Supporting Information

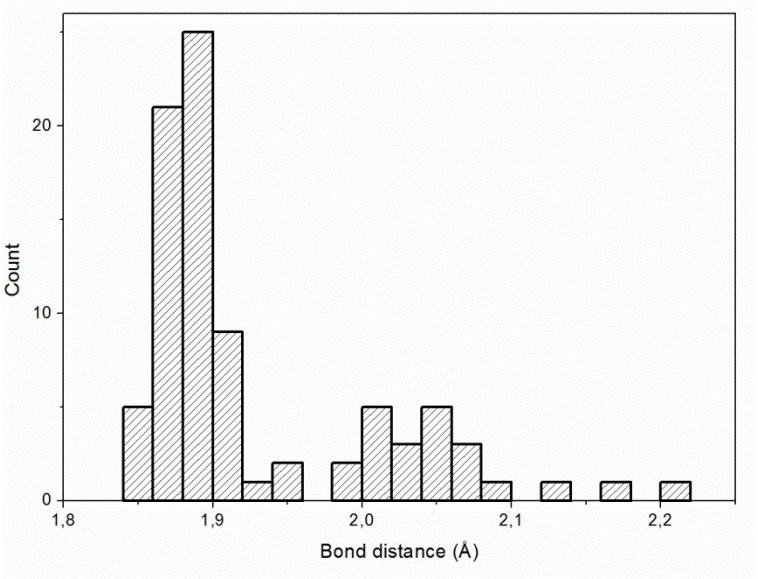

Figure S1. Histogram for 6-coordinate Co-O(DBSQ) bonds from the CSD.

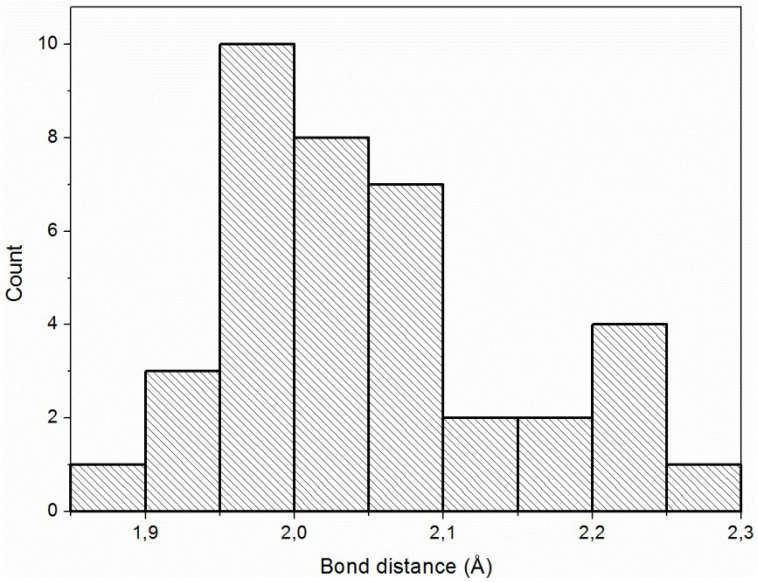

Figure S2. Histogram for 5-coordinate Co-O(DBSQ) bonds from the CSD. Some of the underlying geometries are rather far from the trigonal bipyramidal geometry of **1.**

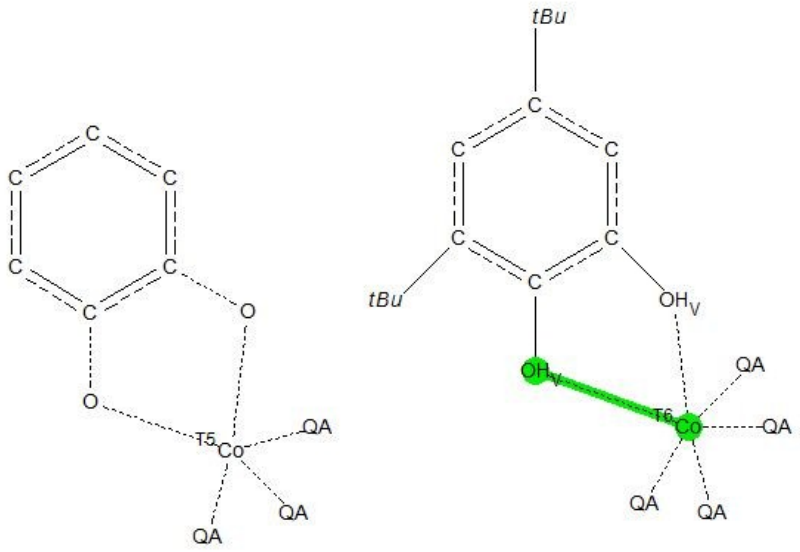

Figure S3. Queries used for the CSD search.

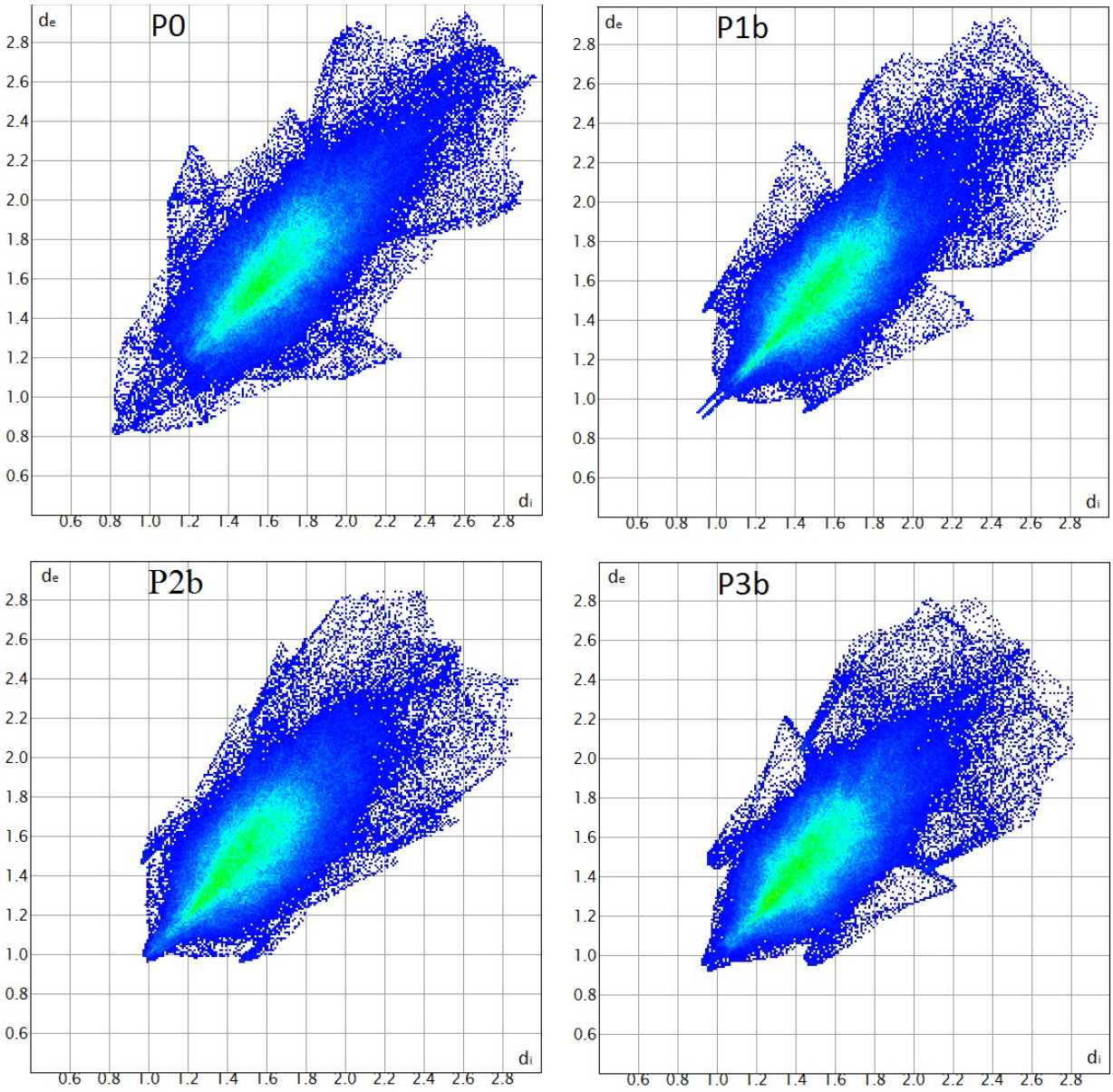

Figure S4: Fingerprint plots of P0, P1b, P2b and P3b. The color indicates – on a relative scale – contribution of the interaction in question, with a small contribution being blue, medium green and a high contribution red.

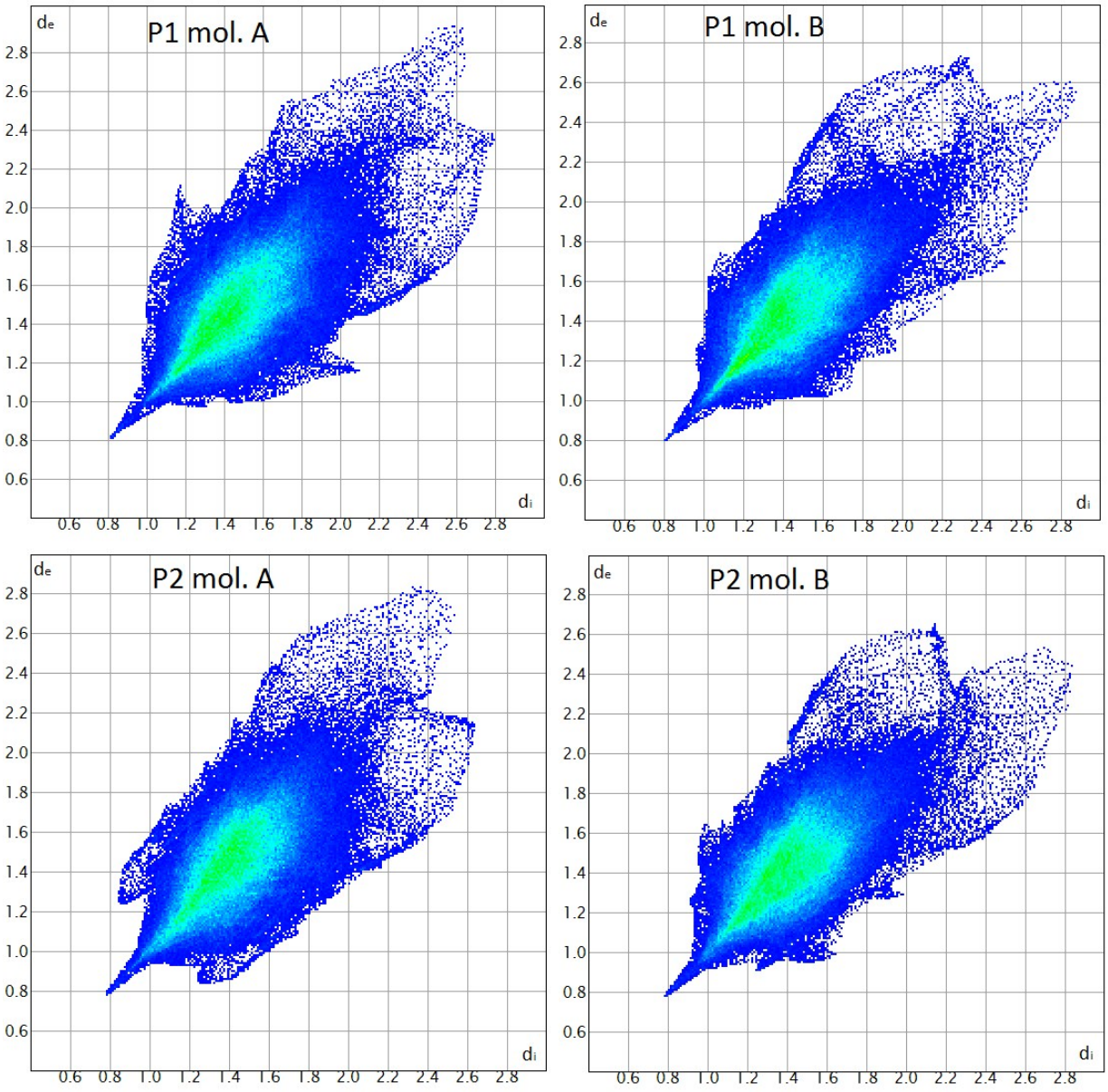

Figure S5: Fingerprint plots of P1 molecule A and B and P2 molecule A and B. The color indicates – on a relative scale – contribution of the interaction in question, with a small contribution being blue, medium green and a high contribution red.

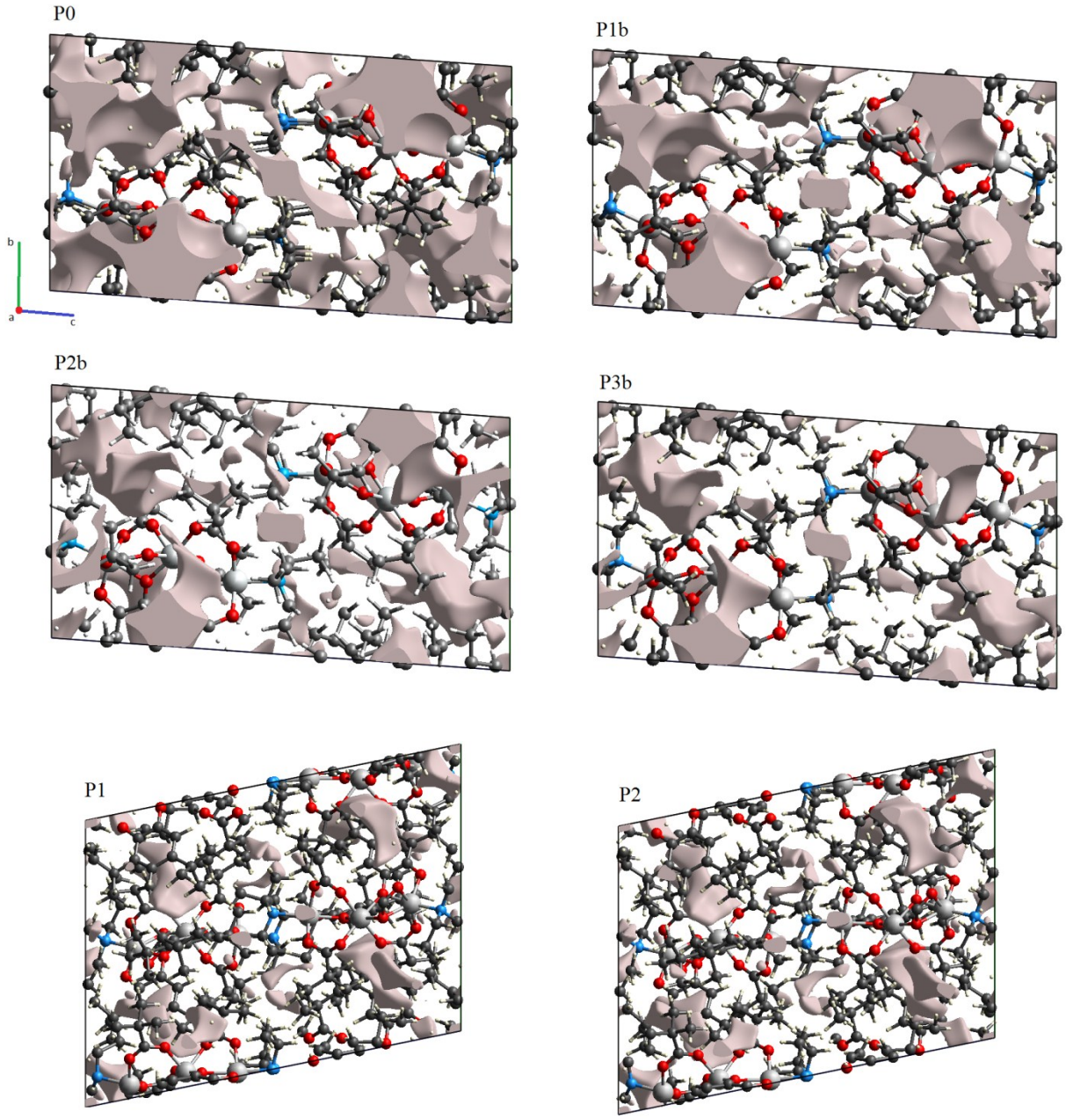

Figure S6: Void spaces calculated with an isovalue of 0.001 e au^{-3} for P0, P1b, P2b, P3b, P1 and P2.

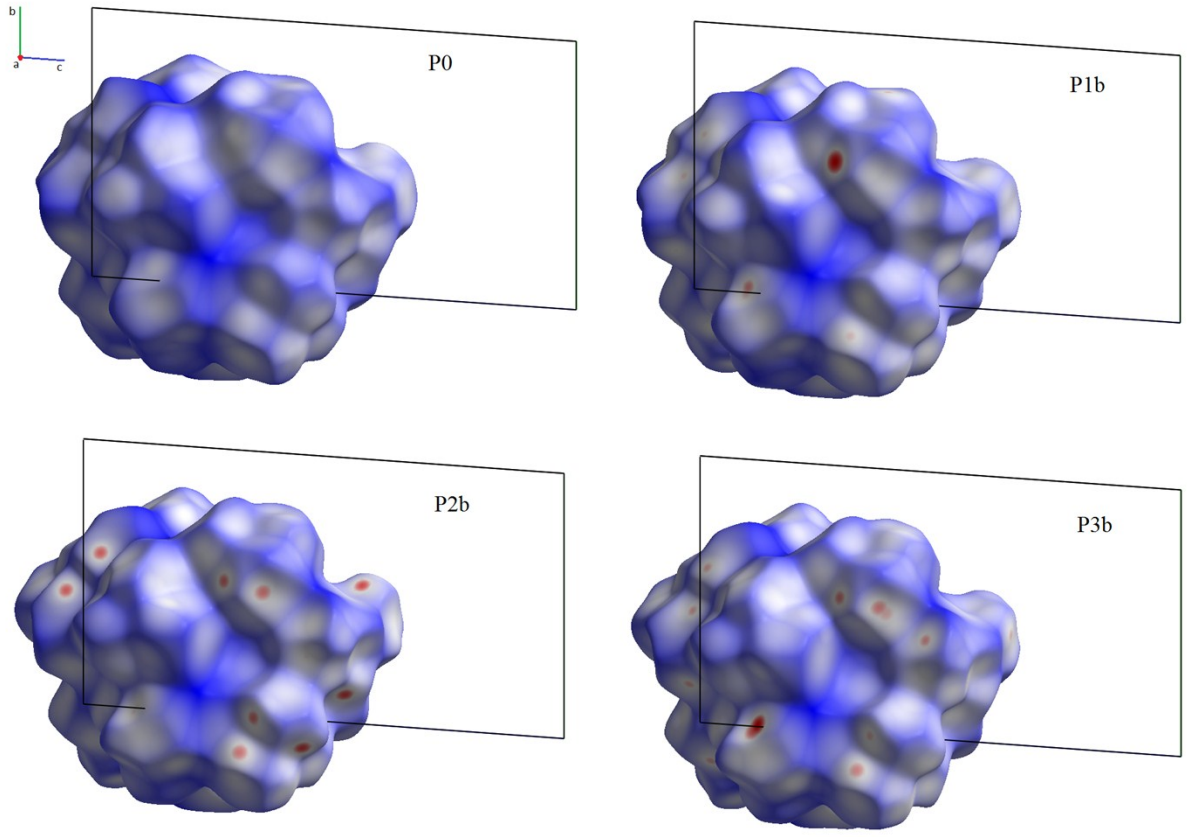

Figure S7: Hirshfeld surfaces for P0, P1b, P2b and P3b (isovalue of 0.5).

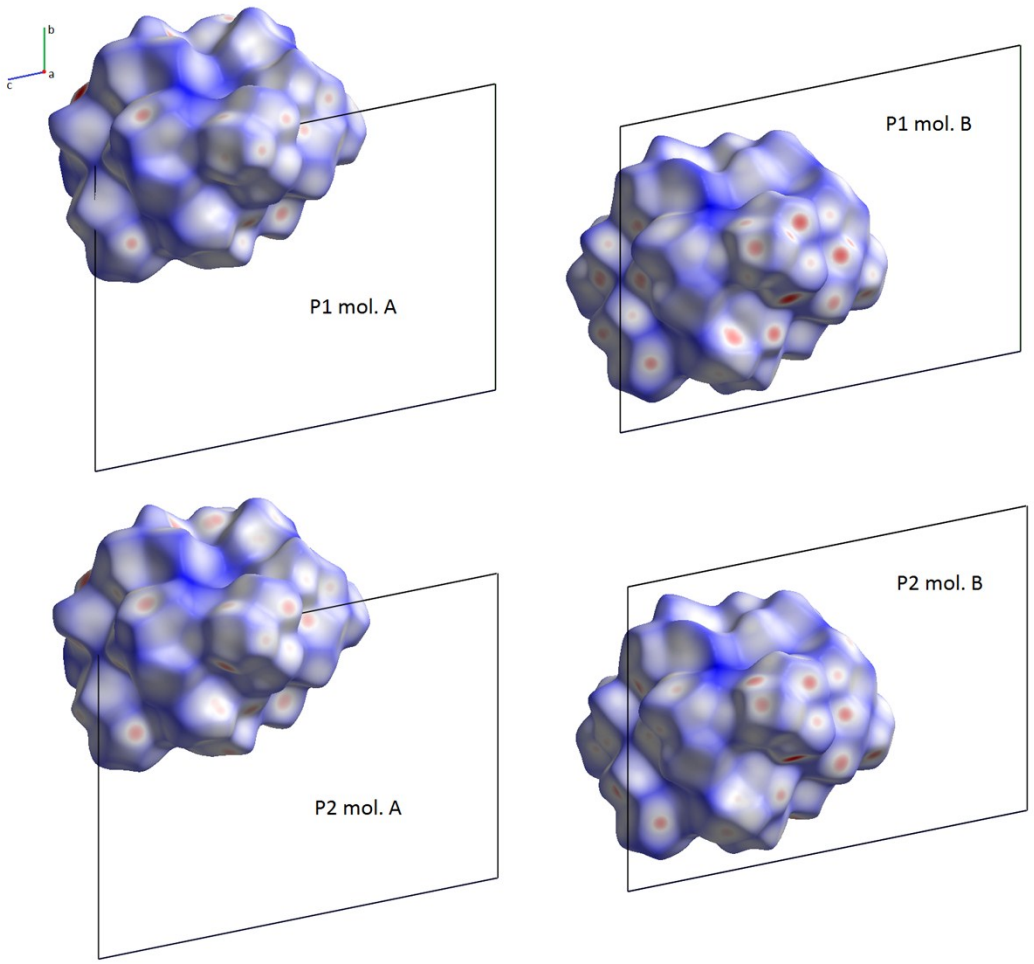

Figure S8: Hirshfeld surfaces for P1 molecule A and B and P2 molecule A and B (isovalue of 0.5).

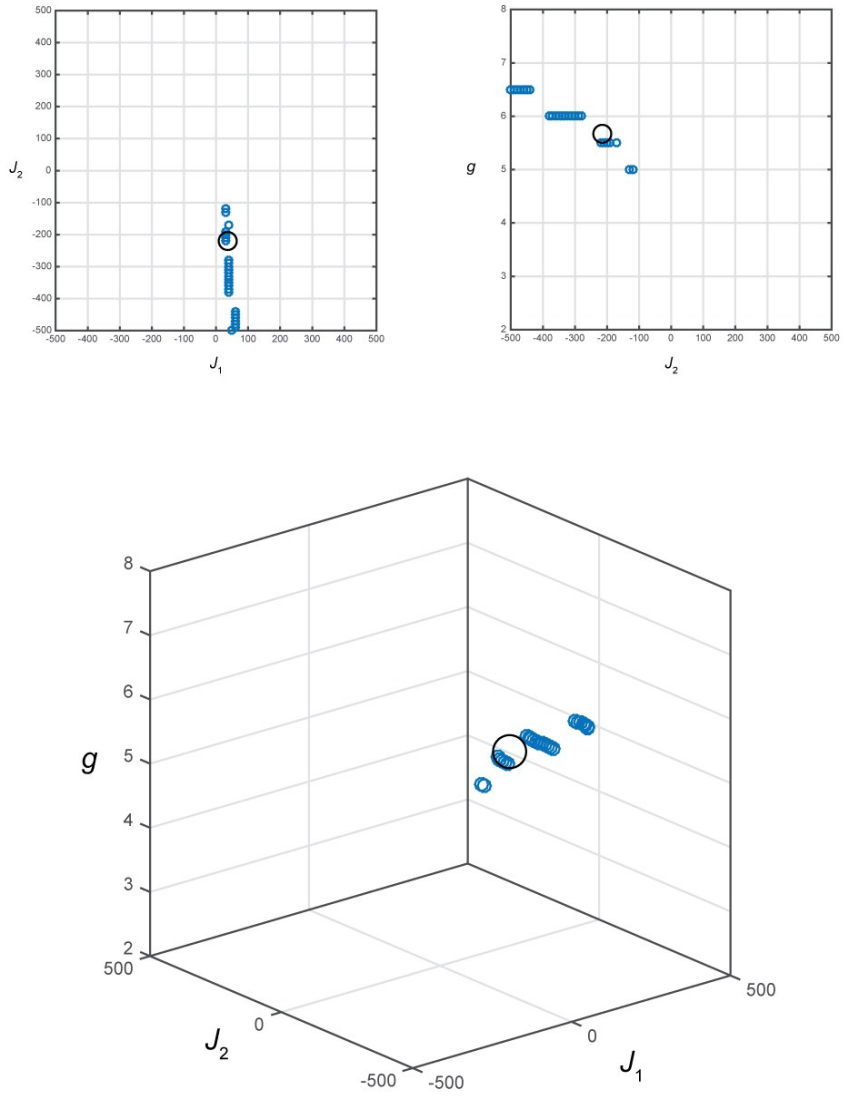

Figure S9: Three dimensional plot of the parameters from the 25 simulations (blue circles) with the lowest residual error against the experimental magnetic susceptibility after a survey over the following parameter space: J_1 = −500 to +500 cm^{-1} in steps of 10 cm^{-1}; J_2 = −500 to +500 cm^{-1} in steps of 10 cm^{-1}; g = 2 to 8 in steps of 0.5. Total number of simulations: 132,613. The black circle indicates the parameters arrived at from the free refinement. The same solution is arrived at when using each of the best survey parameters as starting points, suggesting that this solution is the global minimum within the surveyed parameter range. There appears to be some correlation between J_2 and g, but solutions with g > 6 are physically unrealistic, and are noticeably poorer fits than those close to the global minimum. The survey has been performed using the program PHI.

Table S1: Crystallographic tables for both temperature and high-pressure data.

	20 K	**40 K**	**60 K**	**80 K**
Formula	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$
M_r	1224.25	1224.25	1224.25	1224.25
Crystal system	Triclinic	Triclinic	Triclinic	Triclinic
Space group	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$
a (Å)	11.12620(10)	11.12530(10)	11.1410(5)	11.1496(5)
b (Å)	13.0303(2)	13.0300(2)	13.0395(5)	13.0524(6)
c (Å)	23.6869(2)	23.6966(2)	23.7190(10)	23.7319(11)
α (deg.)	84.9760(10)	84.9960(10)	84.992(6)	84.994(6)
β (deg.)	80.6660(10)	80.7750(10)	80.834(6)	80.842(6)
γ (deg.)	80.6530(10)	80.6510(10)	80.683(6)	80.675(6)
V ($Å^3$)	3337.00(7)	3339.05(7)	3350.2(3)	3357.9(3)
Z	2	2	2	2
λ (Å)	0.4997	0.4997	0.4997	0.4997
μ (mm^{-1})	0.304	0.304	0.303	0.302
T_{max} / T_{min}	-	-	-	-
F(000)	1314	1314	1314	1314
d_{min} (Å)	0.770	0.770	0.770	0.770
Total reflections	32261	32352	32458	32565
Unique reflections	15267	15285	15327	15361
Redundancy	2.113	2.117	2.118	2.120
No. of discarded reflections	30	22	18	13
No. of parameters	862	862	862	862
R_{int}	0.0465	0.0469	0.0555	0.0609
Mean I/σ	10.6344	10.5593	9.4860	8.8580
Completeness	0.9981	0.9986	0.9986	0.9985
GOF	1.139	1.089	1.095	1.145
R_{all}	0.0649	0.0689	0.0694	0.0715
wR_2	0.1179	0.1140	0.1201	0.1176
Max peak	0.87	0.87	0.78	0.72
Min peak	-0.82	-0.86	-0.81	-0.70
CCDC number	1481083	1481084	1481085	1481086

	90 K	**100 K**	**125 K**	**150 K**
Formula	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$
M_r	1224.25	1224.25	1224.25	1224.25
Crystal system	Triclinic	Triclinic	Triclinic	Triclinic
Space group	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$
a (Å)	11.1631(3)	11.1736(3)	11.1802(2)	11.1939(5)
b (Å)	13.0585(3)	13.0734(4)	13.0996(2)	13.1136(6)
c (Å)	23.7733(5)	23.8027(7)	23.8188(4)	23.7996(8)
α (deg.)	84.9778(18)	85.002(2)	84.9528(14)	84.866(3)
β (deg.)	80.6354(18)	80.744(2)	80.7062(16)	80.750(3)
γ (deg.)	80.772(2)	80.730(3)	80.7735(15)	80.803(4)
V ($Å^3$)	3368.36(13)	3380.26(18)	3391.17(11)	3396.5(2)
Z	2	2	2	2
λ (Å)	0.71073	0.71073	0.71073	0.71073
μ (mm^{-1})	0.785	0.782	0.779	0.778
T_{max} / T_{min}	0.873 / 0.820	1.000 / 0.8147	1.000 / 0.9143	1.000 / 0.8617
F(000)	1314	1314	1314	1314
d_{min} (Å)	0.656	0.700	0.655	0.654
Total reflections	86650	87493	86836	87449
Unique reflections	23219	20615	23350	23410
Redundancy	3.732	4.244	3.720	3.736
No. of discarded reflections	17	4447	35	64
No. of parameters	862	862	862	862
R_{int}	0.0420	0.0488	0.0508	0.0386
Mean I/σ	15.3931	14.3932	13.0721	15.4194
Completeness	0.9286	0.9998	0.9257	0.9219
GOF	1.049	1.053	1.225	1.254
R_{all}	0.0572	0.0539	0.0629	0.0634
wR_2	0.0973	0.0966	0.1201	0.1185
Max peak	1.17	1.09	0.79	0.84
Min peak	-0.79	-0.95	-0.69	-0.55
CCDC number	1481087	1481088	1481089	1481090

	175 K	**200 K**	**225 K**
Formula	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$
M_r	1224.25	1224.25	1224.25
Crystal system	Triclinic	Triclinic	Triclinic
Space group	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$
a (Å)	11.1908(2)	11.2147(4)	11.2253(3)
b (Å)	13.1253(2)	13.1564(5)	13.1918(5)
c (Å)	23.8212(4)	23.8767(8)	23.8945(6)
α (deg.)	84.9081(14)	84.904(3)	84.898(3)
β (deg.)	80.7814(17)	80.795(3)	80.845(2)
γ (deg.)	80.7381(16)	80.717(3)	80.685(3)
V (Å^3)	3401.59(11)	3424.8(2)	3439.93(19)
Z	2	2	2
λ (Å)	0.71073	0.71073	0.71073
μ (mm^{-1})	0.777	0.772	0.768
T_{max} / T_{min}	1.000 / 0.8692	1.000 / 0.8777	-
F(000)	1314	1314	1314
d_{min} (Å)	0.655	0.654	0.653
Total reflections	87799	88254	86830
Unique reflections	23526	23633	23793
Redundancy	3.732	3.734	3.650
No. of discarded reflections	36	19	13
No. of parameters	862	862	862
R_{int}	0.0388	0.0372	0.1844
Mean I/σ	14.5528	14.5813	4.5501
Completeness	0.9307	0.9208	0.9187
GOF	1.305	1.353	1.422
R_{all}	0.0705	0.0761	0.1677
wR_2	0.1282	0.1347	0.2399
Max peak	0.88	1.16	1.42
Min peak	-0.45	-0.57	-1.92
CCDC number	1481091	1481092	1481093

	296K (P0)	**0.67GPa (P1b)**	**0.95 GPa (P2b)**	**1.34 GPa (P3b)**
Formula	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$
M_r	1224.25	1224.25	1224.25	1224.25
Crystal system	Triclinic	Triclinic	Triclinic	Triclinic
Space group	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$
a (Å)	11.275(4)	10.9575(16)	10.664(3)	10.472(3)
b (Å)	13.288(4)	12.7764(13)	12.768(2)	12.763(3)
c (Å)	24.031(7)	23.2676(13)	23.2107(19)	23.102(2)
α (deg.)	84.662(9)	84.691(7)	84.712(10)	84.771(12)
β (deg.)	81.022(9)	81.325(8)	81.583(13)	81.525(15)
γ (deg.)	80.548(9)	80.721(10)	81.279(19)	81.41(2)
V ($Å^3$)	3499.4(19)	3170.2(6)	3082.0(11)	3012.4(12)
Z	2	2	2	2
λ (Å)	0.56086	0.71073	0.71073	0.71073
μ (mm^{-1})	0.290	0.329	0.329	0.337
T_{max} / T_{min}	0.7445 / 0.7005	1.000 / 0.6309	1.000 / 0.5115	1.000 / 0.6015
F(000)	1314	1314	1314	1314
d_{min} (Å)	0.799	1.00	1.00	1.00
Total reflections	36372	15479	15433	14705
Unique reflections	11942	2523	2447	2410
Redundancy	3.05	6.14	6.13	6.10
No. of discarded reflections	0	0	0	0
No. of parameters	862	712	712	712
R_{int}	0.0350	0.1184	0.1181	0.1312
Mean I/σ	15.5	8.4	8.4	6.8
Completeness	0.8335	0.3803	0.3794	0.3818
GOF	1.057	1.073	1.068	1.042
R_{all}	0.0683	0.1341	0.1410	0.1705
wR_2	0.1587	0.2194	0.2389	0.3297
Max peak	0.94	0.52	0.41	0.41
Min peak	-0.47	-0.36	-0.29	-0.51
CCDC number	1488918	1481096	1481097	1481098

Crystallographic Data				
	1.82 GPa (P4b)	**2.01 GPa (P5b)**	**2.50 GPa (P6b)**	**2.85 GPa (P7b)**
Formula	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$
M_r	1224.25	1224.25	1224.25	1224.25
Crystal system	Triclinic	Triclinic	Triclinic	Triclinic
Space group	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$
a (Å)	10.650(6)	10.588(4)	10.673(10)	10.588(11)
b (Å)	12.256(5)	12.167(4)	12.082(6)	11.989(7)
c (Å)	22.942(3)	22.914(3)	22.903(5)	22.837(6)
α (deg.)	84.357(17)	84.430(18)	84.13(3)	84.16(3)
β (deg.)	81.72(2)	81.88(2)	81.41(4)	81.64(4)
γ (deg.)	80.93(4)	81.25(3)	80.83(6)	81.17(7)
V ($Å^3$)	2917(2)	2879.5(16)	2874(3)	2825(4)
Z	2	2	2	2
λ (Å)	0.71073	0.71073	0.71073	0.71073

Crystallographic Data			
	3.94 GPa (P8b)	**1.99 GPa (P1)**	**2.57 GPa (P2)**
Formula	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$	$Co_3O_{12}N_2C_{60}H_{106}$
M_r	1224.25	1224.25	1224.25
Crystal system	Triclinic	Triclinic	Triclinic
Space group	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$
a (Å)	9.297(9)	14.6567(16)	14.5270(19)
b (Å)	12.996(8)	17.3207(17)	17.177(2)
c (Å)	22.396(5)	23.808(2)	23.735(2)
α (deg.)	84.19(3)	99.402(6)	99.475(8)
β (deg.)	81.70(4)	105.188(7)	105.533(8)
γ (deg.)	84.08(6)	96.155(6)	95.668(8)
V ($Å^3$)	2653(3)	5682.6(10)	5564.5(12)
Z	2	4	4
λ (Å)	0.71073	0.56086	0.56086
μ (mm^{-1})	-	0.490	0.501
T_{max} / T_{min}	-	0.7440 / 0.6087	0.7440 / 0.6054
F(000)	-	1314	1314
d_{min} (Å)	-	0.992	0.992
Total reflections	-	30706	31445
Unique reflections	-	5177	5043
Redundancy	-	5.93	6.24
No. of discarded reflections	-	0	0
No. of parameters	-	702	692
R_{int}	-	0.1115	0.1682
Mean I/σ	-	10.0	7.3
Completeness	-	0.4246	0.4218
GOF	-	1.026	1.034
R_{all}	-	0.1038	0.1464
wR_2	-	0.1778	0.2534
Max peak	-	0.43	0.62
Min peak	-	-0.37	-0.38
CCDC number		1481094	1481095

Table S2: The different EoS fits performed with V_0 being the unit cell volume at zero applied pressure, K_0 is the bulk modulus also at zero applied pressure, K' is the first derivative of the bulk modulus and χ^2 denotes the agreement factor of the fit.

Model	Data	χ^2	V_0 (Å^3)	K_0 (GPa)	K'
Murnaghan	P1b-P5b	2.54	3499.4(30)	4.61(33)	7.55(69)
Birch-Murnaghan, 3rd order	P1b-P5b	2.09	3499.5(27)	4.08(41)	11.1(18)
Vinet, 3rd order	P1b-P5b	2.23	3499.4(28)	4.31(32)	9.4(9)
Tait, 3rd order	P1b-P5b	2.33	3499.4(28)	4.45(30)	8.42(67)
Natural strain, 3rd order	P1b-P5b	1.80	3499.5(25)	3.54(25)	16.7(10)
Murnaghan	P6b-P8b	More parameters than data points!			
Birch-Murnaghan, 2nd order	P6b-P8b	1.13	2877.9(74)	15.3(10)	4.00 (fixed)
Vinet, 2nd order	P6b-P8b	0.39	2875.5(62)	17.4(9)	1.00 (fixed)
Tait, 2nd order	P6b-P8b	1.12	2877.9(74)	15.3(10)	4.00 (fixed)
Natural strain, 2nd order	P6b-P8b	0.60	2876.3(65)	16.71(97)	2.00 (fixed)

Table S3: The percentage distribution of intermolecular interactions covering the Hirshfeld surface at different pressures.

	H-H (%)	C-H (%)	O-H (%)
P0	98.7	0.3	0.9
P1b	98.4	0.4	1.3
P2b	97.8	0.8	1.4
P3b	97.2	0.9	1.8
P1 mol. A	94.9	1.9	3.2
P1 mol. B	96.1	0.9	2.9
P2 mol. A	93.8	2.4	3.4
P2 mol. B	95.8	1.0	3.2