Table S1. MP2.5/bas-MP2/6-31G* RMS binding energy errors and error ranges (in parentheses) for S66 binding categories (kcal/mol)

	MP2.5	bas-x=	6-31G*	
Total	bas-MP2	CBS	aQZ	aTZ
	0.9	0.56 (1.89)	0.46 (1.99)	0.48 (2.44)
	0.95	0.45 (1.57)	0.39 (1.72)	0.43 (2.15)
	1	0.36 (1.30)	0.33 (1.49)	0.37 (1.89)
	1.05	0.29 (1.10)	0.28 (1.29)	0.32 (1.65)
	1.1	0.24 (0.94)	0.23 (1.12)	0.28 (1.45)
	1.25	0.14 (0.60)	0.14 (0.72)	0.18 (0.94)
	1.5	0.06 (0.28)	0.06 (0.33)	0.08 (0.42)
	2	0.02 (0.06)	0.02 (0.07)	0.02 (0.08)
HB	bas-MP2	CBS	aQZ	aTZ
	0.9	0.09 (0.30)	0.19 (0.38)	0.57 (0.93)
	0.95	0.05 (0.21)	0.19 (0.36)	0.51 (0.85)
	1	0.04 (0.14)	0.19 (0.35)	0.46 (0.78)
	1.05	0.05 (0.14)	0.18 (0.33)	0.41 (0.72)
	1.1	0.06 (0.16)	0.17 (0.32)	0.36 (0.66)
	1.25	0.07 (0.16)	0.13 (0.27)	0.23 (0.48)
	1.5	0.04 (0.12)	0.07 (0.17)	0.11 (0.25)
	2	0.01 (0.04)	0.01 (0.05)	0.02 (0.06)
DISP	bas-MP2	CBS	aQZ	aTZ
	0.9	0.85 (1.89)	0.70 (1.72)	0.54 (1.71)
	0.95	0.68 (1.56)	0.58 (1.43)	0.47 (1.39)
	1	0.55 (1.29)	0.48 (1.19)	0.40 (1.15)
	1.05	0.44 (1.06)	0.40 (0.99)	0.34 (0.96)
	1.1	0.36 (0.87)	0.33 (0.82)	0.29 (0.81)
	1.25	0.19 (0.49)	0.18 (0.47)	0.17 (0.48)
	1.5	0.07 (0.18)	0.07 (0.18)	0.07 (0.18)
	2	0.02 (0.04)	0.02 (0.04)	0.02 (0.04)
MIX	bas-MP2	CBS	aQZ	aTZ
	0.9	0.44 (0.83)	0.32 (0.80)	0.24 (0.80)
	0.95	0.35 (0.68)	0.26 (0.67)	0.20 (0.67)
	1	0.28 (0.56)	0.22 (0.56)	0.17 (0.57)
	1.05	0.23 (0.47)	0.18 (0.47)	0.14 (0.48)
	1.1	0.19 (0.39)	0.15 (0.39)	0.12 (0.40)
	1.25	0.11 (0.23)	0.10 (0.24)	0.08 (0.24)
	1.5	0.05 (0.11)	0.05 (0.11)	0.04 (0.11)
	2	0.02 (0.03)	0.02 (0.03)	0.02 (0.03)

Table S2. MP2.5/bas-MP2/6-311++G** RMS binding energy errors and error ranges (in parentheses) for S66 binding categories (kcal/mol)

	MP2.5	bas-x=	6-311++G**	
Total	bas-MP2	CBS	aQZ	aTZ
	0.90	0.37 (1.33)	0.28 (1.18)	0.39 (1.58)
	0.95	0.30 (1.11)	0.24 (1.06)	0.34 (1.45)
	1.00	0.24 (0.93)	0.21 (0.94)	0.29 (1.32)
	1.05	0.20 (0.77)	0.18 (0.83)	0.26 (1.19)
	1.10	0.16 (0.64)	0.16 (0.73)	0.22 (1.07)
	1.25	0.09 (0.38)	0.09 (0.50)	0.14 (0.72)
	1.50	0.04 (0.20)	0.04 (0.25)	0.07 (0.33)
	2.00	0.02 (0.06)	0.02 (0.06)	0.02 (0.08)
HB	bas-MP2	CBS	aQZ	aTZ
	0.90	0.15 (0.39)	0.13 (0.20)	0.50 (0.75)
	0.95	0.10 (0.29)	0.13 (0.19)	0.45 (0.68)
	1.00	0.06 (0.21)	0.13 (0.19)	0.40 (0.62)
	1.05	0.04 (0.16)	0.13 (0.19)	0.36 (0.58)
	1.10	0.03 (0.12)	0.13 (0.19)	0.32 (0.53)
	1.25	0.04 (0.07)	0.10 (0.17)	0.21 (0.38)
	1.50	0.03 (0.08)	0.06 (0.13)	0.10 (0.21)
	2.00	0.01 (0.03)	0.01 (0.04)	0.02 (0.05)
DISP	bas-MP2	CBS	aQZ	aTZ
	0.90	0.53 (1.33)	0.42 (1.17)	0.38 (1.33)
	0.95	0.43 (1.11)	0.36 (1.02)	0.32 (1.10)
	1.00	0.35 (0.93)	0.30 (0.87)	0.27 (0.91)
	1.05	0.29 (0.77)	0.25 (0.74)	0.23 (0.76)
	1.10	0.24 (0.64)	0.21 (0.62)	0.19 (0.63)
	1.25	0.13 (0.36)	0.12 (0.37)	0.12 (0.37)
	1.50	0.05 (0.14)	0.05 (0.14)	0.05 (0.14)
	2.00	0.02 (0.03)	0.01 (0.03)	0.01 (0.03)
MIX	bas-MP2	CBS	aQZ	aTZ
	0.90	0.32 (0.64)	0.20 (0.61)	0.19 (0.61)
	0.95	0.25 (0.54)	0.17 (0.52)	0.16 (0.52)
	1.00	0.20 (0.45)	0.14 (0.44)	0.13 (0.44)
	1.05	0.17 (0.38)	0.12 (0.37)	0.11 (0.38)
	1.10	0.14 (0.32)	0.10 (0.31)	0.09 (0.32)
	1.25	0.08 (0.19)	0.07 (0.19)	0.06 (0.19)
	1.50	0.04 (0.09)	0.04 (0.09)	0.03 (0.09)
	2.00	0.02 (0.04)	0.01 (0.03)	0.01 (0.02)

Table S3. MP2.5/bas-MP2/aDZ RMS binding energy errors and error ranges (in parentheses) for S66 binding categories (kcal/mol)

	MP2.5	bas-x=	aDZ	
Total	bas-MP2	CBS	aQZ	aTZ
	0.90	0.24 (1.22)	0.27 (1.33)	0.49 (1.0)
	0.95	0.20 (1.03)	0.23 (1.12)	0.42 (1.25)
	1.00	0.17 (0.87)	0.20 (0.94)	0.36 (1.10)
	1.05	0.14 (0.73)	0.18 (0.79)	0.31 (1.02)
	1.10	0.12 (0.61)	0.16 (0.66)	0.26 (0.93)
	1.25	0.08 (0.38)	0.11 (0.45)	0.16 (0.66)
	1.50	0.04 (0.22)	0.05 (0.25)	0.08 (0.33)
	2.00	0.01 (0.07)	0.02 (0.08)	0.02 (0.09)
HB	bas-MP2	CBS	aQZ	aTZ
	0.90	0.08 (0.28)	0.20 (0.22)	0.58 (0.78)
	0.95	0.05 (0.19)	0.20 (0.25)	0.52 (0.72)
	1.00	0.05 (0.14)	0.20 (0.26)	0.47 (0.67)
	1.05	0.06 (0.14)	0.19 (0.27)	0.42 (0.63)
	1.10	0.07 (0.15)	0.18 (0.27)	0.37 (0.58)
	1.25	0.08 (0.18)	0.14 (0.25)	0.25 (0.43)
	1.50	0.06 (0.15)	0.06 (0.17)	0.12 (0.24)
	2.00	0.02 (0.06)	0.02 (0.07)	0.03 (0.07)
DISP	bas-MP2	CBS	aQZ	aTZ
	0.90	0.36 (1.22)	0.37 (1.33)	0.53 (1.50)
	0.95	0.30 (1.03)	0.31 (1.12)	0.42 (1.25)
	1.00	0.25 (0.87)	0.26 (0.94)	0.33 (1.04)
	1.05	0.21 (0.73)	0.22 (0.79)	0.27 (0.87)
	1.10	0.17 (0.61)	0.18 (0.66)	0.21 (0.72)
	1.25	0.10 (0.36)	0.10 (0.39)	0.11 (0.42)
	1.50	0.04 (0.15)	0.04 (0.16)	0.04 (0.17)
	2.00	0.01 (0.04)	0.01 (0.04)	0.01 (0.04)
MIX	bas-MP2	CBS	aQZ	aTZ
	0.90	0.18 (0.60)	0.16 (0.59)	0.31 (0.60)
	0.95	0.15 (0.50)	0.14 (0.50)	0.25 (0.51)
	1.00	0.12 (0.43)	0.12 (0.43)	0.20 (0.43)
	1.05	0.10 (0.36)	0.10 (0.36)	0.16 (0.37)
	1.10	0.08 (0.31)	0.08 (0.31)	0.12 (0.32)
	1.25	0.05 (0.19)	0.05 (0.19)	0.07 (0.20)
	1.50	0.03 (0.09)	0.03 (0.09)	0.02 (0.09)
	2.00	0.01 (0.03)	0.01 (0.03)	0.01 (0.02)

Table S4. MP2.X/bas-MP2/6-31G* RMS binding energy errors and error ranges (in parentheses) for S66 binding categories (kcal/mol)

	MP2.X	bas-x=	6-31G*	
Total	bas-MP2	CBS	aQZ	aTZ
	0.90	0.21 (1.28)	0.30 (1.50)	0.56 (1.83)
	0.95	0.19 (1.03)	0.24 (1.20)	0.44 (1.44)
	1.00	0.17 (0.87)	0.21 (0.96)	0.36 (1.17)
	1.05	0.14 (0.73)	0.18 (0.81)	0.30 (1.05)
	1.10	0.13 (0.61)	0.15 (0.68)	0.25 (0.95)
	1.25	0.08 (0.36)	0.09 (0.40)	0.14 (0.62)
	1.50	0.04 (0.15)	0.04 (0.19)	0.05 (0.27)
	2.00	0.02 (0.11)	0.02 (0.09)	0.01 (0.08)
HB	bas-MP2	CBS	aQZ	aTZ
	0.90	0.13 (0.46)	0.29 (0.34)	0.66 (0.89)
	0.95	0.12 (0.39)	0.26 (0.32)	0.57 (0.80)
	1.00	0.11 (0.34)	0.23 (0.29)	0.49 (0.73)
	1.05	0.10 (0.29)	0.20 (0.27)	0.42 (0.66)
	1.10	0.09 (0.26)	0.17 (0.25)	0.36 (0.59)
	1.25	0.06 (0.18)	0.11 (0.20)	0.21 (0.41)
	1.50	0.03 (0.12)	0.04 (0.11)	0.08 (0.20)
	2.00	0.03 (0.10)	0.02 (0.08)	0.02 (0.07)
DISP	bas-MP2	CBS	aQZ	aTZ
	0.90	0.30 (1.28)	0.38 (1.50)	0.60 (1.83)
	0.95	0.27 (1.03)	0.30 (1.20)	0.43 (1.44)
	1.00	0.23 (0.87)	0.25 (0.96)	0.32 (1.15)
	1.05	0.20 (0.73)	0.21 (0.81)	0.25 (0.92)
	1.10	0.17 (0.61)	0.18 (0.68)	0.20 (0.76)
	1.25	0.10 (0.36)	0.10 (0.39)	0.11 (0.43)
	1.50	0.04 (0.15)	0.04 (0.17)	0.04 (0.18)
	2.00	0.01 (0.04)	0.01 (0.04)	0.01 (0.05)
MIX	bas-MP2	CBS	aQZ	aTZ
	0.90	0.16 (0.47)	0.16 (0.50)	0.33 (0.58)
	0.95	0.14 (0.42)	0.13 (0.42)	0.25 (0.48)
	1.00	0.12 (0.37)	0.11 (0.37)	0.19 (0.38)
	1.05	0.11 (0.32)	0.10 (0.32)	0.14 (0.33)
	1.10	0.09 (0.28)	0.08 (0.28)	0.11 (0.29)
	1.25	0.06 (0.18)	0.05 (0.18)	0.06 (0.19)
	1.50	0.03 (0.09)	0.03 (0.09)	0.03 (0.09)
	2.00	0.01 (0.03)	0.01 (0.03)	0.01 (0.03)

Table S5. MP2.X/bas-MP2/6-311++G** RMS binding energy errors and error ranges (in parentheses) for S66 binding categories (kcal/mol)

	MP2.X	bas-x=	6-311++G**	
Total	bas-MP2	CBS	aQZ	aTZ
	0.90	0.20 (0.88)	0.22 (1.09)	0.47 (1.42)
	0.95	0.17 (0.71)	0.19 (0.88)	0.37 (1.12)
	1.00	0.14 (0.62)	0.16 (0.71)	0.31 (0.90)
	1.05	0.12 (0.53)	0.14 (0.59)	0.26 (0.81)
	1.10	0.10 (0.45)	0.12 (0.50)	0.21 (0.73)
	1.25	0.06 (0.27)	0.08 (0.30)	0.13 (0.52)
	1.50	0.03 (0.12)	0.04 (0.16)	0.06 (0.25)
	2.00	0.01 (0.04)	0.01 (0.04)	0.01 (0.05)
HB	bas-MP2	CBS	aQZ	aTZ
	0.90	0.17 (0.50)	0.16 (0.28)	0.51 (0.70)
	0.95	0.13 (0.41)	0.15 (0.23)	0.45 (0.62)
	1.00	0.10 (0.34)	0.14 (0.18)	0.40 (0.56)
	1.05	0.08 (0.29)	0.13 (0.15)	0.35 (0.51)
	1.10	0.06 (0.24)	0.12 (0.13)	0.30 (0.46)
	1.25	0.05 (0.14)	0.09 (0.12)	0.19 (0.33)
	1.50	0.03 (0.07)	0.05 (0.09)	0.08 (0.17)
	2.00	0.01 (0.04)	0.01 (0.04)	0.02 (0.04)
DISP	bas-MP2	CBS	aQZ	aTZ
	0.90	0.25 (0.87)	0.32 (1.09)	0.54 (1.42)
	0.95	0.22 (0.71)	0.26 (0.88)	0.40 (1.12)
	1.00	0.19 (0.62)	0.21 (0.71)	0.30 (0.90)
	1.05	0.17 (0.53)	0.18 (0.59)	0.23 (0.72)
	1.10	0.14 (0.45)	0.15 (0.50)	0.18 (0.58)
	1.25	0.08 (0.27)	0.09 (0.30)	0.09 (0.33)
	1.50	0.03 (0.11)	0.03 (0.12)	0.03 (0.13)
	2.00	0.01 (0.03)	0.01 (0.03)	0.01 (0.03)
MIX	bas-MP2	CBS	aQZ	aTZ
	0.90	0.17 (0.48)	0.13 (0.47)	0.28 (0.53)
	0.95	0.14 (0.40)	0.11 (0.41)	0.21 (0.44)
	1.00	0.12 (0.34)	0.09 (0.35)	0.16 (0.36)
	1.05	0.10 (0.29)	0.08 (0.30)	0.12 (0.31)
	1.10	0.08 (0.25)	0.07 (0.25)	0.10 (0.26)
	1.25	0.05 (0.16)	0.04 (0.15)	0.05 (0.16)
	1.50	0.03 (0.08)	0.02 (0.07)	0.02 (0.07)
	2.00	0.02 (0.03)	0.01 (0.02)	0.01 (0.02)

Table S6. MP2.X/bas-MP2/aDZ RMS binding energy errors and error ranges (in parentheses) for S66 binding categories (kcal/mol)

	MP2.X	bas-x=	aDZ	
Total	bas-MP2	CBS	aQZ	aTZ
	0.90	0.22 (1.19)	0.27 (1.30)	0.52 (1.55)
	0.95	0.19 (1.01)	0.24 (1.10)	0.43 (1.24)
	1.00	0.16 (0.86)	0.21 (0.93)	0.37 (1.03)
	1.05	0.14 (0.72)	0.18 (0.78)	0.31 (0.95)
	1.10	0.12 (0.61)	0.16 (0.65)	0.27 (0.88)
	1.25	0.08 (0.36)	0.11 (0.43)	0.16 (0.63)
	1.50	0.04 (0.21)	0.05 (0.24)	0.08 (0.31)
	2.00	0.01 (0.06)	0.01 (0.08)	0.02 (0.08)
HB	bas-MP2	CBS	aQZ	aTZ
	0.90	0.08 (0.29)	0.21 (0.22)	0.59 (0.78)
	0.95	0.05 (0.20)	0.21 (0.25)	0.53 (0.71)
	1.00	0.05 (0.14)	0.20 (0.26)	0.47 (0.66)
	1.05	0.06 (0.13)	0.19 (0.27)	0.42 (0.62)
	1.10	0.07 (0.15)	0.18 (0.27)	0.37 (0.57)
	1.25	0.08 (0.18)	0.14 (0.24)	0.25 (0.43)
	1.50	0.05 (0.15)	0.08 (0.17)	0.12 (0.23)
	2.00	0.02 (0.06)	0.02 (0.07)	0.03 (0.07)
DISP	bas-MP2	CBS	aQZ	aTZ
	0.90	0.34 (1.19)	0.38 (1.30)	0.57 (1.55)
	0.95	0.29 (1.01)	0.32 (1.10)	0.44 (1.24)
	1.00	0.24 (0.86)	0.27 (0.93)	0.35 (1.03)
	1.05	0.20 (0.72)	0.22 (0.78)	0.28 (0.86)
	1.10	0.17 (0.61)	0.18 (0.65)	0.22 (0.72)
	1.25	0.10 (0.36)	0.10 (0.39)	0.12 (0.42)
	1.50	0.04 (0.15)	0.04 (0.16)	0.04 (0.17)
	2.00	0.01 (0.04)	0.01 (0.04)	0.01 (0.04)
MIX	bas-MP2	CBS	aQZ	aTZ
	0.90	0.16 (0.57)	0.17 (0.57)	0.33 (0.61)
	0.95	0.13 (0.48)	0.14 (0.48)	0.26 (0.51)
	1.00	0.11 (0.41)	0.12 (0.41)	0.21 (0.43)
	1.05	0.09 (0.35)	0.10 (0.35)	0.17 (0.36)
	1.10	0.08 (0.30)	0.09 (0.30)	0.13 (0.31)
	1.25	0.05 (0.18)	0.05 (0.19)	0.07 (0.19)
	1.50	0.02 (0.09)	0.02 (0.08)	0.02 (0.09)
	2.00	0.01 (0.03)	0.01 (0.02)	0.03 (0.02)

Table S7. %RMS Errors for points along the dissociation curves of the S66 complexes for the MP2.X/CBS/6-31G*, MP2.X/aQZ/6-31G*, MP2.5/CBS/6-31G*, and MP2.5/aQZ/6-31G* methods.

MP2.X/CBS

	Total	H-bond	Dispersion	Mixed
0.9	34.90%	2.20%	58.70%	6.60%
0.95	7.70%	1.90%	12.30%	4.50%
1	5.80%	1.70%	9.00%	3.70%
1.05	5.00%	1.60%	7.80%	3.30%
1.1	4.70%	1.50%	7.20%	3.20%
1.25	4.40%	1.30%	6.70%	3.00%
1.5	4.80%	0.90%	7.50%	3.10%
2	8.40%	1.40%	13.80%	3.80%

MP2.X/aQZ

	Total	H-bond	Dispersion	Mixed
0.9	22.30%	4.60%	37.20%	5.80%
0.95	6.90%	3.70%	10.40%	4.10%
1	5.40%	3.20%	7.90%	3.40%
1.05	4.80%	2.80%	7.10%	3.00%
1.1	4.50%	2.60%	6.70%	2.90%
1.25	4.30%	2.00%	6.50%	2.70%
1.5	4.70%	1.20%	7.50%	2.80%
2	8.90%	1.00%	14.80%	3.40%

MP2.5/CBS

	Total	H-bond	Dispersion	Mixed
0.9	115.90%	1.20%	195.50%	18.80%
0.95	18.10%	0.90%	29.00%	10.80%
1	12.00%	0.80%	18.90%	8.10%
1.05	9.70%	0.80%	15.20%	6.80%
1.1	8.60%	0.80%	13.40%	6.00%
1.25	7.40%	0.90%	11.60%	5.10%
1.5	8.00%	0.70%	12.70%	5.00%
2	15.30%	1.10%	25.40%	5.40%

MP2.5/aQZ

	Total	H-bond	Dispersion	Mixed
0.9	100.60%	2.60%	169.90%	14.30%
0.95	15.70%	2.30%	25.30%	8.40%
1	10.60%	2.10%	16.90%	6.40%
1.05	8.80%	2.00%	13.80%	5.50%
1.1	7.90%	1.90%	12.40%	5.00%
1.25	7.10%	1.60%	11.10%	4.50%
1.5	7.90%	1.10%	12.60%	4.50%
2	15.80%	0.80%	26.40%	4.90%

Table S8. %RMS Errors for points along the dissociation curves of the S66 complexes for the MP2.X/CBS/6-311++G**, MP2.X/aQZ/6-311++G**, MP2.5/CBS/6-311++G**, and MP2.5/aQZ/6-311++G** methods.

MP2.X/CBS

	Total	H-bond	Dispersion	Mixed
0.9	28.50%	1.20%	47.80%	6.80%
0.95	6.50%	0.90%	10.20%	4.40%
1	4.80%	0.90%	7.50%	3.50%
1.05	4.20%	0.90%	6.50%	3.10%
1.1	3.90%	0.90%	6.00%	2.80%
1.25	3.70%	1.00%	5.60%	2.60%
1.5	4.10%	0.90%	6.40%	2.60%
2	13.20%	0.70%	21.90%	4.60%

MP2.X/aQZ

	Total	H-bond	Dispersion	Mixed
0.9	16.10%	2.80%	26.80%	4.60%
0.95	5.70%	2.40%	8.80%	3.40%
1	4.50%	2.20%	6.80%	2.90%
1.05	4.00%	2.00%	6.00%	2.60%
1.1	3.80%	1.90%	5.60%	2.40%
1.25	3.60%	1.70%	5.40%	2.20%
1.5	4.10%	1.40%	6.40%	2.30%
2	8.50%	0.90%	14.10%	2.60%

MP2.5/CBS

	Total	H-bond	Dispersion	Mixed
0.9	73.90%	1.20%	124.50%	13.30%
0.95	12.20%	0.70%	19.40%	7.80%
1	8.20%	0.50%	12.80%	5.90%
1.05	6.80%	0.50%	10.50%	4.90%
1.1	6.00%	0.60%	9.30%	4.40%
1.25	5.30%	0.80%	8.30%	3.80%
1.5	5.90%	0.70%	9.40%	3.70%
2	17.60%	0.50%	29.20%	6.00%

MP2.5/aQZ

	Total	H-bond	Dispersion	Mixed
0.9	58.70%	2.00%	99.10%	8.90%
0.95	10.00%	1.90%	16.10%	5.40%
1	7.00%	1.80%	11.10%	4.30%
1.05	5.90%	1.70%	9.30%	3.70%
1.1	5.40%	1.70%	8.50%	3.50%
1.25	5.10%	1.60%	7.90%	3.20%
1.5	5.80%	1.20%	9.30%	3.30%
2	12.30%	0.60%	20.60%	3.60%

Table S9. %RMS Errors for points along the dissociation curves of the S66 complexes for the MP2.X/CBS/aDZ, MP2.X/aQZ/aDZ, MP2.5/CBS/aDZ, and MP2.5/aQZ/aDZ methods.

MP2.X/CBS

	Total	H-bond	Dispersion	Mixed
0.9	40.90%	0.90%	69.00%	7.00%
0.95	7.40%	0.80%	11.90%	4.40%
1	5.30%	0.90%	8.20%	3.50%
1.05	4.40%	1.00%	6.90%	3.00%
1.1	4.00%	1.00%	6.20%	2.80%
1.25	3.60%	1.10%	5.40%	2.50%
1.5	3.50%	0.90%	5.40%	2.50%
2	4.50%	0.80%	7.10%	2.80%

MP2.X/aQZ

	Total	H-bond	Dispersion	Mixed
0.9	26.70%	3.00%	44.70%	5.90%
0.95	6.80%	2.60%	10.40%	4.30%
1	5.20%	2.40%	7.80%	3.60%
1.05	4.50%	2.20%	6.70%	3.20%
1.1	4.20%	2.10%	6.10%	3.00%
1.25	3.70%	1.90%	5.40%	2.60%
1.5	3.60%	1.40%	5.50%	2.40%
2	4.90%	0.70%	8.00%	2.40%

MP2.5/CBS

	Total	H-bond	Dispersion	Mixed
0.9	48.10%	0.90%	81.10%	7.90%
0.95	8.20%	0.80%	13.20%	4.80%
1	5.70%	0.80%	8.90%	3.70%
1.05	4.70%	0.90%	7.40%	3.20%
1.1	4.20%	1.00%	6.60%	2.90%
1.25	3.70%	1.10%	5.70%	2.60%
1.5	3.70%	0.90%	5.80%	2.60%
2	5.20%	0.80%	8.30%	2.90%

MP2.5/aQZ

	Total	H-bond	Dispersion	Mixed
0.9	33.50%	2.90%	56.30%	6.00%
0.95	7.20%	2.50%	11.20%	4.30%
1	5.40%	2.30%	8.20%	3.60%
1.05	4.70%	2.20%	7.00%	3.20%
1.1	4.30%	2.10%	6.30%	2.90%
1.25	3.80%	1.80%	5.70%	2.60%
1.5	3.80%	1.40%	5.90%	2.40%
2	5.60%	0.70%	9.20%	2.50%

Table S10. Average signed Errors for the MP2.X/CBS/6-31G*, MP2.X/aQZ/6-31G*, MP2.5/CBS/6-31G*, and MP2.5/aQZ/6-31G* methods.
(Positive/negative values indicate under/over-binding, respectively)

MP2.X/CBS

	Total	H-bond	Dispersion	Mixed
0.90	0.01	0.06	0.02	-0.06
0.95	0.00	0.07	-0.01	-0.06
1.00	0.00	0.07	-0.02	-0.05
1.05	0.00	0.07	-0.03	-0.04
1.10	0.00	0.07	-0.03	-0.04
1.25	0.00	0.05	-0.02	-0.03
1.50	-0.01	0.01	-0.01	-0.02
2.00	-0.01	-0.02	0.00	-0.01

MP2.X/aQZ

	Total	H-bond	Dispersion	Mixed
0.90	0.19	0.28	0.19	0.09
0.95	0.14	0.25	0.11	0.05
1.00	0.11	0.22	0.06	0.03
1.05	0.08	0.19	0.03	0.02
1.10	0.07	0.16	0.02	0.01
1.25	0.03	0.10	0.00	-0.01
1.50	0.01	0.03	-0.01	-0.01
2.00	-0.01	-0.01	0.00	-0.01

MP2.5/CBS

	Total	H-bond	Dispersion	Mixed
0.90	-0.33	-0.05	-0.60	-0.35
0.95	-0.25	-0.01	-0.47	-0.27
1.00	-0.19	0.02	-0.37	-0.22
1.05	-0.14	0.04	-0.29	-0.17
1.10	-0.11	0.05	-0.23	-0.14
1.25	-0.05	0.05	-0.12	-0.08
1.50	-0.02	0.02	-0.05	-0.04
2.00	-0.01	-0.01	-0.01	-0.02

MP2.5/aQZ

	Total	H-bond	Dispersion	Mixed
0.90	-0.15	0.17	-0.42	-0.20
0.95	-0.11	0.17	-0.34	-0.16
1.00	-0.08	0.16	-0.28	-0.13
1.05	-0.06	0.16	-0.23	-0.11
1.10	-0.04	0.14	-0.19	-0.09
1.25	-0.02	0.10	-0.11	-0.06
1.50	-0.01	0.05	-0.05	-0.03
2.00	-0.01	0.00	-0.01	-0.01

Table S11. Average signed Errors for the MP2.X/CBS/6-311++G**, MP2.X/aQZ/6-311++G**, MP2.5/CBS/6-311++G**, and MP2.5/aQZ/6-311++G** methods. (Positive/negative values indicate under/over-binding, respectively)

MP2.X/CBS

	Total	H-bond	Dispersion	Mixed
0.90	-0.06	-0.07	0.01	-0.11
0.95	-0.04	-0.04	0.00	-0.09
1.00	-0.03	-0.01	0.00	-0.07
1.05	-0.02	0.01	0.00	-0.06
1.10	-0.01	0.02	-0.01	-0.05
1.25	0.00	0.03	-0.01	-0.03
1.50	0.00	0.02	-0.01	-0.02
2.00	-0.01	0.00	-0.01	-0.01

MP2.X/aQZ

	Total	H-bond	Dispersion	Mixed
0.90	0.13	0.15	0.18	0.04
0.95	0.10	0.14	0.12	0.02
1.00	0.08	0.13	0.08	0.01
1.05	0.07	0.13	0.06	0.01
1.10	0.05	0.12	0.04	0.00
1.25	0.03	0.09	0.01	-0.01
1.50	0.01	0.04	0.00	-0.01
2.00	0.00	0.01	0.00	-0.01

MP2.5/CBS

	Total	H-bond	Dispersion	Mixed
0.90	-0.23	-0.10	-0.33	-0.26
0.95	-0.17	-0.06	-0.25	-0.20
1.00	-0.12	-0.02	-0.20	-0.16
1.05	-0.09	0.00	-0.15	-0.12
1.10	-0.07	0.02	-0.12	-0.10
1.25	-0.03	0.04	-0.07	-0.06
1.50	-0.01	0.03	-0.03	-0.03
2.00	-0.01	0.00	-0.01	-0.02

MP2.5/aQZ

	Total	H-bond	Dispersion	Mixed
0.90	-0.05	0.12	-0.15	-0.11
0.95	-0.03	0.12	-0.13	-0.09
1.00	-0.02	0.13	-0.11	-0.07
1.05	-0.01	0.12	-0.09	-0.06
1.10	0.00	0.12	-0.08	-0.05
1.25	0.00	0.09	-0.05	-0.04
1.50	0.00	0.05	-0.03	-0.02
2.00	0.00	0.01	-0.01	-0.01

Table S12. Average signed Errors for the MP2.X/CBS/aDZ, MP2.X/aQZ/aDZ, MP2.5/CBS/aDZ, and MP2.5/aQZ/aDZ methods.
(Positive/negative values indicate under/over-binding, respectively)

MP2.X/CBS

	Total	H-bond	Dispersion	Mixed
0.90	-0.03	-0.02	0.00	-0.07
0.95	0.00	0.01	0.01	-0.04
1.00	0.01	0.04	0.02	-0.03
1.05	0.02	0.05	0.03	-0.02
1.10	0.03	0.06	0.03	-0.01
1.25	0.03	0.06	0.02	-0.01
1.50	0.01	0.04	0.01	-0.01
2.00	0.00	0.00	0.00	-0.01

MP2.X/aQZ

	Total	H-bond	Dispersion	Mixed
0.90	0.15	0.20	0.17	0.08
0.95	0.14	0.19	0.14	0.07
1.00	0.12	0.18	0.11	0.06
1.05	0.11	0.17	0.09	0.05
1.10	0.09	0.16	0.07	0.04
1.25	0.06	0.12	0.03	0.02
1.50	0.02	0.06	0.01	0.00
2.00	0.00	0.01	0.00	0.00

MP2.5/CBS

	Total	H-bond	Dispersion	Mixed
0.90	-0.06	-0.03	-0.06	-0.09
0.95	-0.03	0.01	-0.03	-0.06
1.00	0.00	0.04	-0.01	-0.04
1.05	0.01	0.05	0.00	-0.03
1.10	0.02	0.06	0.01	-0.02
1.25	0.02	0.06	0.01	-0.01
1.50	0.01	0.04	0.00	-0.01
2.00	0.00	0.00	0.00	-0.01

MP2.5/aQZ

	Total	H-bond	Dispersion	Mixed
0.90	0.12	0.19	0.12	0.05
0.95	0.11	0.19	0.09	0.05
1.00	0.10	0.18	0.08	0.04
1.05	0.09	0.17	0.06	0.03
1.10	0.08	0.16	0.05	0.03
1.25	0.05	0.12	0.02	0.01
1.50	0.02	0.06	0.01	0.00
2.00	0.00	0.01	0.00	-0.01

Table S13. MP2.5 RMS errors in intermolecular distance (in terms of % of the reference equilibrium distance) and ranges of errors (in parentheses) for S66 interaction categories and subcategories.

MP2.5			
	bas-MP2=	CBS	
bas-x	aDZ	6-311++G**	6-31G*
Total	0.53 (2.47)	0.76 (2.56)	1.05 (3.17)
H-bond	0.34 (0.52)	0.44 (0.46)	0.37 (0.60)
HB (single)	0.27 (0.43)	0.39 (0.36)	0.35 (0.60)
HB (cyclic)	0.47 (0.08)	0.58 (0.18)	0.45 (0.15)
Dispersion	0.70 (2.47)	0.96 (2.56)	1.45 (3.17)
Disp (π-π)	0.91 (1.79)	1.35 (1.86)	2.03 (2.45)
Disp (A-A)	0.51 (0.45)	0.25 (0.58)	0.26 (0.66)
Disp (π-A)	0.41 (0.97)	0.57 (0.92)	0.93 (1.20)
Mixed	0.46 (1.37)	0.79 (1.36)	1.01 (1.70)

MP2.5			
	bas-MP2=	aQZ	
bas-x	aDZ	6-311++G**	6-31G*
Total	0.50 (2.33)	0.51 (2.49)	0.79 (3.12)
H-bond	0.23 (0.58)	0.13 (0.51)	0.22 (0.61)
HB (single)	0.26 (0.45)	0.13 (0.38)	0.25 (0.61)
HB (cyclic)	0.03 (0.08)	0.14 (0.20)	0.07 (0.17)
Dispersion	0.70 (2.22)	0.74 (2.49)	1.16 (3.12)
Disp (π-π)	0.62 (1.70)	0.97 (1.77)	1.65 (2.38)
Disp (A-A)	0.92 (0.46)	0.61 (0.59)	0.36 (0.67)
Disp (π-A)	0.64 (1.12)	0.40 (1.06)	0.62 (1.30)
Mixed	0.45 (1.66)	0.46 (1.54)	0.67 (1.89)

MP2.5			
	bas-MP2=	aTZ	
bas-x	aDZ	6-311++G*	6-31G*
Total	1.00 (2.47)	0.75 (2.46)	0.83 (3.19)
H-bond	0.90 (0.88)	0.76 (0.81)	0.87 (0.83)
HB (single)	0.96 (0.74)	0.82 (0.63)	0.92 (0.83)
HB (cyclic)	0.62 (0.13)	0.51 (0.22)	0.65 (0.19)
Dispersion	1.10 (2.10)	0.82 (2.37)	0.95 (3.08)
Disp (π-π)	0.66 (1.58)	0.59 (1.66)	1.15 (2.34)
Disp (A-A)	1.52 (0.51)	1.20 (0.60)	0.91 (0.69)
Disp (π-A)	1.21 (1.35)	0.78 (1.28)	0.58 (1.53)
Mixed	0.99 (2.01)	0.66 (1.85)	0.65 (2.21)

Table S14. MP2.X RMS errors in intermolecular distance (in terms of % of the reference equilibrium distance) and ranges of errors (in parentheses) for S66 interaction categories and subcategories.

MP2.X			
	bas-MP2=	CBS	
bas-x	aDZ	6-311++G**	6-31G*
Total	0.49 (2.19)	0.42 (1.88)	0.40 (2.16)
H-bond	0.31 (0.54)	0.33 (0.52)	0.23 (0.83)
HB (single)	0.25 (0.42)	0.23 (0.28)	0.22 (0.74)
HB (cyclic)	0.46 (0.08)	0.54 (0.17)	0.25 (0.12)
Dispersion	0.64 (2.19)	0.48 (1.88)	0.55 (2.16)
Disp (π-π)	0.80 (1.74)	0.57 (1.53)	0.69 (2.16)
Disp (A-A)	0.56 (0.44)	0.49 (0.49)	0.39 (0.52)
Disp (π-A)	0.43 (0.96)	0.35 (0.86)	0.43 (1.02)
Mixed	0.45 (1.48)	0.43 (1.21)	0.34 (1.25)

MP2.X			
	bas-MP2=	aQZ	
bas-x	aDZ	6-311++G**	6-31G*
Total	0.52 (2.25)	0.45 (1.79)	0.59 (2.24)
H-bond	0.25 (0.61)	0.24 (0.56)	0.56 (0.84)
HB (single)	0.28 (0.45)	0.26 (0.31)	0.63 (0.73)
HB (cyclic)	0.03 (0.07)	0.11 (0.19)	0.22 (0.14)
Dispersion	0.71 (2.11)	0.63 (1.79)	0.67 (2.08)
Disp (π-π)	0.55 (1.65)	0.46 (1.43)	0.63 (2.08)
Disp (A-A)	0.97 (0.44)	0.89 (0.50)	0.79 (0.53)
Disp (π-A)	0.70 (1.09)	0.61 (0.87)	0.65 (0.96)
Mixed	0.48 (1.65)	0.38 (1.38)	0.49 (1.40)

MP2.X			
	bas-MP2=	aTZ	
bas-x	aDZ	6-311++G*	6-31G*
Total	1.05 (2.39)	1.00 (1.90)	1.17 (2.39)
H-bond	0.92 (0.89)	0.89 (0.85)	1.25 (0.99)
HB (single)	0.99 (0.73)	0.97 (0.54)	1.34 (0.83)
HB (cyclic)	0.62 (0.13)	0.55 (0.19)	0.84 (0.14)
Dispersion	1.16 (1.99)	1.14 (1.66)	1.15 (1.98)
Disp (π-π)	0.74 (1.53)	0.86 (1.30)	0.96 (1.98)
Disp (A-A)	1.58 (0.50)	1.49 (0.50)	1.38 (0.54)
Disp (π-A)	1.29 (1.31)	1.20 (1.01)	1.21 (1.10)
Mixed	1.05 (2.00)	0.94 (1.72)	1.11 (1.74)