Supporting Information

Nonlocal Van der Waals Approach Merged with Double-Hybrid Density Functionals: Towards the Accurate Treatment of Noncovalent Interactions

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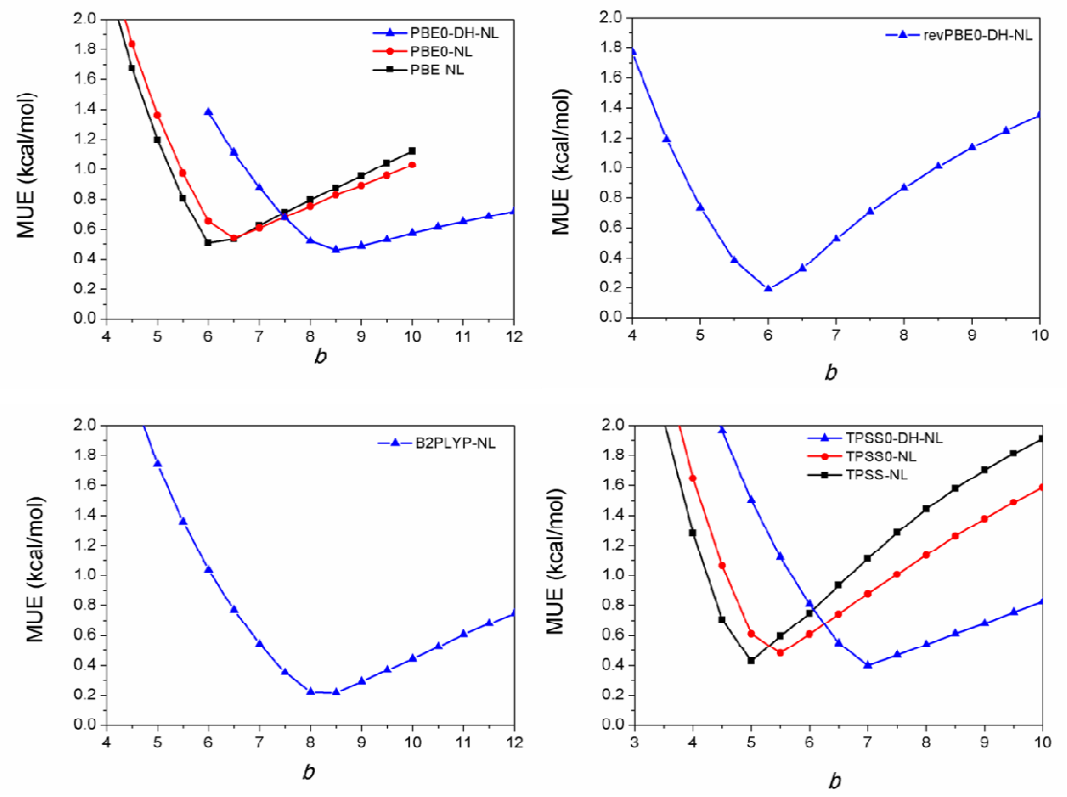

Figure S1. MUE values computed with the different DFT-NL functionals for the S22 dataset as a function of the attenuation parameter *b*.

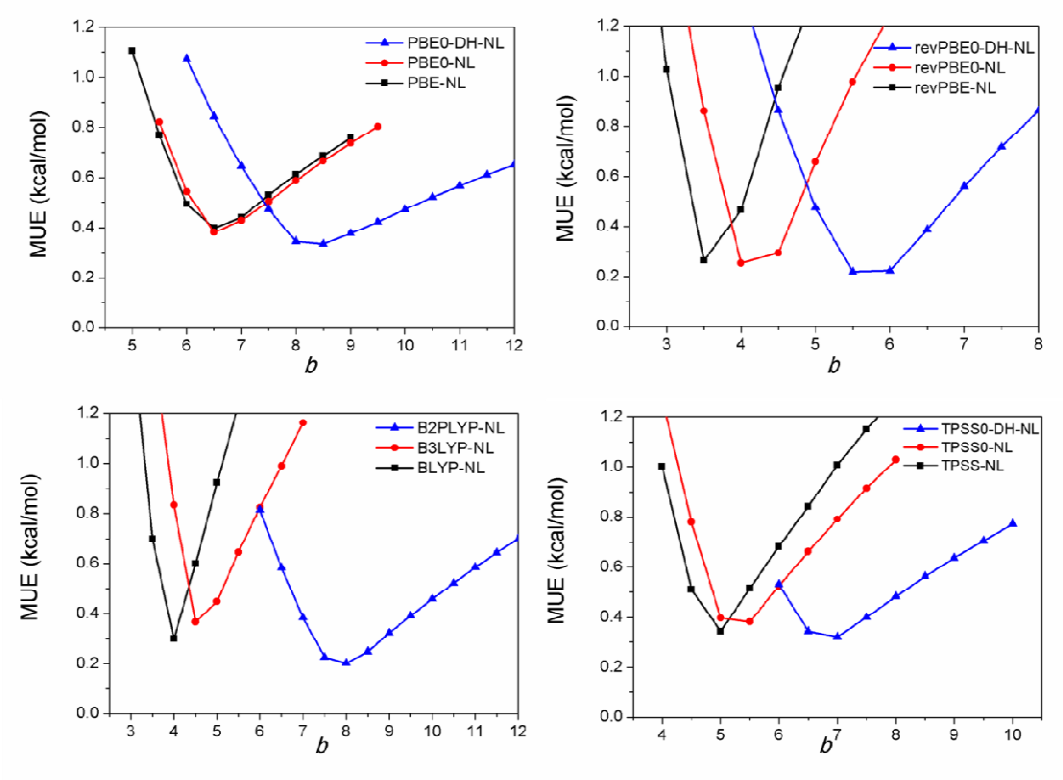

Figure S2. MUE values computed with the different DFT-NL functionals for the S66 dataset as a function of the attenuation parameter *b*.

Table S1. Interactions energies (kcal/mol) computed for the S22 dataset with the PBE0-DH-NL, PBE0-NL, and PBE-NL functionals at their optimum *b* value. The def2-QZVP basis set is employed for the calculations. ME, MUE and MURE errors are also included.

	Complexes	PBE0-DH-NL (b = 8.6)	PBE0-NL (b = 6.5)	PBE-NL (b = 6.2)
	Hydrogen Bonds			
1	Ammonia dimer, C_{2v}	-3.355	-3.511	-3.698
2	Water dimer, Cs	-5.527	-5.725	-5.902
3	Formic acid dimer, C_{2h}	-20.973	-21.182	-20.648
4	Formamide dimer, C_{2h}	-17.248	-17.422	-17.061
5	H-donded uracil dimer, C_{2h}	-21.785	-21.887	-21.395
6	2-pyridone 2-aminopyridine, C_1	-18.080	-18.343	-18.513
7	Adenine thymine WC, C_1	-17.510	-17.759	-17.819
	Dispersion			
8	Methane dimer, D_{3d}	-0.624	-0.631	-0.636
9	Ethene dimer, D_{2d}	-1.350	-1.445	-1.488
10	Benzene methane C_3	-1.495	-1.521	-1.515
11	Parallel displaced benzene dimer, C_{2h}	-2.454	-2.434	-2.641
12	Pyrazine dimer, C_s	-3.889	-3.789	-3.901
13	Stacked uracil dimer, C_2	-9.697	-9.743	-9.635
14	Stacked indole benzene, C_1	-4.023	-3.945	-4.270
15	Stacked adenina thymine, C_1	-11.466	-11.289	-11.170
	Mixed			
16	Ethene ethine, C_{2v}	-1.700	-1.782	-1.794
17	Benzene water, C_s	-3.574	-3.699	-3.766
18	Benzene ammonia, C_s	-2.425	-2.481	-2.493
19	Benzene hydrogen cyanide, C_s	-4.972	-4.890	-4.546
20	T-shaped benzene dimer, C_{2v}	-2.746	-2.724	-2.653
21	T-shaped indole benzene, C_1	-5.674	-5.614	-5.552
22	Phenol dimer, C_1	-7.128	-7.145	-7.092
	Errors			
	ME (kcal/mol)	0.344	0.409	0.372
	MUE (kcal/mol)	0.462	0.542	0.479
	MURE (%)	7.0	8.2	7.6

Table S2. Interactions energies (kcal/mol) computed for the S22 dataset with the revPBE0-DH-NL, functional at their optimum *b* value. The def2-QZVP basis set is employed for the calculations. ME, MUE and MURE errors are also included.

	Complexes	revPBE0-DH-NL (b = 6.0)
	Hydrogen Bonds	
1	Ammonia dimer, C_{2v}	-2.988
2	Water dimer, Cs	-5.047
3	Formic acid dimer, C_{2h}	-19.853
4	Formamide dimer, C_{2h}	-16.311
5	H-donded uracil dimer, C_{2h}	-20.887
6	2-pyridone 2-aminopyridine, C_1	-17.294
7	Adenine thymine WC, C_1	-16.708
	Dispersion	
8	Methane dimer, D_{3d}	-0.549
9	Ethene dimer, D_{2d}	-1.096
10	Benzene methane C_3	-1.372
11	Parallel displaced benzene dimer, C_{2h}	-2.583
12	Pyrazine dimer, C_s	-3.947
13	Stacked uracil dimer, C_2	-9.810
14	Stacked indole benzene, C_1	-4.303
15	Stacked adenina thymine, C_1	-11.760
	Mixed	
16	Ethene ethine, C_{2v}	-1.549
17	Benzene water, C_s	-3.363
18	Benzene ammonia, C_s	-2.276
19	Benzene hydrogen cyanide, C_s	-4.726
20	T-shaped benzene dimer, C_{2v}	-2.662
21	T-shaped indole benzene, C_1	-5.579
22	Phenol dimer, C_1	-6.707
	Errors	
	ME (kcal/mol)	0.155
	MUE (kcal/mol)	0.190
	MURE (%)	4.0

Table S3. Interactions energies (kcal/mol) computed for the S22 dataset with the B2PLYP-NL, functional at their optimum *b* value. The def2-QZVP basis set is employed for the calculations. ME, MUE and MURE errors are also included.

	Complexes	B2PLYP-NL (b = 8.3)
	Hydrogen Bonds	
1	Ammonia dimer, C_{2v}	-3.217
2	Water dimer, Cs	-5.358
3	Formic acid dimer, C_{2h}	-19.612
4	Formamide dimer, C_{2h}	-16.381
5	H-donded uracil dimer, C_{2h}	-21.030
6	2-pyridone 2-aminopyridine, C_1	-17.452
7	Adenine thymine WC, C_1	-16.867
	Dispersion	
8	Methane dimer, D_{3d}	-0.533
9	Ethene dimer, D_{2d}	-1.156
10	Benzene methane C_3	-1.336
11	Parallel displaced benzene dimer, C_{2h}	-2.556
12	Pyrazine dimer, C_s	-4.106
13	Stacked uracil dimer, C_2	-9.895
14	Stacked indole benzene, C_1	-4.392
15	Stacked adenina thymine, C_1	-11.839
	Mixed	
16	Ethene ethine, C_{2v}	-1.461
17	Benzene water, C_s	-3.257
18	Benzene ammonia, C_s	-2.200
19	Benzene hydrogen cyanide, C_s	-4.371
20	T-shaped benzene dimer, C_{2v}	-2.547
21	T-shaped indole benzene, C_1	-5.424
22	Phenol dimer, C_1	-7.053
	Errors	
	ME (kcal/mol)	0.186
	MUE (kcal/mol)	0.205
	MURE (%)	4.0

Table S4. Interactions energies (kcal/mol) computed for the S22 dataset with the TPSS0-DH-NL, TPSS0-NL, and TPSS-NL functionals at their optimum *b* value. The def2-QZVP basis set is employed for the calculations. ME, MUE and MURE errors are also included.

	Complexes	TSS0-DH-NL (b = 7.1)	TPSS0-NL (b = 5.4)	TPSS-NL (b = 5.0)
	Hydrogen Bonds			
1	Ammonia dimer, C_{2v}	-3.187	-3.227	-3.314
2	Water dimer, Cs	-5.400	-5.506	-5.637
3	Formic acid dimer, C_{2h}	-20.951	-21.221	-20.879
4	Formamide dimer, C_{2h}	-17.151	-17.268	-16.957
5	H-donded uracil dimer, C_{2h}	-21.766	-21.801	-21.358
6	2-pyridone 2-aminopyridine, C_1	-17.967	-18.132	-18.291
7	Adenine thymine WC, C_1	-17.391	-17.494	-17.522
	Dispersion			
8	Methane dimer, D_{3d}	-0.594	-0.541	-0.496
9	Ethene dimer, D_{2d}	-1.222	-1.200	-1.144
10	Benzene methane C_3	-1.426	-1.389	-1.340
11	Parallel displaced benzene dimer, C_{2h}	-2.458	-2.453	-2.598
12	Pyrazine dimer, C_s	-3.897	-3.815	-3.862
13	Stacked uracil dimer, C_2	-9.708	-9.738	-9.547
14	Stacked indole benzene, C_1	-4.116	-4.139	-4.431
15	Stacked adenina thymine, C_1	-11.614	-11.626	-11.494
	Mixed			
16	Ethene ethine, C_{2v}	-1.649	-1.659	-1.627
17	Benzene water, C_s	-3.448	-3.522	-3.546
18	Benzene ammonia, C_s	-2.331	-2.333	-2.299
19	Benzene hydrogen cyanide, C_s	-4.887	-4.805	-4.464
20	T-shaped benzene dimer, C_{2v}	-2.708	-2.647	-2.558
21	T-shaped indole benzene, C_1	-5.599	-5.523	-5.447
22	Phenol dimer, C_1	-6.996	-6.911	-6.790
	Errors			
	ME (kcal/mol)	0.339	0.393	0.392
	MUE (kcal/mol)	0.408	0.458	0.429
	MURE (%)	5.8	6.2	6.3

Table S5. Interactions energies (kcal/mol) computed for the S66 dataset with the PBE0-DH-NL, PBE0-NL, and PBE-NL functionals at their optimum *b* value. The def2-QZVP basis set is employed for the calculations. ME, MUE and MURE errors are also included.

	Complexes	PBE0-DH-NL (b = 8.3)	PBE0-NL (b = 6.6)	PBE-NL (b = 6.5)
	Hydrogen Bonds			
1	water···water	-5.55	-5.69	-5.77
2	water···MeOH	-6.10	-6.21	-6.26
3	water···$MeNH_2$	-7.74	-7.96	-8.18
4	water···peptide	-8.77	-8.86	-8.72
5	MeOH···MeOH	-6.27	-6.39	-6.48
6	MeOH···$MeNH_2$	-8.30	-8.54	-8.77
7	MeOH···peptide	-8.99	-9.13	-9.11
8	MeOH···water	-5.61	-5.76	-5.86
9	$MeNH_2$···MeOH	-3.22	-3.36	-3.49
10	$MeNH_2$···$MeNH_2$	-4.36	-4.49	-4.63
11	$MeNH_2$···peptide	-5.59	-5.67	-5.63
12	$MeNH_2$···water	-8.04	-8.20	-8.33
13	peptide···MeOH	-6.50	-6.53	-6.44
14	peptide···$MeNH_2$	-7.98	-8.12	-8.19
15	peptide···peptide	-9.05	-9.08	-8.88
16	peptide···water	-5.57	-5.65	-5.62
17	uracil···uracil (BP)	-18.63	-18.68	-18.25
18	water···pyridine	-7.53	-7.73	-7.98
19	MeOH···pyridine	-8.03	-8.22	-8.50
20	AcOH···AcOH	-21.49	-21.63	-21.08
21	$AcNH_2$···$AcNH_2$	-17.61	-17.70	-17.33
22	AcOH···uracil	-21.42	-21.47	-20.89
23	$AcNH_2$···uracil	-20.78	-20.80	-20.23
	Dispersion			
24	benzene···benzene	-2.64	-2.48	-2.50
25	pyridine···pyridine	-3.73	-3.51	-3.44
26	uracil···uracil	-9.91	-9.62	-9.12
27	benzene···pyridine	-3.27	-3.07	-3.03
28	benzene···uracil	-5.48	-5.18	-4.92
29	pyridine···uracil	-6.62	-6.31	-6.02
30	benzene···ethene	-1.24	-1.26	-1.38
31	uracil···ethene	-3.26	-3.20	-3.08
32	uracil···ethyne	-3.70	-3.64	-3.49
33	pyridine···ethene	-1.68	-1.68	-1.77
34	pentane···pentane	-3.49	-3.64	-3.73
35	neopentane···pentane	-2.51	-2.66	-2.78
36	neopentane···neopentane	-1.75	-1.91	-2.09
37	cyclopentane···neopentane	-2.28	-2.45	-2.59
38	cyclopentane···cyclopentane	-2.77	-2.94	-3.07
39	benzene···cyclopentane	-3.47	-3.41	-3.35
40	benzene···neopentane	-2.90	-2.89	-2.86
41	uracil···pentane	-4.59	-4.55	-4.45
42	uracil···cyclopentane	-3.88	-3.84	-3.80
43	uracil···neopentane	-3.61	-3.61	-3.54
44	ethene···pentane	-1.84	-1.92	-1.99
45	ethyne···pentane	-1.66	-1.75	-1.84
46	peptide···pentane	-4.03	-4.10	-4.08
	Mixed			
47	benzene···benzene (TS)	-2.83	-2.71	-2.59
48	pyridine···pyridine (TS)	-3.51	-3.36	-3.18

49	benzene···pyridine (TS)	-3.35	-3.20	-3.02
50	benzene···ethyne (CH···π)	-3.07	-2.97	-2.75
51	ethyne···ethyne (TS)	-1.70	-1.76	-1.77
52	benzene···AcOH (OH···π)	-5.14	-5.03	-4.74
53	benzene···$AcNH_2$ (NH···π)	-4.63	-4.59	-4.40
54	benzene···water (OH···π)	-3.75	-3.76	-3.65
55	benzene···MeOH (OH···π)	-4.40	-4.40	-4.31
56	benzene···$MeNH_2$ (NH···π)	-3.22	-3.20	-3.14
57	benzene···peptide (NH···π)	-5.41	-5.28	-5.00
58	pyridine···pyridine (CH···N)	-4.20	-4.14	-4.07
59	ethyne···water (CH···O)	-3.29	-3.35	-3.30
60	ethyne···AcOH (OH···π)	-5.57	-5.65	-5.50
61	pentane···AcOH	-2.76	-2.88	-2.94
62	pentane···$AcNH_2$	-3.43	-3.54	-3.60
63	benzene···AcOH	-3.83	-3.72	-3.50
64	peptide···ethene	-2.95	-3.00	-2.96
65	pyridine···ethyne	-4.44	-4.50	-4.51
66	$MeNH_2$···pyridine	-3.89	-3.92	-3.99
	Errors			
	ME (kcal/mol)	-0.24	-0.27	-0.21
	MUE (kcal/mol)	0.33	0.38	0.40
	MURE (%)	5.3	6.2	7.1

Table S6. Interactions energies (kcal/mol) computed for the S66 dataset with the revPBE0-DH-NL, revPBE0-NL, and revPBE-NL functionals at their optimum *b* value. The def2-QZVP basis set is employed for the calculations. ME, MUE and MURE errors are also included.

	Complexes	revPBE0-DH-NL (b = 5.7)	revPBE0-NL (b = 4.2)	revPBE-NL (b = 3.6)
	Hydrogen Bonds			
1	water···water	-5.11	-5.07	-4.99
2	water···MeOH	-5.65	-5.58	-5.50
3	water···$MeNH_2$	-7.26	-7.30	-7.38
4	water···peptide	-8.20	-8.09	-7.82
5	MeOH···MeOH	-5.87	-5.84	-5.84
6	MeOH···$MeNH_2$	-7.86	-7.96	-8.12
7	MeOH···peptide	-8.50	-8.46	-8.36
8	MeOH···water	-5.21	-5.20	-5.17
9	$MeNH_2$···MeOH	-2.98	-3.02	-3.10
10	$MeNH_2$···$MeNH_2$	-4.01	-4.04	-4.14
11	$MeNH_2$···peptide	-5.27	-5.26	-5.24
12	$MeNH_2$···water	-7.52	-7.51	-7.52
13	peptide···MeOH	-6.16	-6.06	-5.92
14	peptide···$MeNH_2$	-7.65	-7.69	-7.75
15	peptide···peptide	-8.71	-8.64	-8.45
16	peptide···water	-5.23	-5.16	-5.04
17	uracil···uracil (BP)	-17.87	-17.64	-17.06
18	water···pyridine	-7.08	-7.11	-7.23
19	MeOH···pyridine	-7.65	-7.71	-7.91
20	AcOH···AcOH	-20.49	-20.29	-19.53
21	$AcNH_2$···$AcNH_2$	-16.78	-16.58	-16.03
22	AcOH···uracil	-20.51	-20.25	-19.49
23	$AcNH_2$···uracil	-19.96	-19.69	-18.95
	Dispersion			
24	benzene···benzene	-2.91	-2.93	-3.30
25	pyridine···pyridine	-3.99	-3.95	-4.26
26	uracil···uracil	-10.22	-10.22	-10.34
27	benzene···pyridine	-3.54	-3.51	-3.85
28	benzene···uracil	-5.80	-5.74	-6.02
29	pyridine···uracil	-6.90	-6.82	-7.04
30	benzene···ethene	-1.27	-1.35	-1.64
31	uracil···ethene	-3.25	-3.26	-3.35
32	uracil···ethyne	-3.66	-3.65	-3.68
33	pyridine···ethene	-1.68	-1.74	-2.01
34	pentane···pentane	-3.34	-3.50	-3.79
35	neopentane···pentane	-2.38	-2.50	-2.72
36	neopentane···neopentane	-1.66	-1.76	-1.97
37	cyclopentane···neopentane	-2.16	-2.29	-2.53
38	cyclopentane···cyclopentane	-2.65	-2.78	-3.03
39	benzene···cyclopentane	-3.55	-3.58	-3.80
40	benzene···neopentane	-2.93	-2.96	-3.12
41	uracil···pentane	-4.63	-4.70	-4.95
42	uracil···cyclopentane	-3.94	-4.00	-4.26
43	uracil···neopentane	-3.64	-3.70	-3.87
44	ethene···pentane	-1.66	-1.71	-1.83
45	ethyne···pentane	-1.58	-1.66	-1.82
46	peptide···pentane	-3.89	-3.99	-4.20
	Mixed			
47	benzene···benzene (TS)	-2.82	-2.74	-2.78
48	pyridine···pyridine (TS)	-3.44	-3.30	-3.26

49	benzene···pyridine (TS)	-3.32	-3.21	-3.19
50	benzene···ethyne (CH···π)	-2.95	-2.85	-2.71
51	ethyne···ethyne (TS)	-1.56	-1.56	-1.53
52	benzene···AcOH (OH···π)	-5.02	-4.93	-4.76
53	benzene···$AcNH_2$ (NH···π)	-4.44	-4.36	-4.24
54	benzene···water (OH···π)	-3.57	-3.55	-3.50
55	benzene···MeOH (OH···π)	-4.28	-4.30	-4.36
56	benzene···$MeNH_2$ (NH···π)	-3.15	-3.17	-3.25
57	benzene···peptide (NH···π)	-5.41	-5.34	-5.31
58	pyridine···pyridine (CH···N)	-3.89	-3.72	-3.60
59	ethyne···water (CH···O)	-3.02	-2.96	-2.82
60	ethyne···AcOH (OH···π)	-5.19	-5.15	-4.94
61	pentane···AcOH	-2.66	-2.76	-2.96
62	pentane···$AcNH_2$	-3.27	-3.36	-3.56
63	benzene···AcOH	-3.83	-3.81	-3.82
64	peptide···ethene	-2.77	-2.78	-2.80
65	pyridine···ethyne	-4.18	-4.13	-4.07
66	$MeNH_2$···pyridine	-3.71	-3.71	-3.88
	Errors			
	ME (kcal/mol)	-0.04	-0.02	-0.04
	MUE (kcal/mol)	0.19	0.16	0.20
	MURE (%)	4.1	3.3	4.6

Table S7. Interactions energies (kcal/mol) computed for the S66 dataset with the B2PLYP-NL, B3LYP-NL, and BLYP-NL functionals at their optimum *b* value. The def2-QZVP basis set is employed for the calculations. ME, MUE and MURE errors are also included.

	Complexes	B2PLYP-NL (b = 7.8)	B3LYP-NL (b = 4.6)	BLYP-NL (b = 4.0)
	Hydrogen Bonds			
1	water···water	-5.41	-5.65	-5.52
2	water···MeOH	-6.02	-6.25	-6.09
3	water···$MeNH_2$	-7.43	-7.80	-7.78
4	water···peptide	-8.48	-8.86	-8.51
5	MeOH···MeOH	-6.22	-6.45	-6.33
6	MeOH···$MeNH_2$	-7.95	-8.35	-8.31
7	MeOH···peptide	-8.74	-9.14	-8.90
8	MeOH···water	-5.49	-5.74	-5.63
9	$MeNH_2$···MeOH	-3.19	-3.33	-3.22
10	$MeNH_2$···$MeNH_2$	-4.22	-4.39	-4.29
11	$MeNH_2$···peptide	-5.47	-5.71	-5.49
12	$MeNH_2$···water	-7.74	-8.05	-7.95
13	peptide···MeOH	-6.46	-6.58	-6.30
14	peptide···$MeNH_2$	-7.71	-7.94	-7.81
15	peptide···peptide	-8.83	-9.07	-8.72
16	peptide···water	-5.46	-5.60	-5.37
17	uracil···uracil (BP)	-18.07	-18.72	-18.10
18	water···pyridine	-7.40	-7.71	-7.72
19	MeOH···pyridine	-7.93	-8.23	-8.26
20	AcOH···AcOH	-20.31	-21.35	-20.57
21	$AcNH_2$···$AcNH_2$	-16.90	-17.59	-16.99
22	AcOH···uracil	-20.50	-21.35	-20.59
23	$AcNH_2$···uracil	-19.98	-20.69	-19.95
	Dispersion			
24	benzene···benzene	-2.93	-2.60	-2.62
25	pyridine···pyridine	-3.99	-3.63	-3.59
26	uracil···uracil	-10.26	-10.34	-10.09
27	benzene···pyridine	-3.54	-3.19	-3.18
28	benzene···uracil	-5.80	-5.51	-5.42
29	pyridine···uracil	-6.99	-6.75	-6.62
30	benzene···ethene	-1.28	-1.20	-1.20
31	uracil···ethene	-3.25	-3.22	-3.04
32	uracil···ethyne	-3.67	-3.66	-3.45
33	pyridine···ethene	-1.73	-1.64	-1.61
34	pentane···pentane	-3.54	-3.77	-3.60
35	neopentane···pentane	-2.47	-2.62	-2.49
36	neopentane···neopentane	-1.69	-1.79	-1.69
37	cyclopentane···neopentane	-2.26	-2.43	-2.33
38	cyclopentane···cyclopentane	-2.89	-3.06	-2.98
39	benzene···cyclopentane	-3.53	-3.44	-3.33
40	benzene···neopentane	-2.84	-2.79	-2.64
41	uracil···pentane	-4.77	-4.83	-4.67
42	uracil···cyclopentane	-4.15	-4.16	-4.05
43	uracil···neopentane	-3.72	-3.76	-3.59
44	ethene···pentane	-1.78	-1.82	-1.65
45	ethyne···pentane	-1.60	-1.62	-1.49
46	peptide···pentane	-4.09	-4.26	-4.06
	Mixed			
47	benzene···benzene (TS)	-2.78	-2.56	-2.38
48	pyridine···pyridine (TS)	-3.49	-3.28	-3.03

49	benzene···pyridine (TS)	-3.24	-3.02	-2.79
50	benzene···ethyne (CH···π)	-2.77	-2.66	-2.38
51	ethyne···ethyne (TS)	-1.51	-1.54	-1.39
52	benzene···AcOH (OH···π)	-4.72	-4.68	-4.35
53	benzene···$AcNH_2$ (NH···π)	-4.36	-4.38	-4.07
54	benzene···water (OH···π)	-3.43	-3.46	-3.26
55	benzene···MeOH (OH···π)	-4.14	-4.16	-4.00
56	benzene···$MeNH_2$ (NH···π)	-3.06	-3.04	-2.88
57	benzene···peptide (NH···π)	-5.17	-5.04	-4.75
58	pyridine···pyridine (CH···N)	-4.16	-4.06	-3.80
59	ethyne···water (CH···O)	-3.14	-3.24	-3.01
60	ethyne···AcOH (OH···π)	-5.05	-5.32	-4.98
61	pentane···AcOH	-2.80	-2.93	-2.78
62	pentane···$AcNH_2$	-3.45	-3.61	-3.45
63	benzene···AcOH	-3.69	-3.64	-3.39
64	peptide···ethene	-2.87	-2.94	-2.72
65	pyridine···ethyne	-4.27	-4.36	-4.19
66	$MeNH_2$···pyridine	-3.94	-3.96	-3.89
	Errors			
	ME (kcal/mol)	-0.12	-0.24	-0.04
	MUE (kcal/mol)	0.20	0.34	0.30
	MURE (%)	3.6	5.3	6.2

Table S8. Interactions energies (kcal/mol) computed for the S66 dataset with the TPSS0-DH-NL, TPSS0-NL, and TPSS-NL functionals at their optimum *b* value. The def2-QZVP basis set is employed for the calculations. ME, MUE and MURE errors are also included.

	Complexes	TPSS0-DH-NL (b = 6.8)	TPSS0-NL (b = 5.2)	TPSS-NL (b = 5.0)
	Hydrogen Bonds			
1	water···water	-5.44	-5.52	-5.53
2	water···MeOH	-6.00	-6.06	-6.04
3	water···$MeNH_2$	-7.63	-7.82	-7.97
4	water···peptide	-8.63	-8.70	-8.48
5	MeOH···MeOH	-6.18	-6.27	-6.30
6	MeOH···$MeNH_2$	-8.18	-8.41	-8.56
7	MeOH···peptide	-8.87	-8.98	-8.88
8	MeOH···water	-5.51	-5.60	-5.64
9	$MeNH_2$···MeOH	-3.14	-3.21	-3.25
10	$MeNH_2$···$MeNH_2$	-4.20	-4.28	-4.30
11	$MeNH_2$···peptide	-5.45	-5.50	-5.33
12	$MeNH_2$···water	-7.89	-8.03	-8.09
13	peptide···MeOH	-6.40	-6.39	-6.19
14	peptide···$MeNH_2$	-7.87	-7.99	-7.98
15	peptide···peptide	-8.96	-9.00	-8.72
16	peptide···water	-5.47	-5.48	-5.36
17	uracil···uracil (BP)	-18.63	-18.74	-18.32
18	water···pyridine	-7.45	-7.61	-7.80
19	MeOH···pyridine	-7.96	-8.13	-8.35
20	AcOH···AcOH	-21.47	-21.76	-21.32
21	$AcNH_2$···$AcNH_2$	-17.53	-17.67	-17.30
22	AcOH···uracil	-21.40	-21.56	-21.05
23	$AcNH_2$···uracil	-20.75	-20.83	-20.28
	Dispersion			
24	benzene···benzene	-2.72	-2.72	-2.67
25	pyridine···pyridine	-3.81	-3.78	-3.65
26	uracil···uracil	-10.01	-10.09	-9.53
27	benzene···pyridine	-3.35	-3.33	-3.22
28	benzene···uracil	-5.59	-5.59	-5.28
29	pyridine···uracil	-6.73	-6.71	-6.37
30	benzene···ethene	-1.23	-1.31	-1.34
31	uracil···ethene	-3.24	-3.27	-3.08
32	uracil···ethyne	-3.68	-3.71	-3.49
33	pyridine···ethene	-1.65	-1.72	-1.73
34	pentane···pentane	-3.35	-3.50	-3.37
35	neopentane···pentane	-2.41	-2.52	-2.48
36	neopentane···neopentane	-1.70	-1.80	-1.85
37	cyclopentane···neopentane	-2.19	-2.32	-2.30
38	cyclopentane···cyclopentane	-2.67	-2.78	-2.74
39	benzene···cyclopentane	-3.47	-3.53	-3.37
40	benzene···neopentane	-2.89	-2.93	-2.81
41	uracil···pentane	-4.54	-4.65	-4.42
42	uracil···cyclopentane	-3.85	-3.92	-3.75
43	uracil···neopentane	-3.59	-3.65	-3.48
44	ethene···pentane	-1.72	-1.77	-1.70
45	ethyne···pentane	-1.60	-1.66	-1.64
46	peptide···pentane	-3.90	-4.02	-3.83
	Mixed			
47	benzene···benzene (TS)	-2.81	-2.77	-2.59
48	pyridine···pyridine (TS)	-3.46	-3.36	-3.11

49	benzene···pyridine (TS)	-3.33	-3.26	-3.03
50	benzene···ethyne (CH···π)	-3.01	-2.97	-2.72
51	ethyne···ethyne (TS)	-1.66	-1.67	-1.63
52	benzene···AcOH (OH···π)	-5.07	-5.10	-4.77
53	benzene···$AcNH_2$ (NH···π)	-4.52	-4.51	-4.25
54	benzene···water (OH···π)	-3.64	-3.69	-3.52
55	benzene···MeOH (OH···π)	-4.31	-4.40	-4.24
56	benzene···$MeNH_2$ (NH···π)	-3.15	-3.22	-3.07
57	benzene···peptide (NH···π)	-5.38	-5.39	-5.06
58	pyridine···pyridine (CH···N)	-4.10	-3.98	-3.81
59	ethyne···water (CH···O)	-3.22	-3.22	-3.11
60	ethyne···AcOH (OH···π)	-5.46	-5.56	-5.36
61	pentane···AcOH	-2.67	-2.78	-2.68
62	pentane···$AcNH_2$	-3.30	-3.41	-3.30
63	benzene···AcOH	-3.82	-3.84	-3.55
64	peptide···ethene	-2.84	-2.86	-2.70
65	pyridine···ethyne	-4.42	-4.43	-4.39
66	$MeNH_2$···pyridine	-3.79	-3.82	-3.78
	Errors			
	ME(kcal/mol)	-0.18	-0.25	-0.11
	MUE (kcal/mol)	0.31	0.33	0.34
	MURE (%)	5.2	5.1	6.2

Table S9. Interactions energies (kcal/mol) computed for the NCC31 dataset with the revPBE0-DH-NL, revPBE0-NL, and revPBE-NL functionals at their optimum *b* value. The def2-QZVP basis set is employed for the calculations. ME, MUE and MURE errors are also included.

	Complexes	revPBE0-DH-NL (b = 5.7)	revPBE0-NL (b = 4.2)	revPBE-NL (b = 3.6)
	Hydrogen Bonds			
1	$(NH_3)_2$	-3.14	-3.17	-3.23
2	$(HF)_2$	-4.57	-4.57	-4.56
3	$(H_2O)_2$	-5.05	-5.06	-5.07
4	NH_3··· H_2O	-6.55	-6.65	-6.79
5	$(HCONH_2)_2$	-14.99	-14.95	-14.83
6	$(HCOOH)_2$	-17.17	-17.10	-16.70
	Charge Transfer			
7	C_2H_4··· F_2	-1.04	-1.55	-3.07
8	NH_3··· F_2	-1.54	-2.34	-4.92
9	C_2H_2··· ClF	-4.53	-5.30	-6.81
10	HCN··· ClF	-4.84	-5.03	-5.88
11	NH_3··· Cl_2	-5.36	-6.22	-8.14
12	H_2O ··· ClF	-5.68	-6.16	-7.41
13	NH_3··· ClF	-12.40	-13.91	-17.13
	Dipole Interaction			
14	$(H_2S)_2$	-1.62	-1.76	-2.05
15	$(HCl)_2$	-1.76	-1.94	-2.24
16	HCl··· H_2S	-3.55	-3.83	-4.33
17	CH_3Cl··· HCl	-3.23	-3.49	-3.94
18	HCN··· CH_3SH	-3.77	-3.87	-4.01
19	CH_3SH ···HCl	-5.14	-5.55	-6.27
	Weak Interaction			
20	HeNe	-0.09	-0.07	-0.11
21	HeAr	-0.14	-0.11	-0.12
22	Ne_2	-0.10	-0.14	-0.18
23	NeAr	-0.13	-0.18	-0.23
24	CH_4··· Ne	-0.18	-0.24	-0.30
25	C_6H_6··· Ne	-0.48	-0.58	-0.69
	π-π Stacking			
26	$(CH_4)_2$	-0.33	-0.38	-0.44
27	$(C_2H_4)_2$	-1.38	-1.37	-1.37
28	$(C_2H_2)_2$	-1.14	-1.14	-1.15
29	Sandwich $(C_6H_6)_2$	-1.83	-1.91	-2.38
30	T-shaped $(C_6H_6)_2$	-2.74	-2.67	-2.70
31	Parallel-displaced $(C_6H_6)_2$	-2.83	-2.86	-3.24
	Errors			
	ME (kcal/mol)	-0.15	-0.37	-0.90
	MUE (kcal/mol)	0.25	0.41	0.93
	MURE (%)	16.4	20.3	46.0

Table S10. Interactions energies (kcal/mol) computed for the NCC31 dataset with the B2PLYP-NL, B3LYP-NL, and BLYP-NL functionals at their optimum *b* value. The def2-QZVP basis set is employed for the calculations. ME, MUE and MURE errors are also included.

	Complexes	B2PLYP-NL (b = 7.8)	B3LYP-NL (b = 4.6)	BLYP-NL (b = 4.0)
	Hydrogen Bond			
1	$(NH_3)_2$	-3.30	-3.44	-3.36
2	$(HF)_2$	-5.02	-5.30	-5.22
3	$(H_2O)_2$	-5.40	-5.67	-5.58
4	NH_3··· H_2O	-6.79	-7.17	-7.17
5	$(HCONH_2)_2$	-15.37	-15.99	-15.72
6	$(HCOOH)_2$	-17.22	-18.25	-17.72
	Charge Transfer			
7	C_2H_4··· F_2	-1.58	-2.17	-3.53
8	NH_3··· F_2	-2.49	-3.42	-5.68
9	C_2H_2··· ClF	-4.79	-5.80	-6.93
10	HCN··· ClF	-5.60	-5.99	-6.60
11	NH_3··· Cl_2	-5.96	-7.03	-8.50
12	H_2O ··· ClF	-6.36	-7.16	-8.06
13	NH_3··· ClF	-13.24	-14.89	-17.35
	Dipole Interaction			
14	$(H_2S)_2$	-1.59	-1.75	-1.85
15	$(HCl)_2$	-1.87	-2.12	-2.25
16	HCl··· H_2S	-3.51	-3.91	-4.18
17	CH_3Cl··· HCl	-3.38	-3.77	-3.99
18	HCN··· CH_3SH	-3.71	-3.90	-3.85
19	CH_3SH ···HCl	-5.04	-5.58	-5.99
	Weak interaction			
20	HeNe	-0.02	-0.01	0.02
21	HeAr	-0.06	-0.03	0.03
22	Ne_2	-0.08	-0.09	-0.06
23	NeAr	-0.11	-0.12	-0.09
24	CH_4··· Ne	-0.13	-0.16	-0.11
25	C_6H_6··· Ne	-0.47	-0.56	-0.49
	π-π Stacking			
26	$(CH_4)_2$	-0.31	-0.32	-0.21
27	$(C_2H_4)_2$	-1.35	-1.31	-1.16
28	$(C_2H_2)_2$	-1.20	-1.19	-0.98
29	Sandwich $(C_6H_6)_2$	-1.91	-1.64	-1.73
30	T-shaped $(C_6H_6)_2$	-2.68	-2.47	-2.26
31	Parallel-displaced $(C_6H_6)_2$	-2.87	-2.58	-2.59
	Errors			
	ME (kcal/mol)	-0.35	-0.69	-0.99
	MUE (kcal/mol)	0.42	0.77	1.12
	MURE (%)	14.5	26.3	47.0

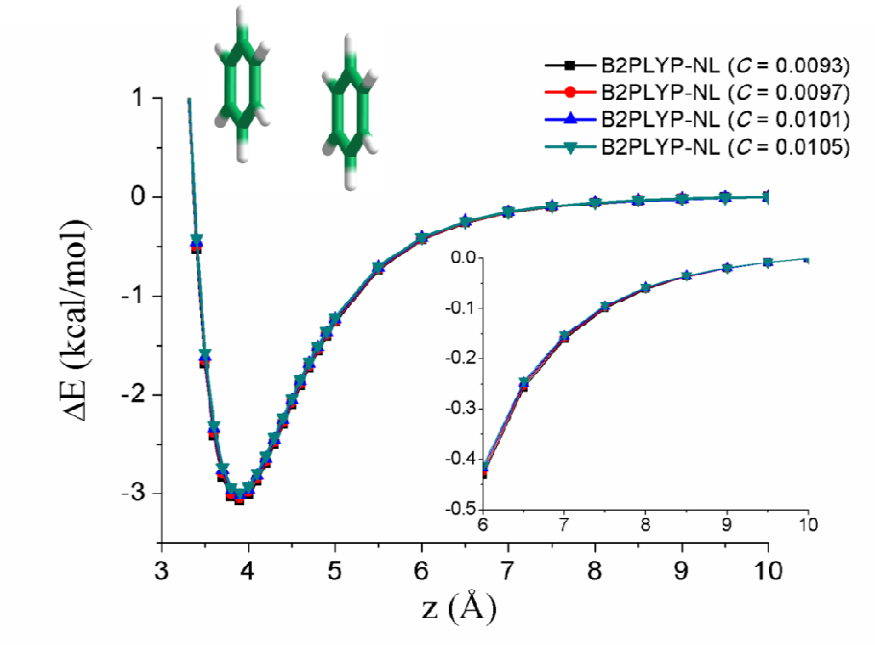

Figure S3. Potential energy curves computed for the parallel-displaced benzene-benzene dimer at the B2PLYP-NL/def2-QZVP level as a function of the long-range parameter *C*.