−5.39
G21IP	−0.11	1.96	2.81	0.01	16.37	10.82	−5.54
G2RC	−1.98	2.77	3.30	0.05	11.99	4.23	−7.77
HAL59	−0.82	0.82	0.94	0.18	2.65	0.15	−2.50
HEAVY28	−0.55	0.55	0.57	0.44	0.52	−0.29	−0.81
HEAVYSB11	−1.52	1.55	1.78	0.03	3.36	0.18	−3.18
ICONF	0.04	0.20	0.25	0.06	0.94	0.48	−0.46
IDISP	−0.27	2.95	3.43	0.21	9.54	4.71	−4.83
IL16	1.68	1.68	1.73	0.02	1.49	2.68	1.19
INV24	0.69	1.17	2.22	0.04	11.59	8.95	−2.64
ISO34	−0.13	0.90	1.24	0.06	5.66	2.67	−2.99
ISOL24	0.23	1.45	1.99	0.07	9.07	4.35	−4.72
MB16-43	−17.47	18.19	21.97	0.04	70.55	11.41	−59.14
MCONF	−0.59	0.62	0.71	0.12	1.42	0.29	−1.13
NBPRC	0.74	1.18	1.61	0.04	4.76	3.59	−1.17
PA26	2.86	2.86	3.11	0.02	4.89	5.84	0.95
PArel	0.36	0.45	0.66	0.10	2.19	1.69	−0.50
PCONF21	−0.01	1.17	1.40	0.72	4.19	2.50	−1.70
PNICO23	−0.69	0.69	0.74	0.16	1.23	−0.27	−1.50
PX13	0.35	0.60	0.75	0.02	1.99	1.45	−0.54
RC21	−1.28	2.36	2.83	0.07	10.29	4.55	−5.74
RG18	−0.33	0.33	0.41	0.57	0.78	−0.02	−0.80
RSE43	1.45	1.45	2.38	0.19	10.34	10.40	0.06
S22	−1.40	1.40	1.79	0.19	4.38	−0.10	−4.48
S66	−1.30	1.30	1.51	0.24	3.41	−0.10	−3.51
SCONF	−0.24	0.48	0.55	0.10	1.76	1.03	−0.73
TAUT15	−0.27	0.48	0.53	0.16	1.31	0.53	−0.78
UPU23	1.04	1.19	1.61	0.21	3.97	3.60	−0.37
W4-11	−3.70	4.70	7.01	0.02	37.25	5.50	−31.75
WATER27	−4.34	4.39	7.24	0.05	20.26	0.30	−19.96
WCPT18	1.38	1.38	1.54	0.04	3.09	3.56	0.46
YBDE18	0.68	1.27	1.49	0.03	4.62	2.94	−1.68
BH76RC	−0.43	1.41	1.90	0.07	7.89	2.96	−4.93

Table S111: Statistical analysis of the SOS0-PBE0-DH DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.19	7.19	8.39	0.21	16.13	18.06	1.93
ACONF	0.39	0.39	0.48	0.21	0.89	0.87	−0.02
ADIM6	−3.02	3.02	3.33	0.90	4.08	−1.08	−5.16
AHB21	−0.66	0.92	1.27	0.04	4.39	1.12	−3.27
AL2X6	−1.72	1.79	2.02	0.05	3.07	0.23	−2.84
ALK8	1.48	1.63	2.40	0.03	5.99	5.40	−0.59
ALKBDE10	−7.27	7.27	8.28	0.07	15.16	−1.48	−16.64
AMINO20x4	0.01	0.38	0.48	0.16	2.53	1.41	−1.12
BH76	−0.74	1.42	1.73	0.08	7.71	4.01	−3.70
BHDIV10	−1.17	1.66	2.02	0.04	6.04	2.02	−4.02
BHPERI	1.34	1.54	1.97	0.07	7.37	5.20	−2.17
BHROT27	0.50	0.51	0.70	0.08	1.51	1.39	−0.12
BSR36	−7.10	7.10	7.97	0.44	14.94	−2.52	−17.46
BUT14DIOL	−0.21	0.27	0.30	0.10	0.85	0.32	−0.52
C60ISO	3.30	3.30	3.96	0.03	7.68	7.78	0.10
CARBHB12	0.33	0.47	0.69	0.08	2.28	1.77	−0.50
CDIE20	0.87	0.97	1.20	0.24	3.00	2.51	−0.49
CHB6	−0.94	1.20	1.48	0.04	3.70	0.78	−2.92
DARC	−2.86	2.86	3.75	0.09	7.94	−0.45	−8.39
DC13	−1.73	8.03	9.39	0.15	32.73	19.06	−13.68
DIPCS10	−0.78	2.73	3.52	0.00	12.29	4.54	−7.74
FH51	−1.52	2.45	3.10	0.08	14.21	5.96	−8.25
G21EA	−1.75	2.72	3.38	0.08	9.49	2.43	−7.06
G21IP	−0.04	2.89	3.53	0.01	14.41	7.56	−6.85
G2RC	−3.62	5.67	7.13	0.11	27.81	10.56	−17.25
HAL59	−0.87	0.98	1.26	0.21	5.86	1.30	−4.57
HEAVY28	−0.63	0.63	0.68	0.51	0.96	−0.18	−1.13
HEAVYSB11	−3.24	3.24	3.46	0.06	3.42	−1.73	−5.16
ICONF	0.21	0.34	0.47	0.10	1.55	1.09	−0.45
IDISP	0.23	6.68	7.22	0.47	18.35	8.70	−9.66
IL16	1.63	1.63	1.74	0.01	2.38	3.16	0.78
INV24	−0.01	1.51	2.38	0.05	11.42	7.16	−4.26
ISO34	−0.21	1.49	2.01	0.10	8.62	3.34	−5.28
ISOL24	0.05	2.48	3.52	0.11	17.11	7.59	−9.52
MB16-43	−12.29	16.72	20.59	0.04	71.75	29.32	−42.42
MCONF	−1.18	1.26	1.43	0.25	2.97	0.57	−2.40
NBPRC	0.32	2.30	2.72	0.08	8.45	5.50	−2.95
PA26	2.97	3.01	3.47	0.02	8.07	7.53	−0.55
PArel	0.78	1.08	1.46	0.23	5.79	4.26	−1.53
PCONF21	−0.22	2.33	2.66	1.44	7.44	4.29	−3.14
PNICO23	−0.47	0.59	0.73	0.14	3.38	1.27	−2.11
PX13	−2.79	2.79	2.97	0.08	3.73	−0.91	−4.64
RC21	2.70	3.29	4.16	0.09	14.65	12.32	−2.33
RG18	−0.35	0.36	0.45	0.62	0.96	0.01	−0.95
RSE43	−0.42	0.44	0.57	0.06	2.04	0.23	−1.80
S22	−1.94	1.98	2.78	0.27	7.97	0.44	−7.54
S66	−1.72	1.74	2.17	0.32	5.72	0.39	−5.33
SCONF	−0.15	0.37	0.47	0.08	1.87	1.15	−0.71
TAUT15	0.22	0.75	1.02	0.25	3.32	2.13	−1.19
UPU23	1.43	1.68	2.39	0.29	6.56	5.89	−0.68
W4-11	−6.09	6.36	8.33	0.02	31.20	2.90	−28.30
WATER27	−1.66	2.52	4.18	0.03	14.69	1.87	−12.83
WCPT18	−0.96	1.46	1.82	0.04	6.01	2.30	−3.71
YBDE18	−1.37	1.74	2.26	0.04	6.08	1.05	−5.04
BH76RC	−0.52	2.04	2.56	0.10	8.44	3.29	−5.15

Table S112: Statistical analysis of the SOS1-PBE-QIDH DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	2.89	3.01	3.94	0.09	9.91	9.22	−0.68
ACONF	0.21	0.25	0.31	0.14	0.80	0.63	−0.17
ADIM6	−2.55	2.55	2.81	0.76	3.42	−0.91	−4.33
AHB21	−0.34	0.72	0.99	0.03	3.94	1.05	−2.89
AL2X6	−1.66	1.66	1.81	0.05	2.10	−0.48	−2.58
ALK8	1.02	1.21	1.93	0.02	5.20	4.69	−0.51
ALKBDE10	−6.70	6.70	7.27	0.07	10.86	−2.34	−13.20
AMINO20x4	−0.03	0.29	0.37	0.12	1.98	1.05	−0.93
BH76	1.40	1.66	2.47	0.09	13.93	12.13	−1.80
BHDIV10	0.39	0.95	1.06	0.02	3.10	1.60	−1.50
BHPERI	2.14	2.19	2.45	0.10	6.24	5.58	−0.66
BHROT27	0.35	0.37	0.50	0.06	1.16	1.03	−0.14
BSR36	−5.90	5.90	6.54	0.36	11.70	−2.29	−13.98
BUT14DIOL	−0.29	0.31	0.34	0.11	0.74	0.20	−0.54
C60ISO	1.21	2.32	2.74	0.02	8.07	5.44	−2.63
CARBHB12	0.05	0.34	0.44	0.06	1.71	1.12	−0.59
CDIE20	0.51	0.61	0.80	0.15	2.21	1.80	−0.41
CHB6	−0.98	1.09	1.34	0.04	2.79	0.31	−2.47
DARC	−3.34	3.34	3.65	0.10	4.85	−1.77	−6.62
DC13	−1.87	5.71	7.05	0.10	25.91	12.51	−13.40
DIPCS10	−1.18	2.88	3.52	0.00	11.97	4.32	−7.65
FH51	−1.47	2.00	2.55	0.06	12.91	4.52	−8.39
G21EA	−2.13	2.58	3.21	0.08	11.22	4.52	−6.71
G21IP	−0.03	2.04	2.71	0.01	11.67	5.65	−6.02
G2RC	−2.95	4.23	5.15	0.08	18.82	7.20	−11.62
HAL59	−0.85	0.87	1.05	0.19	3.95	0.63	−3.32
HEAVY28	−0.59	0.59	0.62	0.48	0.66	−0.26	−0.93
HEAVYSB11	−2.29	2.29	2.42	0.04	2.65	−0.61	−3.26
ICONF	0.12	0.26	0.33	0.08	1.10	0.69	−0.41
IDISP	−0.26	4.25	4.74	0.30	12.74	6.15	−6.59
IL16	1.60	1.60	1.67	0.01	1.84	2.82	0.98
INV24	0.55	1.37	2.45	0.04	12.33	8.71	−3.62
ISO34	−0.13	1.15	1.61	0.08	7.08	3.24	−3.84
ISOL24	0.37	1.80	2.53	0.08	11.07	5.97	−5.10
MB16-43	−14.56	16.47	20.63	0.04	70.26	15.55	−54.71
MCONF	−0.81	0.86	0.98	0.17	2.01	0.41	−1.59
NBPRC	0.45	1.64	2.09	0.06	6.18	4.55	−1.63
PA26	3.02	3.02	3.37	0.02	6.02	6.69	0.66
PArel	0.59	0.75	1.03	0.16	3.71	2.78	−0.93
PCONF21	−0.07	1.61	1.88	0.99	5.40	3.17	−2.23
PNICO23	−0.59	0.59	0.67	0.14	1.71	−0.03	−1.74
PX13	−0.67	0.89	1.01	0.03	2.34	0.75	−1.59
RC21	0.74	1.78	2.75	0.05	13.21	8.83	−4.38
RG18	−0.35	0.35	0.43	0.60	0.84	−0.01	−0.85
RSE43	0.64	0.68	1.09	0.09	5.04	4.53	−0.51
S22	−1.58	1.58	2.14	0.22	5.64	0.02	−5.63
S66	−1.44	1.44	1.75	0.26	4.18	0.00	−4.17
SCONF	−0.20	0.43	0.51	0.09	1.77	1.08	−0.70
TAUT15	−0.07	0.47	0.52	0.15	1.62	0.69	−0.94
UPU23	1.21	1.38	1.92	0.24	4.98	4.49	−0.49
W4-11	−5.49	5.96	8.24	0.02	33.39	4.59	−28.80
WATER27	−3.11	3.42	5.76	0.04	17.54	0.87	−16.66
WCPT18	0.65	0.97	1.24	0.03	4.23	3.54	−0.69
YBDE18	−0.11	1.42	1.76	0.03	6.54	2.65	−3.90
BH76RC	−0.55	1.68	2.22	0.09	8.87	3.21	−5.66

Table S113: Statistical analysis of the SOS0-PBE-CIDH DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	5.96	5.96	7.09	0.18	14.34	15.63	1.29
ACONF	0.35	0.36	0.44	0.20	0.87	0.81	−0.06
ADIM6	−2.92	2.92	3.22	0.87	3.93	−1.04	−4.98
AHB21	−0.61	0.89	1.24	0.04	4.41	1.10	−3.32
AL2X6	−1.62	1.66	1.89	0.05	2.77	0.12	−2.66
ALK8	1.43	1.54	2.26	0.02	5.32	4.89	−0.44
ALKBDE10	−7.43	7.43	8.31	0.07	14.51	−1.89	−16.41
AMINO20x4	0.01	0.36	0.46	0.15	2.40	1.31	−1.09
BH76	−0.17	1.17	1.59	0.06	9.08	5.85	−3.23
BHDIV10	−0.73	1.30	1.62	0.03	5.21	1.82	−3.40
BHPERI	1.62	1.75	2.15	0.08	7.35	5.57	−1.78
BHROT27	0.47	0.49	0.66	0.08	1.45	1.34	−0.11
BSR36	−6.83	6.83	7.65	0.42	14.18	−2.49	−16.67
BUT14DIOL	−0.22	0.28	0.31	0.10	0.82	0.29	−0.53
C60ISO	3.16	3.17	3.88	0.03	7.82	7.78	−0.04
CARBHB12	0.27	0.42	0.62	0.07	2.15	1.63	−0.52
CDIE20	0.78	0.88	1.10	0.22	2.83	2.35	−0.48
CHB6	−0.98	1.20	1.48	0.04	3.54	0.65	−2.89
DARC	−3.23	3.23	3.91	0.10	7.23	−1.02	−8.25
DC13	−1.86	7.59	8.97	0.14	31.96	17.81	−14.15
DIPCS10	−0.77	2.78	3.54	0.00	12.57	4.82	−7.75
FH51	−1.60	2.40	3.04	0.08	14.32	5.62	−8.71
G21EA	−1.92	2.66	3.39	0.08	9.41	2.15	−7.27
G21IP	−0.02	2.70	3.32	0.01	13.76	7.16	−6.61
G2RC	−3.60	5.46	6.81	0.11	26.27	9.96	−16.31
HAL59	−0.88	0.96	1.21	0.21	5.43	1.14	−4.28
HEAVY28	−0.62	0.62	0.67	0.50	0.88	−0.20	−1.08
HEAVYSB11	−3.08	3.08	3.24	0.05	3.02	−1.67	−4.69
ICONF	0.19	0.32	0.44	0.10	1.37	0.94	−0.43
IDISP	0.05	6.08	6.59	0.43	17.14	8.09	−9.05
IL16	1.61	1.61	1.71	0.01	2.26	3.08	0.81
INV24	0.17	1.51	2.46	0.05	11.52	7.27	−4.25
ISO34	−0.18	1.43	1.94	0.10	8.37	3.43	−4.94
ISOL24	0.20	2.26	3.24	0.10	15.39	7.42	−7.98
MB16-43	−12.59	16.58	20.51	0.04	71.01	25.86	−45.15
MCONF	−1.09	1.16	1.33	0.23	2.75	0.54	−2.21
NBPRC	0.31	2.20	2.62	0.08	8.03	5.43	−2.60
PA26	3.02	3.04	3.49	0.02	7.66	7.39	−0.27
PArel	0.76	1.03	1.38	0.22	5.33	3.93	−1.40
PCONF21	−0.18	2.16	2.48	1.33	6.96	4.03	−2.93
PNICO23	−0.50	0.59	0.70	0.14	2.98	0.96	−2.02
PX13	−2.20	2.20	2.39	0.07	3.33	−0.43	−3.75
RC21	2.35	2.96	3.86	0.08	14.75	11.92	−2.83
RG18	−0.36	0.36	0.45	0.62	0.92	0.00	−0.92
RSE43	−0.18	0.27	0.40	0.04	2.20	0.71	−1.49
S22	−1.85	1.88	2.63	0.26	7.48	0.39	−7.09
S66	−1.65	1.67	2.07	0.31	5.40	0.34	−5.06
SCONF	−0.16	0.39	0.48	0.08	1.85	1.14	−0.71
TAUT15	0.16	0.67	0.88	0.22	2.89	1.80	−1.09
UPU23	1.39	1.61	2.29	0.28	6.20	5.57	−0.63
W4-11	−6.35	6.63	8.70	0.02	30.94	3.39	−27.55
WATER27	−1.97	2.67	4.49	0.03	15.31	1.65	−13.65
WCPT18	−0.50	1.24	1.53	0.04	5.64	2.78	−2.86
YBDE18	−1.09	1.71	2.17	0.03	6.45	1.38	−5.06
BH76RC	−0.56	1.99	2.51	0.10	8.06	2.86	−5.20

Table S114: Statistical analysis of the SOS0-PBE0-2(b) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	4.43	4.43	4.91	0.13	7.46	8.53	1.07
ACONF	−0.26	0.26	0.28	0.14	0.40	0.00	−0.41
ADIM6	−1.07	1.07	1.18	0.32	1.40	−0.39	−1.79
AHB21	−0.44	0.51	0.66	0.02	2.45	0.38	−2.07
AL2X6	0.08	0.62	0.78	0.02	2.33	0.72	−1.60
ALK8	2.51	3.56	6.05	0.06	18.93	15.41	−3.51
ALKBDE10	9.86	9.86	11.33	0.10	14.87	18.69	3.82
AMINO20x4	−0.06	0.13	0.16	0.05	0.68	0.28	−0.40
BH76	0.58	2.13	3.87	0.11	25.46	21.05	−4.41
BHDIV10	−2.04	2.51	2.85	0.06	7.19	2.32	−4.88
BHPERI	−2.93	3.08	3.32	0.15	6.82	1.93	−4.89
BHROT27	−0.01	0.13	0.17	0.02	0.74	0.23	−0.50
BSR36	−1.76	1.76	1.85	0.11	2.21	−0.94	−3.15
BUT14DIOL	−0.04	0.06	0.08	0.02	0.28	0.14	−0.14
C60ISO	−13.22	13.22	15.56	0.13	23.88	−3.40	−27.28
CARBHB12	0.41	0.41	0.58	0.07	1.30	1.28	−0.02
CDIE20	0.29	0.32	0.40	0.08	1.15	1.00	−0.15
CHB6	−1.00	1.00	1.18	0.04	1.91	−0.30	−2.21
DARC	−5.00	5.00	5.07	0.15	2.51	−3.64	−6.15
DC13	−2.48	9.20	12.04	0.17	48.20	21.28	−26.93
DIPCS10	6.79	6.79	7.24	0.01	7.48	10.69	3.21
FH51	−0.69	1.48	2.11	0.05	11.96	5.61	−6.35
G21EA	4.53	5.41	6.53	0.16	20.55	16.50	−4.05
G21IP	5.18	5.72	7.37	0.02	32.47	24.24	−8.23
G2RC	−0.57	2.46	3.32	0.05	15.83	7.11	−8.72
HAL59	0.06	0.32	0.47	0.07	2.40	1.82	−0.58
HEAVY28	−0.12	0.14	0.18	0.11	0.51	0.13	−0.38
HEAVYSB11	6.65	6.65	6.70	0.11	3.12	7.63	4.50
ICONF	−0.03	0.18	0.24	0.06	1.07	0.39	−0.68
IDISP	−0.74	2.47	3.48	0.17	11.40	4.30	−7.10
IL16	−0.09	0.30	0.38	0.00	1.44	0.62	−0.82
INV24	−0.18	0.80	1.60	0.03	8.84	1.63	−7.21
ISO34	−0.29	1.03	1.53	0.07	7.18	3.25	−3.93
ISOL24	0.98	2.04	2.74	0.09	9.63	6.34	−3.29
MB16-43	9.36	12.24	14.85	0.03	50.29	31.89	−18.40
MCONF	0.16	0.21	0.27	0.04	0.80	0.53	−0.28
NBPRC	−0.92	2.00	2.39	0.07	7.50	2.24	−5.26
PA26	1.67	1.72	1.84	0.01	3.70	3.06	−0.64
PArel	−0.02	0.72	1.14	0.16	4.85	1.87	−2.98
PCONF21	−0.10	0.40	0.47	0.25	1.53	0.53	−1.01
PNICO23	0.01	0.24	0.37	0.06	1.97	1.46	−0.51
PX13	−4.19	4.19	4.30	0.13	4.13	−2.16	−6.29
RC21	−1.02	2.69	3.32	0.08	12.88	7.84	−5.03
RG18	−0.18	0.22	0.28	0.38	0.90	0.30	−0.61
RSE43	1.52	1.57	3.46	0.21	16.08	15.63	−0.46
S22	−0.35	0.36	0.53	0.05	1.37	0.07	−1.30
S66	−0.44	0.46	0.57	0.08	1.48	0.16	−1.31
SCONF	0.14	0.28	0.31	0.06	1.02	0.37	−0.64
TAUT15	−0.28	0.64	0.80	0.21	2.46	0.88	−1.59
UPU23	0.44	0.56	0.70	0.10	1.85	1.52	−0.32
W4-11	28.42	28.48	32.75	0.09	71.44	68.61	−2.83
WATER27	−0.04	1.14	1.64	0.01	6.50	1.51	−4.98
WCPT18	−2.70	2.70	2.80	0.08	2.69	−1.82	−4.50
YBDE18	9.18	9.18	9.82	0.19	11.79	16.82	5.03
BH76RC	−0.23	1.84	2.50	0.09	12.14	8.87	−3.27

Table S115: Statistical analysis of the SOS0-PBE0-DH(b) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	8.30	8.30	9.42	0.25	16.75	19.09	2.34
ACONF	0.28	0.30	0.36	0.16	0.73	0.66	−0.07
ADIM6	−2.67	2.67	2.95	0.79	3.60	−0.95	−4.56
AHB21	−0.76	0.90	1.22	0.04	3.71	0.91	−2.79
AL2X6	−1.11	1.36	1.49	0.04	2.82	0.73	−2.09
ALK8	2.06	2.27	3.12	0.04	6.62	5.79	−0.83
ALKBDE10	−2.74	3.65	4.36	0.04	12.04	4.24	−7.80
AMINO20x4	0.02	0.32	0.41	0.13	2.11	1.23	−0.89
BH76	−1.63	2.09	2.34	0.11	10.36	3.68	−6.68
BHDIV10	−2.18	2.54	2.90	0.06	6.22	1.41	−4.81
BHPERI	−0.13	1.01	1.33	0.05	6.18	3.69	−2.49
BHROT27	0.42	0.44	0.58	0.07	1.23	1.11	−0.12
BSR36	−6.08	6.08	6.82	0.38	12.68	−2.18	−14.86
BUT14DIOL	−0.11	0.18	0.21	0.06	0.71	0.33	−0.38
C60ISO	−0.69	2.33	2.95	0.02	8.25	2.06	−6.19
CARBHB12	0.49	0.56	0.80	0.09	2.24	1.94	−0.30
CDIE20	0.86	0.93	1.13	0.23	2.68	2.29	−0.39
CHB6	−0.94	1.12	1.39	0.04	3.25	0.54	−2.71
DARC	−3.36	3.36	4.04	0.10	7.16	−0.99	−8.15
DC13	−1.87	7.56	9.37	0.14	34.52	17.53	−16.99
DIPCS10	1.59	2.52	2.86	0.00	7.59	4.32	−3.27
FH51	−1.32	2.32	2.98	0.07	12.81	4.92	−7.89
G21EA	0.11	1.45	1.69	0.04	5.65	2.70	−2.95
G21IP	1.34	2.45	2.96	0.01	12.59	7.85	−4.74
G2RC	−2.97	5.24	6.67	0.10	25.34	9.09	−16.25
HAL59	−0.60	0.84	1.08	0.18	5.65	1.73	−3.93
HEAVY28	−0.50	0.50	0.56	0.40	0.89	−0.07	−0.95
HEAVYSB11	−0.94	1.36	1.69	0.02	3.57	0.83	−2.74
ICONF	0.19	0.27	0.38	0.08	1.17	0.88	−0.29
IDISP	0.14	5.12	5.52	0.36	13.87	6.80	−7.07
IL16	1.10	1.10	1.22	0.01	2.25	2.43	0.18
INV24	−0.29	1.13	1.60	0.04	7.06	4.09	−2.97
ISO34	−0.28	1.34	1.88	0.09	7.74	3.38	−4.36
ISOL24	0.16	2.23	3.10	0.10	13.75	6.99	−6.76
MB16-43	−5.01	11.97	15.01	0.03	60.56	32.29	−28.27
MCONF	−0.96	1.03	1.18	0.21	2.67	0.61	−2.06
NBPRC	−0.14	2.48	2.78	0.09	8.48	4.79	−3.69
PA26	2.64	2.69	3.08	0.01	7.55	6.83	−0.72
PArel	0.66	1.02	1.38	0.22	6.14	4.00	−2.14
PCONF21	−0.26	2.10	2.35	1.30	6.31	3.68	−2.63
PNICO23	−0.26	0.49	0.69	0.11	3.82	2.01	−1.81
PX13	−4.22	4.22	4.38	0.13	4.58	−2.08	−6.66
RC21	2.96	3.51	4.25	0.10	13.36	11.10	−2.26
RG18	−0.31	0.31	0.40	0.53	0.88	0.03	−0.85
RSE43	−0.56	0.58	0.71	0.08	2.41	0.53	−1.88
S22	−1.64	1.69	2.39	0.23	7.01	0.51	−6.50
S66	−1.47	1.50	1.89	0.27	5.18	0.48	−4.70
SCONF	−0.03	0.18	0.24	0.04	1.06	0.65	−0.40
TAUT15	0.17	0.87	1.10	0.29	3.58	2.22	−1.35
UPU23	1.25	1.49	2.11	0.26	5.81	5.15	−0.66
W4-11	3.28	5.18	6.70	0.02	34.04	18.74	−15.30
WATER27	−0.36	1.82	2.72	0.02	10.30	2.35	−7.95
WCPT18	−2.31	2.33	2.78	0.07	5.64	0.20	−5.44
YBDE18	0.92	1.44	1.61	0.03	5.66	3.04	−2.62
BH76RC	−0.42	1.90	2.52	0.09	11.12	6.34	−4.78

Table S116: Statistical analysis of the SOS0-PBE-QIDH(b) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	5.37	5.37	6.02	0.16	10.10	11.63	1.53
ACONF	−0.05	0.10	0.13	0.05	0.43	0.15	−0.28
ADIM6	−1.72	1.72	1.90	0.51	2.28	−0.62	−2.90
AHB21	−0.64	0.71	0.94	0.03	3.01	0.55	−2.46
AL2X6	−0.18	0.48	0.69	0.01	2.13	0.71	−1.43
ALK8	2.48	2.64	4.62	0.04	12.16	11.53	−0.63
ALKBDE10	3.96	4.02	5.36	0.04	11.49	11.18	−0.31
AMINO20x4	−0.02	0.15	0.20	0.06	1.06	0.63	−0.43
BH76	−0.26	1.70	2.53	0.09	15.34	11.41	−3.93
BHDIV10	−1.90	2.20	2.55	0.05	6.00	1.51	−4.49
BHPERI	−1.47	1.86	1.98	0.09	5.75	2.83	−2.93
BHROT27	0.18	0.20	0.27	0.03	0.69	0.62	−0.07
BSR36	−3.50	3.50	3.82	0.22	6.33	−1.49	−7.82
BUT14DIOL	−0.07	0.09	0.11	0.03	0.37	0.15	−0.22
C60ISO	−7.42	7.42	9.28	0.08	16.72	−0.82	−17.54
CARBHB12	0.44	0.47	0.66	0.08	1.67	1.55	−0.12
CDIE20	0.49	0.55	0.66	0.14	1.52	1.32	−0.20
CHB6	−1.04	1.04	1.25	0.04	1.79	−0.25	−2.05
DARC	−4.96	4.96	5.10	0.15	3.31	−3.31	−6.62
DC13	−2.49	7.42	10.12	0.13	34.75	18.11	−16.64
DIPCS10	4.81	4.81	5.09	0.01	5.68	7.40	1.72
FH51	−1.15	1.84	2.46	0.06	12.57	4.55	−8.02
G21EA	2.43	2.74	3.32	0.08	10.93	8.92	−2.01
G21IP	3.55	3.79	4.55	0.01	16.71	13.70	−3.01
G2RC	−1.78	3.44	4.54	0.07	16.69	6.37	−10.32
HAL59	−0.22	0.48	0.63	0.10	3.33	1.58	−1.74
HEAVY28	−0.29	0.29	0.33	0.23	0.51	0.01	−0.50
HEAVYSB11	3.36	3.36	3.47	0.06	2.90	4.51	1.61
ICONF	0.07	0.13	0.17	0.04	0.53	0.33	−0.19
IDISP	−0.55	1.39	1.71	0.10	4.43	1.66	−2.78
IL16	0.34	0.44	0.52	0.00	1.56	1.11	−0.45
INV24	−0.07	0.70	1.02	0.02	5.24	3.45	−1.79
ISO34	−0.26	1.11	1.60	0.08	7.14	3.55	−3.59
ISOL24	0.82	1.84	2.48	0.08	9.28	6.68	−2.60
MB16-43	3.83	8.87	10.96	0.02	41.30	22.09	−19.21
MCONF	−0.28	0.33	0.40	0.07	1.27	0.49	−0.78
NBPRC	−0.71	2.18	2.58	0.08	7.80	3.11	−4.70
PA26	2.21	2.21	2.42	0.01	4.36	4.84	0.48
PArel	0.32	0.78	1.03	0.17	4.53	1.98	−2.55
PCONF21	−0.15	1.04	1.14	0.64	3.12	1.76	−1.36
PNICO23	−0.09	0.32	0.48	0.07	2.69	1.66	−1.03
PX13	−3.94	3.94	4.07	0.12	4.20	−1.87	−6.07
RC21	1.13	2.35	2.98	0.07	11.22	7.92	−3.30
RG18	−0.24	0.25	0.32	0.43	0.69	0.03	−0.66
RSE43	0.54	0.73	1.58	0.10	7.59	6.76	−0.83
S22	−0.85	0.87	1.19	0.12	3.36	0.22	−3.14
S66	−0.84	0.87	1.07	0.16	2.86	0.23	−2.64
SCONF	0.08	0.13	0.17	0.03	0.63	0.31	−0.32
TAUT15	−0.11	0.71	0.78	0.23	2.15	0.98	−1.17
UPU23	0.78	0.94	1.25	0.16	3.16	2.77	−0.39
W4-11	17.01	17.16	20.29	0.06	54.84	47.57	−7.28
WATER27	−0.05	1.41	1.98	0.02	7.26	1.92	−5.33
WCPT18	−2.33	2.33	2.56	0.07	3.53	−1.01	−4.54
YBDE18	5.70	5.70	6.02	0.12	6.85	9.13	2.29
BH76RC	−0.38	1.53	2.16	0.07	11.04	7.10	−3.93

Table S117: Statistical analysis of the SOS0-PBE-CIDH(b) DFA for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	7.38	7.38	8.37	0.22	14.82	16.96	2.14
ACONF	0.21	0.24	0.29	0.13	0.67	0.55	−0.12
ADIM6	−2.47	2.47	2.72	0.74	3.31	−0.88	−4.20
AHB21	−0.75	0.88	1.19	0.04	3.57	0.82	−2.75
AL2X6	−0.84	1.09	1.20	0.03	2.47	0.76	−1.72
ALK8	2.20	2.39	3.36	0.04	7.79	7.05	−0.74
ALKBDE10	−1.60	3.01	3.51	0.03	10.37	5.16	−5.21
AMINO20x4	0.01	0.28	0.36	0.11	1.87	1.07	−0.80
BH76	−1.26	1.86	2.13	0.10	9.69	3.93	−5.77
BHDIV10	−2.02	2.35	2.70	0.05	5.64	1.05	−4.59
BHPERI	−0.29	1.02	1.33	0.05	5.88	3.65	−2.22
BHROT27	0.37	0.39	0.52	0.06	1.12	1.01	−0.11
BSR36	−5.52	5.52	6.16	0.34	11.26	−2.05	−13.30
BUT14DIOL	−0.10	0.16	0.19	0.06	0.63	0.28	−0.35
C60ISO	−1.86	2.65	3.77	0.03	9.74	1.57	−8.17
CARBHB12	0.48	0.54	0.76	0.09	2.11	1.85	−0.26
CDIE20	0.77	0.84	1.02	0.21	2.44	2.08	−0.36
CHB6	−0.99	1.10	1.37	0.04	2.96	0.33	−2.63
DARC	−3.94	3.94	4.41	0.12	6.26	−1.78	−8.03
DC13	−2.08	7.44	9.29	0.14	32.36	15.96	−16.41
DIPCS10	2.34	2.73	3.15	0.00	6.80	4.85	−1.95
FH51	−1.36	2.25	2.90	0.07	12.60	4.28	−8.31
G21EA	0.51	1.21	1.46	0.04	5.63	3.69	−1.94
G21IP	1.81	2.36	2.92	0.01	11.46	7.55	−3.91
G2RC	−2.81	4.90	6.26	0.10	23.26	8.07	−15.18
HAL59	−0.52	0.76	0.98	0.17	5.14	1.69	−3.45
HEAVY28	−0.46	0.46	0.51	0.37	0.79	−0.07	−0.85
HEAVYSB11	−0.07	1.14	1.22	0.02	3.27	1.53	−1.74
ICONF	0.16	0.24	0.32	0.07	0.98	0.71	−0.27
IDISP	−0.07	4.05	4.41	0.28	11.35	5.63	−5.72
IL16	0.92	0.92	1.05	0.01	2.10	2.13	0.04
INV24	−0.18	1.02	1.46	0.03	6.94	4.28	−2.66
ISO34	−0.26	1.27	1.81	0.09	7.27	3.51	−3.76
ISOL24	0.37	2.02	2.78	0.09	11.78	6.72	−5.06
MB16-43	−3.02	10.60	13.42	0.03	54.96	29.86	−25.10
MCONF	−0.80	0.87	1.00	0.18	2.36	0.59	−1.76
NBPRC	−0.29	2.44	2.75	0.09	8.11	4.55	−3.56
PA26	2.59	2.63	2.98	0.01	6.95	6.47	−0.48
PArel	0.61	0.96	1.29	0.21	5.78	3.57	−2.21
PCONF21	−0.23	1.85	2.07	1.14	5.51	3.25	−2.26
PNICO23	−0.23	0.45	0.63	0.11	3.54	1.90	−1.64
PX13	−4.03	4.03	4.18	0.12	4.43	−1.91	−6.34
RC21	2.65	3.23	3.97	0.09	12.76	10.45	−2.31
RG18	−0.30	0.30	0.39	0.52	0.84	0.03	−0.81
RSE43	−0.33	0.46	0.59	0.06	2.85	1.27	−1.58
S22	−1.46	1.51	2.12	0.21	6.24	0.49	−5.75
S66	−1.33	1.36	1.71	0.25	4.70	0.47	−4.23
SCONF	0.00	0.15	0.19	0.03	0.82	0.51	−0.31
TAUT15	0.11	0.82	1.00	0.27	3.21	1.93	−1.27
UPU23	1.15	1.37	1.92	0.24	5.19	4.61	−0.58
W4-11	5.78	6.90	8.83	0.02	35.79	24.72	−11.07
WATER27	−0.29	1.72	2.55	0.02	9.64	2.25	−7.39
WCPT18	−2.21	2.22	2.63	0.06	5.21	0.14	−5.07
YBDE18	1.92	2.10	2.30	0.04	5.40	3.95	−1.45
BH76RC	−0.44	1.79	2.41	0.08	10.78	6.17	−4.61

SI.8.6 Results for dispersion-uncorrected and -corrected MP2 and its spin-scaled variants

Table S118: Statistical analysis of MP2 for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	−1.29	1.54	2.32	0.05	5.84	0.84	−5.00
ACONF	−0.49	0.49	0.51	0.27	0.74	−0.04	−0.78
ADIM6	0.26	0.26	0.29	0.08	0.37	0.45	0.08
AHB21	−0.40	0.43	0.70	0.02	2.91	0.19	−2.73
AL2X6	−0.54	0.73	1.01	0.02	2.57	0.38	−2.19
ALK8	1.14	3.29	5.89	0.05	20.62	15.62	−4.99
ALKBDE10	5.40	6.33	8.09	0.06	17.73	14.59	−3.13
AMINO20x4	−0.08	0.26	0.33	0.11	1.64	0.72	−0.92
BH76	3.78	4.44	6.87	0.24	34.13	28.00	−6.13
BHDIV10	−1.26	2.24	2.99	0.05	10.65	3.07	−7.58
BHPERI	−7.64	7.64	8.05	0.37	11.75	−1.09	−12.84
BHROT27	0.17	0.25	0.30	0.04	1.15	0.65	−0.50
BSR36	2.81	2.82	3.63	0.17	9.74	9.68	−0.06
BUT14DIOL	0.07	0.10	0.14	0.04	0.67	0.45	−0.22
C60ISO	−18.26	18.26	22.07	0.19	35.34	−3.92	−39.26
CARBHB12	0.54	0.54	0.73	0.09	1.35	1.41	0.06
CDIE20	0.50	0.50	0.66	0.12	1.76	1.85	0.09
CHB6	−0.90	0.92	1.24	0.03	2.38	0.04	−2.34
DARC	−3.57	3.57	3.94	0.11	4.25	−1.29	−5.55
DC13	2.17	8.28	10.86	0.15	35.75	23.06	−12.69
DIPCS10	−4.70	5.05	6.30	0.01	13.22	1.30	−11.93
FH51	−1.17	2.58	3.59	0.08	17.92	4.80	−13.11
G21EA	−0.87	3.80	6.44	0.11	41.20	21.19	−20.00
G21IP	−0.46	3.21	5.01	0.01	31.71	20.40	−11.31
G2RC	−1.20	3.09	3.89	0.06	13.17	4.27	−8.90
HAL59	0.55	0.59	0.89	0.13	3.69	3.07	−0.62
HEAVY28	0.20	0.24	0.29	0.19	0.96	0.60	−0.36
HEAVYSB11	3.69	3.69	3.99	0.06	4.34	6.09	1.75
ICONF	0.02	0.23	0.34	0.07	1.56	0.40	−1.15
IDISP	−0.87	6.55	7.43	0.46	22.27	10.69	−11.58
IL16	−0.32	0.41	0.52	0.00	1.49	0.32	−1.17
INV24	−0.83	0.96	1.17	0.03	3.77	1.20	−2.57
ISO34	0.62	1.13	1.68	0.08	7.52	5.46	−2.06
ISOL24	1.87	2.73	3.70	0.12	16.15	11.38	−4.76
MB16-43	21.65	22.70	25.52	0.05	65.49	46.99	−18.50
MCONF	1.00	1.08	1.23	0.22	2.55	2.03	−0.52
NBPRC	−0.66	1.10	1.38	0.04	4.09	1.64	−2.44
PA26	−1.67	1.85	3.01	0.01	10.73	1.44	−9.30
PArel	0.10	0.93	1.37	0.20	6.81	3.83	−2.97
PCONF21	0.20	0.86	1.12	0.53	3.86	2.14	−1.72
PNICO23	0.24	0.24	0.36	0.06	1.14	1.08	−0.06
PX13	−2.70	2.70	2.81	0.08	2.87	−1.41	−4.28
RC21	−4.92	6.73	7.95	0.19	21.73	6.98	−14.75
RG18	−0.02	0.20	0.33	0.34	1.61	1.03	−0.59
RSE43	3.53	3.53	6.71	0.46	27.04	27.49	0.45
S22	0.93	1.00	1.54	0.14	4.14	3.84	−0.30
S66	0.55	0.56	0.82	0.10	2.34	2.26	−0.08
SCONF	0.37	0.44	0.49	0.10	1.23	0.83	−0.39
TAUT15	0.66	0.76	0.94	0.25	2.36	1.72	−0.64
UPU23	−0.23	0.54	0.77	0.09	3.27	0.78	−2.50
W4-11	3.33	7.54	9.52	0.02	46.30	23.51	−22.80
WATER27	0.14	0.63	0.93	0.01	5.31	1.70	−3.61
WCPT18	−2.54	2.54	2.65	0.07	3.09	−1.27	−4.36
YBDE18	6.44	6.44	7.52	0.13	15.16	15.63	0.47
BH76RC	0.06	3.03	4.13	0.14	19.58	11.53	−8.05

Table S119: Statistical analysis of SCS-MP2 for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	−2.13	2.15	2.50	0.06	3.38	0.08	−3.30
ACONF	−0.22	0.22	0.26	0.12	0.50	0.04	−0.45
ADIM6	−1.01	1.01	1.12	0.30	1.37	−0.37	−1.73
AHB21	0.74	0.74	0.78	0.03	1.01	1.17	0.16
AL2X6	−2.97	2.97	2.98	0.08	0.75	−2.71	−3.45
ALK8	−1.12	3.45	4.68	0.06	15.84	9.32	−6.52
ALKBDE10	1.02	2.91	3.88	0.03	12.19	7.45	−4.74
AMINO20x4	−0.20	0.30	0.40	0.12	1.78	0.42	−1.35
BH76	5.36	5.46	7.78	0.29	31.07	29.54	−1.53
BHDIV10	1.57	1.57	1.82	0.03	2.80	2.83	0.03
BHPERI	−0.79	1.23	1.50	0.06	5.64	2.94	−2.70
BHROT27	−0.06	0.18	0.24	0.03	0.99	0.38	−0.60
BSR36	−1.59	1.59	1.66	0.10	2.13	−0.77	−2.90
BUT14DIOL	−0.34	0.35	0.38	0.13	0.82	0.19	−0.63
C60ISO	−12.20	12.20	14.46	0.12	23.40	−2.37	−25.77
CARBHB12	−0.40	0.40	0.50	0.07	1.04	−0.05	−1.09
CDIE20	0.08	0.21	0.31	0.05	1.38	1.08	−0.30
CHB6	−0.53	0.59	0.85	0.02	2.05	0.19	−1.87
DARC	0.70	0.83	1.10	0.03	2.90	2.54	−0.36
DC13	1.15	4.18	5.18	0.08	18.02	9.81	−8.21
DIPCS10	−5.79	5.79	6.34	0.01	8.29	−0.63	−8.93
FH51	0.17	1.57	2.26	0.05	12.89	4.05	−8.84
G21EA	−2.19	4.48	7.15	0.13	44.39	23.69	−20.70
G21IP	−0.94	3.45	5.67	0.01	37.01	24.89	−12.12
G2RC	0.47	2.27	2.58	0.04	9.60	5.19	−4.40
HAL59	−0.42	0.48	0.61	0.10	2.44	0.63	−1.81
HEAVY28	−0.27	0.27	0.30	0.22	0.55	−0.02	−0.56
HEAVYSB11	1.02	1.66	1.85	0.03	4.57	2.92	−1.65
ICONF	−0.13	0.24	0.33	0.07	1.19	0.37	−0.82
IDISP	−0.08	1.77	2.31	0.12	7.66	4.23	−3.43
IL16	1.52	1.52	1.55	0.01	1.06	2.32	1.25
INV24	−0.83	0.96	1.17	0.03	3.77	1.20	−2.57
ISO34	0.06	0.48	0.73	0.03	4.24	2.67	−1.57
ISOL24	0.01	0.95	1.28	0.04	5.89	2.54	−3.36
MB16-43	−14.48	16.03	19.90	0.04	59.31	13.50	−45.81
MCONF	0.17	0.28	0.32	0.06	1.22	0.67	−0.55
NBPRC	1.27	1.59	1.86	0.06	4.76	3.80	−0.96
PA26	0.87	1.28	1.62	0.01	6.30	3.89	−2.40
PArel	−0.22	0.58	0.83	0.13	3.66	1.08	−2.59
PCONF21	0.15	0.27	0.36	0.17	1.15	0.86	−0.28
PNICO23	−0.70	0.70	0.80	0.16	1.74	−0.31	−2.05
PX13	1.15	1.15	1.27	0.03	1.79	2.18	0.39
RC21	−6.33	7.04	8.04	0.20	17.66	4.82	−12.84
RG18	−0.19	0.25	0.34	0.43	1.36	0.58	−0.77
RSE43	4.01	4.01	7.11	0.53	29.66	30.25	0.58
S22	−0.52	0.71	0.96	0.10	2.87	0.72	−2.15
S66	−0.58	0.62	0.75	0.11	2.24	0.42	−1.82
SCONF	−0.11	0.33	0.40	0.07	1.39	0.90	−0.49
TAUT15	−0.31	0.69	0.91	0.23	2.92	0.99	−1.94
UPU23	0.42	0.50	0.63	0.09	1.60	1.13	−0.47
W4-11	0.48	4.09	6.56	0.01	49.07	15.03	−34.04
WATER27	−6.14	6.14	9.19	0.08	22.52	−0.45	−22.97
WCPT18	1.42	1.42	1.54	0.04	1.92	2.28	0.36
YBDE18	3.18	3.18	3.64	0.06	7.29	8.55	1.26
BH76RC	0.07	1.45	2.22	0.07	11.80	9.31	−2.49

Table S120: Statistical analysis of SOS-MP2 for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	−2.56	2.57	3.13	0.08	5.00	0.08	−4.92
ACONF	−0.08	0.15	0.19	0.08	0.56	0.17	−0.39
ADIM6	−1.65	1.65	1.82	0.49	2.24	−0.59	−2.83
AHB21	1.31	1.31	1.34	0.06	1.15	1.85	0.70
AL2X6	−4.18	4.18	4.24	0.12	2.12	−3.25	−5.37
ALK8	−2.25	3.81	4.94	0.06	13.61	6.18	−7.44
ALKBDE10	−1.17	2.66	3.22	0.03	10.64	4.02	−6.62
AMINO20x4	−0.27	0.40	0.54	0.16	2.19	0.45	−1.74
BH76	6.14	6.17	8.47	0.33	31.32	30.61	−0.71
BHDIV10	2.98	3.06	3.25	0.07	4.51	4.11	−0.39
BHPERI	2.64	2.64	2.91	0.13	5.77	6.00	0.23
BHROT27	−0.18	0.25	0.34	0.04	0.94	0.28	−0.66
BSR36	−3.78	3.78	4.06	0.23	6.55	−1.49	−8.04
BUT14DIOL	−0.54	0.55	0.59	0.20	1.11	0.24	−0.87
C60ISO	−9.17	9.17	10.69	0.09	17.43	−1.59	−19.02
CARBHB12	−0.86	0.86	1.04	0.14	1.93	−0.31	−2.23
CDIE20	−0.13	0.26	0.33	0.06	1.44	0.82	−0.62
CHB6	−0.35	0.45	0.72	0.02	1.89	0.26	−1.63
DARC	2.83	2.83	2.91	0.09	2.56	4.46	1.90
DC13	0.64	4.82	5.75	0.09	18.02	9.35	−8.67
DIPCS10	−6.34	6.34	6.92	0.01	9.95	−0.05	−10.00
FH51	0.84	1.82	2.47	0.06	13.64	5.07	−8.57
G21EA	−2.85	5.21	7.90	0.15	45.99	24.94	−21.05
G21IP	−1.18	3.87	6.37	0.02	41.63	27.13	−14.50
G2RC	1.30	2.60	3.48	0.05	14.69	10.17	−4.52
HAL59	−0.90	0.90	1.05	0.20	2.67	−0.14	−2.81
HEAVY28	−0.51	0.51	0.53	0.41	0.68	−0.12	−0.80
HEAVYSB11	−0.32	1.46	1.79	0.03	5.47	2.12	−3.35
ICONF	−0.20	0.29	0.40	0.09	1.33	0.38	−0.94
IDISP	0.32	1.47	1.84	0.10	5.91	2.47	−3.44
IL16	2.45	2.45	2.47	0.02	1.31	3.34	2.03
INV24	−0.83	0.96	1.17	0.03	3.77	1.20	−2.57
ISO34	−0.21	0.62	0.85	0.04	4.54	2.42	−2.12
ISOL24	−0.93	1.82	2.78	0.08	13.61	2.88	−10.73
MB16-43	−32.54	32.56	36.99	0.08	75.75	0.43	−75.31
MCONF	−0.25	0.28	0.35	0.06	1.00	0.30	−0.69
NBPRC	2.23	2.67	3.28	0.10	8.48	6.92	−1.56
PA26	2.14	2.14	2.45	0.01	4.88	5.45	0.57
PArel	−0.38	0.78	0.95	0.17	3.62	1.23	−2.40
PCONF21	0.12	0.48	0.58	0.30	1.98	1.30	−0.68
PNICO23	−1.18	1.18	1.35	0.28	3.13	−0.48	−3.61
PX13	3.08	3.08	3.22	0.09	2.99	4.74	1.75
RC21	−7.03	7.46	8.46	0.21	17.50	4.49	−13.01
RG18	−0.27	0.31	0.39	0.53	1.23	0.36	−0.87
RSE43	4.24	4.24	7.33	0.56	31.20	31.62	0.43
S22	−1.24	1.25	1.57	0.17	3.15	0.08	−3.08
S66	−1.15	1.15	1.29	0.21	2.41	−0.33	−2.74
SCONF	−0.35	0.72	0.81	0.16	2.62	1.54	−1.07
TAUT15	−0.79	1.22	1.59	0.40	4.65	1.24	−3.41
UPU23	0.74	0.83	1.05	0.15	2.40	1.99	−0.42
W4-11	−0.95	4.28	7.25	0.01	53.52	13.85	−39.67
WATER27	−9.29	9.37	13.80	0.12	35.09	1.13	−33.96
WCPT18	3.40	3.40	3.56	0.10	3.50	5.16	1.66
YBDE18	1.55	1.58	2.11	0.03	5.25	5.01	−0.24
BH76RC	0.08	1.47	2.19	0.07	11.99	8.87	−3.13

Table S121: Statistical analysis of MP2-D3(BJ) for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	−1.35	1.56	2.33	0.05	5.76	0.75	−5.01
ACONF	−0.36	0.36	0.39	0.20	0.57	0.00	−0.57
ADIM6	−0.80	0.80	0.88	0.24	1.05	−0.29	−1.33
AHB21	−0.30	0.39	0.66	0.02	2.99	0.34	−2.65
AL2X6	−1.13	1.13	1.29	0.03	1.98	−0.42	−2.40
ALK8	0.77	3.45	5.84	0.06	20.41	15.25	−5.16
ALKBDE10	5.37	6.32	8.07	0.06	17.71	14.56	−3.15
AMINO20x4	−0.08	0.21	0.27	0.09	1.20	0.54	−0.66
BH76	3.86	4.50	6.91	0.24	34.07	28.02	−6.05
BHDIV10	−1.19	2.17	2.93	0.05	10.51	3.07	−7.45
BHPERI	−7.17	7.17	7.54	0.34	10.59	−1.06	−11.64
BHROT27	0.17	0.25	0.30	0.04	1.16	0.66	−0.50
BSR36	1.82	1.88	2.57	0.12	8.17	7.62	−0.55
BUT14DIOL	0.02	0.09	0.12	0.03	0.59	0.34	−0.25
C60ISO	−18.32	18.32	22.10	0.19	35.27	−4.01	−39.28
CARBHB12	0.36	0.42	0.59	0.07	1.57	1.29	−0.28
CDIE20	0.52	0.52	0.68	0.13	1.83	1.88	0.05
CHB6	−0.87	0.89	1.21	0.03	2.35	0.04	−2.31
DARC	−2.71	2.71	3.17	0.08	4.25	−0.42	−4.66
DC13	2.42	7.96	10.59	0.14	35.37	22.87	−12.50
DIPCS10	−4.70	5.05	6.30	0.01	13.22	1.30	−11.93
FH51	−1.01	2.53	3.56	0.08	18.07	4.98	−13.10
G21EA	−0.87	3.80	6.44	0.11	41.20	21.19	−20.00
G21IP	−0.46	3.21	5.01	0.01	31.71	20.40	−11.31
G2RC	−1.14	3.07	3.86	0.06	13.15	4.25	−8.91
HAL59	0.32	0.42	0.63	0.09	3.24	2.45	−0.79
HEAVY28	0.08	0.16	0.21	0.13	0.91	0.48	−0.44
HEAVYSB11	3.28	3.28	3.53	0.06	3.67	5.33	1.66
ICONF	0.02	0.24	0.35	0.07	1.54	0.42	−1.12
IDISP	−0.82	4.52	5.37	0.32	16.39	8.58	−7.81
IL16	0.13	0.42	0.49	0.00	1.71	0.93	−0.79
INV24	−0.98	1.11	1.32	0.03	4.01	1.20	−2.80
ISO34	0.59	1.11	1.65	0.08	7.65	5.57	−2.09
ISOL24	1.60	2.50	3.09	0.11	11.99	7.01	−4.97
MB16-43	19.09	20.39	23.42	0.05	65.94	44.36	−21.57
MCONF	0.55	0.63	0.71	0.13	1.70	1.22	−0.49
NBPRC	−0.34	0.88	1.05	0.03	3.43	1.70	−1.73
PA26	−1.74	1.91	3.07	0.01	10.87	1.38	−9.48
PArel	0.10	0.92	1.36	0.20	6.81	3.86	−2.95
PCONF21	0.16	0.40	0.50	0.25	1.80	1.18	−0.62
PNICO23	−0.01	0.16	0.23	0.04	1.04	0.83	−0.21
PX13	−2.64	2.64	2.75	0.08	2.77	−1.36	−4.13
RC21	−5.17	6.87	8.09	0.19	21.43	6.62	−14.81
RG18	−0.12	0.22	0.32	0.38	1.50	0.75	−0.75
RSE43	3.50	3.50	6.69	0.46	27.01	27.44	0.43
S22	0.21	0.55	0.81	0.08	3.11	2.10	−1.01
S66	−0.09	0.33	0.44	0.06	2.04	1.06	−0.98
SCONF	0.37	0.38	0.43	0.08	0.83	0.72	−0.11
TAUT15	0.68	0.78	0.97	0.26	2.41	1.76	−0.64
UPU23	0.16	0.32	0.42	0.06	1.34	0.81	−0.53
W4-11	3.22	7.51	9.48	0.02	46.19	23.34	−22.85
WATER27	−1.29	1.49	2.50	0.02	7.51	0.55	−6.96
WCPT18	−2.42	2.42	2.52	0.07	2.80	−1.27	−4.07
YBDE18	6.08	6.08	7.17	0.12	14.97	15.30	0.32
BH76RC	0.07	3.02	4.12	0.14	19.55	11.49	−8.05

Table S122: Statistical analysis of the alternative MP2-D3(BJ) variant for all 55 test sets of the GMTKN55 database where damping parameters were taken from Ref. S12. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	−1.29	1.54	2.32	0.05	5.84	0.84	−5.00
ACONF	−0.24	0.25	0.29	0.14	0.54	0.06	−0.48
ADIM6	0.01	0.06	0.07	0.02	0.17	0.09	−0.07
AHB21	−0.10	0.29	0.39	0.01	1.87	0.82	−1.04
AL2X6	−53.15	53.15	53.54	1.48	19.08	−43.22	−62.31
ALK8	−44.33	47.68	62.43	0.76	149.12	13.42	−135.70
ALKBDE10	0.20	8.72	13.42	0.09	50.16	14.28	−35.87
AMINO20x4	−0.09	0.21	0.27	0.09	1.19	0.40	−0.78
BH76	3.90	4.54	6.91	0.24	33.84	27.99	−5.85
BHDIV10	−0.14	2.28	2.89	0.05	10.52	4.38	−6.14
BHPERI	−6.24	6.24	6.52	0.30	9.10	−0.94	−10.04
BHROT27	0.13	0.27	0.35	0.04	1.75	0.65	−1.09
BSR36	0.31	0.47	0.73	0.03	3.37	2.51	−0.87
BUT14DIOL	−0.04	0.10	0.12	0.04	0.48	0.22	−0.26
C60ISO	−18.38	18.38	22.11	0.19	34.99	−4.19	−39.18
CARBHB12	0.12	0.31	0.39	0.05	1.42	0.85	−0.57
CDIE20	0.48	0.48	0.65	0.12	1.91	1.87	−0.04
CHB6	10.17	10.17	13.68	0.38	24.35	25.03	0.68
DARC	−1.52	1.97	2.30	0.06	4.76	0.89	−3.87
DC13	6.26	9.29	14.85	0.17	53.17	44.93	−8.24
DIPCS10	−4.70	5.05	6.30	0.01	13.22	1.30	−11.93
FH51	−1.13	2.46	3.51	0.08	17.36	4.48	−12.88
G21EA	−0.85	3.79	6.44	0.11	41.20	21.19	−20.00
G21IP	−0.45	3.21	5.00	0.01	31.71	20.40	−11.31
G2RC	−2.72	7.41	13.18	0.14	71.90	40.45	−31.45
HAL59	−2.57	2.65	3.49	0.58	14.03	0.87	−13.16
HEAVY28	−2.94	2.94	3.58	2.37	9.12	−0.48	−9.60
HEAVYSB11	−68.86	68.86	90.46	1.19	178.63	−0.63	−179.26
ICONF	0.19	0.91	1.28	0.28	5.59	3.35	−2.24
IDISP	−0.25	3.38	3.64	0.24	10.22	5.11	−5.10
IL16	0.51	0.54	0.80	0.00	2.80	2.60	−0.20
INV24	−1.24	1.34	1.54	0.04	4.10	1.20	−2.90
ISO34	0.54	1.11	1.66	0.08	8.16	5.98	−2.18
ISOL24	1.17	2.29	2.75	0.10	10.23	4.77	−5.46
MB16-43	−158.81	158.95	206.87	0.38	621.50	3.00	−618.50
MCONF	0.57	0.59	0.66	0.12	1.38	1.19	−0.19
NBPRC	0.89	2.62	4.41	0.09	19.63	12.27	−7.36
PA26	−0.84	2.65	5.25	0.01	31.41	22.04	−9.36
PArel	−0.10	1.01	1.58	0.22	8.60	3.65	−4.94
PCONF21	−0.07	0.38	0.45	0.23	1.59	0.69	−0.90
PNICO23	−2.75	2.76	3.78	0.65	9.96	0.03	−9.93
PX13	−2.69	2.69	2.80	0.08	2.88	−1.40	−4.28
RC21	−5.29	6.88	8.08	0.19	20.30	5.83	−14.46
RG18	−0.38	0.39	0.61	0.67	2.00	0.06	−1.94
RSE43	3.60	3.60	6.74	0.47	27.00	27.47	0.47
S22	0.24	0.46	0.59	0.06	1.97	1.40	−0.56
S66	0.10	0.22	0.28	0.04	1.12	0.81	−0.31
SCONF	0.38	0.38	0.42	0.08	0.60	0.62	0.02
TAUT15	0.65	0.76	0.94	0.25	2.36	1.71	−0.66
UPU23	−0.16	0.43	0.52	0.08	1.66	0.62	−1.04
W4-11	1.00	8.57	12.30	0.03	92.48	22.27	−70.21
WATER27	0.29	0.65	1.02	0.01	6.40	2.73	−3.67
WCPT18	−2.41	2.41	2.50	0.07	2.90	−1.28	−4.17
YBDE18	3.45	3.45	4.88	0.07	14.41	14.79	0.38
BH76RC	0.02	3.04	4.12	0.14	19.52	11.47	−8.05

Table S123: Statistical analysis of SCS-MP2-D3(BJ) for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	−1.30	2.35	2.54	0.07	7.17	3.97	−3.20
ACONF	−0.42	0.42	0.44	0.23	0.64	−0.03	−0.66
ADIM6	0.04	0.06	0.07	0.02	0.16	0.13	−0.03
AHB21	−0.46	0.51	0.81	0.02	3.03	0.24	−2.80
AL2X6	2.24	2.24	2.31	0.06	1.63	3.28	1.65
ALK8	4.00	4.67	7.15	0.07	16.80	14.79	−2.01
ALKBDE10	4.86	5.13	6.70	0.05	14.75	13.42	−1.33
AMINO20x4	−0.08	0.18	0.23	0.07	1.02	0.45	−0.57
BH76	4.15	4.43	6.92	0.24	31.16	29.23	−1.93
BHDIV10	0.45	1.48	1.91	0.03	6.40	3.46	−2.93
BHPERI	−1.89	2.02	2.42	0.10	5.28	1.33	−3.95
BHROT27	0.12	0.20	0.25	0.03	0.93	0.56	−0.37
BSR36	0.32	0.59	0.96	0.04	3.91	2.92	−1.00
BUT14DIOL	0.04	0.09	0.12	0.03	0.58	0.34	−0.24
C60ISO	−8.00	8.16	10.50	0.08	21.39	0.73	−20.65
CARBHB12	0.24	0.29	0.39	0.05	1.07	0.90	−0.16
CDIE20	0.21	0.53	0.62	0.13	2.20	1.32	−0.88
CHB6	2.74	4.26	5.20	0.16	10.75	8.74	−2.01
DARC	0.12	0.75	0.95	0.02	3.00	1.39	−1.61
DC13	0.64	6.75	8.82	0.12	34.80	13.95	−20.85
DIPCS10	−5.46	5.46	5.98	0.01	5.98	−2.53	−8.51
FH51	−0.94	1.45	2.21	0.05	10.03	3.14	−6.89
G21EA	−2.53	4.77	7.31	0.14	44.37	23.65	−20.72
G21IP	−0.97	3.44	5.66	0.01	36.87	24.43	−12.43
G2RC	−1.63	2.27	2.88	0.04	9.82	2.48	−7.34
HAL59	0.07	0.36	0.53	0.08	3.09	1.89	−1.20
HEAVY28	0.02	0.13	0.18	0.10	0.83	0.33	−0.49
HEAVYSB11	4.71	4.71	4.95	0.08	5.09	7.60	2.51
ICONF	0.13	0.24	0.34	0.07	1.42	0.65	−0.77
IDISP	−0.07	3.44	3.57	0.24	8.94	4.53	−4.41
IL16	−0.28	0.51	0.60	0.00	2.19	1.52	−0.67
INV24	0.01	1.64	2.25	0.05	11.09	7.69	−3.39
ISO34	0.38	0.59	0.90	0.04	3.95	3.31	−0.64
ISOL24	0.83	1.76	2.55	0.08	11.00	7.60	−3.40
MB16-43	2.44	13.12	17.83	0.03	100.87	39.21	−61.66
MCONF	0.65	0.70	0.79	0.14	1.54	1.22	−0.32
NBPRC	−1.54	2.24	2.68	0.08	7.82	2.37	−5.45
PA26	3.74	3.83	4.23	0.02	9.58	8.46	−1.13
PArel	−0.17	1.22	1.88	0.26	8.92	4.42	−4.51
PCONF21	0.08	0.51	0.65	0.31	2.41	1.35	−1.06
PNICO23	−0.05	0.12	0.15	0.03	0.67	0.46	−0.20
PX13	−3.27	3.27	3.41	0.10	3.51	−1.36	−4.87
RC21	−4.46	6.21	7.20	0.17	21.86	11.11	−10.75
RG18	−0.06	0.20	0.30	0.34	1.42	0.88	−0.54
RSE43	3.52	3.52	6.92	0.46	30.74	30.81	0.08
S22	0.54	0.60	0.88	0.08	2.46	2.16	−0.30
S66	0.29	0.31	0.45	0.06	1.50	1.33	−0.17
SCONF	0.22	0.23	0.25	0.05	0.46	0.39	−0.07
TAUT15	0.51	0.72	0.92	0.24	2.58	2.04	−0.54
UPU23	0.15	0.42	0.54	0.07	2.31	0.90	−1.41
W4-11	11.72	13.11	15.27	0.04	62.44	33.43	−29.01
WATER27	0.31	0.90	1.31	0.01	6.89	1.92	−4.96
WCPT18	−0.73	1.24	1.49	0.04	4.80	1.89	−2.91
YBDE18	8.79	8.79	8.93	0.18	5.76	11.62	5.87
BH76RC	0.23	1.75	2.17	0.08	9.54	6.25	−3.29

Table S124: Statistical analysis of SOS-MP2-D3(BJ) for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	−1.73	2.59	2.85	0.08	6.98	2.49	−4.49
ACONF	−0.45	0.45	0.47	0.25	0.66	−0.04	−0.71
ADIM6	0.17	0.17	0.19	0.05	0.20	0.28	0.09
AHB21	−0.35	0.39	0.66	0.02	2.59	0.14	−2.45
AL2X6	6.43	6.43	6.58	0.18	4.67	8.45	3.78
ALK8	8.16	8.76	14.32	0.14	38.96	36.75	−2.21
ALKBDE10	6.20	6.20	7.91	0.06	12.67	13.66	0.99
AMINO20x4	−0.07	0.17	0.22	0.07	1.04	0.36	−0.68
BH76	4.53	4.70	7.20	0.25	31.39	29.19	−2.20
BHDIV10	0.95	1.58	2.15	0.03	6.51	4.79	−1.72
BHPERI	−0.13	1.56	1.76	0.07	6.29	3.50	−2.80
BHROT27	0.06	0.22	0.36	0.04	1.64	1.31	−0.34
BSR36	−0.61	0.68	0.83	0.04	2.44	0.85	−1.59
BUT14DIOL	0.06	0.10	0.13	0.04	0.61	0.37	−0.24
C60ISO	−3.37	4.27	5.60	0.04	14.79	2.75	−12.04
CARBHB12	0.18	0.24	0.31	0.04	0.93	0.74	−0.19
CDIE20	0.21	0.63	0.75	0.16	2.42	1.47	−0.95
CHB6	2.65	5.27	6.16	0.20	14.83	11.11	−3.72
DARC	−1.75	1.75	2.14	0.05	3.62	−0.76	−4.38
DC13	−2.88	7.06	12.25	0.13	45.09	7.63	−37.46
DIPCS10	−5.97	5.97	6.44	0.01	8.79	−1.02	−9.81
FH51	−0.85	1.37	2.09	0.04	10.62	2.91	−7.71
G21EA	−3.38	5.64	8.22	0.17	45.97	24.91	−21.06
G21IP	−1.26	3.89	6.38	0.02	41.05	26.40	−14.65
G2RC	−1.09	2.48	3.67	0.05	16.97	5.35	−11.62
HAL59	0.04	0.40	0.57	0.09	3.24	1.64	−1.60
HEAVY28	0.05	0.17	0.22	0.14	1.03	0.48	−0.55
HEAVYSB11	8.59	8.59	8.88	0.15	6.45	11.97	5.52
ICONF	0.29	0.39	0.58	0.12	1.75	1.41	−0.34
IDISP	0.29	3.77	3.99	0.27	10.04	5.88	−4.17
IL16	−0.42	0.64	0.73	0.01	2.60	1.62	−0.98
INV24	0.37	2.26	3.26	0.07	15.75	11.18	−4.56
ISO34	0.30	0.56	0.72	0.04	2.41	1.72	−0.69
ISOL24	1.21	1.77	2.57	0.08	12.46	8.88	−3.58
MB16-43	10.65	19.70	24.22	0.05	114.62	55.76	−58.87
MCONF	0.58	0.62	0.71	0.12	1.34	1.06	−0.28
NBPRC	−3.21	3.73	4.49	0.13	12.55	2.45	−10.10
PA26	4.81	4.81	5.23	0.03	10.57	10.91	0.34
PArel	−0.34	1.53	2.66	0.33	13.80	6.44	−7.36
PCONF21	0.03	0.55	0.66	0.34	2.36	1.23	−1.12
PNICO23	0.05	0.16	0.32	0.04	1.59	1.42	−0.17
PX13	−1.32	1.32	1.42	0.04	1.63	−0.49	−2.11
RC21	−3.14	5.57	6.44	0.16	23.05	13.84	−9.20
RG18	−0.04	0.19	0.30	0.33	1.42	0.89	−0.53
RSE43	3.36	3.44	6.95	0.45	32.77	31.80	−0.96
S22	0.54	0.57	0.77	0.08	1.94	1.74	−0.20
S66	0.33	0.33	0.43	0.06	1.17	1.11	−0.06
SCONF	0.23	0.25	0.27	0.05	0.62	0.47	−0.15
TAUT15	0.40	0.76	1.05	0.25	3.80	2.77	−1.03
UPU23	0.28	0.47	0.58	0.08	2.27	1.00	−1.28
W4-11	13.26	14.96	17.85	0.05	93.16	58.98	−34.18
WATER27	1.13	1.37	1.64	0.02	7.17	3.89	−3.27
WCPT18	0.19	1.04	1.29	0.03	4.74	2.77	−1.97
YBDE18	12.05	12.05	12.33	0.24	8.92	15.73	6.81
BH76RC	0.66	1.37	1.83	0.06	9.28	6.11	−3.17

Table S125: Statistical analysis of MP2-D3(0) for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	−1.34	1.55	2.33	0.05	5.69	0.65	−5.03
ACONF	−0.39	0.39	0.41	0.21	0.59	−0.01	−0.59
ADIM6	−0.29	0.29	0.32	0.09	0.41	−0.06	−0.48
AHB21	−0.30	0.39	0.67	0.02	3.09	0.43	−2.66
AL2X6	−0.99	0.99	1.20	0.03	2.02	−0.38	−2.40
ALK8	0.80	3.25	5.53	0.05	19.51	14.42	−5.09
ALKBDE10	5.39	6.33	8.09	0.06	17.73	14.59	−3.14
AMINO20x4	−0.09	0.23	0.29	0.09	1.31	0.61	−0.70
BH76	3.86	4.50	6.91	0.24	34.11	28.02	−6.09
BHDIV10	−1.19	2.19	2.92	0.05	10.50	3.09	−7.40
BHPERI	−7.21	7.21	7.59	0.35	10.78	−1.10	−11.88
BHROT27	0.16	0.24	0.30	0.04	1.21	0.67	−0.54
BSR36	2.23	2.27	3.07	0.14	9.35	8.97	−0.38
BUT14DIOL	0.03	0.10	0.12	0.04	0.61	0.35	−0.26
C60ISO	−18.19	18.19	21.99	0.19	35.24	−3.90	−39.14
CARBHB12	0.40	0.43	0.60	0.07	1.43	1.29	−0.14
CDIE20	0.54	0.54	0.70	0.13	1.86	1.91	0.05
CHB6	−0.80	0.83	1.12	0.03	2.27	0.09	−2.18
DARC	−2.90	2.90	3.33	0.09	4.23	−0.58	−4.81
DC13	2.33	8.02	10.56	0.15	34.99	22.31	−12.68
DIPCS10	−4.70	5.04	6.29	0.01	13.24	1.31	−11.93
FH51	−1.01	2.55	3.57	0.08	18.10	5.00	−13.10
G21EA	−0.87	3.80	6.44	0.11	41.20	21.19	−20.00
G21IP	−0.46	3.21	5.01	0.01	31.71	20.40	−11.31
G2RC	−1.16	3.08	3.87	0.06	13.15	4.25	−8.90
HAL59	0.35	0.45	0.70	0.10	3.24	2.44	−0.80
HEAVY28	0.04	0.14	0.19	0.11	0.83	0.43	−0.41
HEAVYSB11	3.40	3.40	3.65	0.06	3.81	5.45	1.64
ICONF	0.02	0.23	0.33	0.07	1.46	0.46	−1.00
IDISP	−0.84	5.30	6.20	0.37	19.10	9.31	−9.79
IL16	0.09	0.40	0.47	0.00	1.60	0.79	−0.81
INV24	−0.94	1.08	1.28	0.03	3.81	1.21	−2.60
ISO34	0.60	1.11	1.64	0.08	7.63	5.58	−2.05
ISOL24	1.65	2.54	3.23	0.12	13.30	8.35	−4.96
MB16-43	19.28	20.54	23.47	0.05	65.31	44.47	−20.85
MCONF	0.75	0.83	0.95	0.17	2.12	1.61	−0.51
NBPRC	−0.36	0.94	1.14	0.03	3.71	1.87	−1.85
PA26	−1.72	1.90	3.06	0.01	10.84	1.37	−9.47
PArel	0.09	0.93	1.37	0.20	6.78	3.87	−2.90
PCONF21	0.15	0.55	0.69	0.34	2.60	1.54	−1.05
PNICO23	0.07	0.17	0.26	0.04	1.15	0.94	−0.21
PX13	−2.67	2.67	2.79	0.08	2.94	−1.35	−4.28
RC21	−5.14	6.87	8.09	0.19	21.61	6.83	−14.78
RG18	−0.11	0.22	0.32	0.38	1.53	0.82	−0.72
RSE43	3.47	3.47	6.68	0.46	27.02	27.42	0.40
S22	0.50	0.73	1.11	0.10	3.64	2.95	−0.69
S66	0.18	0.36	0.54	0.07	2.15	1.72	−0.44
SCONF	0.34	0.35	0.39	0.08	0.78	0.69	−0.09
TAUT15	0.71	0.81	1.00	0.27	2.47	1.82	−0.65
UPU23	−0.05	0.38	0.51	0.07	2.27	0.77	−1.50
W4-11	3.28	7.52	9.50	0.02	46.25	23.42	−22.83
WATER27	−1.04	1.28	2.04	0.02	6.25	0.58	−5.67
WCPT18	−2.43	2.43	2.52	0.07	2.81	−1.25	−4.06
YBDE18	6.12	6.12	7.19	0.12	14.87	15.22	0.35
BH76RC	0.07	3.02	4.12	0.14	19.56	11.50	−8.05

Table S126: Statistical analysis of SCS-MP2-D3(0) for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	−1.47	2.22	2.41	0.07	5.84	2.58	−3.26
ACONF	−0.49	0.49	0.51	0.27	0.66	−0.05	−0.71
ADIM6	0.01	0.05	0.06	0.01	0.18	0.11	−0.06
AHB21	−0.37	0.49	0.79	0.02	2.83	0.38	−2.45
AL2X6	1.19	1.19	1.27	0.03	1.29	1.70	0.41
ALK8	2.51	3.80	5.56	0.06	15.65	11.69	−3.95
ALKBDE10	3.20	3.66	4.87	0.04	11.68	10.09	−1.60
AMINO20x4	−0.15	0.23	0.31	0.09	1.27	0.39	−0.88
BH76	3.74	4.00	6.68	0.21	30.76	28.27	−2.50
BHDIV10	−0.40	1.12	1.42	0.02	4.09	1.39	−2.70
BHPERI	−2.93	3.08	3.38	0.15	7.33	1.99	−5.34
BHROT27	−0.12	0.25	0.32	0.04	1.07	0.37	−0.70
BSR36	0.31	0.58	0.91	0.04	3.75	2.65	−1.11
BUT14DIOL	0.08	0.10	0.14	0.04	0.56	0.41	−0.15
C60ISO	−11.98	11.98	14.26	0.12	23.45	−2.12	−25.57
CARBHB12	0.15	0.21	0.30	0.03	0.99	0.75	−0.24
CDIE20	0.00	0.20	0.33	0.05	1.53	1.17	−0.35
CHB6	−0.63	1.02	1.14	0.04	2.70	1.00	−1.70
DARC	−4.88	4.88	4.94	0.15	2.88	−2.95	−5.83
DC13	−1.58	7.91	10.03	0.14	36.04	18.61	−17.43
DIPCS10	−6.20	6.20	6.53	0.01	6.43	−2.19	−8.62
FH51	−1.05	1.44	2.13	0.05	13.83	4.72	−9.11
G21EA	−2.10	4.47	7.12	0.13	44.40	23.73	−20.68
G21IP	−0.99	3.43	5.70	0.01	36.93	25.36	−11.57
G2RC	−1.12	1.81	2.35	0.04	9.78	4.92	−4.85
HAL59	0.02	0.36	0.53	0.08	3.15	1.88	−1.27
HEAVY28	0.00	0.13	0.16	0.10	0.76	0.28	−0.48
HEAVYSB11	3.58	3.58	3.83	0.06	5.04	6.16	1.12
ICONF	0.00	0.22	0.31	0.07	1.36	0.48	−0.88
IDISP	−1.18	5.26	6.11	0.37	18.07	6.95	−11.12
IL16	−0.10	0.36	0.46	0.00	1.89	1.36	−0.52
INV24	−0.64	0.87	1.13	0.03	4.39	1.65	−2.73
ISO34	−0.04	1.09	1.39	0.07	5.38	2.83	−2.54
ISOL24	1.65	2.42	3.24	0.11	13.32	9.39	−3.93
MB16-43	2.27	11.64	14.87	0.03	80.53	32.66	−47.87
MCONF	0.60	0.69	0.77	0.14	1.82	1.31	−0.51
NBPRC	−1.33	2.10	3.02	0.08	10.24	2.10	−8.14
PA26	2.19	2.32	2.63	0.01	5.79	4.74	−1.05
PArel	−0.30	1.09	1.69	0.24	7.66	2.51	−5.15
PCONF21	−0.07	0.68	0.83	0.42	2.89	1.38	−1.51
PNICO23	−0.17	0.18	0.20	0.04	0.36	0.03	−0.33
PX13	−9.10	9.10	9.90	0.27	12.81	−1.93	−14.74
RC21	−4.02	5.76	6.56	0.16	16.87	6.80	−10.07
RG18	−0.06	0.20	0.30	0.34	1.42	0.87	−0.55
RSE43	3.85	3.85	7.10	0.51	30.29	30.42	0.13
S22	0.49	0.60	0.88	0.08	2.72	2.22	−0.50
S66	0.24	0.29	0.43	0.05	1.62	1.34	−0.28
SCONF	0.26	0.39	0.42	0.08	1.19	0.55	−0.64
TAUT15	0.43	0.60	0.76	0.20	2.21	1.71	−0.50
UPU23	0.19	0.42	0.55	0.07	2.42	1.07	−1.35
W4-11	4.29	6.57	8.33	0.02	49.33	17.57	−31.76
WATER27	0.75	1.38	2.06	0.02	11.06	3.57	−7.48
WCPT18	−3.06	3.12	3.52	0.09	6.34	0.51	−5.83
YBDE18	7.12	7.12	7.30	0.14	5.79	10.70	4.91
BH76RC	0.13	1.74	2.35	0.08	11.94	8.08	−3.86

Table S127: Statistical analysis of SOS-MP2-D3(0) for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	−1.94	2.34	2.75	0.07	5.89	1.40	−4.49
ACONF	−0.52	0.52	0.54	0.28	0.70	−0.07	−0.76
ADIM6	0.19	0.19	0.21	0.06	0.21	0.31	0.10
AHB21	0.10	0.18	0.26	0.01	1.12	0.75	−0.36
AL2X6	0.21	0.96	1.20	0.03	3.53	2.02	−1.50
ALK8	2.40	3.68	5.34	0.06	12.78	10.60	−2.18
ALKBDE10	−0.40	2.12	2.79	0.02	9.95	4.51	−5.44
AMINO20x4	−0.14	0.22	0.29	0.09	1.24	0.34	−0.89
BH76	4.87	5.21	7.53	0.28	31.47	28.58	−2.89
BHDIV10	1.07	1.52	1.77	0.03	4.31	3.14	−1.17
BHPERI	−0.61	1.47	1.66	0.07	4.81	2.22	−2.59
BHROT27	−0.17	0.23	0.29	0.04	0.87	0.19	−0.68
BSR36	−0.56	0.64	0.77	0.04	2.41	1.03	−1.38
BUT14DIOL	0.06	0.09	0.13	0.03	0.57	0.39	−0.18
C60ISO	−8.66	8.66	10.28	0.09	17.72	−1.00	−18.72
CARBHB12	−0.01	0.15	0.19	0.02	0.73	0.39	−0.34
CDIE20	−0.23	0.30	0.37	0.07	1.30	0.66	−0.64
CHB6	−1.59	1.59	1.79	0.06	2.56	−0.33	−2.89
DARC	−2.59	2.59	2.74	0.08	3.02	−0.41	−3.43
DC13	−1.85	4.80	6.50	0.09	23.60	7.60	−16.00
DIPCS10	−6.02	6.24	6.69	0.01	10.86	1.10	−9.75
FH51	−0.18	1.11	1.75	0.04	11.60	2.76	−8.84
G21EA	−2.81	5.24	7.90	0.16	45.99	24.94	−21.05
G21IP	−1.16	3.87	6.37	0.02	41.64	27.21	−14.43
G2RC	0.34	1.20	1.58	0.02	7.36	3.65	−3.71
HAL59	0.01	0.33	0.48	0.07	2.81	1.70	−1.11
HEAVY28	−0.02	0.15	0.18	0.12	0.79	0.27	−0.52
HEAVYSB11	2.07	2.57	2.96	0.04	6.60	5.10	−1.49
ICONF	−0.19	0.26	0.44	0.08	1.79	0.19	−1.60
IDISP	−0.82	4.66	5.15	0.33	14.10	5.26	−8.84
IL16	−0.07	0.40	0.52	0.00	2.05	1.45	−0.60
INV24	−0.20	0.79	1.03	0.02	4.48	2.00	−2.47
ISO34	0.10	0.82	1.07	0.06	4.85	2.65	−2.20
ISOL24	1.17	1.78	2.28	0.08	8.21	6.08	−2.13
MB16-43	−9.36	15.97	20.14	0.04	92.11	26.76	−65.34
MCONF	0.58	0.65	0.73	0.13	1.61	1.18	−0.44
NBPRC	0.05	1.17	1.41	0.04	4.56	2.19	−2.37
PA26	3.11	3.11	3.32	0.02	5.17	5.79	0.62
PArel	−0.42	0.71	1.04	0.15	3.17	0.85	−2.33
PCONF21	−0.06	0.70	0.84	0.43	2.82	1.32	−1.50
PNICO23	−0.27	0.27	0.38	0.06	1.36	0.01	−1.35
PX13	1.72	1.72	2.10	0.05	4.00	4.14	0.14
RC21	−4.79	5.93	6.69	0.17	14.63	4.58	−10.04
RG18	−0.04	0.19	0.29	0.33	1.42	0.90	−0.52
RSE43	4.15	4.15	7.25	0.55	30.89	31.35	0.46
S22	0.50	0.56	0.79	0.08	2.23	1.86	−0.36
S66	0.29	0.30	0.42	0.05	1.34	1.17	−0.17
SCONF	0.18	0.33	0.36	0.07	1.15	0.50	−0.65
TAUT15	−0.42	0.95	1.13	0.31	3.13	1.04	−2.09
UPU23	0.29	0.48	0.61	0.08	2.53	1.24	−1.29
W4-11	0.65	4.77	7.37	0.02	53.55	15.32	−38.24
WATER27	0.63	0.85	1.06	0.01	5.36	2.84	−2.52
WCPT18	0.88	0.88	1.08	0.03	2.33	2.36	0.03
YBDE18	4.45	4.45	4.67	0.09	5.31	8.08	2.77
BH76RC	−0.01	1.50	2.10	0.07	11.37	8.12	−3.25

Table S128: Statistical analysis of SCS-MP2-D3(CSO) for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	−2.10	2.12	2.47	0.06	3.39	0.09	−3.30
ACONF	−0.31	0.31	0.33	0.17	0.50	0.01	−0.49
ADIM6	−0.52	0.52	0.59	0.15	0.73	−0.18	−0.92
AHB21	0.64	0.64	0.69	0.03	1.02	1.04	0.02
AL2X6	−2.17	2.17	2.19	0.06	0.86	−1.60	−2.46
ALK8	−0.80	3.47	4.87	0.06	16.75	10.31	−6.44
ALKBDE10	1.12	2.90	3.87	0.03	11.89	7.56	−4.33
AMINO20x4	−0.19	0.30	0.40	0.12	1.83	0.54	−1.28
BH76	5.29	5.40	7.74	0.29	31.15	29.52	−1.63
BHDIV10	1.45	1.50	1.74	0.03	3.11	2.82	−0.29
BHPERI	−1.22	1.59	1.85	0.08	6.24	2.62	−3.62
BHROT27	−0.06	0.18	0.24	0.03	0.99	0.39	−0.59
BSR36	−0.83	0.88	0.97	0.05	2.65	0.86	−1.79
BUT14DIOL	−0.29	0.30	0.33	0.11	0.76	0.18	−0.58
C60ISO	−12.13	12.13	14.40	0.12	23.46	−2.27	−25.72
CARBHB12	−0.27	0.28	0.37	0.05	0.88	0.05	−0.83
CDIE20	0.06	0.22	0.30	0.05	1.38	1.04	−0.34
CHB6	−0.74	0.74	1.04	0.03	2.19	−0.02	−2.21
DARC	−0.05	0.82	0.93	0.03	3.23	2.07	−1.17
DC13	0.87	4.37	5.47	0.08	17.80	9.61	−8.19
DIPCS10	−5.79	5.79	6.34	0.01	8.29	−0.63	−8.92
FH51	0.05	1.45	2.17	0.05	12.67	3.84	−8.83
G21EA	−2.19	4.48	7.15	0.13	44.39	23.69	−20.70
G21IP	−0.94	3.45	5.67	0.01	37.01	24.89	−12.12
G2RC	0.44	2.16	2.44	0.04	8.84	4.66	−4.18
HAL59	−0.18	0.40	0.55	0.09	3.05	1.44	−1.61
HEAVY28	−0.11	0.15	0.19	0.12	0.65	0.12	−0.52
HEAVYSB11	1.68	2.18	2.49	0.04	5.25	3.91	−1.33
ICONF	−0.13	0.23	0.34	0.07	1.36	0.32	−1.04
IDISP	−0.18	2.76	3.48	0.19	11.43	5.77	−5.66
IL16	1.21	1.21	1.23	0.01	1.01	1.92	0.91
INV24	−0.70	0.84	1.08	0.03	3.77	1.20	−2.56
ISO34	0.10	0.51	0.74	0.04	4.18	2.66	−1.52
ISOL24	0.26	0.78	1.07	0.04	4.63	2.86	−1.77
MB16-43	−10.48	13.22	16.81	0.03	57.30	15.80	−41.50
MCONF	0.41	0.51	0.57	0.10	1.71	1.10	−0.61
NBPRC	0.97	1.28	1.49	0.05	3.75	2.84	−0.91
PA26	0.92	1.30	1.63	0.01	6.19	3.94	−2.26
PArel	−0.22	0.59	0.83	0.13	3.68	1.14	−2.54
PCONF21	0.18	0.50	0.61	0.31	2.02	1.23	−0.80
PNICO23	−0.47	0.47	0.57	0.11	1.51	−0.12	−1.64
PX13	1.09	1.09	1.20	0.03	1.62	1.99	0.38
RC21	−6.12	6.91	7.87	0.19	17.39	4.82	−12.57
RG18	−0.12	0.21	0.32	0.36	1.46	0.78	−0.68
RSE43	4.02	4.02	7.12	0.53	29.69	30.29	0.60
S22	−0.12	0.69	0.93	0.09	3.50	1.67	−1.84
S66	−0.25	0.43	0.56	0.08	2.54	1.03	−1.51
SCONF	−0.09	0.26	0.31	0.06	1.05	0.66	−0.39
TAUT15	−0.31	0.70	0.91	0.23	2.88	0.98	−1.90
UPU23	0.25	0.39	0.52	0.07	1.82	1.13	−0.70
W4-11	0.66	4.14	6.58	0.01	49.28	15.29	−33.98
WATER27	−5.33	5.33	7.94	0.07	19.35	−0.33	−19.69
WCPT18	1.32	1.32	1.41	0.04	1.73	2.09	0.36
YBDE18	3.60	3.60	4.02	0.07	7.26	8.88	1.62
BH76RC	0.08	1.44	2.21	0.07	11.65	9.28	−2.37

Table S129: Statistical analysis of SOS-MP2-D3(CSO) for all 55 test sets of the GMTKN55 database. The statistical key data are: mean deviation (MD), mean absolute deviation (MAD), root-mean-square deviation (RMSD), normalised MAD (NMAD), deviation span (Δ_{error}), maximum (max) and minimum deviation (min). The NMAD is defined as the ratio between the MAD for a set and its average absolute reaction energy. For all test sets, the def2-QZVP atomic-orbital basis set was used, with additional diffuse functions for the G21EA, WATER27, AHB21, and IL16 sets. All values are in kcal/mol, except for the dimensionless NMAD.

set	MD	MAD	RMSD	NMAD	Δ_{error}	max	min
SIE4x4	−2.52	2.54	3.09	0.08	4.95	0.09	−4.86
ACONF	−0.17	0.18	0.23	0.10	0.48	0.05	−0.43
ADIM6	−1.15	1.15	1.28	0.34	1.59	−0.41	−2.00
AHB21	1.21	1.21	1.25	0.05	1.08	1.71	0.62
AL2X6	−3.39	3.39	3.42	0.09	1.37	−2.79	−4.16
ALK8	−1.94	3.84	4.98	0.06	14.35	7.15	−7.20
ALKBDE10	−1.07	2.59	3.13	0.03	10.35	4.14	−6.21
AMINO20x4	−0.26	0.38	0.51	0.16	2.11	0.44	−1.67
BH76	6.07	6.11	8.42	0.33	31.39	30.58	−0.81
BHDIV10	2.86	2.94	3.12	0.06	4.42	4.01	−0.41
BHPERI	2.21	2.21	2.51	0.11	5.43	5.69	0.26
BHROT27	−0.17	0.25	0.33	0.04	0.94	0.29	−0.65
BSR36	−3.04	3.04	3.22	0.19	4.74	−1.35	−6.09
BUT14DIOL	−0.49	0.51	0.54	0.18	1.01	0.20	−0.81
C60ISO	−9.10	9.10	10.64	0.09	17.48	−1.49	−18.98
CARBHB12	−0.74	0.74	0.90	0.12	1.74	−0.23	−1.97
CDIE20	−0.15	0.27	0.34	0.07	1.44	0.78	−0.66
CHB6	−0.55	0.57	0.87	0.02	2.02	0.06	−1.97
DARC	2.10	2.10	2.23	0.06	2.67	3.99	1.32
DC13	0.37	4.62	5.67	0.08	17.88	9.15	−8.73
DIPCS10	−6.33	6.33	6.91	0.01	9.95	−0.05	−9.99
FH51	0.72	1.65	2.30	0.05	13.41	4.86	−8.55
G21EA	−2.85	5.21	7.90	0.15	45.99	24.94	−21.05
G21IP	−1.18	3.87	6.37	0.02	41.63	27.13	−14.50
G2RC	1.27	2.48	3.31	0.05	13.73	9.43	−4.31
HAL59	−0.67	0.69	0.87	0.15	2.83	0.21	−2.62
HEAVY28	−0.35	0.35	0.39	0.28	0.59	−0.05	−0.64
HEAVYSB11	0.34	1.90	2.04	0.03	5.94	2.90	−3.04
ICONF	−0.21	0.28	0.40	0.09	1.23	0.34	−0.89
IDISP	0.22	1.36	1.55	0.10	4.38	2.52	−1.85
IL16	2.13	2.13	2.15	0.02	1.25	2.95	1.71
INV24	−0.70	0.85	1.08	0.03	3.77	1.20	−2.56
ISO34	−0.18	0.60	0.82	0.04	4.48	2.40	−2.08
ISOL24	−0.68	1.50	2.19	0.07	10.64	2.87	−7.77
MB16-43	−28.61	28.73	33.23	0.07	72.36	2.70	−69.66
MCONF	−0.01	0.22	0.26	0.04	1.04	0.42	−0.62
NBPRC	1.94	2.36	2.87	0.09	7.44	5.98	−1.47
PA26	2.19	2.19	2.49	0.01	4.81	5.39	0.59
PArel	−0.39	0.78	0.95	0.17	3.64	1.29	−2.35
PCONF21	0.16	0.33	0.43	0.20	1.33	1.04	−0.29
PNICO23	−0.94	0.94	1.12	0.22	2.89	−0.32	−3.20
PX13	3.02	3.02	3.15	0.09	2.90	4.63	1.73
RC21	−6.82	7.25	8.27	0.20	17.23	4.49	−12.74
RG18	−0.20	0.27	0.35	0.47	1.33	0.56	−0.78
RSE43	4.26	4.26	7.34	0.56	31.22	31.67	0.45
S22	−0.85	0.90	1.25	0.12	3.02	0.26	−2.76
S66	−0.81	0.82	0.97	0.15	2.54	0.12	−2.42
SCONF	−0.33	0.64	0.71	0.14	2.28	1.31	−0.97
TAUT15	−0.80	1.22	1.60	0.40	4.63	1.25	−3.38
UPU23	0.57	0.63	0.79	0.11	1.71	1.30	−0.41
W4-11	−0.77	4.26	7.23	0.01	53.62	14.01	−39.61
WATER27	−8.47	8.56	12.54	0.11	31.90	1.25	−30.65
WCPT18	3.30	3.30	3.43	0.09	3.27	4.92	1.66
YBDE18	1.96	1.96	2.37	0.04	4.88	5.34	0.45
BH76RC	0.08	1.45	2.17	0.07	11.84	8.84	−3.01

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