Exploring the Binding of Barbital to a Synthetic Macrocyclic Receptor; a Charge Density Study†

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Spectroscopic Details of 14,16-dioxa-2,6,8,22-tetraaza-1,7(2,6)-dipyridina-15(2,7)-naphthalena-4(1,3)-benzenacyclodocosaphane-3,5,9,21-tetraone (**1**)

^{1}H-NMR (d^{6}-DMSO) δ ppm 10.28 (s, 2H, NH isophthal), 10.06 (s, 2H, pyr-NHCO), 8.50 (s, 1H, isophthal C(2)-H), 8.10 (dd, J = 1.5, 7.7, 2H, isophthal C(4)-H and C(6)-H), 7.83 (m, 4H, pyr C(3)-H and C(5)-H), 7.77 (d, J=1.9, 7, 2H, pyr C(4)-H) 7.69 (d, J=7.7, 1H, isophth-5H) 7.65 (d, J= 9, 2H, naph-C(4)-H and naph-C(5)-H), 7.19 (d, J= 2.3, 2H naph-C(1)-H and naph-C(8)-H), 6.95 (dd, J = 2.3, 8.8, 2H, naph-C(3)-H and C(6)-H), 4.05 (t, J= 6.3, 4H, OC$\underline{H}_2$), 2.30 (t, J=6.2, 4H, COC$\underline{H}_2$), 1.67-1.85 (8H, m, OCH$_2$C$\underline{H}_2$C$\underline{H}_2$CH$_2$)

^{13}C-NMR (d^{6}-DMSO) δ ppm 172.30 (pyr-NH$\underline{C}$O), 165.79 (isophthal-$\underline{C}$O), 157.31 (napth-$\underline{C}$O), 150.90 and 150.47 (pyr C2 & C6), 140.55 (pyr C4), 136.20 (napth C9), 134.26 (napth C4 & C5), 132.33 (napth C4 & C5), 129.46 (isophthal C5), 129.38 (napth C10), 124.08 (isophthal C2), 116.36 (napth C1 & C8), 110.29 and 109.99 (pyr C), 106.50 (napth C3 & C5), 67.50 (O$\underline{C}$H$_2$), 33.73 ($\underline{C}$H$_2$CO), 28.52 (OCH$_2\underline{C}$H$_2$CH$_2$), 21.70 ($\underline{C}$H$_2$CH$_2$CO).

Anisotropic Temperature Refinement of Hydrogen Atoms

An alternative multipole refinement was carried out using anisotropic temperature factors for the hydrogen atoms as well as the heavy atoms, as discussed by Hoser *et al.*[1] regarding the increased accuracy of the multipole model obtained from multipole refinement involving the use of anisotropic temperature factors for hydrogen.[1-2] Anisotropic temperature factors for the hydrogen atoms were calculated using the SHADE3 server developed by Madsen[3]. Multipole analyses with anisotropic temperature factors for hydrogen were truncated at the same level as above (l_{max} = 3) for heavy atoms and up to the (l_{max} = 1) for hydrogen atoms). The multipole refinement for hydrogen atoms was stopped at the dipolar expansion as the observations reported by Nguyen *et al.*[4] illustrated that refinement of hydrogen atoms in the multipole model past the dipolar to the quadrupolar level was generally unnecessary and would not lead to improved modelling of the electron density. However, it should be noted that the decision to refine hydrogen atoms anisotropically should be made on a per case basis.

Table S1: Calculated hydrogen anisotropic displacement parameters for (**1**)

Atom	U11	U22	U33	U12	U13	U23
H1	0.016809	0.01007	0.012283	-0.001387	-0.006679	-0.00228
H3	0.013246	0.014243	0.015196	0.003922	-0.007011	0.004152
H4	0.016506	0.010328	0.012314	-0.001364	-0.006771	-0.002263
H5	0.018448	0.00582	0.017293	0.004041	-0.002877	0.001126
H9	0.017596	0.015216	0.006662	0.004804	0.000121	-0.002916
H10	0.012206	0.015184	0.014651	0.009057	0.000482	0.002048
H11	0.015168	0.019252	0.008285	0.004872	-0.004434	0.003702
H14A	0.019162	0.013014	0.013779	-0.004888	0.004167	0.005955
H14B	0.015661	0.014525	0.015284	0.006867	-0.004154	0.004361
H15A	0.014939	0.014658	0.012491	0.001149	0.006864	-0.00169
H15B	0.012	0.013046	0.021931	0.003361	-0.004046	0.008124
H16A	0.012647	0.015079	0.016611	-0.004431	-0.007948	0.004278
H16B	0.02109	0.013754	0.009319	0.00463	0.006512	0.000976
H17A	0.018661	0.013907	0.015124	0.000614	0.00713	0.008434
H17B	0.008333	0.014694	0.018978	-0.002358	-0.004203	0.000001
H19	0.017339	0.015432	0.010218	0.00416	-0.006115	0.004118
H20	0.019995	0.010088	0.009003	0.002222	-0.003196	-0.003474
H22	0.019833	0.009802	0.009413	0.001662	-0.00374	-0.003426
H23	0.01811	0.008289	0.016122	0.006496	-0.002489	0.002643
H25	0.019922	0.010275	0.008887	0.002127	-0.003339	-0.003448
H27	0.020014	0.010824	0.008327	0.002434	-0.003069	-0.003361
H28A	0.020486	0.009101	0.017136	-0.003381	-0.002791	0.007355
H28B	0.009136	0.022984	0.012416	-0.000289	0.004623	0.003988
H29A	0.021005	0.007456	0.017566	-0.002144	-0.003236	0.005831
H29B	0.008887	0.023279	0.012744	-0.000489	0.004408	0.004761
H30A	0.011387	0.022637	0.011706	-0.00264	0.004158	0.00627
H30B	0.019989	0.00694	0.018867	0.000211	-0.003775	0.005247
H31A	0.012331	0.021544	0.012394	-0.00411	0.003377	0.00739
H31B	0.01934	0.006797	0.018546	0.003196	-0.003328	0.002823
H34	0.0181	0.009945	0.014761	0.008267	-0.000465	0.002857
H35	0.018749	0.016544	0.008008	0.004734	-0.004236	0.004359
H36	0.020153	0.012119	0.007443	0.004919	0.000055	-0.002814
H01	0.014102	0.014057	0.011315	0.008647	0.000864	-0.000202
H03	0.010013	0.018139	0.012303	0.007694	-0.001135	0.002407
H04	0.013188	0.01623	0.011029	0.006456	-0.004425	0.002293
H06	0.019223	0.012912	0.00656	0.005329	-0.001299	-0.001801
H01A	0.008799	0.03308	0.017571	0.007539	-0.002861	0.004005

H01B	0.031467	0.018534	0.015361	0.010938	0.002636	0.011443
H01C	0.018809	0.018422	0.017363	-0.011171	0.000105	-0.004174
H001	0.011954	0.00945	0.008901	0.002248	-0.000755	-0.002112
H00A	0.012861	0.00922	0.023745	0.004639	0.000811	0.00544
H00B	0.017394	0.019071	0.006167	-0.002403	0.001654	-0.000264
H00C	0.016744	0.014863	0.02668	0.004811	0.013052	0.00034
H00D	0.019691	0.029318	0.011425	-0.009287	-0.008795	0.00691
H00E	0.017552	0.011333	0.035817	0.005688	0.003592	0.012641

Table S2: Calculated hydrogen anisotropic displacement parameters for (**2**)

Atom	U11	U22	U33	U12	U13	U23
H1	0.018321	0.010368	0.012587	-0.001799	0.00809	0.002454
H4A	0.014539	0.018292	0.012156	-0.005286	0.002126	0.006208
H4B	0.013429	0.01722	0.01433	0.00516	0.001954	-0.006789
H5A	0.015936	0.024879	0.019996	-0.002332	0.004773	0.014026
H5B	0.020176	0.013352	0.026455	-0.005258	-0.012097	-0.003288
H5C	0.023566	0.021792	0.015295	0.01315	0.001551	-0.006759

Table S3: Calculated hydrogen anisotropic displacement parameters for (**3**)

Atom	U11	U22	U33	U12	U13	U23
H1	0.018167	0.014545	0.010935	-0.005255	-0.006292	0.007441
H3	0.014305	0.013499	0.016933	-0.009752	-0.004717	0.004007
H4	0.018287	0.01378	0.011568	-0.00522	-0.006377	0.007559
H5	0.014213	0.01777	0.014374	-0.006016	-0.010029	0.001859
H9	0.013706	0.007865	0.021016	0.001111	-0.007079	0.004496
H10	0.006702	0.015272	0.022008	-0.002066	-0.006714	0.001674
H11	0.013979	0.008233	0.021893	-0.006299	-0.005294	0.002761
H14A	0.010651	0.015885	0.020805	-0.007052	-0.006456	0.002582
H14B	0.022831	0.012031	0.010608	0.000316	-0.00444	-0.005553
H15A	0.008793	0.017198	0.020703	-0.00586	-0.005665	0.002942
H15B	0.022992	0.012155	0.009833	0.000115	-0.003228	-0.005268
H16A	0.013375	0.016482	0.014043	-0.005691	-0.000762	0.007271
H16B	0.015934	0.017497	0.011333	0.002382	-0.002566	-0.008287
H17A	0.012493	0.017708	0.013313	-0.005326	0.000227	0.006715
H17B	0.016663	0.016619	0.011603	0.002637	-0.003134	-0.008189
H19	0.015237	0.008166	0.022014	-0.005183	-0.007658	-0.001637
H20	0.014299	0.007087	0.02172	0.002543	-0.008217	0.001375
H22	0.01347	0.007277	0.022169	0.002937	-0.007765	0.000799

H23	0.0089	0.014599	0.02172	-0.001681	-0.009199	-0.000728
H25	0.013322	0.007062	0.022407	0.002958	-0.007397	0.000547
H27	0.014278	0.006854	0.021908	0.002777	-0.007986	0.000732
H28A	0.008206	0.014322	0.019153	0.000954	0.003669	-0.002163
H28B	0.023479	0.015985	0.007061	-0.00447	-0.005239	-0.001132
H29A	0.008117	0.014064	0.01972	0.001128	0.003269	-0.002883
H29B	0.02325	0.016441	0.006487	-0.004923	-0.004214	-0.000738
H30A	0.016516	0.014673	0.01741	-0.009108	-0.007755	0.000732
H30B	0.00824	0.018775	0.014459	0.003451	0.002034	0.000012
H31A	0.010151	0.015985	0.014954	0.004827	0.002104	-0.000722
H31B	0.014792	0.016991	0.016843	-0.008787	-0.007913	0.000947
H34	0.011188	0.014569	0.019645	-0.001285	-0.010764	0.001495
H35	0.015909	0.00835	0.021344	-0.005826	-0.00803	-0.00071
H36	0.016163	0.006839	0.020415	0.000553	-0.008555	0.003666
H5'A	0.011875	0.01211	0.020216	-0.006649	0.001673	-0.000452
H5'B	0.014595	0.018975	0.01375	0.001153	-0.009363	-0.003913
H6'A	0.012594	0.013218	0.034233	-0.007646	-0.001098	0.003545
H6'B	0.026434	0.019859	0.008624	0.00428	0.005454	-0.001981
H6'C	0.025786	0.019079	0.020723	0.001993	-0.018497	-0.002896
H7'A	0.015382	0.018162	0.014027	0.000817	-0.009761	-0.004087
H7'B	0.01153	0.012356	0.019779	-0.006141	0.002305	0.000548
H8'A	0.014995	0.033806	0.014762	0.000092	-0.009434	-0.00777
H8'B	0.025174	0.012945	0.017909	0.009735	-0.00298	0.003192
H8'C	0.0178	0.019432	0.0224	-0.012984	0.006167	-0.003744
H01	0.007415	0.012486	0.022168	-0.000824	-0.007044	0.002412
H03	0.005254	0.01392	0.020642	-0.001318	-0.001429	0.003232
H04	0.009352	0.011758	0.020909	-0.005855	-0.004423	0.004276
H06	0.017407	0.006957	0.01967	-0.00451	-0.009232	0.00017
HB1	0.014559	0.016567	0.013323	-0.006265	-0.009612	0.003479
HB2	0.014826	0.012955	0.015565	-0.009222	-0.005749	0.004862
HOS1	0.013387	0.006172	0.011547	0.001565	-0.000336	-0.000511
H1SA	0.011028	0.010675	0.0207	0.003996	-0.001881	0.002123
H1SB	0.019854	0.016277	0.010253	-0.006268	-0.001847	-0.006054
H2SA	0.019042	0.026794	0.016212	-0.005057	-0.000453	-0.014853
H2SB	0.020893	0.026022	0.014471	-0.011078	-0.004778	0.012438
H2SC	0.013378	0.009864	0.036697	0.004783	-0.007151	-0.002034

Table S4: Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for (**1**) . U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
C(1)	2908(1)	1643(1)	2364(1)	18(1)
C(2)	1762(1)	1900(1)	1854(1)	18(1)
C(3)	1075(1)	1200(1)	1161(1)	22(1)
C(4)	1530(1)	256(1)	979(1)	25(1)
C(5)	2668(1)	2(1)	1485(1)	23(1)
C(6)	3359(1)	695(1)	2184(1)	18(1)
C(7)	4582(1)	368(1)	2698(1)	19(1)
C(8)	5776(1)	743(1)	4189(1)	20(1)
C(9)	6957(1)	96(1)	4015(1)	24(1)
C(10)	7881(1)	47(1)	4719(1)	26(1)
C(11)	7631(1)	611(1)	5553(1)	24(1)
C(12)	6399(1)	1229(1)	5653(1)	20(1)
C(13)	6682(1)	1943(1)	7247(1)	22(1)
C(14)	5850(1)	2566(1)	8009(1)	21(1)
C(15)	6972(1)	3229(1)	8664(1)	23(1)
C(16)	7822(1)	3960(1)	8280(1)	27(1)
C(17)	6823(1)	4746(1)	8141(1)	27(1)
C(18)	5888(1)	6233(1)	8983(1)	24(1)
C(19)	6074(1)	6976(1)	9767(1)	27(1)
C(20)	5376(1)	7829(1)	9849(1)	26(1)
C(21)	4439(1)	7977(1)	9162(1)	22(1)
C(22)	3689(1)	8849(1)	9228(1)	25(1)
C(23)	2769(1)	8964(1)	8561(1)	25(1)
C(24)	2563(1)	8209(1)	7785(1)	23(1)
C(25)	3309(1)	7363(1)	7688(1)	23(1)
C(26)	4251(1)	7229(1)	8381(1)	21(1)
C(27)	5008(1)	6358(1)	8298(1)	24(1)
C(28)	1345(1)	7678(1)	6375(1)	27(1)
C(29)	-22(1)	7943(1)	5902(1)	27(1)

C(30)	-269(1)	7234(1)	5003(1)	32(1)
C(31)	-1858(1)	7303(1)	4619(1)	30(1)
C(32)	-2159(1)	6783(1)	3668(1)	27(1)
C(33)	-1369(1)	5412(1)	2473(1)	20(1)
C(34)	-2124(1)	5626(1)	1774(1)	23(1)
C(35)	-1997(1)	4983(1)	957(1)	26(1)
C(36)	-1143(1)	4172(1)	861(1)	25(1)
C(37)	-446(1)	4026(1)	1611(1)	19(1)
C(38)	1251(1)	2913(1)	2123(1)	19(1)
N(1)	4760(1)	891(1)	3550(1)	23(1)
N(2)	5500(1)	1299(1)	4984(1)	21(1)
N(3)	5962(1)	1818(1)	6452(1)	22(1)
N(4)	-1318(1)	6007(1)	3332(1)	25(1)
N(5)	-557(1)	4623(1)	2398(1)	20(1)
N(6)	390(1)	3191(1)	1514(1)	21(1)
O(1)	5338(1)	-330(1)	2370(1)	24(1)
O(2)	7900(1)	1583(1)	7348(1)	33(1)
O(3)	6650(1)	5421(1)	8976(1)	30(1)
O(4)	1585(1)	8402(1)	7180(1)	28(1)
O(5)	-3140(1)	7067(1)	3247(1)	47(1)
O(6)	1590(1)	3426(1)	2855(1)	25(1)
C(01)	11226(1)	1375(1)	7958(1)	44(1)
C(02)	10671(1)	1802(1)	8825(1)	30(1)
N(01)	10259(1)	2126(1)	9517(1)	36(1)
C(001)	2393(1)	5173(1)	4604(1)	51(1)
C(002)	3697(2)	5802(1)	4394(1)	77(1)
O(001)	1052(1)	5120(1)	4085(1)	62(1)

Table S5: Bond lengths [Å] and angles [°] for (**1**).

C(1)-C(6)	1.3941(6)
C(1)-C(2)	1.3967(6)
C(1)-H(1)	1.0826(11)
C(2)-C(3)	1.3940(8)
C(2)-C(38)	1.4970(7)
C(3)-C(4)	1.3910(7)
C(3)-H(3)	1.0825(10)
C(4)-C(5)	1.3861(7)
C(4)-H(4)	1.0824(11)
C(5)-C(6)	1.3962(8)
C(5)-H(5)	1.0824(10)
C(6)-C(7)	1.4954(7)
C(7)-O(1)	1.2232(7)
C(7)-N(1)	1.3690(7)
C(8)-N(2)	1.3352(7)
C(8)-C(9)	1.3933(7)
C(8)-N(1)	1.4014(6)
C(9)-C(10)	1.3871(7)
C(9)-H(9)	1.0824(11)
C(10)-C(11)	1.3850(8)
C(10)-H(10)	1.0827(11)
C(11)-C(12)	1.4007(7)
C(11)-H(11)	1.0828(10)
C(12)-N(2)	1.3389(6)
C(12)-N(3)	1.3950(7)
C(13)-O(2)	1.2225(6)
C(13)-N(3)	1.3676(6)
C(13)-C(14)	1.5129(8)
C(14)-C(15)	1.5260(9)
C(14)-H(14A)	1.0910(11)
C(14)-H(14B)	1.0912(10)
C(15)-C(16)	1.5271(9)
C(15)-H(15A)	1.0916(11)
C(15)-H(15B)	1.0911(10)
C(16)-C(17)	1.5126(9)

C(16)-H(16A)	1.0918(11)
C(16)-H(16B)	1.0917(11)
C(17)-O(3)	1.4364(9)
C(17)-H(17A)	1.0915(11)
C(17)-H(17B)	1.0913(11)
C(18)-O(3)	1.3641(7)
C(18)-C(27)	1.3771(8)
C(18)-C(19)	1.4157(9)
C(19)-C(20)	1.3690(9)
C(19)-H(19)	1.0826(11)
C(20)-C(21)	1.4184(8)
C(20)-H(20)	1.0826(11)
C(21)-C(26)	1.4174(8)
C(21)-C(22)	1.4181(8)
C(22)-C(23)	1.3685(8)
C(22)-H(22)	1.0825(11)
C(23)-C(24)	1.4174(9)
C(23)-H(23)	1.0827(10)
C(24)-O(4)	1.3641(7)
C(24)-C(25)	1.3778(8)
C(25)-C(26)	1.4220(7)
C(25)-H(25)	1.0825(11)
C(26)-C(27)	1.4167(8)
C(27)-H(27)	1.0825(11)
C(28)-O(4)	1.4299(8)
C(28)-C(29)	1.5119(8)
C(28)-H(28A)	1.0915(10)
C(28)-H(28B)	1.0911(11)
C(29)-C(30)	1.5280(9)
C(29)-H(29A)	1.0918(10)
C(29)-H(29B)	1.0918(11)
C(30)-C(31)	1.5193(9)
C(30)-H(30A)	1.0914(11)
C(30)-H(30B)	1.0917(10)
C(31)-C(32)	1.5132(8)
C(31)-H(31A)	1.0919(11)

C(31)-H(31B)	1.0916(10)
C(32)-O(5)	1.2248(7)
C(32)-N(4)	1.3521(7)
C(33)-N(5)	1.3407(6)
C(33)-C(34)	1.3924(7)
C(33)-N(4)	1.4079(7)
C(34)-C(35)	1.3911(8)
C(34)-H(34)	1.0824(11)
C(35)-C(36)	1.3831(7)
C(35)-H(35)	1.0824(10)
C(36)-C(37)	1.3994(7)
C(36)-H(36)	1.0825(11)
C(37)-N(5)	1.3251(7)
C(37)-N(6)	1.4042(7)
C(38)-O(6)	1.2277(7)
C(38)-N(6)	1.3669(6)
N(1)-H(01)	1.0083(11)
N(3)-H(03)	1.0082(10)
N(4)-H(04)	1.0087(10)
N(6)-H(06)	1.0085(11)
C(01)-C(02)	1.4483(10)
C(01)-H(01A)	1.0586(10)
C(01)-H(01B)	1.0587(10)
C(01)-H(01C)	1.0589(11)
C(02)-N(01)	1.1439(8)
C(001)-O(001)	1.4061(11)
C(001)-C(002)	1.521(2)
C(001)-H(00A)	1.0923(11)
C(001)-H(00B)	1.0922(10)
C(002)-H(00C)	1.0588(11)
C(002)-H(00D)	1.0596(11)
C(002)-H(00E)	1.0589(10)
O(001)-H(001)	0.9671(11)

C(6)-C(1)-C(2)	120.47(4)
C(6)-C(1)-H(1)	119.8(5)

C(2)-C(1)-H(1)	119.8(5)
C(3)-C(2)-C(1)	119.36(4)
C(3)-C(2)-C(38)	122.83(4)
C(1)-C(2)-C(38)	117.67(4)
C(4)-C(3)-C(2)	120.17(4)
C(4)-C(3)-H(3)	122.3(6)
C(2)-C(3)-H(3)	117.5(6)
C(5)-C(4)-C(3)	120.40(5)
C(5)-C(4)-H(4)	121.0(7)
C(3)-C(4)-H(4)	118.6(7)
C(4)-C(5)-C(6)	119.95(4)
C(4)-C(5)-H(5)	119.2(6)
C(6)-C(5)-H(5)	120.8(6)
C(1)-C(6)-C(5)	119.65(4)
C(1)-C(6)-C(7)	123.44(4)
C(5)-C(6)-C(7)	116.91(4)
O(1)-C(7)-N(1)	123.47(5)
O(1)-C(7)-C(6)	121.30(4)
N(1)-C(7)-C(6)	115.22(4)
N(2)-C(8)-C(9)	123.60(4)
N(2)-C(8)-N(1)	112.59(4)
C(9)-C(8)-N(1)	123.81(4)
C(10)-C(9)-C(8)	116.69(5)
C(10)-C(9)-H(9)	122.6(6)
C(8)-C(9)-H(9)	120.6(6)
C(11)-C(10)-C(9)	121.41(5)
C(11)-C(10)-H(10)	118.7(6)
C(9)-C(10)-H(10)	119.9(6)
C(10)-C(11)-C(12)	116.96(5)
C(10)-C(11)-H(11)	123.1(8)
C(12)-C(11)-H(11)	119.8(8)
N(2)-C(12)-N(3)	113.06(4)
N(2)-C(12)-C(11)	122.89(4)
N(3)-C(12)-C(11)	124.04(4)
O(2)-C(13)-N(3)	123.45(5)
O(2)-C(13)-C(14)	121.67(5)

N(3)-C(13)-C(14)	114.88(4)
C(13)-C(14)-C(15)	111.68(5)
C(13)-C(14)-H(14A)	108.4(6)
C(15)-C(14)-H(14A)	109.3(6)
C(13)-C(14)-H(14B)	109.0(6)
C(15)-C(14)-H(14B)	111.3(6)
H(14A)-C(14)-H(14B)	107.1(9)
C(14)-C(15)-C(16)	112.51(4)
C(14)-C(15)-H(15A)	108.2(6)
C(16)-C(15)-H(15A)	109.3(6)
C(14)-C(15)-H(15B)	108.1(6)
C(16)-C(15)-H(15B)	109.3(6)
H(15A)-C(15)-H(15B)	109.3(8)
C(17)-C(16)-C(15)	113.92(5)
C(17)-C(16)-H(16A)	109.8(6)
C(15)-C(16)-H(16A)	108.9(6)
C(17)-C(16)-H(16B)	109.6(7)
C(15)-C(16)-H(16B)	108.6(7)
H(16A)-C(16)-H(16B)	105.6(9)
O(3)-C(17)-C(16)	107.53(5)
O(3)-C(17)-H(17A)	109.0(6)
C(16)-C(17)-H(17A)	110.9(6)
O(3)-C(17)-H(17B)	113.0(6)
C(16)-C(17)-H(17B)	108.3(6)
H(17A)-C(17)-H(17B)	108.1(9)
O(3)-C(18)-C(27)	124.70(5)
O(3)-C(18)-C(19)	114.67(5)
C(27)-C(18)-C(19)	120.63(5)
C(20)-C(19)-C(18)	119.98(5)
C(20)-C(19)-H(19)	122.3(7)
C(18)-C(19)-H(19)	117.7(7)
C(19)-C(20)-C(21)	120.93(5)
C(19)-C(20)-H(20)	121.3(6)
C(21)-C(20)-H(20)	117.7(6)
C(26)-C(21)-C(22)	118.74(5)
C(26)-C(21)-C(20)	118.85(5)

C(22)-C(21)-C(20)	122.41(5)
C(23)-C(22)-C(21)	121.08(5)
C(23)-C(22)-H(22)	119.6(6)
C(21)-C(22)-H(22)	119.3(7)
C(22)-C(23)-C(24)	120.08(5)
C(22)-C(23)-H(23)	123.4(6)
C(24)-C(23)-H(23)	116.4(6)
O(4)-C(24)-C(25)	125.09(5)
O(4)-C(24)-C(23)	114.41(5)
C(25)-C(24)-C(23)	120.49(5)
C(24)-C(25)-C(26)	119.94(5)
C(24)-C(25)-H(25)	120.6(7)
C(26)-C(25)-H(25)	119.4(7)
C(27)-C(26)-C(21)	119.59(5)
C(27)-C(26)-C(25)	120.77(5)
C(21)-C(26)-C(25)	119.63(5)
C(18)-C(27)-C(26)	119.99(5)
C(18)-C(27)-H(27)	121.8(6)
C(26)-C(27)-H(27)	118.2(6)
O(4)-C(28)-C(29)	108.01(5)
O(4)-C(28)-H(28A)	108.8(6)
C(29)-C(28)-H(28A)	110.5(6)
O(4)-C(28)-H(28B)	108.8(6)
C(29)-C(28)-H(28B)	109.2(6)
H(28A)-C(28)-H(28B)	111.3(9)
C(28)-C(29)-C(30)	110.61(5)
C(28)-C(29)-H(29A)	110.5(7)
C(30)-C(29)-H(29A)	111.2(7)
C(28)-C(29)-H(29B)	110.0(6)
C(30)-C(29)-H(29B)	108.6(6)
H(29A)-C(29)-H(29B)	105.8(9)
C(31)-C(30)-C(29)	111.75(5)
C(31)-C(30)-H(30A)	108.7(7)
C(29)-C(30)-H(30A)	110.7(7)
C(31)-C(30)-H(30B)	105.8(7)
C(29)-C(30)-H(30B)	111.5(7)

H(30A)-C(30)-H(30B) 108.2(10)
C(32)-C(31)-C(30) 117.52(5)
C(32)-C(31)-H(31A) 111.3(9)
C(30)-C(31)-H(31A) 107.2(9)
C(32)-C(31)-H(31B) 105.9(8)
C(30)-C(31)-H(31B) 111.8(8)
H(31A)-C(31)-H(31B) 102.1(12)
O(5)-C(32)-N(4) 123.30(5)
O(5)-C(32)-C(31) 119.81(5)
N(4)-C(32)-C(31) 116.89(5)
N(5)-C(33)-C(34) 123.67(4)
N(5)-C(33)-N(4) 111.87(4)
C(34)-C(33)-N(4) 124.43(4)
C(35)-C(34)-C(33) 117.12(4)
C(35)-C(34)-H(34) 117.5(6)
C(33)-C(34)-H(34) 125.4(6)
C(36)-C(35)-C(34) 120.27(5)
C(36)-C(35)-H(35) 117.4(6)
C(34)-C(35)-H(35) 122.3(6)
C(35)-C(36)-C(37) 117.61(5)
C(35)-C(36)-H(36) 124.6(7)
C(37)-C(36)-H(36) 117.8(7)
N(5)-C(37)-C(36) 123.40(4)
N(5)-C(37)-N(6) 119.16(4)
C(36)-C(37)-N(6) 117.43(4)
O(6)-C(38)-N(6) 124.64(4)
O(6)-C(38)-C(2) 119.89(4)
N(6)-C(38)-C(2) 115.46(4)
C(7)-N(1)-C(8) 128.12(4)
C(7)-N(1)-H(01) 121.3(7)
C(8)-N(1)-H(01) 110.1(7)
C(8)-N(2)-C(12) 118.44(4)
C(13)-N(3)-C(12) 127.33(4)
C(13)-N(3)-H(03) 121.7(7)
C(12)-N(3)-H(03) 110.8(7)
C(32)-N(4)-C(33) 127.84(4)

C(32)-N(4)-H(04)	121.3(8)
C(33)-N(4)-H(04)	110.7(8)
C(37)-N(5)-C(33)	117.92(4)
C(38)-N(6)-C(37)	128.91(4)
C(38)-N(6)-H(06)	115.5(7)
C(37)-N(6)-H(06)	115.5(7)
C(18)-O(3)-C(17)	116.85(5)
C(24)-O(4)-C(28)	117.38(5)
C(02)-C(01)-H(01A)	100.1(10)
C(02)-C(01)-H(01B)	107.3(10)
H(01A)-C(01)-H(01B)	123.1(15)
C(02)-C(01)-H(01C)	111.8(13)
H(01A)-C(01)-H(01C)	120.0(16)
H(01B)-C(01)-H(01C)	94.5(16)
N(01)-C(02)-C(01)	178.30(7)
O(001)-C(001)-C(002)	114.04(10)
O(001)-C(001)-H(00A)	119.2(9)
C(002)-C(001)-H(00A)	99.6(10)
O(001)-C(001)-H(00B)	117.2(10)
C(002)-C(001)-H(00B)	102.8(10)
H(00A)-C(001)-H(00B)	101.2(13)
C(001)-C(002)-H(00C)	112.7(15)
C(001)-C(002)-H(00D)	105.3(9)
H(00C)-C(002)-H(00D)	97.2(16)
C(001)-C(002)-H(00E)	111.4(13)
H(00C)-C(002)-H(00E)	124(2)
H(00D)-C(002)-H(00E)	102.1(16)
C(001)-O(001)-H(001)	107.6(15)

Table S6: Anisotropic displacement parameters (Å ^{2}x 10^3) for (**1**). The anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U^{11} + ... + 2 h k a^* b^* U^{12}]$

	U11	U22	U33	U23	U13	U12
C(1)	20(1)	18(1)	15(1)	3(1)	-2(1)	3(1)
C(2)	20(1)	17(1)	16(1)	3(1)	-1(1)	4(1)
C(3)	25(1)	20(1)	19(1)	2(1)	-6(1)	4(1)
C(4)	32(1)	20(1)	21(1)	0(1)	-9(1)	5(1)
C(5)	30(1)	18(1)	19(1)	1(1)	-5(1)	6(1)
C(6)	22(1)	18(1)	15(1)	3(1)	-1(1)	5(1)
C(7)	21(1)	19(1)	17(1)	4(1)	-1(1)	4(1)
C(8)	21(1)	22(1)	16(1)	5(1)	-1(1)	6(1)
C(9)	23(1)	29(1)	20(1)	3(1)	-1(1)	10(1)
C(10)	23(1)	30(1)	23(1)	3(1)	-3(1)	11(1)
C(11)	22(1)	28(1)	21(1)	5(1)	-4(1)	9(1)
C(12)	21(1)	22(1)	17(1)	4(1)	-3(1)	6(1)
C(13)	23(1)	25(1)	18(1)	4(1)	-5(1)	5(1)
C(14)	22(1)	24(1)	17(1)	3(1)	-3(1)	2(1)
C(15)	25(1)	23(1)	19(1)	5(1)	-6(1)	2(1)
C(16)	22(1)	26(1)	32(1)	8(1)	-2(1)	1(1)
C(17)	29(1)	25(1)	27(1)	9(1)	-2(1)	1(1)
C(18)	26(1)	26(1)	23(1)	9(1)	-2(1)	2(1)
C(19)	31(1)	31(1)	21(1)	9(1)	-4(1)	3(1)
C(20)	30(1)	30(1)	18(1)	6(1)	-3(1)	3(1)
C(21)	23(1)	26(1)	17(1)	5(1)	0(1)	2(1)
C(22)	27(1)	27(1)	18(1)	3(1)	0(1)	3(1)
C(23)	27(1)	26(1)	21(1)	4(1)	-1(1)	4(1)
C(24)	22(1)	26(1)	19(1)	5(1)	-1(1)	2(1)
C(25)	24(1)	25(1)	20(1)	4(1)	-2(1)	2(1)
C(26)	21(1)	24(1)	19(1)	6(1)	-1(1)	1(1)
C(27)	26(1)	24(1)	22(1)	6(1)	-3(1)	1(1)
C(28)	26(1)	32(1)	20(1)	3(1)	-3(1)	6(1)

C(29)	29(1)	33(1)	19(1)	4(1)	-3(1)	7(1)
C(30)	27(1)	43(1)	21(1)	-1(1)	-4(1)	12(1)
C(31)	32(1)	30(1)	23(1)	-1(1)	-6(1)	14(1)
C(32)	28(1)	29(1)	21(1)	1(1)	-5(1)	14(1)
C(33)	21(1)	21(1)	18(1)	4(1)	-3(1)	6(1)
C(34)	25(1)	25(1)	20(1)	5(1)	-5(1)	9(1)
C(35)	31(1)	28(1)	20(1)	5(1)	-7(1)	10(1)
C(36)	31(1)	25(1)	17(1)	3(1)	-6(1)	8(1)
C(37)	20(1)	19(1)	17(1)	4(1)	-3(1)	4(1)
C(38)	21(1)	18(1)	17(1)	4(1)	-2(1)	4(1)
N(1)	26(1)	26(1)	16(1)	4(1)	-2(1)	11(1)
N(2)	23(1)	23(1)	16(1)	4(1)	-2(1)	7(1)
N(3)	23(1)	27(1)	16(1)	3(1)	-4(1)	9(1)
N(4)	29(1)	26(1)	18(1)	1(1)	-5(1)	13(1)
N(5)	22(1)	20(1)	17(1)	4(1)	-2(1)	6(1)
N(6)	26(1)	20(1)	17(1)	3(1)	-3(1)	7(1)
O(1)	27(1)	21(1)	23(1)	2(1)	-3(1)	9(1)
O(2)	29(1)	43(1)	24(1)	2(1)	-9(1)	17(1)
O(3)	37(1)	28(1)	27(1)	9(1)	-4(1)	7(1)
O(4)	30(1)	30(1)	21(1)	3(1)	-6(1)	7(1)
O(5)	51(1)	60(1)	26(1)	-3(1)	-12(1)	41(1)
O(6)	34(1)	20(1)	20(1)	0(1)	-8(1)	7(1)
C(01)	36(1)	67(1)	27(1)	5(1)	3(1)	21(1)
C(02)	29(1)	35(1)	25(1)	5(1)	-2(1)	10(1)
N(01)	43(1)	39(1)	24(1)	2(1)	-2(1)	12(1)
C(001)	60(1)	45(1)	44(1)	7(1)	-25(1)	13(1)
C(002)	67(1)	82(1)	82(1)	24(1)	-8(1)	12(1)
O(001)	74(1)	46(1)	51(1)	-18(1)	-36(1)	36(1)

Table S7: Hydrogen coordinates (x 10^4) and isotropic displacement parameters (Å ^{2}x 10^3) for (**1**).

	x	y	z	U(eq)
H(1)	3463(10)	2185(5)	2896(4)	27(2)
H(3)	160(9)	1418(8)	794(6)	38(3)
H(4)	960(13)	-276(7)	444(5)	47(3)
H(5)	2976(13)	-742(3)	1346(8)	40(3)
H(9)	7089(13)	-369(6)	3365(3)	36(3)
H(10)	8822(8)	-439(6)	4620(7)	34(2)
H(11)	8290(14)	560(10)	6120(6)	59(4)
H(14A)	5217(12)	2097(7)	8332(7)	41(3)
H(14B)	5001(10)	2974(7)	7763(7)	36(3)
H(15A)	6318(11)	3608(7)	9224(4)	33(2)
H(15B)	7808(9)	2786(6)	8875(6)	30(2)
H(16A)	8808(8)	4277(7)	8705(6)	34(3)
H(16B)	8309(13)	3579(8)	7660(4)	47(3)
H(17A)	7362(13)	5116(8)	7699(6)	44(3)
H(17B)	5727(6)	4416(7)	7845(7)	37(3)
H(19)	6757(13)	6835(10)	10290(6)	57(4)
H(20)	5513(13)	8411(5)	10438(4)	36(3)
H(22)	3874(14)	9438(6)	9808(5)	43(3)
H(23)	2210(12)	9625(4)	8574(7)	38(3)
H(25)	3156(15)	6786(6)	7095(4)	46(3)
H(27)	4866(14)	5803(6)	7690(4)	40(3)
H(28A)	1119(12)	6987(4)	6516(7)	33(2)
H(28B)	2368(8)	7665(9)	5981(6)	43(3)
H(29A)	128(14)	8679(3)	5850(8)	46(3)
H(29B)	-1064(7)	7929(8)	6276(6)	36(3)
H(30A)	598(11)	7364(10)	4552(7)	57(4)
H(30B)	-197(15)	6492(3)	5037(8)	49(3)
H(31A)	-2699(14)	7076(11)	5036(9)	81(5)
H(31B)	-2148(17)	8048(3)	4686(10)	64(4)

H(34)	-2764(12)	6265(5)	1811(8)	45(3)
H(35)	-2520(12)	5100(8)	374(4)	39(3)
H(36)	-1001(14)	3636(7)	250(4)	45(3)
H(01)	4025(11)	1406(6)	3821(7)	41(3)
H(03)	4966(7)	2133(7)	6387(7)	38(3)
H(04)	-481(11)	5811(10)	3688(7)	56(4)
H(06)	409(15)	2756(7)	902(3)	45(3)
H(01A)	12376(7)	1249(13)	8114(11)	85(5)
H(01B)	10940(20)	1833(10)	7565(9)	94(6)
H(01C)	10490(20)	796(11)	7609(14)	130(8)
H(001)	1130(30)	4574(11)	3582(10)	136(9)
H(00A)	3012(18)	4519(7)	4569(12)	87(5)
H(00B)	2290(20)	5457(13)	5308(2)	93(6)
H(00C)	4520(20)	5399(17)	3982(14)	159(11)
H(00D)	4401(16)	6011(12)	4968(6)	74(5)
H(00E)	3300(30)	6469(8)	4342(15)	126(8)

Table S8: Torsion angles [°] for (**1**).

C(6)-C(1)-C(2)-C(3)	-0.19(7)
C(6)-C(1)-C(2)-C(38)	175.57(4)
C(1)-C(2)-C(3)-C(4)	-0.11(7)
C(38)-C(2)-C(3)-C(4)	-175.65(5)
C(2)-C(3)-C(4)-C(5)	0.05(9)
C(3)-C(4)-C(5)-C(6)	0.31(9)
C(2)-C(1)-C(6)-C(5)	0.56(7)
C(2)-C(1)-C(6)-C(7)	179.61(4)
C(4)-C(5)-C(6)-C(1)	-0.62(8)
C(4)-C(5)-C(6)-C(7)	-179.73(5)
C(1)-C(6)-C(7)-O(1)	-153.07(5)
C(5)-C(6)-C(7)-O(1)	26.01(7)
C(1)-C(6)-C(7)-N(1)	28.16(7)
C(5)-C(6)-C(7)-N(1)	-152.76(5)
N(2)-C(8)-C(9)-C(10)	-0.38(8)

N(1)-C(8)-C(9)-C(10)	178.80(5)
C(8)-C(9)-C(10)-C(11)	0.45(9)
C(9)-C(10)-C(11)-C(12)	0.21(9)
C(10)-C(11)-C(12)-N(2)	-1.05(8)
C(10)-C(11)-C(12)-N(3)	177.69(5)
O(2)-C(13)-C(14)-C(15)	40.19(7)
N(3)-C(13)-C(14)-C(15)	-140.15(5)
C(13)-C(14)-C(15)-C(16)	64.33(6)
C(14)-C(15)-C(16)-C(17)	72.17(6)
C(15)-C(16)-C(17)-O(3)	77.71(6)
O(3)-C(18)-C(19)-C(20)	179.34(5)
C(27)-C(18)-C(19)-C(20)	-0.12(9)
C(18)-C(19)-C(20)-C(21)	1.22(9)
C(19)-C(20)-C(21)-C(26)	-0.90(8)
C(19)-C(20)-C(21)-C(22)	179.45(5)
C(26)-C(21)-C(22)-C(23)	1.56(8)
C(20)-C(21)-C(22)-C(23)	-178.79(5)
C(21)-C(22)-C(23)-C(24)	-0.59(8)
C(22)-C(23)-C(24)-O(4)	178.17(5)
C(22)-C(23)-C(24)-C(25)	-1.33(8)
O(4)-C(24)-C(25)-C(26)	-177.24(5)
C(23)-C(24)-C(25)-C(26)	2.21(8)
C(22)-C(21)-C(26)-C(27)	179.17(5)
C(20)-C(21)-C(26)-C(27)	-0.50(7)
C(22)-C(21)-C(26)-C(25)	-0.66(7)
C(20)-C(21)-C(26)-C(25)	179.67(5)
C(24)-C(25)-C(26)-C(27)	178.97(5)
C(24)-C(25)-C(26)-C(21)	-1.20(7)
O(3)-C(18)-C(27)-C(26)	179.32(5)
C(19)-C(18)-C(27)-C(26)	-1.28(8)
C(21)-C(26)-C(27)-C(18)	1.57(8)
C(25)-C(26)-C(27)-C(18)	-178.60(5)
O(4)-C(28)-C(29)-C(30)	175.89(5)
C(28)-C(29)-C(30)-C(31)	164.02(6)
C(29)-C(30)-C(31)-C(32)	168.76(6)
C(30)-C(31)-C(32)-O(5)	-154.10(8)

C(30)-C(31)-C(32)-N(4)	26.73(9)
N(5)-C(33)-C(34)-C(35)	-0.86(8)
N(4)-C(33)-C(34)-C(35)	176.83(5)
C(33)-C(34)-C(35)-C(36)	-0.27(9)
C(34)-C(35)-C(36)-C(37)	0.83(9)
C(35)-C(36)-C(37)-N(5)	-0.36(8)
C(35)-C(36)-C(37)-N(6)	178.56(5)
C(3)-C(2)-C(38)-O(6)	160.71(5)
C(1)-C(2)-C(38)-O(6)	-14.89(7)
C(3)-C(2)-C(38)-N(6)	-18.05(7)
C(1)-C(2)-C(38)-N(6)	166.35(4)
O(1)-C(7)-N(1)-C(8)	-0.44(8)
C(6)-C(7)-N(1)-C(8)	178.30(5)
N(2)-C(8)-N(1)-C(7)	-170.95(5)
C(9)-C(8)-N(1)-C(7)	9.79(9)
C(9)-C(8)-N(2)-C(12)	-0.40(8)
N(1)-C(8)-N(2)-C(12)	-179.66(4)
N(3)-C(12)-N(2)-C(8)	-177.72(4)
C(11)-C(12)-N(2)-C(8)	1.14(7)
O(2)-C(13)-N(3)-C(12)	4.74(9)
C(14)-C(13)-N(3)-C(12)	-174.92(5)
N(2)-C(12)-N(3)-C(13)	-176.80(5)
C(11)-C(12)-N(3)-C(13)	4.36(9)
O(5)-C(32)-N(4)-C(33)	1.54(11)
C(31)-C(32)-N(4)-C(33)	-179.33(6)
N(5)-C(33)-N(4)-C(32)	-170.35(6)
C(34)-C(33)-N(4)-C(32)	11.72(9)
C(36)-C(37)-N(5)-C(33)	-0.71(7)
N(6)-C(37)-N(5)-C(33)	-179.61(4)
C(34)-C(33)-N(5)-C(37)	1.34(8)
N(4)-C(33)-N(5)-C(37)	-176.61(5)
O(6)-C(38)-N(6)-C(37)	-9.43(8)
C(2)-C(38)-N(6)-C(37)	169.26(5)
N(5)-C(37)-N(6)-C(38)	-5.67(8)
C(36)-C(37)-N(6)-C(38)	175.36(5)
C(27)-C(18)-O(3)-C(17)	14.39(8)

C(19)-C(18)-O(3)-C(17)	-165.04(5)
C(16)-C(17)-O(3)-C(18)	174.70(5)
C(25)-C(24)-O(4)-C(28)	-1.58(8)
C(23)-C(24)-O(4)-C(28)	178.94(5)
C(29)-C(28)-O(4)-C(24)	167.35(5)

Table S9: Hydrogen bonds for (**1**).

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
C(9)-H(9)...O(1)	1.0824(11)	2.192(10)	2.8610(10)	117.8(7)
C(11)-H(11)...O(2)	1.0828(11)	2.149(13)	2.8295(11)	118.4(9)
C(14)-H(14A)...O(1)#1	1.0910(11)	2.525(10)	3.2513(11)	123.1(8)
C(14)-H(14B)...O(5)#2	1.0912(10)	2.252(6)	3.2149(11)	146.0(8)
C(17)-H(17A)...O(6)#3	1.0915(11)	2.630(5)	3.6619(12)	157.5(9)
C(34)-H(34)...O(5)	1.0824(10)	2.305(11)	2.8625(12)	110.1(7)
C(36)-H(36)...N(01)#4	1.0825(11)	2.484(7)	3.4298(14)	145.3(9)
N(3)-H(03)...O(5)#2	1.0081(10)	2.002(5)	2.9552(10)	156.7(9)
N(4)-H(04)...O(001)	1.0087(10)	1.890(4)	2.8713(10)	163.3(12)
N(6)-H(06)...N(01)#4	1.0085(11)	2.150(3)	3.1390(12)	166.4(10)
C(01)-H(01A)...O(1)#5	1.0586(10)	2.456(10)	3.3756(12)	144.7(13)
O(001)-H(001)...N(5)	0.9671(11)	2.39(2)	2.9103(11)	113.3(17)
O(001)-H(001)...O(6)	0.9671(11)	1.814(7)	2.7567(12)	164(2)

Symmetry transformations used to generate equivalent atoms:
#1 -x+1,-y,-z+1 #2 -x,-y+1,-z+1 #3 -x+1,-y+1,-z+1
#4 x-1,y,z-1 #5 -x+2,-y,-z+1

Table S10: Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for (**2**). U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
O(1)	5000	896(1)	2500	16(1)
O(2)	3053(1)	3643(1)	4455(1)	18(1)
N(1)	4016(1)	2282(1)	3457(1)	13(1)
C(1)	5000	1760(1)	2500	11(1)
C(2)	3950(1)	3257(1)	3545(1)	12(1)
C(3)	5000	3847(1)	2500	11(1)
C(4)	6447(1)	4485(1)	3285(1)	17(1)
C(5)	7889(1)	3937(1)	4160(1)	29(1)

Table S11: Bond lengths [Å°] and angles [°] for (**2**).

O(1)-C(1)	1.2106(4)
O(2)-C(2)	1.2254(3)
N(1)-C(2)	1.3680(3)
N(1)-C(1)	1.3827(2)
N(1)-H(1)	0.939(9)
C(1)-N(1)#1	1.3827(2)
C(2)-C(3)	1.5164(3)
C(3)-C(2)#1	1.5165(3)
C(3)-C(4)	1.5466(3)
C(3)-C(4)#1	1.5466(3)
C(4)-C(5)	1.5204(6)
C(4)-H(4A)	1.012(7)
C(4)-H(4B)	1.022(8)
C(5)-H(5A)	1.026(9)
C(5)-H(5B)	1.017(9)
C(5)-H(5C)	0.983(9)

C(2)-N(1)-C(1)	126.01(2)
C(2)-N(1)-H(1)	116.6(6)
C(1)-N(1)-H(1)	117.3(6)
O(1)-C(1)-N(1)#1	121.925(13)
O(1)-C(1)-N(1)	121.925(13)
N(1)#1-C(1)-N(1)	116.15(3)
O(2)-C(2)-N(1)	120.33(2)
O(2)-C(2)-C(3)	120.71(2)
N(1)-C(2)-C(3)	118.962(19)
C(2)-C(3)-C(2)#1	113.89(2)
C(2)-C(3)-C(4)	108.518(14)
C(2)#1-C(3)-C(4)	108.214(15)
C(2)-C(3)-C(4)#1	108.214(15)
C(2)#1-C(3)-C(4)#1	108.520(14)
C(4)-C(3)-C(4)#1	109.42(3)
C(5)-C(4)-C(3)	114.31(3)
C(5)-C(4)-H(4A)	109.4(4)
C(3)-C(4)-H(4A)	107.6(4)
C(5)-C(4)-H(4B)	110.0(5)
C(3)-C(4)-H(4B)	106.1(5)
H(4A)-C(4)-H(4B)	109.3(7)
C(4)-C(5)-H(5A)	112.1(5)
C(4)-C(5)-H(5B)	111.0(6)
H(5A)-C(5)-H(5B)	107.5(9)
C(4)-C(5)-H(5C)	112.8(6)
H(5A)-C(5)-H(5C)	103.2(8)
H(5B)-C(5)-H(5C)	109.8(7)

Symmetry transformations used to generate equivalent atoms:

#1 -x+1,y,-z+1/2

Table S12: Anisotropic displacement parameters ($Å^2 x 10^3$) for (**2**). The anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U^{11} + ... + 2 h k a^* b^* U^{12}]$

	U11	U22	U33	U23	U13	U12
O(1)	20(1)	9(1)	20(1)	0	7(1)	0
O(2)	25(1)	13(1)	17(1)	-2(1)	13(1)	-1(1)
N(1)	16(1)	10(1)	13(1)	0(1)	7(1)	-1(1)
C(1)	13(1)	10(1)	12(1)	0	4(1)	0
C(2)	14(1)	10(1)	11(1)	-1(1)	5(1)	-1(1)
C(3)	13(1)	10(1)	11(1)	0	4(1)	0
C(4)	20(1)	16(1)	16(1)	-2(1)	3(1)	-6(1)
C(5)	24(1)	34(1)	28(1)	1(1)	-8(1)	-7(1)

Table S13: Hydrogen coordinates ($x 10^4$) and isotropic displacement parameters ($Å^2 x 10^3$) for (**2**).

	x	y	z	U(eq)
H(1)	3375(12)	1939(7)	4134(9)	40(2)
H(4A)	7120(11)	4882(6)	2578(8)	24(2)
H(4B)	5678(11)	4924(6)	3899(8)	30(2)
H(5A)	7267(14)	3521(7)	4892(10)	47(3)
H(5B)	8792(14)	4390(7)	4662(11)	53(3)
H(5C)	8605(13)	3473(6)	3621(10)	44(2)

Table S14: Torsion angles [°] for (**2**).

C(2)-N(1)-C(1)-O(1)	179.288(19)
C(2)-N(1)-C(1)-N(1)#1	-0.712(19)
C(1)-N(1)-C(2)-O(2)	-178.75(2)
C(1)-N(1)-C(2)-C(3)	1.33(4)
O(2)-C(2)-C(3)-C(2)#1	179.47(3)
N(1)-C(2)-C(3)-C(2)#1	-0.609(17)
O(2)-C(2)-C(3)-C(4)	58.88(3)
N(1)-C(2)-C(3)-C(4)	-121.20(3)
O(2)-C(2)-C(3)-C(4)#1	-59.77(3)
N(1)-C(2)-C(3)-C(4)#1	120.15(3)
C(2)-C(3)-C(4)-C(5)	59.14(3)
C(2)#1-C(3)-C(4)-C(5)	-64.91(3)
C(4)#1-C(3)-C(4)-C(5)	177.02(3)

Symmetry transformations used to generate equivalent atoms:
#1 -x+1,y,-z+1/2

Table S15: Hydrogen bonds for (**2**). [Å and °].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
N(1)-H(1)...O(2)#2	0.939(9)	1.905(9)	2.8415(3)	174.4(9)

Symmetry transformations used to generate equivalent atoms:
#1 -x+1,y,-z+1/2 #2 -x+1/2,-y+1/2,-z+1

Table S16: Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for (**3**). U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
O(1)	-1042(1)	7508(1)	5384(1)	28(1)
O(2)	-1426(1)	1483(1)	7279(1)	24(1)
O(3)	3237(1)	-1896(1)	8558(1)	19(1)
O(4)	8360(1)	1003(1)	9608(1)	18(1)
O(5)	9586(1)	5812(1)	7784(1)	26(1)
O(6)	4239(1)	9993(1)	6168(1)	25(1)
N(1)	401(1)	6157(1)	5859(1)	15(1)
N(2)	234(1)	4289(1)	6243(1)	14(1)
N(3)	285(1)	2398(1)	6612(1)	15(1)
N(4)	7423(1)	5781(1)	7662(1)	16(1)
N(5)	5995(1)	7017(1)	7087(1)	14(1)
N(6)	4468(1)	8108(1)	6488(1)	14(1)
C(1)	2008(1)	8066(1)	5868(1)	14(1)
C(2)	2886(1)	8889(1)	5702(1)	14(1)
C(3)	2821(1)	9656(1)	5096(1)	18(1)
C(4)	1893(1)	9601(1)	4654(1)	20(1)
C(5)	1015(1)	8789(1)	4821(1)	18(1)
C(6)	1072(1)	8018(1)	5424(1)	15(1)
C(7)	40(1)	7212(1)	5560(1)	16(1)
C(8)	-382(1)	5259(1)	5989(1)	14(1)
C(9)	-1689(1)	5369(1)	5867(1)	17(1)
C(10)	-2372(1)	4426(1)	6028(1)	19(1)
C(11)	-1768(1)	3414(1)	6292(1)	17(1)
C(12)	-447(1)	3385(1)	6383(1)	14(1)
C(13)	-227(1)	1522(1)	7052(1)	15(1)
C(14)	824(1)	576(1)	7223(1)	17(1)
C(15)	335(1)	-96(1)	7940(1)	19(1)
C(16)	1295(1)	-1143(1)	8094(1)	19(1)

C(17)	2502(1)	-853(1)	8360(1)	17(1)
C(18)	4359(1)	-1830(1)	8832(1)	15(1)
C(19)	5041(1)	-2890(1)	9020(1)	18(1)
C(20)	6208(1)	-2936(1)	9274(1)	17(1)
C(21)	6756(1)	-1935(1)	9359(1)	15(1)
C(22)	7986(1)	-1954(1)	9597(1)	17(1)
C(23)	8479(1)	-972(1)	9683(1)	17(1)
C(24)	7765(1)	84(1)	9515(1)	15(1)
C(25)	6579(1)	136(1)	9271(1)	15(1)
C(26)	6050(1)	-875(1)	9190(1)	14(1)
C(27)	4840(1)	-840(1)	8927(1)	16(1)
C(28)	7866(1)	2067(1)	9291(1)	16(1)
C(29)	8831(1)	2890(1)	9360(1)	19(1)
C(30)	8446(1)	4080(1)	9039(1)	19(1)
C(31)	8577(1)	4119(1)	8194(1)	17(1)
C(32)	8594(1)	5318(1)	7863(1)	16(1)
C(33)	7090(1)	6883(1)	7392(1)	14(1)
C(34)	7809(1)	7771(1)	7460(1)	17(1)
C(35)	7323(1)	8843(1)	7226(1)	18(1)
C(36)	6189(1)	9010(1)	6911(1)	17(1)
C(37)	5573(1)	8062(1)	6841(1)	13(1)
C(38)	3905(1)	9045(1)	6145(1)	15(1)
O(1')	3206(1)	5793(1)	6403(1)	19(1)
O(2')	1681(1)	2814(1)	7833(1)	18(1)
O(3')	5317(1)	4504(1)	8272(1)	21(1)
N(1')	2434(1)	4307(1)	7126(1)	15(1)
N(2')	4248(1)	5137(1)	7344(1)	17(1)
C(1')	3278(1)	5116(1)	6931(1)	15(1)
C(2')	2446(1)	3528(1)	7716(1)	14(1)
C(3')	3392(1)	3624(1)	8233(1)	14(1)
C(4')	4404(1)	4438(1)	7952(1)	15(1)
C(5')	2519(1)	4093(1)	8965(1)	20(1)
C(6')	1835(1)	5302(1)	8897(1)	24(1)
C(7')	4128(1)	2446(1)	8387(1)	18(1)
C(8')	5042(1)	1947(1)	7698(1)	24(1)
O(1S)	3127(1)	12217(1)	6134(1)	21(1)

C(1S)	3608(1)	12823(1)	5464(1)	24(1)
C(2S)	4950(1)	13186(1)	5465(1)	40(1)

Table S17: Bond lengths [Å] and angles [°] for (**3**)

O(1)-C(7)	1.2231(5)
O(2)-C(13)	1.2244(5)
O(3)-C(18)	1.3658(5)
O(3)-C(17)	1.4305(4)
O(4)-C(24)	1.3649(5)
O(4)-C(28)	1.4312(5)
O(5)-C(32)	1.2250(5)
O(6)-C(38)	1.2318(5)
N(1)-C(7)	1.3712(5)
N(1)-C(8)	1.4044(5)
N(1)-H(01)	1.00900(2)
N(2)-C(8)	1.3432(5)
N(2)-C(12)	1.3466(5)
N(3)-C(13)	1.3707(5)
N(3)-C(12)	1.3962(5)
N(3)-H(03)	1.00900(6)
N(4)-C(32)	1.3669(5)
N(4)-C(33)	1.3948(4)
N(4)-H(04)	1.00900(2)
N(5)-C(37)	1.3420(4)
N(5)-C(33)	1.3465(5)
N(6)-C(38)	1.3629(4)
N(6)-C(37)	1.4161(5)
N(6)-H(06)	1.00900(3)
C(1)-C(6)	1.3969(5)
C(1)-C(2)	1.3992(5)
C(1)-H(1)	1.08300(3)
C(2)-C(3)	1.3982(5)
C(2)-C(38)	1.4954(5)
C(3)-C(4)	1.3898(6)

C(3)-H(3)	1.08300(4)
C(4)-C(5)	1.3888(6)
C(4)-H(4)	1.082999(18)
C(5)-C(6)	1.3964(5)
C(5)-H(5)	1.083001(19)
C(6)-C(7)	1.4996(6)
C(8)-C(9)	1.3965(5)
C(9)-C(10)	1.3882(5)
C(9)-H(9)	1.083000(16)
C(10)-C(11)	1.3847(5)
C(10)-H(10)	1.083000(18)
C(11)-C(12)	1.3994(5)
C(11)-H(11)	1.08300(2)
C(13)-C(14)	1.5177(5)
C(14)-C(15)	1.5252(4)
C(14)-H(14A)	1.092000(17)
C(14)-H(14B)	1.09200(2)
C(15)-C(16)	1.5323(5)
C(15)-H(15A)	1.092000(18)
C(15)-H(15B)	1.09200(3)
C(16)-C(17)	1.5167(5)
C(16)-H(16A)	1.092000(17)
C(16)-H(16B)	1.092001(15)
C(17)-H(17A)	1.092000(16)
C(17)-H(17B)	1.09200(2)
C(18)-C(27)	1.3778(5)
C(18)-C(19)	1.4179(5)
C(19)-C(20)	1.3697(5)
C(19)-H(19)	1.083001(15)
C(20)-C(21)	1.4195(5)
C(20)-H(20)	1.08300(2)
C(21)-C(26)	1.4189(5)
C(21)-C(22)	1.4204(5)
C(22)-C(23)	1.3711(5)
C(22)-H(22)	1.083000(16)
C(23)-C(24)	1.4174(5)

C(23)-H(23)	1.08300(2)
C(24)-C(25)	1.3773(5)
C(25)-C(26)	1.4197(5)
C(25)-H(25)	1.083000(18)
C(26)-C(27)	1.4213(5)
C(27)-H(27)	1.083000(17)
C(28)-C(29)	1.5174(5)
C(28)-H(28A)	1.09200(3)
C(28)-H(28B)	1.092001(15)
C(29)-C(30)	1.5309(5)
C(29)-H(29A)	1.09200(2)
C(29)-H(29B)	1.09200(3)
C(30)-C(31)	1.5358(5)
C(30)-H(30A)	1.09200(3)
C(30)-H(30B)	1.09200(2)
C(31)-C(32)	1.5153(5)
C(31)-H(31A)	1.09200(2)
C(31)-H(31B)	1.09200(3)
C(33)-C(34)	1.4004(5)
C(34)-C(35)	1.3816(5)
C(34)-H(34)	1.08300(5)
C(35)-C(36)	1.3919(5)
C(35)-H(35)	1.08300(4)
C(36)-C(37)	1.3957(5)
C(36)-H(36)	1.08300(3)
O(1')-C(1')	1.2263(5)
O(2')-C(2')	1.2151(5)
O(3')-C(4')	1.2222(5)
N(1')-C(1')	1.3656(5)
N(1')-C(2')	1.3808(5)
N(1')-HB1	1.009000(17)
N(2')-C(4')	1.3713(5)
N(2')-C(1')	1.3795(5)
N(2')-HB2	1.00900(3)
C(2')-C(3')	1.5170(5)
C(3')-C(4')	1.5076(5)

C(3')-C(7')	1.5406(5)
C(3')-C(5')	1.5568(5)
C(5')-C(6')	1.5256(5)
C(5')-H(5'A)	1.09200(3)
C(5')-H(5'B)	1.09200(3)
C(6')-H(6'A)	1.05900(3)
C(6')-H(6'B)	1.058999(15)
C(6')-H(6'C)	1.05900(2)
C(7')-C(8')	1.5250(4)
C(7')-H(7'A)	1.09200(2)
C(7')-H(7'B)	1.09200(2)
C(8')-H(8'A)	1.05900(2)
C(8')-H(8'B)	1.059000(15)
C(8')-H(8'C)	1.05900(2)
O(1S)-C(1S)	1.4181(5)
O(1S)-HOS1	0.96700(3)
C(1S)-C(2S)	1.5026(6)
C(1S)-H(1SA)	1.09200(2)
C(1S)-H(1SB)	1.092001(15)
C(2S)-H(2SA)	1.05900(3)
C(2S)-H(2SB)	1.05900(3)
C(2S)-H(2SC)	1.05900(6)

C(18)-O(3)-C(17)	117.23(3)
C(24)-O(4)-C(28)	117.07(3)
C(7)-N(1)-C(8)	125.80(3)
C(7)-N(1)-H(01)	119.75(16)
C(8)-N(1)-H(01)	114.15(16)
C(8)-N(2)-C(12)	118.07(3)
C(13)-N(3)-C(12)	126.22(3)
C(13)-N(3)-H(03)	115.7(4)
C(12)-N(3)-H(03)	117.6(3)
C(32)-N(4)-C(33)	128.14(3)
C(32)-N(4)-H(04)	116.1(3)
C(33)-N(4)-H(04)	115.3(3)
C(37)-N(5)-C(33)	117.77(3)

C(38)-N(6)-C(37)	125.70(3)
C(38)-N(6)-H(06)	118.21(6)
C(37)-N(6)-H(06)	115.43(6)
C(6)-C(1)-C(2)	119.46(3)
C(6)-C(1)-H(1)	121.43(14)
C(2)-C(1)-H(1)	119.02(13)
C(3)-C(2)-C(1)	120.01(3)
C(3)-C(2)-C(38)	115.52(3)
C(1)-C(2)-C(38)	124.45(3)
C(4)-C(3)-C(2)	120.31(3)
C(4)-C(3)-H(3)	119.4(3)
C(2)-C(3)-H(3)	120.3(3)
C(5)-C(4)-C(3)	119.73(3)
C(5)-C(4)-H(4)	121.55(16)
C(3)-C(4)-H(4)	118.72(16)
C(4)-C(5)-C(6)	120.44(3)
C(4)-C(5)-H(5)	122.02(7)
C(6)-C(5)-H(5)	117.50(7)
C(5)-C(6)-C(1)	120.04(3)
C(5)-C(6)-C(7)	115.20(3)
C(1)-C(6)-C(7)	124.70(3)
O(1)-C(7)-N(1)	123.94(4)
O(1)-C(7)-C(6)	119.29(4)
N(1)-C(7)-C(6)	116.73(3)
N(2)-C(8)-C(9)	123.02(3)
N(2)-C(8)-N(1)	114.20(3)
C(9)-C(8)-N(1)	122.77(3)
C(10)-C(9)-C(8)	117.53(3)
C(10)-C(9)-H(9)	121.03(5)
C(8)-C(9)-H(9)	121.44(6)
C(11)-C(10)-C(9)	120.93(3)
C(11)-C(10)-H(10)	119.2(2)
C(9)-C(10)-H(10)	119.9(2)
C(10)-C(11)-C(12)	117.18(3)
C(10)-C(11)-H(11)	123.4(2)
C(12)-C(11)-H(11)	119.34(19)

N(2)-C(12)-N(3)	114.40(3)
N(2)-C(12)-C(11)	123.25(3)
N(3)-C(12)-C(11)	122.34(3)
O(2)-C(13)-N(3)	123.66(4)
O(2)-C(13)-C(14)	122.22(3)
N(3)-C(13)-C(14)	114.11(3)
C(13)-C(14)-C(15)	112.12(2)
C(13)-C(14)-H(14A)	109.56(5)
C(15)-C(14)-H(14A)	111.68(7)
C(13)-C(14)-H(14B)	104.09(12)
C(15)-C(14)-H(14B)	108.56(5)
H(14A)-C(14)-H(14B)	110.57(17)
C(14)-C(15)-C(16)	114.21(2)
C(14)-C(15)-H(15A)	109.73(8)
C(16)-C(15)-H(15A)	110.05(5)
C(14)-C(15)-H(15B)	109.49(7)
C(16)-C(15)-H(15B)	109.24(15)
H(15A)-C(15)-H(15B)	103.6(2)
C(17)-C(16)-C(15)	112.87(2)
C(17)-C(16)-H(16A)	107.43(5)
C(15)-C(16)-H(16A)	109.37(5)
C(17)-C(16)-H(16B)	109.6(2)
C(15)-C(16)-H(16B)	110.48(14)
H(16A)-C(16)-H(16B)	106.84(12)
O(3)-C(17)-C(16)	107.35(3)
O(3)-C(17)-H(17A)	110.70(5)
C(16)-C(17)-H(17A)	109.84(5)
O(3)-C(17)-H(17B)	108.89(15)
C(16)-C(17)-H(17B)	111.0(3)
H(17A)-C(17)-H(17B)	109.07(18)
O(3)-C(18)-C(27)	125.09(3)
O(3)-C(18)-C(19)	114.63(3)
C(27)-C(18)-C(19)	120.27(3)
C(20)-C(19)-C(18)	120.04(3)
C(20)-C(19)-H(19)	123.45(5)
C(18)-C(19)-H(19)	116.48(4)

C(19)-C(20)-C(21)	121.31(3)
C(19)-C(20)-H(20)	121.04(5)
C(21)-C(20)-H(20)	117.65(5)
C(26)-C(21)-C(20)	118.44(3)
C(26)-C(21)-C(22)	118.81(3)
C(20)-C(21)-C(22)	122.75(3)
C(23)-C(22)-C(21)	121.32(3)
C(23)-C(22)-H(22)	118.37(5)
C(21)-C(22)-H(22)	120.30(6)
C(22)-C(23)-C(24)	119.47(3)
C(22)-C(23)-H(23)	122.03(12)
C(24)-C(23)-H(23)	118.49(12)
O(4)-C(24)-C(25)	124.65(3)
O(4)-C(24)-C(23)	114.45(3)
C(25)-C(24)-C(23)	120.90(3)
C(24)-C(25)-C(26)	120.14(3)
C(24)-C(25)-H(25)	120.93(6)
C(26)-C(25)-H(25)	118.84(5)
C(21)-C(26)-C(25)	119.35(3)
C(21)-C(26)-C(27)	119.67(3)
C(25)-C(26)-C(27)	120.97(3)
C(18)-C(27)-C(26)	120.21(3)
C(18)-C(27)-H(27)	120.91(5)
C(26)-C(27)-H(27)	118.86(6)
O(4)-C(28)-C(29)	106.29(3)
O(4)-C(28)-H(28A)	110.50(8)
C(29)-C(28)-H(28A)	110.21(7)
O(4)-C(28)-H(28B)	109.49(9)
C(29)-C(28)-H(28B)	111.95(7)
H(28A)-C(28)-H(28B)	108.40(9)
C(28)-C(29)-C(30)	112.89(2)
C(28)-C(29)-H(29A)	108.16(6)
C(30)-C(29)-H(29A)	110.53(7)
C(28)-C(29)-H(29B)	107.55(10)
C(30)-C(29)-H(29B)	108.87(13)
H(29A)-C(29)-H(29B)	108.71(11)

C(29)-C(30)-C(31)	112.63(3)
C(29)-C(30)-H(30A)	107.98(4)
C(31)-C(30)-H(30A)	107.20(5)
C(29)-C(30)-H(30B)	109.87(8)
C(31)-C(30)-H(30B)	110.40(7)
H(30A)-C(30)-H(30B)	108.62(6)
C(32)-C(31)-C(30)	111.64(3)
C(32)-C(31)-H(31A)	105.79(6)
C(30)-C(31)-H(31A)	109.96(7)
C(32)-C(31)-H(31B)	111.67(4)
C(30)-C(31)-H(31B)	112.17(5)
H(31A)-C(31)-H(31B)	105.21(5)
O(5)-C(32)-N(4)	123.93(4)
O(5)-C(32)-C(31)	121.86(3)
N(4)-C(32)-C(31)	114.21(3)
N(5)-C(33)-N(4)	113.99(3)
N(5)-C(33)-C(34)	123.47(3)
N(4)-C(33)-C(34)	122.50(3)
C(35)-C(34)-C(33)	117.29(3)
C(35)-C(34)-H(34)	122.9(4)
C(33)-C(34)-H(34)	119.7(4)
C(34)-C(35)-C(36)	120.54(3)
C(34)-C(35)-H(35)	121.40(5)
C(36)-C(35)-H(35)	118.04(5)
C(35)-C(36)-C(37)	117.76(3)
C(35)-C(36)-H(36)	122.36(6)
C(37)-C(36)-H(36)	119.86(6)
N(5)-C(37)-C(36)	123.09(3)
N(5)-C(37)-N(6)	113.72(3)
C(36)-C(37)-N(6)	123.17(3)
O(6)-C(38)-N(6)	122.57(4)
O(6)-C(38)-C(2)	119.62(3)
N(6)-C(38)-C(2)	117.77(3)
C(1')-N(1')-C(2')	125.28(3)
C(1')-N(1')-HB1	113.78(11)
C(2')-N(1')-HB1	120.91(10)

C(4')-N(2')-C(1')	125.30(3)
C(4')-N(2')-HB2	118.69(19)
C(1')-N(2')-HB2	115.85(18)
O(1')-C(1')-N(1')	122.12(4)
O(1')-C(1')-N(2')	120.40(3)
N(1')-C(1')-N(2')	117.46(3)
O(2')-C(2')-N(1')	120.65(3)
O(2')-C(2')-C(3')	121.13(3)
N(1')-C(2')-C(3')	118.17(3)
C(4')-C(3')-C(2')	114.25(3)
C(4')-C(3')-C(7')	109.34(3)
C(2')-C(3')-C(7')	109.53(3)
C(4')-C(3')-C(5')	106.59(3)
C(2')-C(3')-C(5')	106.87(3)
C(7')-C(3')-C(5')	110.17(3)
O(3')-C(4')-N(2')	120.44(3)
O(3')-C(4')-C(3')	121.01(3)
N(2')-C(4')-C(3')	118.51(3)
C(6')-C(5')-C(3')	115.17(2)
C(6')-C(5')-H(5'A)	109.83(4)
C(3')-C(5')-H(5'A)	106.65(11)
C(6')-C(5')-H(5'B)	106.99(14)
C(3')-C(5')-H(5'B)	109.79(6)
H(5'A)-C(5')-H(5'B)	108.26(16)
C(5')-C(6')-H(6'A)	112.09(4)
C(5')-C(6')-H(6'B)	107.44(7)
H(6'A)-C(6')-H(6'B)	108.05(19)
C(5')-C(6')-H(6'C)	111.8(5)
H(6'A)-C(6')-H(6'C)	105.0(4)
H(6'B)-C(6')-H(6'C)	112.46(14)
C(8')-C(7')-C(3')	113.23(3)
C(8')-C(7')-H(7'A)	109.60(5)
C(3')-C(7')-H(7'A)	109.84(9)
C(8')-C(7')-H(7'B)	112.86(7)
C(3')-C(7')-H(7'B)	105.04(4)
H(7'A)-C(7')-H(7'B)	105.94(10)

C(7')-C(8')-H(8'A)	109.93(4)
C(7')-C(8')-H(8'B)	109.69(6)
H(8'A)-C(8')-H(8'B)	112.70(10)
C(7')-C(8')-H(8'C)	111.80(8)
H(8'A)-C(8')-H(8'C)	108.64(11)
H(8'B)-C(8')-H(8'C)	103.97(6)
C(1S)-O(1S)-HOS1	108.0(2)
O(1S)-C(1S)-C(2S)	111.11(3)
O(1S)-C(1S)-H(1SA)	106.47(6)
C(2S)-C(1S)-H(1SA)	110.40(4)
O(1S)-C(1S)-H(1SB)	108.07(4)
C(2S)-C(1S)-H(1SB)	116.23(6)
H(1SA)-C(1S)-H(1SB)	103.914(9)
C(1S)-C(2S)-H(2SA)	108.44(11)
C(1S)-C(2S)-H(2SB)	110.15(10)
H(2SA)-C(2S)-H(2SB)	106.44(5)
C(1S)-C(2S)-H(2SC)	108.53(6)
H(2SA)-C(2S)-H(2SC)	113.5(2)
H(2SB)-C(2S)-H(2SC)	109.76(19)

Table S18: Anisotropic displacement parameters ($Å^2 x\ 10^3$) for (**3**). The anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U^{11} + ... + 2 h k a^* b^* U^{12}]$

	U11	U22	U33	U23	U13	U12
O(1)	25(1)	17(1)	49(1)	11(1)	-23(1)	-5(1)
O(2)	15(1)	21(1)	33(1)	10(1)	-3(1)	-5(1)
O(3)	19(1)	13(1)	26(1)	2(1)	-10(1)	-2(1)
O(4)	20(1)	13(1)	23(1)	4(1)	-9(1)	-3(1)
O(5)	16(1)	22(1)	41(1)	8(1)	-12(1)	-5(1)
O(6)	25(1)	10(1)	45(1)	5(1)	-18(1)	-4(1)
N(1)	16(1)	11(1)	19(1)	4(1)	-7(1)	-2(1)
N(2)	15(1)	11(1)	16(1)	2(1)	-5(1)	-2(1)
N(3)	14(1)	11(1)	19(1)	3(1)	-4(1)	-3(1)
N(4)	15(1)	12(1)	22(1)	4(1)	-7(1)	-2(1)
N(5)	14(1)	10(1)	17(1)	3(1)	-5(1)	-2(1)
N(6)	16(1)	10(1)	19(1)	3(1)	-6(1)	-2(1)
C(1)	16(1)	10(1)	15(1)	3(1)	-5(1)	-1(1)
C(2)	15(1)	10(1)	17(1)	3(1)	-4(1)	-1(1)
C(3)	19(1)	14(1)	20(1)	6(1)	-4(1)	-3(1)
C(4)	24(1)	17(1)	18(1)	7(1)	-7(1)	-3(1)
C(5)	24(1)	14(1)	17(1)	4(1)	-9(1)	-2(1)
C(6)	18(1)	10(1)	16(1)	2(1)	-6(1)	-1(1)
C(7)	19(1)	11(1)	20(1)	3(1)	-9(1)	-2(1)
C(8)	15(1)	12(1)	15(1)	2(1)	-5(1)	-2(1)
C(9)	16(1)	16(1)	22(1)	3(1)	-8(1)	-2(1)
C(10)	16(1)	18(1)	25(1)	3(1)	-9(1)	-3(1)
C(11)	16(1)	15(1)	21(1)	2(1)	-6(1)	-4(1)
C(12)	14(1)	12(1)	16(1)	1(1)	-4(1)	-3(1)
C(13)	16(1)	12(1)	18(1)	2(1)	-3(1)	-3(1)
C(14)	18(1)	12(1)	19(1)	2(1)	-1(1)	-1(1)
C(15)	15(1)	20(1)	20(1)	6(1)	-4(1)	-3(1)
C(16)	18(1)	16(1)	23(1)	6(1)	-7(1)	-5(1)
C(17)	17(1)	14(1)	22(1)	3(1)	-7(1)	-2(1)
C(18)	17(1)	12(1)	17(1)	2(1)	-5(1)	-1(1)

C(19)	22(1)	11(1)	21(1)	2(1)	-8(1)	-2(1)
C(20)	23(1)	10(1)	20(1)	2(1)	-8(1)	0(1)
C(21)	18(1)	11(1)	16(1)	3(1)	-5(1)	0(1)
C(22)	19(1)	12(1)	20(1)	4(1)	-7(1)	0(1)
C(23)	17(1)	14(1)	20(1)	4(1)	-7(1)	-1(1)
C(24)	16(1)	12(1)	16(1)	3(1)	-5(1)	-2(1)
C(25)	16(1)	11(1)	19(1)	2(1)	-5(1)	-1(1)
C(26)	15(1)	11(1)	15(1)	2(1)	-4(1)	0(1)
C(27)	16(1)	11(1)	20(1)	2(1)	-6(1)	-1(1)
C(28)	18(1)	12(1)	20(1)	2(1)	-5(1)	-2(1)
C(29)	25(1)	14(1)	20(1)	2(1)	-10(1)	-5(1)
C(30)	27(1)	12(1)	19(1)	0(1)	-6(1)	-3(1)
C(31)	18(1)	13(1)	20(1)	1(1)	-7(1)	1(1)
C(32)	15(1)	15(1)	19(1)	2(1)	-6(1)	-1(1)
C(33)	14(1)	12(1)	16(1)	3(1)	-5(1)	-3(1)
C(34)	18(1)	14(1)	20(1)	4(1)	-8(1)	-6(1)
C(35)	22(1)	14(1)	22(1)	4(1)	-9(1)	-7(1)
C(36)	21(1)	11(1)	21(1)	3(1)	-9(1)	-5(1)
C(37)	14(1)	10(1)	15(1)	2(1)	-4(1)	-2(1)
C(38)	14(1)	10(1)	21(1)	3(1)	-5(1)	-1(1)
O(1')	23(1)	15(1)	22(1)	8(1)	-9(1)	-3(1)
O(2')	18(1)	17(1)	21(1)	4(1)	-6(1)	-8(1)
O(3')	21(1)	20(1)	28(1)	8(1)	-13(1)	-9(1)
N(1')	15(1)	13(1)	17(1)	3(1)	-6(1)	-3(1)
N(2')	16(1)	13(1)	21(1)	7(1)	-8(1)	-5(1)
C(1')	16(1)	11(1)	17(1)	4(1)	-5(1)	-2(1)
C(2')	14(1)	12(1)	16(1)	2(1)	-4(1)	-3(1)
C(3')	15(1)	12(1)	16(1)	3(1)	-5(1)	-4(1)
C(4')	15(1)	12(1)	19(1)	4(1)	-7(1)	-4(1)
C(5')	21(1)	22(1)	16(1)	0(1)	-4(1)	-5(1)
C(6')	22(1)	23(1)	26(1)	-7(1)	-6(1)	0(1)
C(7')	19(1)	13(1)	22(1)	5(1)	-7(1)	-3(1)
C(8')	24(1)	18(1)	29(1)	1(1)	-6(1)	3(1)
O(1S)	21(1)	15(1)	24(1)	4(1)	-1(1)	-1(1)
C(1S)	26(1)	23(1)	23(1)	6(1)	-5(1)	-7(1)
C(2S)	31(1)	39(1)	48(1)	-17(1)	12(1)	-18(1)

Table S19: Hydrogen coordinates (x 10^4) and isotropic displacement parameters (Å ^{2}x 10 ^{3}) for (**3**).

	x	y	z	U(eq)
H(1)	2035(3)	7509(1)	6356(1)	28(2)
H(3)	3504(1)	10290(1)	4962(3)	30(3)
H(4)	1869(3)	10203(1)	4186(1)	36(3)
H(5)	291(1)	8707(1)	4484(1)	32(3)
H(9)	-2159(1)	6161(1)	5660(1)	33(3)
H(10)	-3400(1)	4484(3)	5961(1)	29(3)
H(11)	-2244(1)	2646(1)	6396(5)	34(3)
H(14A)	1747(1)	925(1)	7236(1)	33(3)
H(14B)	975(3)	20(1)	6762(1)	42(3)
H(15A)	-650(1)	-338(1)	7938(1)	34(3)
H(15B)	149(4)	456(1)	8406(1)	32(3)
H(16A)	770(1)	-1690(1)	8525(1)	28(2)
H(16B)	1627(4)	-1637(1)	7600(1)	25(2)
H(17A)	3119(1)	-384(1)	7920(1)	27(2)
H(17B)	2195(6)	-341(1)	8847(1)	25(2)
H(19)	4604(1)	-3635(1)	8937(1)	32(3)
H(20)	6754(1)	-3739(1)	9410(1)	25(2)
H(22)	8567(1)	-2752(1)	9707(1)	29(3)
H(23)	9413(1)	-974(2)	9867(1)	35(3)
H(25)	6008(1)	942(1)	9169(1)	31(3)
H(27)	4297(1)	-25(1)	8815(1)	23(2)
H(28A)	6862(1)	2360(1)	9592(1)	32(3)
H(28B)	7817(1)	1970(1)	8715(1)	33(3)
H(29A)	9832(1)	2547(1)	9081(1)	47(3)
H(29B)	8827(2)	2938(2)	9948(1)	28(2)
H(30A)	9132(1)	4631(1)	9157(1)	38(3)
H(30B)	7429(1)	4401(1)	9305(1)	36(3)
H(31A)	9529(1)	3654(1)	7939(1)	25(2)

H(31B)	7811(1)	3699(1)	8029(1)	28(2)
H(34)	8672(1)	7608(6)	7721(1)	36(3)
H(35)	7786(1)	9579(1)	7297(1)	30(3)
H(36)	5797(1)	9835(1)	6709(1)	26(2)
H(5'A)	1767(1)	3518(1)	9160(1)	42(3)
H(5'B)	3136(1)	4078(2)	9381(1)	27(2)
H(6'A)	2535(1)	5892(1)	8737(2)	36(3)
H(6'B)	1262(1)	5507(1)	9426(1)	40(3)
H(6'C)	1251(1)	5399(8)	8482(1)	37(3)
H(7'A)	4708(1)	2490(2)	8814(1)	34(3)
H(7'B)	3337(1)	1928(1)	8632(1)	29(3)
H(8'A)	4478(1)	1879(1)	7288(1)	42(3)
H(8'B)	5559(1)	1161(1)	7844(1)	37(3)
H(8'C)	5814(1)	2451(1)	7475(1)	43(3)
H(01)	1337(1)	5949(3)	5954(1)	34(3)
H(03)	1292(1)	2361(3)	6501(7)	37(3)
H(04)	6666(1)	5299(1)	7778(6)	44(3)
H(06)	4177(1)	7353(1)	6426(1)	32(3)
HB1	1788(1)	4300(2)	6783(1)	40(3)
HB2	4838(1)	5760(1)	7200(2)	37(3)
HOS1	3523(1)	11437(1)	6089(2)	44(3)
H(1SA)	2869(1)	13561(1)	5424(1)	48(4)
H(1SB)	3553(1)	12319(1)	5004(1)	40(3)
H(2SA)	4857(2)	13682(1)	5929(1)	68(5)
H(2SB)	5243(2)	13717(1)	4990(1)	57(4)
H(2SC)	5658(1)	12454(1)	5463(2)	58(4)

Table S20: Torsion angles [°] for (**3**).

C(6)-C(1)-C(2)-C(3)	0.06(6)
C(6)-C(1)-C(2)-C(38)	178.32(4)
C(1)-C(2)-C(3)-C(4)	-0.39(6)
C(38)-C(2)-C(3)-C(4)	-178.80(4)
C(2)-C(3)-C(4)-C(5)	0.76(6)
C(3)-C(4)-C(5)-C(6)	-0.81(6)

C(4)-C(5)-C(6)-C(1)	0.49(6)
C(4)-C(5)-C(6)-C(7)	177.96(4)
C(2)-C(1)-C(6)-C(5)	-0.11(6)
C(2)-C(1)-C(6)-C(7)	-177.33(4)
C(8)-N(1)-C(7)-O(1)	1.74(8)
C(8)-N(1)-C(7)-C(6)	-176.06(4)
C(5)-C(6)-C(7)-O(1)	-29.36(7)
C(1)-C(6)-C(7)-O(1)	147.98(5)
C(5)-C(6)-C(7)-N(1)	148.55(4)
C(1)-C(6)-C(7)-N(1)	-34.11(6)
C(12)-N(2)-C(8)-C(9)	0.26(6)
C(12)-N(2)-C(8)-N(1)	179.92(4)
C(7)-N(1)-C(8)-N(2)	176.01(4)
C(7)-N(1)-C(8)-C(9)	-4.33(6)
N(2)-C(8)-C(9)-C(10)	0.72(6)
N(1)-C(8)-C(9)-C(10)	-178.91(4)
C(8)-C(9)-C(10)-C(11)	-0.61(7)
C(9)-C(10)-C(11)-C(12)	-0.40(7)
C(8)-N(2)-C(12)-N(3)	177.11(4)
C(8)-N(2)-C(12)-C(11)	-1.39(6)
C(13)-N(3)-C(12)-N(2)	152.64(4)
C(13)-N(3)-C(12)-C(11)	-28.85(7)
C(10)-C(11)-C(12)-N(2)	1.46(7)
C(10)-C(11)-C(12)-N(3)	-176.92(4)
C(12)-N(3)-C(13)-O(2)	2.97(7)
C(12)-N(3)-C(13)-C(14)	-178.43(4)
O(2)-C(13)-C(14)-C(15)	-25.96(5)
N(3)-C(13)-C(14)-C(15)	155.41(3)
C(13)-C(14)-C(15)-C(16)	173.38(3)
C(14)-C(15)-C(16)-C(17)	75.52(3)
C(18)-O(3)-C(17)-C(16)	-178.51(3)
C(15)-C(16)-C(17)-O(3)	173.91(3)
C(17)-O(3)-C(18)-C(27)	-0.48(6)
C(17)-O(3)-C(18)-C(19)	179.85(4)
O(3)-C(18)-C(19)-C(20)	177.54(4)
C(27)-C(18)-C(19)-C(20)	-2.15(7)

C(18)-C(19)-C(20)-C(21)	0.36(7)
C(19)-C(20)-C(21)-C(26)	1.37(6)
C(19)-C(20)-C(21)-C(22)	-177.83(4)
C(26)-C(21)-C(22)-C(23)	1.70(6)
C(20)-C(21)-C(22)-C(23)	-179.10(4)
C(21)-C(22)-C(23)-C(24)	-1.52(6)
C(28)-O(4)-C(24)-C(25)	-13.11(6)
C(28)-O(4)-C(24)-C(23)	166.07(3)
C(22)-C(23)-C(24)-O(4)	-178.80(4)
C(22)-C(23)-C(24)-C(25)	0.42(6)
O(4)-C(24)-C(25)-C(26)	179.60(4)
C(23)-C(24)-C(25)-C(26)	0.46(6)
C(20)-C(21)-C(26)-C(25)	179.98(4)
C(22)-C(21)-C(26)-C(25)	-0.79(6)
C(20)-C(21)-C(26)-C(27)	-1.35(6)
C(22)-C(21)-C(26)-C(27)	177.88(4)
C(24)-C(25)-C(26)-C(21)	-0.26(6)
C(24)-C(25)-C(26)-C(27)	-178.92(4)
O(3)-C(18)-C(27)-C(26)	-177.50(4)
C(19)-C(18)-C(27)-C(26)	2.15(6)
C(21)-C(26)-C(27)-C(18)	-0.40(6)
C(25)-C(26)-C(27)-C(18)	178.26(4)
C(24)-O(4)-C(28)-C(29)	-170.82(3)
O(4)-C(28)-C(29)-C(30)	179.62(3)
C(28)-C(29)-C(30)-C(31)	-67.08(3)
C(29)-C(30)-C(31)-C(32)	-163.21(3)
C(33)-N(4)-C(32)-O(5)	-5.15(8)
C(33)-N(4)-C(32)-C(31)	175.14(4)
C(30)-C(31)-C(32)-O(5)	76.01(5)
C(30)-C(31)-C(32)-N(4)	-104.27(4)
C(37)-N(5)-C(33)-N(4)	177.30(4)
C(37)-N(5)-C(33)-C(34)	-0.50(6)
C(32)-N(4)-C(33)-N(5)	165.67(4)
C(32)-N(4)-C(33)-C(34)	-16.51(7)
N(5)-C(33)-C(34)-C(35)	2.67(6)
N(4)-C(33)-C(34)-C(35)	-174.94(4)

C(33)-C(34)-C(35)-C(36)	-2.26(6)
C(34)-C(35)-C(36)-C(37)	-0.13(6)
C(33)-N(5)-C(37)-C(36)	-2.16(6)
C(33)-N(5)-C(37)-N(6)	176.29(4)
C(35)-C(36)-C(37)-N(5)	2.47(6)
C(35)-C(36)-C(37)-N(6)	-175.82(4)
C(38)-N(6)-C(37)-N(5)	-175.18(4)
C(38)-N(6)-C(37)-C(36)	3.26(6)
C(37)-N(6)-C(38)-O(6)	-6.85(7)
C(37)-N(6)-C(38)-C(2)	170.53(4)
C(3)-C(2)-C(38)-O(6)	27.89(6)
C(1)-C(2)-C(38)-O(6)	-150.44(5)
C(3)-C(2)-C(38)-N(6)	-149.57(4)
C(1)-C(2)-C(38)-N(6)	32.10(6)
C(2')-N(1')-C(1')-O(1')	-179.19(4)
C(2')-N(1')-C(1')-N(2')	2.06(6)
C(4')-N(2')-C(1')-O(1')	178.65(4)
C(4')-N(2')-C(1')-N(1')	-2.57(6)
C(1')-N(1')-C(2')-O(2')	-177.61(4)
C(1')-N(1')-C(2')-C(3')	5.14(6)
O(2')-C(2')-C(3')-C(4')	171.68(4)
N(1')-C(2')-C(3')-C(4')	-11.09(5)
O(2')-C(2')-C(3')-C(7')	48.64(5)
N(1')-C(2')-C(3')-C(7')	-134.13(3)
O(2')-C(2')-C(3')-C(5')	-70.70(5)
N(1')-C(2')-C(3')-C(5')	106.54(4)
C(1')-N(2')-C(4')-O(3')	178.26(4)
C(1')-N(2')-C(4')-C(3')	-4.25(6)
C(2')-C(3')-C(4')-O(3')	-171.84(4)
C(7')-C(3')-C(4')-O(3')	-48.70(6)
C(5')-C(3')-C(4')-O(3')	70.37(5)
C(2')-C(3')-C(4')-N(2')	10.69(6)
C(7')-C(3')-C(4')-N(2')	133.83(4)
C(5')-C(3')-C(4')-N(2')	-107.10(4)
C(4')-C(3')-C(5')-C(6')	56.28(3)
C(2')-C(3')-C(5')-C(6')	-66.28(3)

C(7')-C(3')-C(5')-C(6')	174.80(2)
C(4')-C(3')-C(7')-C(8')	-61.26(4)
C(2')-C(3')-C(7')-C(8')	64.64(4)
C(5')-C(3')-C(7')-C(8')	-178.08(2)

Table S21: Hydrogen bonds for (**3).** [Å and °].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
C(9)-H(9)...O(1)	1.0830(1)	2.0751(8)	2.7636(5)	118.70(6)
C(11)-H(11)...O(2)	1.0830(1)	2.281(7)	2.8595(5)	111.4(5)
C(14)-H(14A)...O(2')	1.0920(1)	2.5707(8)	3.2685(4)	120.88(5)
C(19)-H(19)...O(3')#1	1.0830(1)	2.5653(7)	3.4514(5)	138.45(6)
C(31)-H(31A)...O(2')#2	1.0920(1)	2.2802(6)	3.3181(4)	158.02(9)
C(31)-H(31B)...O(3')	1.0920(1)	2.5758(6)	3.2985(4)	122.87(4)
C(34)-H(34)...O(5)	1.0830(1)	2.227(7)	2.8734(5)	116.1(5)
C(36)-H(36)...O(6)	1.0830(1)	2.0386(9)	2.7471(5)	120.15(6)
N(1)-H(01)...O(1')	1.0090(1)	2.2282(7)	3.2137(4)	165.10(19)
N(3)-H(03)...O(1S)#1	1.0090(1)	1.860(2)	2.8591(4)	169.9(10)
N(4)-H(04)...O(3')	1.0090(1)	1.835(4)	2.8153(4)	163.0(10)
N(6)-H(06)...O(1')	1.0090(1)	2.2370(5)	3.2365(4)	170.50(8)
N(1')-HB1...N(2)	1.0090(1)	2.0537(4)	3.0534(4)	170.51(4)
N(2')-HB2...N(5)	1.0090(1)	2.0116(7)	3.0121(4)	170.9(3)
O(1S)-HOS1...O(6)	0.9670(1)	1.7887(8)	2.7470(4)	170.5(4)

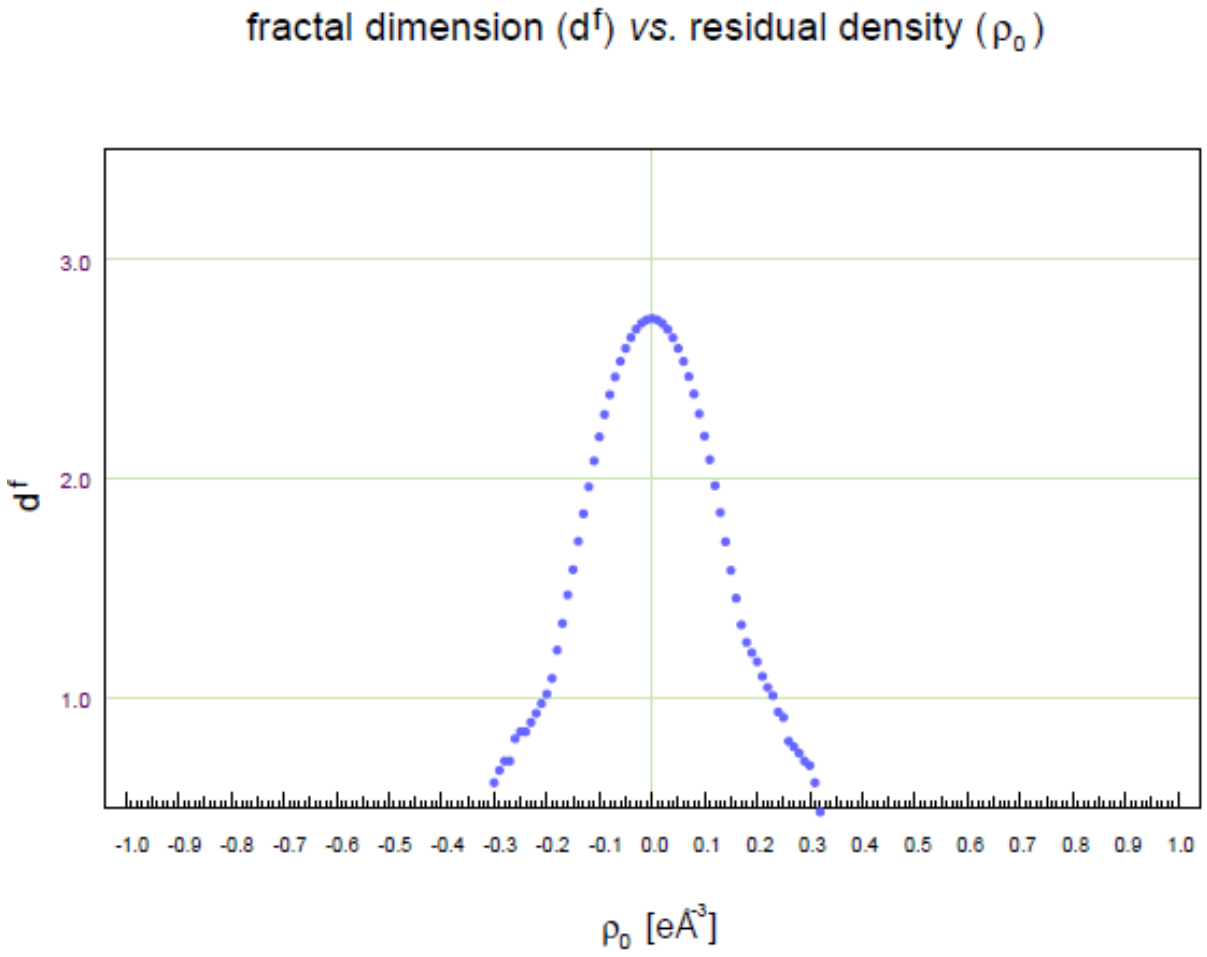
Symmetry transformations used to generate equivalent atoms:
#1 x,y-1,z #2 x+1,y,z

Data quality

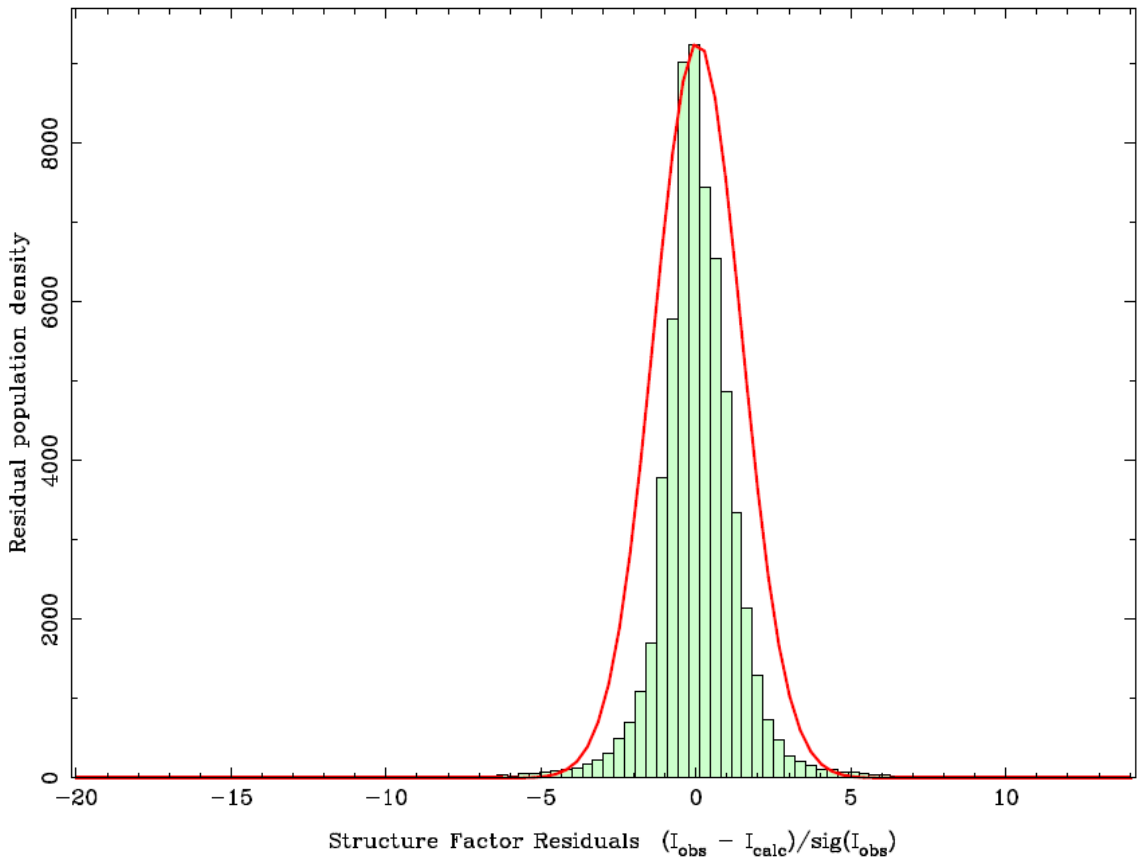
Residual density analysis

The residual density analysis introduced by Meindl *et al.*[5-6] was also performed on the data for (**1**), (**2**) and (**3**). The results of the analysis for all structures show that the residual density can be fitted to a gaussian distribution implying the remaining residual density can be attributed to noise. The parabolic shape of the curve with the absence of "shoulders" in the data explains that the electron density has been modelled well, which can be expected as the structures contain C, H, N and O. The smaller number of points on the fractal curve for (**2**) can be attributed to a smaller number of atoms in the asymmetric unit. Fractal plots and residual density histograms can be found in Figures S1-S3 for (**1**), (**2**) and (**3**) respectively.

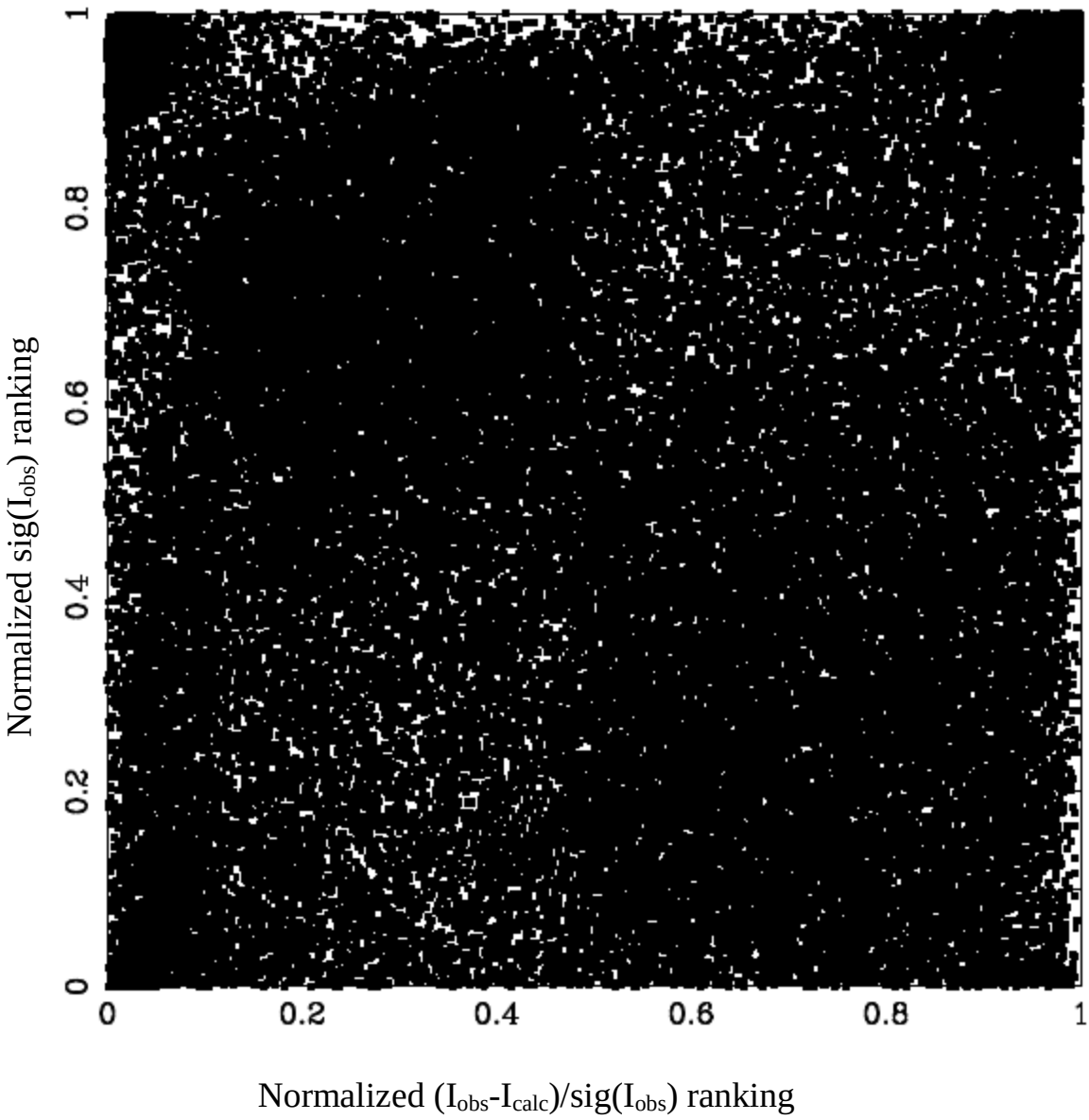
Residual density maps of the complexes of (**1**), (**2**) and (**3**) can be found in Figures S4 – S6 respectively. All the maps are largely featureless with no significant regions of positive or negative electron density indicating a good fit of the multipole model with the collected data.



(a) fractal plot for (**1**); residual density vs. fractal dimension

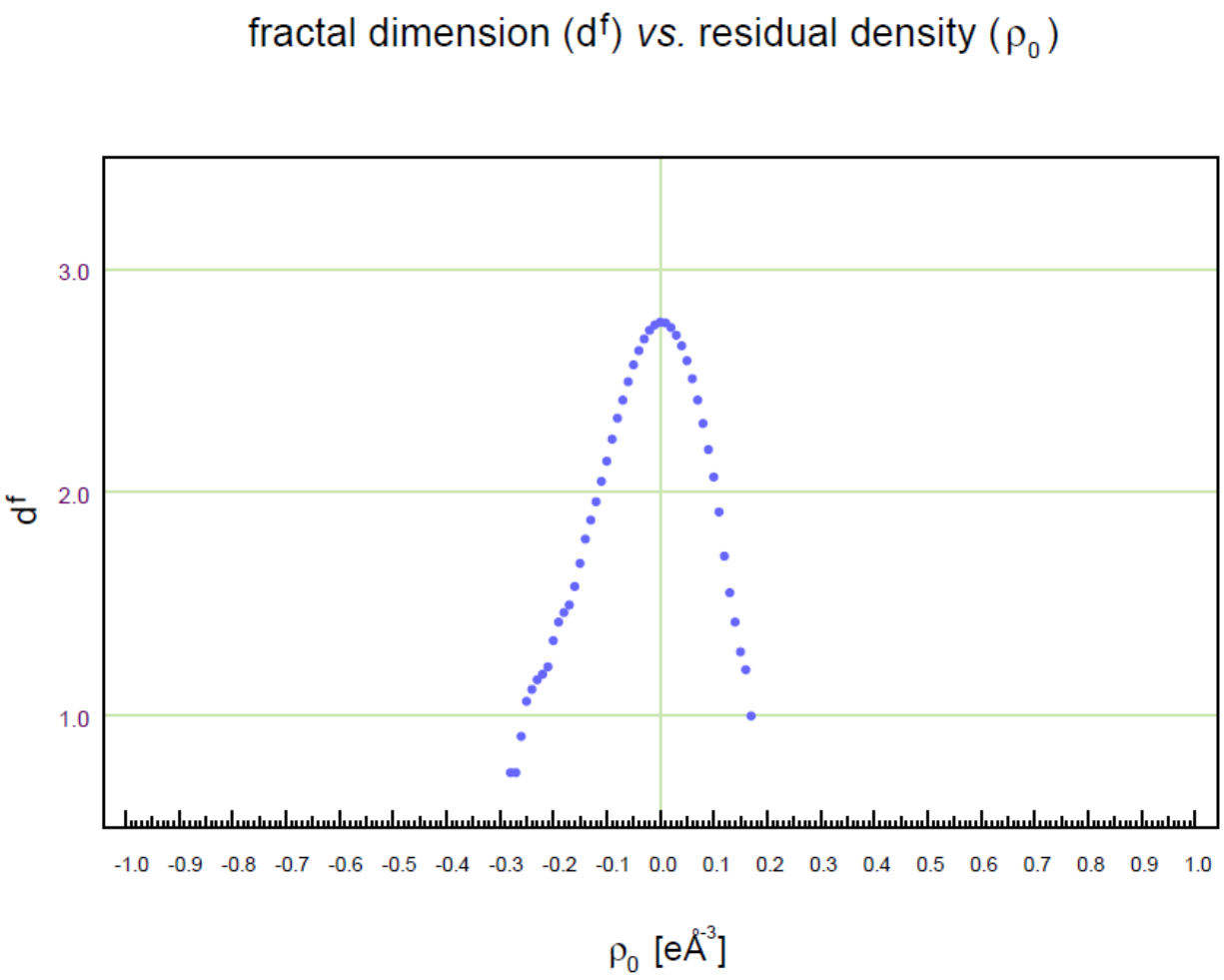


(b) histogram showing residual density of multipole refinement for (**1**)

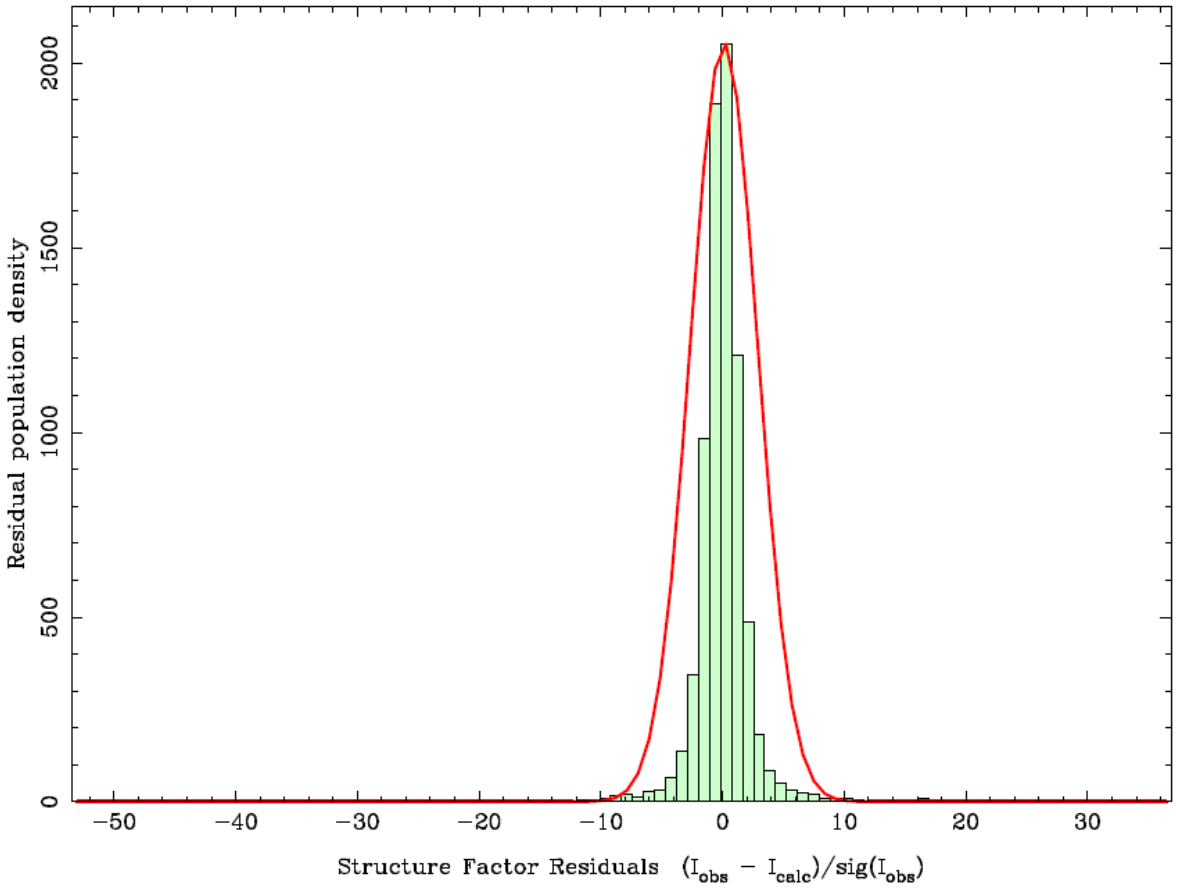


(c) structure factor residuals vs. standard uncertainties for (**1**)

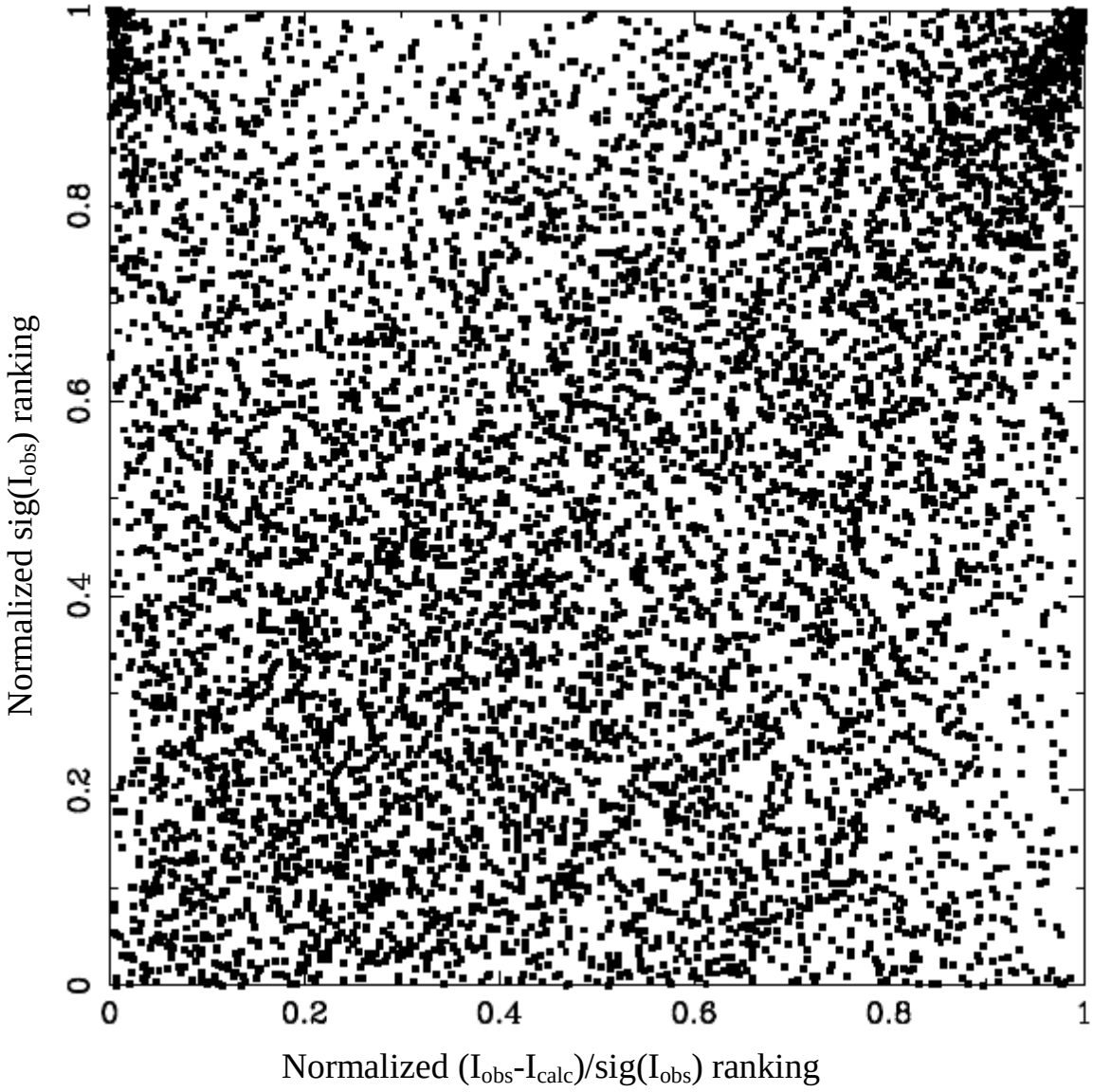
Figure S1: Results of the residual density analysis on the data sets for (**1**)



(a) fractal plot for (**2**); residual density vs. fractal dimension

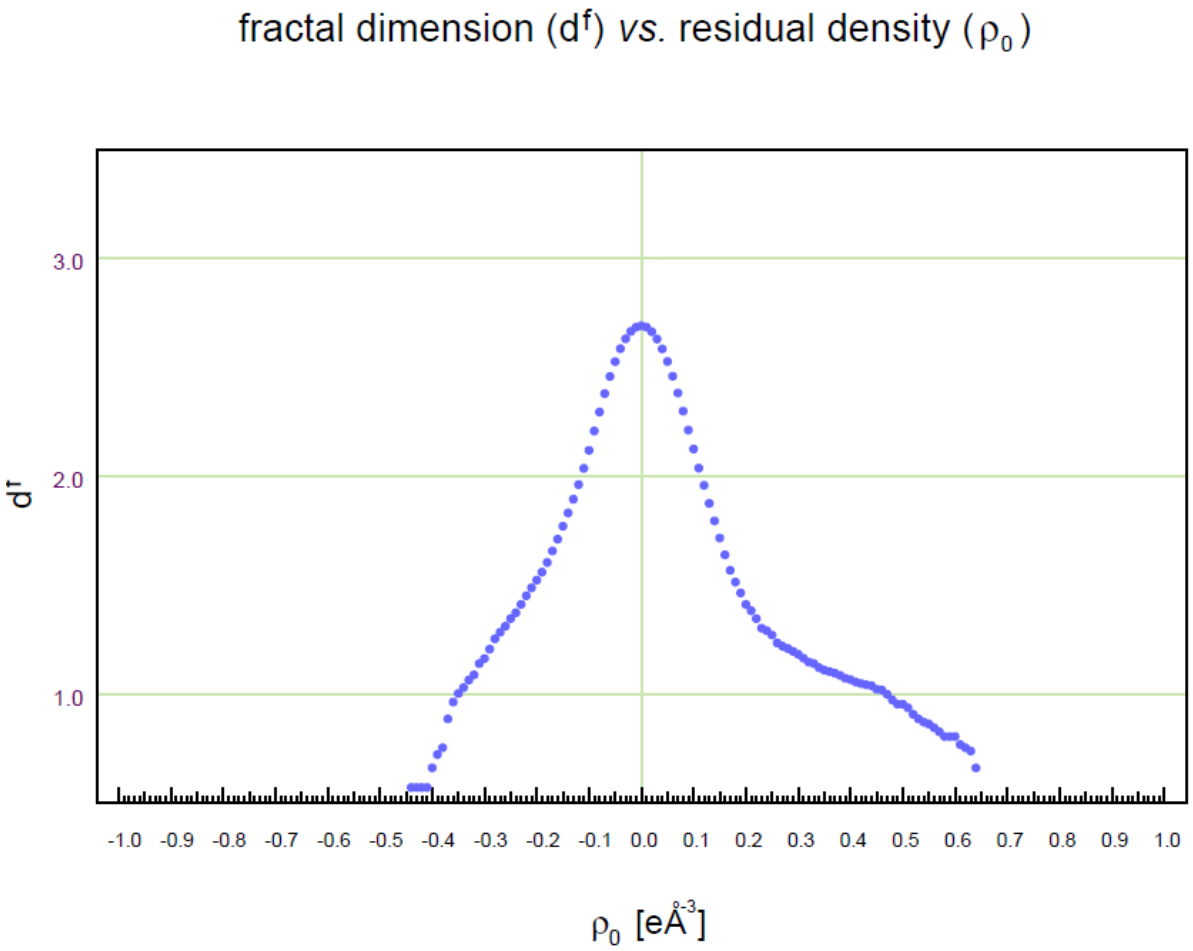


(b) histogram showing residual density of multipole refinement for (**2**)

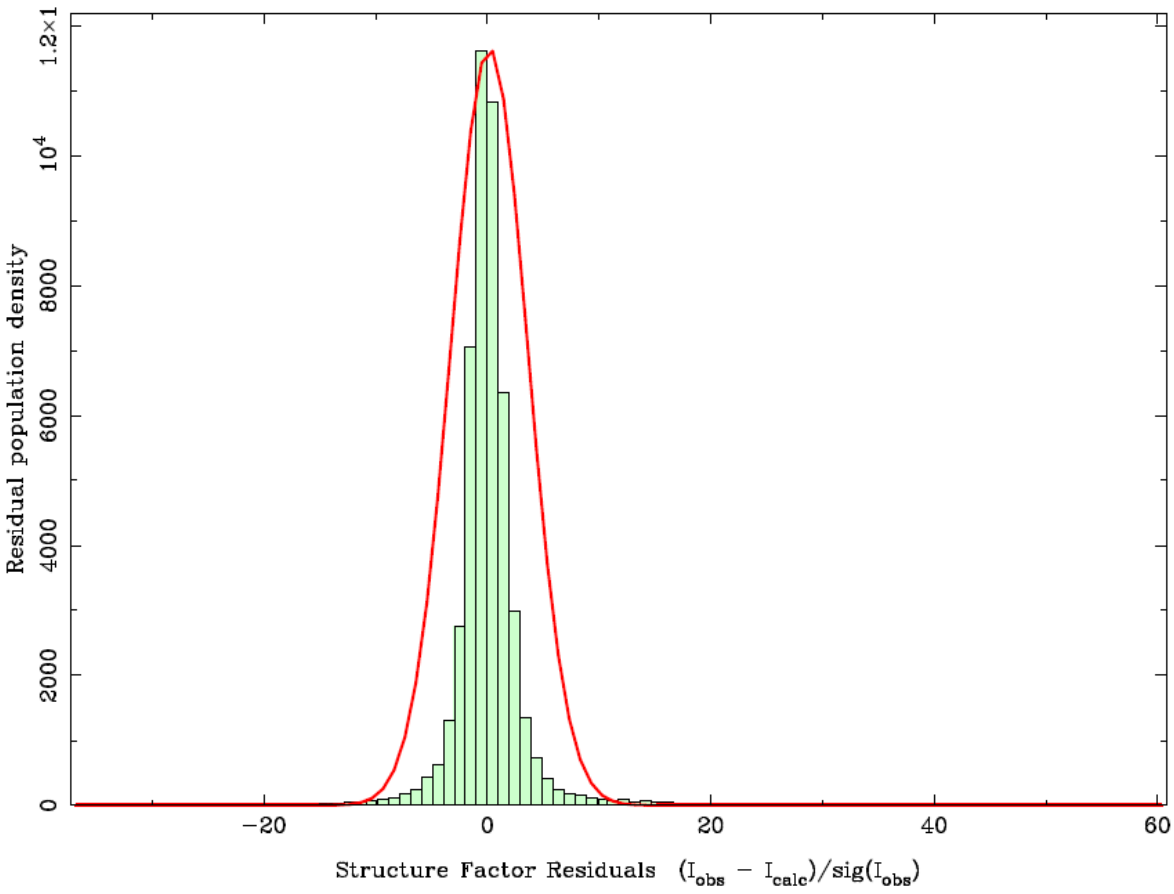


(c) structure factor residuals vs. standard uncertainties for (**2**)

Figure S2: Results of the residual density analysis on the data sets for (**2**)



(a) fractal plot for (**3**); residual density vs. fractal dimension



(b) histogram showing residual density of multipole refinement for (**3**)

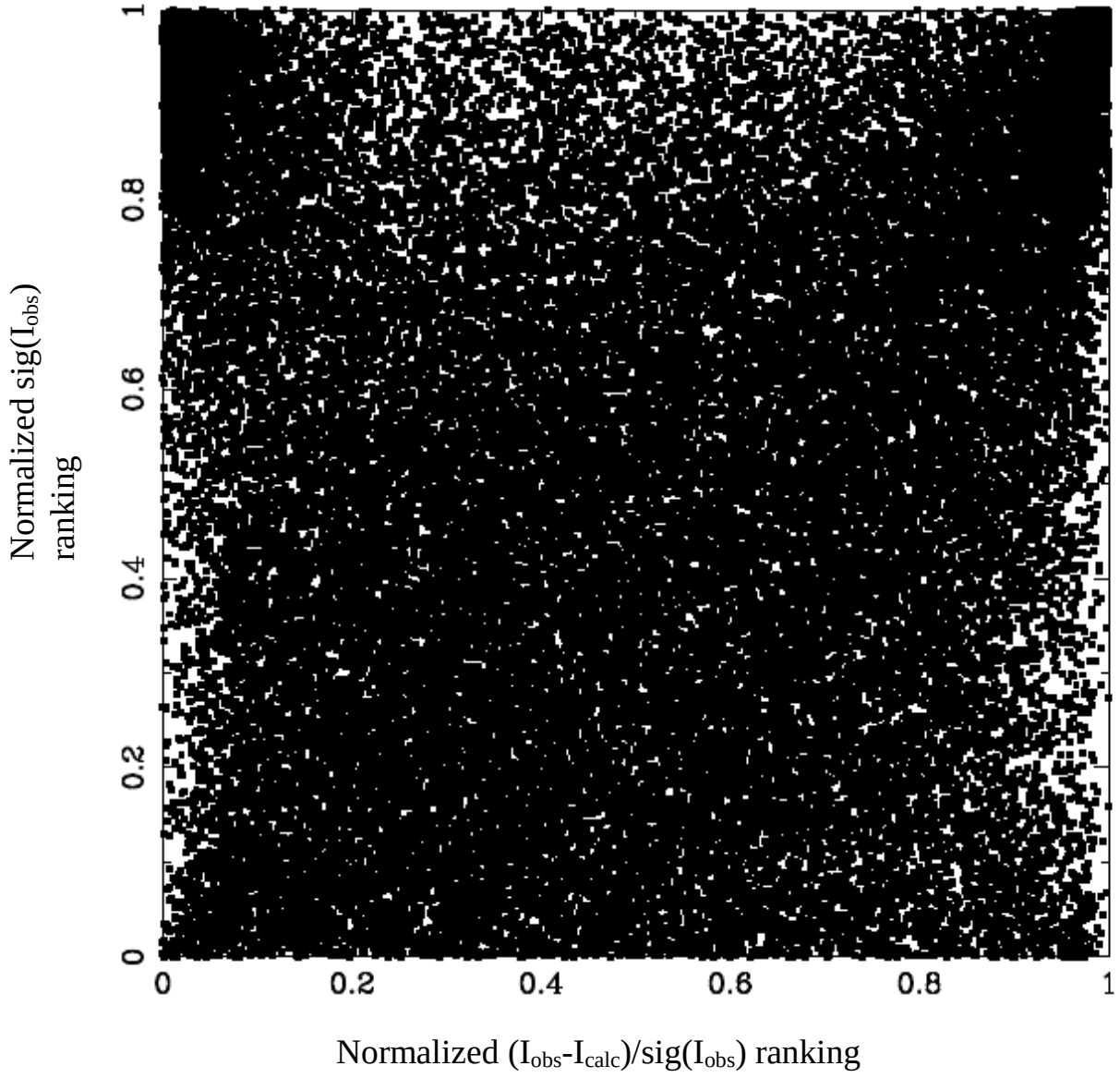


Figure (c) structure factor residuals vs. standard uncertaintes for (**3**).

Figure S3: Results of the residual density analysis on the data sets for (**3**)

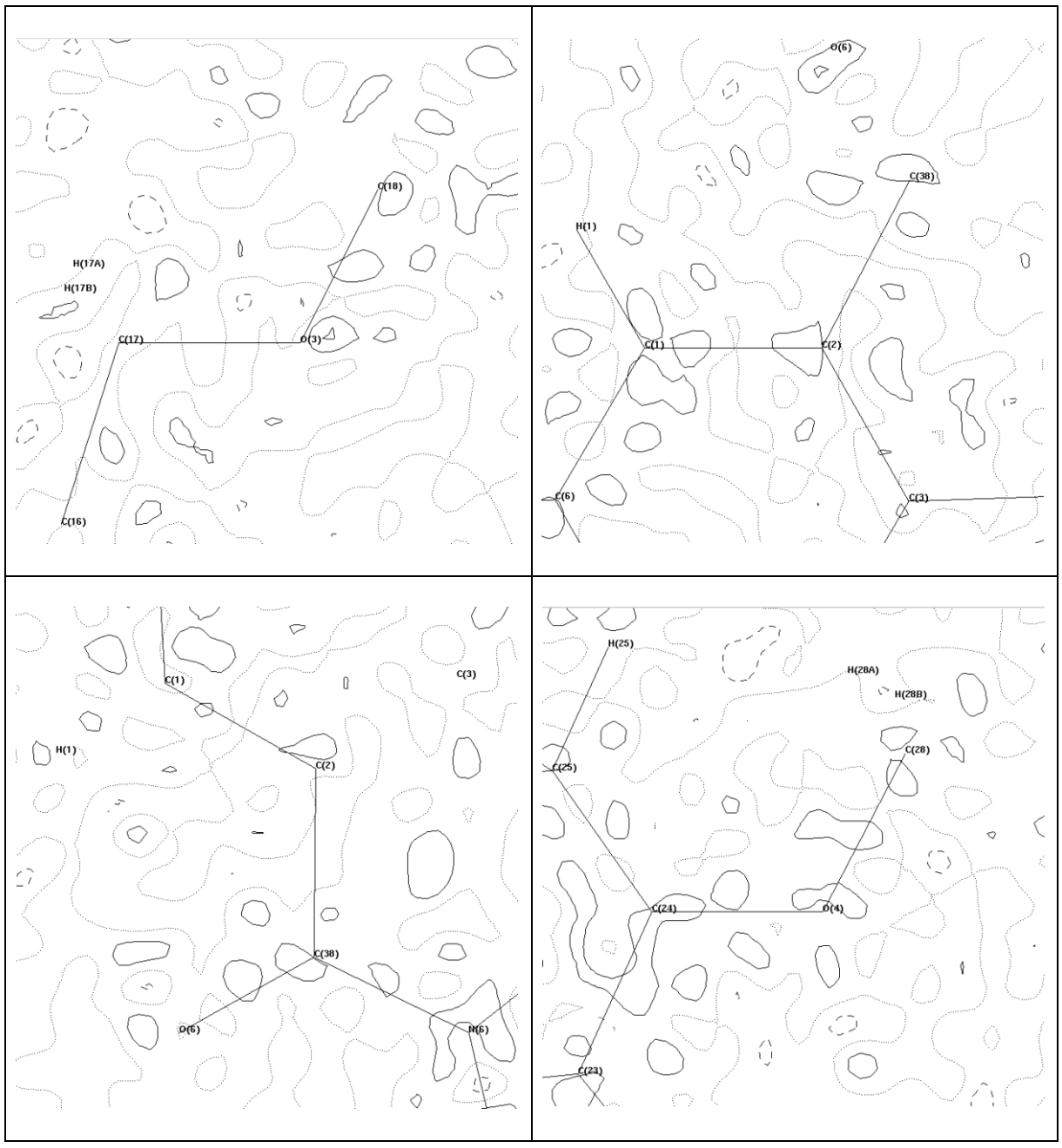


Figure S4: Residual density maps of (**1**). Contours are shown at 0.1 eÅ^{-3}. Solid lines indicate regions of positive electron density and dashed lines indicate regions of negative electron density.

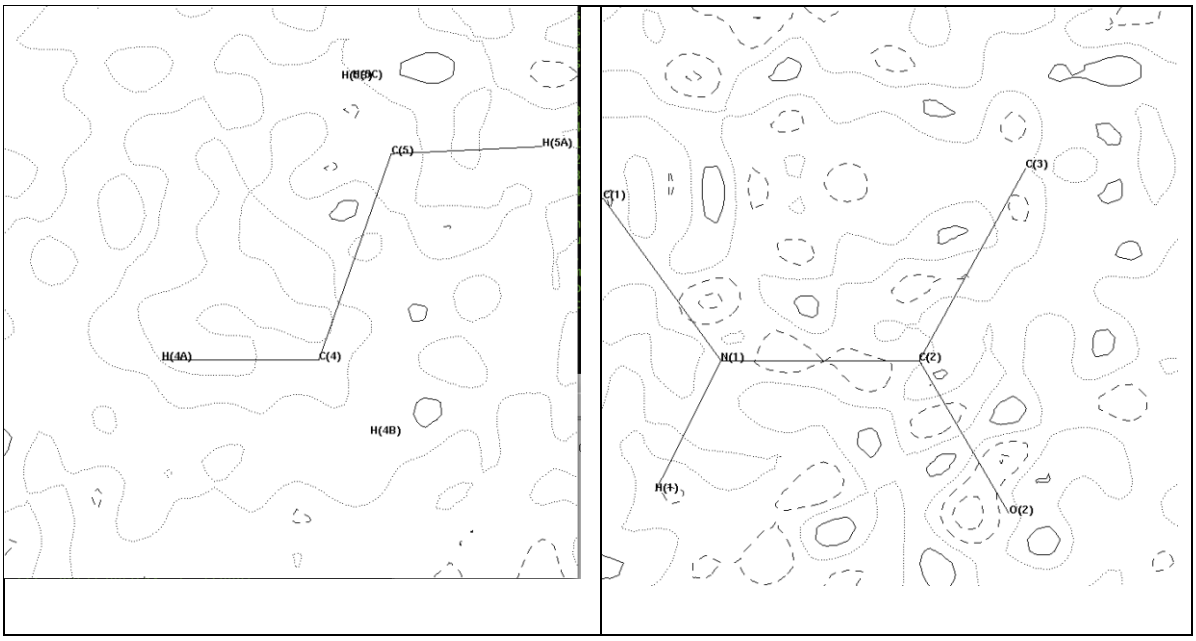


Figure S5: Residual density maps of (**2**). Contours are shown at 0.1 eÅ^{-3}. Solid lines indicate regions of positive electron density and dashed lines indicate regions of negative electron density.

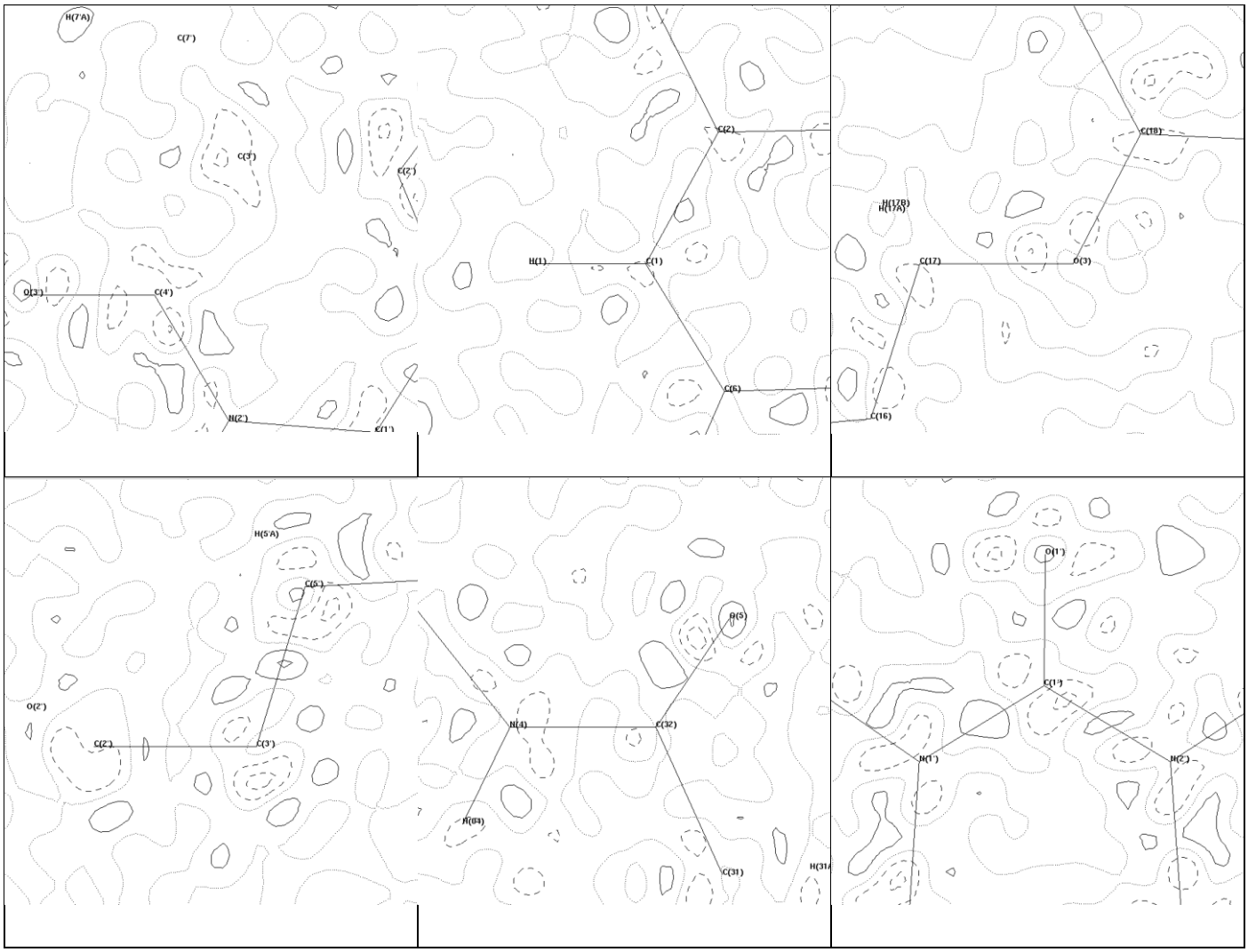


Figure S6: Residual density maps of (**3**). Contours are shown at 0.1 eÅ^{-3}. Solid lines indicate regions of positive electron density and dashed lines indicate regions of negative electron density.

Topological Analysis

Table S22: List of bond critical points (BCP's) found from topological analysis of experimental and theoretical models for (**1**).

	ρ (eÅ^{-3})		$\nabla^2\rho$ (eÅ^{-5})		ε	
Bond	***Exp***	**SP**	***Exp***	**SP**	***Exp***	**SP**
O(1)-C(7)	2.993	2.743	-37.54	-1.66	0.07	0.10
O(2)-C(13)	2.929	2.734	-33.60	-0.51	0.06	0.07
O(3)-C(17)	1.692	1.643	-8.13	-9.91	0.11	0.02
O(3)-C(18)	1.988	1.940	-16.11	-8.65	0.03	0.02
O(4)-C(24)	2.010	1.939	-14.03	-8.66	0.08	0.02
O(4)-C(28)	1.738	1.661	-6.48	-9.15	0.09	0.03
O(5) -C(32)	2.781	2.735	-26.85	-1.81	0.21	0.09
O(6) -C(38)	3.016	2.705	-37.99	-1.88	0.08	0.09
O(001)-C(001)	1.311	1.784	-2.26	-11.66	1.12	0.02
O(001) -H(001)	2.971	2.289	-43.84	-46.99	0.04	0.02
N(1) -C(7)	2.221	2.142	-19.04	-23.63	0.27	0.10
N(1) -C(8)	1.981	2.016	-16.20	-23.23	0.20	0.09
N(1) -H(01)	2.118	2.246	-32.01	-40.88	0.03	0.04
N(2) -C(8)	2.469	2.367	-22.30	-28.15	0.14	0.17
N(2) -C(12)	2.406	2.349	-20.96	-28.11	0.13	0.17
N(3) -C(12)	2.144	2.051	-17.40	-23.96	0.20	0.09
N(3) -C(13)	2.197	2.140	-18.76	-22.88	0.13	0.08
N(3) -H(03)	2.068	2.245	-26.80	-40.88	0.04	0.04
N(4) -C(32)	2.276	2.212	-21.47	-23.05	0.15	0.08
N(4) -C(33)	2.089	2.017	-14.16	-23.91	0.16	0.11
N(4) -H(04)	2.076	2.225	-22.44	-40.84	0.05	0.03
N(5) -C(33)	2.362	2.341	-18.85	-28.22	0.07	0.15
N(5) -C(37)	2.540	2.422	-22.32	-29.36	0.12	0.19
N(6) -C(37)	1.956	1.977	-16.77	-21.70	0.12	0.08
N(6) -C(38)	2.155	2.161	-21.16	-24.41	0.15	0.11
N(6) -H(06)	2.107	2.260	-25.29	-40.91	0.06	0.04
N(01) -C(02)	3.578	3.250	-41.72	10.66	0.05	0.00
C(1) -C(2)	2.086	2.107	-17.24	-20.60	0.17	0.19
C(1) -C(6)	2.064	2.113	-16.82	-20.55	0.18	0.20
C(1) -H(1)	1.759	1.900	-15.79	-24.77	0.06	0.01
C(2) -C(3)	2.108	2.119	-16.87	-20.73	0.22	0.20
C(2) -C(38)	1.754	1.812	-11.49	-16.51	0.15	0.10
C(3) -C(4)	2.128	2.126	-16.95	-21.10	0.18	0.20

C(3) -H(3)	1.773	1.890	-16.37	-23.95	0.03	0.01
C(4) -C(5)	2.167	2.151	-18.96	-21.56	0.16	0.20
C(4) -H(4)	1.788	1.893	-15.90	-24.37	0.05	0.01
C(5) -C(6)	2.067	2.114	-16.85	-20.81	0.16	0.18
C(5) -H(5)	1.740	1.905	-16.55	-25.13	0.02	0.01
C(6) -C(7)	1.773	1.821	-11.32	-16.73	0.13	0.09
C(8) -C(9)	2.138	2.126	-19.04	-21.40	0.25	0.24
C(9) -C(10)	2.127	2.141	-19.01	-21.34	0.19	0.20
C(9) -H(9)	1.834	1.897	-17.98	-25.17	0.08	0.02
C(10) -C(11)	2.136	2.151	-18.55	-21.53	0.21	0.20
C(10) -H(10)	1.783	1.899	-16.56	-24.62	0.05	0.00
C(11) -C(12)	2.051	2.097	-17.78	-20.77	0.15	0.23
C(11) -H(11)	1.813	1.895	-16.89	-24.99	0.05	0.02
C(13) -C(14)	1.760	1.760	-10.91	-15.68	0.15	0.06
C(14) -C(15)	1.650	1.657	-9.66	-13.59	0.01	0.01
C(14) -H(14A)	1.722	1.838	-13.55	-22.34	0.04	0.01
C(14) -H(14B)	1.774	1.844	-13.62	-21.94	0.03	0.01
C(15) -C(16)	1.620	1.670	-9.33	-13.80	0.06	0.01
C(15) -H(15A)	1.729	1.852	-14.24	-22.51	0.04	0.01
C(15) -H(15B)	1.729	1.872	-13.97	-23.42	0.05	0.01
C(16) -C(17)	1.689	1.748	-10.66	-15.36	0.06	0.03
C(16) -H(16A)	1.754	1.851	-13.94	-22.55	0.06	0.01
C(16) -H(16B)	1.753	1.840	-12.85	-21.95	0.06	0.01
C(17) -H(17A)	1.779	1.896	-15.17	-23.97	0.05	0.04
C(17) -H(17B)	1.712	1.890	-13.94	-23.57	0.03	0.04
C(18) -C(19)	2.069	2.067	-16.71	-20.52	0.25	0.19
C(18) -C(27)	2.146	2.185	-19.37	-21.91	0.23	0.31
C(19) -C(20)	2.196	2.200	-20.25	-22.08	0.16	0.27
C(19) -H(19)	1.822	1.879	-16.92	-23.97	0.03	0.02
C(20) -C(21)	2.014	2.031	-16.15	-19.45	0.18	0.16
C(20) -H(20)	1.861	1.886	-17.70	-24.02	0.07	0.01
C(21) -C(22)	2.027	2.030	-16.64	-19.46	0.15	0.15
C(21) -C(26)	2.052	2.044	-17.16	-19.51	0.18	0.17
C(22) -C(23)	2.207	2.203	-19.96	-22.12	0.25	0.27
C(22) -H(22)	1.777	1.885	-16.39	-24.01	0.05	0.01
C(23) -C(24)	2.037	2.064	-17.70	-20.49	0.28	0.19
C(23) -H(23)	1.823	1.878	-17.87	-23.93	0.07	0.02
C(24) -C(25)	2.177	2.181	-20.89	-21.83	0.25	0.31
C(25) -C(26)	1.992	2.004	-16.84	-18.87	0.18	0.17
C(25) -H(25)	1.768	1.869	-17.12	-23.21	0.11	0.03
C(26) -C(27)	2.024	2.026	-16.60	-19.28	0.16	0.17

C(27) -H(27)	1.813	1.868	-17.03	-23.22	0.06	0.02
C(28) -C(29)	1.657	1.751	-10.26	-15.44	0.15	0.05
C(28) -H(28A)	1.700	1.899	-13.05	-24.01	0.09	0.04
C(28) -H(28B)	1.733	1.896	-13.94	-23.97	0.08	0.04
C(29) -C(30)	1.563	1.658	-7.65	-13.61	0.20	0.01
C(29) -H(29A)	1.731	1.848	-15.21	-22.42	0.03	0.01
C(29) -H(29B)	1.634	1.850	-12.63	-22.30	0.03	0.01
C(30) -C(31)	1.495	1.685	-5.02	-13.95	0.56	0.02
C(30) -H(30A)	1.610	1.849	-9.21	-22.21	0.26	0.01
C(30) -H(30B)	1.358	1.860	-4.21	-22.57	0.20	0.00
C(31) -C(32)	1.559	1.755	-7.31	-15.64	0.63	0.07
C(31) -H(31A)	1.299	1.831	-2.87	-21.93	0.12	0.01
C(31) -H(31B)	1.742	1.852	-9.33	-22.57	0.81	0.02
C(33) -C(34)	2.114	2.132	-17.55	-21.43	0.23	0.24
C(34) -C(35)	2.158	2.124	-19.71	-21.00	0.16	0.20
C(34) -H(34)	1.817	1.896	-16.82	-25.15	0.07	0.02
C(35) -C(36)	2.152	2.154	-18.54	-21.52	0.21	0.21
C(35) -H(35)	1.899	1.898	-18.94	-24.62	0.07	0.00
C(36) -C(37)	2.115	2.118	-17.69	-21.02	0.23	0.25
C(36) -H(36)	1.678	1.867	-15.97	-23.24	0.06	0.02
C(01) -C(02)	1.901	1.821	-15.49	-17.16	0.20	0.01
C(01) -H(01A)	1.754	1.961	-13.64	-26.34	0.67	0.01
C(01) -H(01B)	1.942	1.959	-20.29	-26.19	0.16	0.03
C(01) -H(01C)	1.890	1.960	-16.55	-26.22	0.73	0.03
C(001) -C(002)	0.978	1.712	3.59	-14.46	0.49	0.05
C(001) -H(00A)	1.402	1.877	-8.35	-23.11	0.29	0.05
C(001) -H(00B)	1.410	1.882	-6.61	-23.28	0.19	0.05
C(002) -H(00C)	1.883	1.951	-7.92	-25.14	0.70	0.03
C(002) -H(00D)	1.361	1.951	-0.98	-24.49	0.17	0.02
C(002) -H(00E)	1.652	1.947	-9.01	-25.24	0.37	0.02

Table S23: List of ring and cage critical points (RCP, CCP) found from topological analysis of experimental and theoretical models for (**1**).

	ρ (eÅ^{-3})		$\nabla^2\rho$ (eÅ^{-5})	
Bond	***Exp***	**SP**	***Exp***	**SP**
RCP				
C(1)-C(6)-C(5)-C(4)-C(3)-C(2)	0.175	0.14	3.300	3.97
O(2)-C(13)-N(3)-C(12)-C(11)-H(11)	0.111	0.09	1.600	1.75
C(21)-C(20)-C(19)-C(18)-C(27)-C(26)	0.159	0.13	3.200	3.73
C(24)-C(23)-C(22)-C(21)-C(26)-C(25)	0.142	0.13	3.100	3.71
H(25)-C(25)-C(26)-O(4)-C(28)-H(28A)		0.09		1.75
N(5)-C(37)-C(36)-C(35)-C(34)-C(33)	0.196	0.16	3.700	4.24
N(6)-H(06)-H(3)-C(3)-C(2)-C(38)		0.09		1.88
O(1)-C(7)-N(1)-C(8)-C(9)-H(9)	0.098	0.08	1.500	1.65
N(2)-C(12)-C(11)-C(10)-C(9)-C(8)	0.185	0.16	3.600	4.22
C(8)-N(1))-C(13)-C(14)-C(17)-C(18)-C(25)-C(26)-C(27)-C(12)-N(2)-N(3)-O(3)H(14B)-H(17B)-H(25)-H(28A)-C(001)	0.000	0.00	0.000	0.00
C(01)-C(02)-O(2)-C(13)-C(14)-C(15)	0.021	0.06	0.300	0.93
H(34)-C(34)-C(33)-N(4)-C(32)-O(5)	0.104	0.08	1.500	1.54
N(5)-O(6)-H(001)		0.08		1.07
C(30)-H(30B)-O(001)-C(001)-C(002)-H(001E)-H(30A)	0.021	0.02	0.200	0.23
H(001B)-H(28A)-C(28)-C(29)-C(30)-H(30B)-O(001)-C(001)	0.021	0.02	0.200	0.17
H(28A)-C(28)-H(28B)-H(001E)-C(002)-C(001)-H(001B)	0.021	0.01	0.200	0.16
O(001)-H(001)-N(5)-C(33)-N(4)-H(04)	0.080	0.07	0.900	0.98
C(38)-N(6)-C(37)-N(5)-O(6)		0.09		1.60
C(13)-C(14)-C(15)-H(15)-N(01)-C(01)-C(02)-O(2)	0.021	0.02	0.300	0.30
H(14A)-C(14)-C(15)-C(16)-C(17)-H(17A)		0.07		1.09
H(30A)-H(001E)-C(30)-C929)-C(28)-H(26B)	0.021	0.01	0.200	0.14
CCP				
C(28)- C(29)-C(30)-H(28A)-H(28B)-H(30A0 H(30B)-C(001)-C(002)-O(001)-H(001B)-H(001E)	0.053	0.011432	0.700	0.142785

Table S24: List of bond critical points (BCP's) found from topological analysis of experimental and theoretical models for (**2**).

	ρ (eÅ^{-3})		$\nabla^2\rho$ (eÅ^{-5})		ε	
Bond	***Exp***	**SP**	***Exp***	**SP**	***Exp***	**SP**
O(1) -C(1)	3.133	2.840	-46.97	-9.41	0.13	0.11
O(2) -C(2)	3.055	2.726	-43.15	-8.48	0.18	0.08
N(1) -C(1)	2.159	2.101	-22.71	-23.11	0.25	0.12
N(1) -C(2)	2.246	2.115	-24.15	-21.43	0.27	0.04
N(1) -H(1)	1.899	2.745	-27.83	-63.65	0.05	0.04
C(2) -C(3)	1.820	1.728	-18.73	-15.04	0.22	0.06
C(3) -C(4)	1.630	1.562	-10.92	-11.96	0.36	0.01
C(4) -C(5)	1.650	1.638	-10.23	-13.38	0.14	0.02
C(4) -H(4A)	1.805	2.219	-15.81	-31.27	0.06	0.02
C(4) -H(4B)	1.752	2.157	-14.63	-29.57	0.05	0.01
C(5) -H(5A)	1.843	2.161	-14.34	-29.62	0.06	0.02
C(5) -H(5B)	1.865	2.302	-18.86	-33.26	0.17	0.02
C(5) -H(5C)	1.792	2.054	-12.70	-26.71	0.06	0.02

Table S25: List of ring and cage critical points (RCP, CCP) found from topological analysis of experimental and theoretical models for (**2**).

	ρ (eÅ^{-3})		$\nabla^2\rho$ (eÅ^{-5})	
Bond	***Exp***	**SP**	***Exp***	**SP**
C(2) - C(3) - C(2) - N(1) - C(1) - N(1)		0.127		3.29

Table S26: List of bond critical points (BCP's) found from topological analysis of experimental and theoretical models for (**3**).

	ρ (eÅ^{-3})		$\nabla^2\rho$ (eÅ^{-5})		ε	
Bond	***Exp***	**SP**	***Exp***	**SP**	***Exp***	**SP**
O(1) -C(7)	2.722	2.734	-14.54	-1.76	0.18	0.09
O(2) -C(13)	2.906	2.719	-27.74	-2.23	0.18	0.08
O(3) -C(17)	1.703	1.665	-9.77	-9.16	0.12	0.03
O(3) -C(18)	2.016	1.940	-14.48	-8.74	0.17	0.02
O(4) -C(24)	2.055	1.947	-17.81	-8.63	0.07	0.02
O(4) -C(28)	1.650	1.663	-9.57	-9.40	0.12	0.02
O(5) -C(32)	2.969	2.729	-32.70	-1.75	0.16	0.09
O(6) -C(38)	2.771	2.678	-24.97	-2.67	0.09	0.08
O(6) -H(OS1)	0.157	0.232	4.99	3.06	0.06	0.02
O(1') -C(1')	2.943	2.752	-33.15	-5.75	0.16	0.12
O(2') -C(2')	3.014	2.790	-37.74	0.44	0.17	0.09
O(3') -C(4')	3.061	2.740	-32.79	-0.78	0.12	0.08
O(3') -H(04)	0.185	0.238	3.08	2.84	0.01	0.02
O(1S) -C(1S)	1.766	1.765	-10.72	-12.39	0.11	0.01
O(1S) -H(OS1)	2.422	2.319	-22.44	-45.64	0.04	0.02
N(1) -C(7)	2.146	2.145	-19.18	-24.18	0.13	0.10
N(1) -C(8)	2.091	2.009	-19.56	-23.33	0.07	
N(1) -H(01)	1.921	2.233	-24.09	-40.73	0.06	0.03
N(2) -C(8)	2.341	2.327	-23.38	-27.84	0.19	0.17
N(2) -C(12)	2.349	2.309	-21.36	-27.37	0.12	0.18
N(3) -C(12)	2.062	2.048	-16.35	-24.02	0.18	0.09
N(3) -C(13)	2.226	2.141	-21.53	-23.39	0.12	0.08
N(3) -H(03)	1.825	2.245	-23.11	-40.67	0.05	0.04
N(4) -C(32)	2.151	2.163	-20.89	-23.90	0.12	0.09
N(4) -C(33)	2.124	2.064	-19.32	-24.68	0.15	0.10
N(4) -H(04)	1.887	2.233	-26.42	-40.66	0.05	0.03
N(5) -C(33)	2.263	2.311	-21.67	-27.20	0.23	0.17
N(5) -C(37)	2.337	2.335	-21.30	-28.00	0.24	0.18
N(6) -C(37)	1.923	1.961	-15.79	-22.45	0.21	0.10
N(6) -C(38)	2.185	2.183	-20.71	-24.47	0.14	0.11
N(6) -H(06)	1.917	2.235	-23.61	-40.79	0.07	0.03
N(1') -C(1')	2.173	-8.015	-20.04	3.19	0.14	0.13
N(1') -C(2')	2.074	2.104	-19.22	24.02	0.19	0.05
N(1') -H(B1)	1.942	2.190	-22.17	-40.17	0.04	0.03
N(2') -C(1')	2.055	2.176	-19.31	-27.63	0.10	0.13
N(2') -C(4')	2.200	2.150	-20.17	-24.58	0.19	0.05
N(2') -H(B2)	1.921	2.189	-26.17	-40.16	0.03	0.03

C(1) -C(2)	2.142	2.083	-17.93	-19.94	0.18	0.20
C(1) -C(6)	2.124	2.091	-17.46	-20.11	0.25	0.20
C(1) -H(1)	1.820	1.898	-17.43	-24.50	0.02	0.01
C(2) -C(3)	2.137	2.102	-18.13	-20.50	0.20	0.19
C(2) -C(38)	1.771	1.818	-11.49	-16.66	0.13	0.09
C(3) -C(4)	2.184	2.138	-18.90	-21.32	0.23	0.19
C(3) -H(3)	1.775	1.901	-15.59	-24.81	0.06	0.01
C(4) -C(5)	2.224	2.136	-19.31	-21.29	0.26	0.19
C(4) -H(4)	1.879	1.890	-19.38	-24.29	0.04	0.01
C(5) -C(6)	2.169	2.111	-18.30	-20.69	0.25	0.19
C(5) -H(5)	1.901	1.904	-18.33	-25.05	0.04	0.01
C(6) -C(7)	1.830	1.805	-12.71	-16.42	0.09	0.09
C(8) -C(9)	2.207	2.110	-20.35	-21.08	0.24	0.24
C(9) -C(10)	2.295	2.137	-23.10	-21.28	0.22	0.19
C(9) -H(9)	1.854	1.901	-19.30	-25.46	0.10	0.02
C(10) -C(11)	2.255	2.148	-22.32	-21.47	0.23	0.20
C(10) -H(10)	1.884	1.899	-19.51	-24.70	0.12	0.00
C(11) -C(12)	2.205	2.108	-19.23	-21.01	0.22	0.24
C(11) -H(11)	1.849	1.894	-18.46	-24.98	0.08	0.02
C(13) -C(14)	1.817	1.742	-13.86	-15.36	0.03	0.06
C(14) -C(15)	1.700	1.668	-11.77	-13.80	0.12	0.02
C(14) -H(14A)	1.849	1.843	-17.30	-22.12	0.10	0.01
C(14) -H(14B)	1.693	1.831	-15.09	-21.89	0.05	0.01
C(15) -C(16)	1.687	1.646	-11.58	-13.41	0.03	0.01
C(15) -H(15A)	1.782	1.861	-17.00	-22.93	0.20	0.01
C(15) -H(15B)	1.710	1.850	-14.60	-22.31	0.01	0.01
C(16) -C(17)	1.783	1.736	-12.80	-15.17	0.07	0.05
C(16) -H(16A)	1.724	1.853	-16.57	-22.50	0.03	0.01
C(16) -H(16B)	1.799	1.844	-18.15	-22.22	0.03	0.01
C(17) -H(17A)	1.760	1.889	-15.59	-23.66	0.09	0.04
C(17) -H(17B)	1.699	1.894	-13.98	-23.85	0.06	0.04
C(18) -C(19)	2.101	2.060	-17.35	-20.39	0.27	0.19
C(18) -C(27)	2.267	2.175	-21.71	-21.69	0.26	0.31
C(19) -C(20)	2.233	2.187	-20.06	-21.78	0.31	0.27
C(19) -H(19)	1.753	1.877	-15.52	-23.94	0.08	0.02
C(20) -C(21)	2.108	2.026	-18.63	-19.37	0.18	0.15
C(20) -H(20)	1.816	1.885	-17.51	-24.02	0.10	0.01
C(21) -C(22)	2.027	2.020	-15.76	-19.25	0.24	0.15
C(21) -C(26)	2.056	2.041	-16.80	-19.44	0.19	0.17
C(22) -C(23)	2.218	2.185	-21.01	-21.76	0.24	0.27
C(22) -H(22)	1.811	1.885	-16.06	-24.01	0.13	0.01

C(23) -C(24)	2.102	2.059	-18.54	-20.38	0.25	0.19
C(23) -H(23)	1.827	1.877	-18.51	-23.95	0.19	0.02
C(24) -C(25)	2.262	2.179	-21.30	-21.79	0.30	0.31
C(25) -C(26)	2.059	2.010	-17.92	-18.96	0.19	0.17
C(25) -H(25)	1.797	1.865	-17.41	-23.06	0.09	0.03
C(26) -C(27)	2.091	2.005	-17.94	-18.86	0.20	0.17
C(27) -H(27)	1.845	1.867	-18.46	-23.11	0.12	0.03
C(28) -C(29)	1.762	1.736	-11.76	-15.13	0.13	0.05
C(28) -H(28A)	1.862	1.891	-15.89	-23.71	0.06	0.04
C(28) -H(28B)	1.729	1.891	-16.42	-23.77	0.05	0.04
C(29) -C(30)	1.701	1.653	-11.69	-13.54	0.08	0.01
C(29) -H(29A)	1.747	1.850	-15.49	-22.41	0.05	0.01
C(29) -H(29B)	1.779	1.851	-15.78	-22.48	0.06	0.01
C(30) -C(31)	1.609	1.622	-8.96	-12.98	0.10	0.01
C(30) -H(30A)	1.728	1.859	-15.78	-22.74	0.02	0.01
C(30) -H(30B)	1.839	1.848	-16.22	-22.30	0.06	0.01
C(31) -C(32)	1.727	1.751	-11.68	-15.56	0.10	0.05
C(31) -H(31A)	1.809	1.848	-16.74	-22.48	0.04	0.01
C(31) -H(31B)	1.827	1.847	-17.16	-22.21	0.08	0.01
C(33) -C(34)	2.257	2.104	-22.14	-20.94	0.23	0.23
C(34) -C(35)	2.218	2.156	-21.54	-21.62	0.25	0.20
C(34) -H(34)	1.834	1.896	-20.51	-25.17	0.10	0.02
C(35) -C(36)	2.192	2.126	-20.06	-21.05	0.20	0.19
C(35) -H(35)	1.831	1.899	-18.00	-24.74	0.05	0.00
C(36) -C(37)	2.206	2.113	-19.60	-21.14	0.25	0.24
C(36) -H(36)	1.847	1.903	-18.67	-25.48	0.09	0.02
C(2') -C(3')	1.628	1.755	-10.07	-15.44	0.12	0.07
C(3') -C(4')	1.764	1.788	-12.73	-16.04	0.08	0.07
C(3') -C(5')	1.521	1.550	-6.83	-11.87	0.03	0.01
C(3') -C(7')	1.532	1.617	-7.17	-12.91	0.07	0.01
C(5') -C(6')	1.717	1.648	-11.73	-13.42	0.05	0.01
C(5') -H(5'A)	1.725	1.864	-15.38	-22.93	0.04	0.01
C(5') -H(5'B)	1.710	1.860	-15.05	-22.84	0.06	0.02
C(6') -H(6'A)	1.825	1.967	-17.46	-25.50	0.13	0.01
C(6') -H(6'B)	1.699	1.966	-13.99	-25.88	0.05	0.01
C(6') -H(6'C)	1.761	1.964	-15.83	-25.47	0.19	0.01
C(7') -C(8')	1.733	1.661	-11.08	-13.63	0.03	0.01
C(7') -H(7'A)	1.663	1.859	-13.78	-22.74	0.03	0.01
C(7') -H(7'B)	1.845	1.861	-17.53	-22.80	0.03	0.01
C(8') -H(8'A)	1.693	1.961	-11.65	-25.39	0.15	0.01
C(8') -H(8'B)	1.800	1.970	-16.34	-25.98	0.11	0.01

C(8') -H(8'C)	2.005	1.965	-19.07	-25.43	0.11	0.01
C(1S) -C(2S)	1.562	1.729	-4.71	-14.89	0.59	0.04
C(1S) -H(1SA)	1.370	1.892	-5.51	-23.80	0.15	0.04
C(1S) -H(1SB)	1.967	1.878	-16.95	-23.07	0.09	0.04
C(2S) -H(2SA)	1.042	1.964	2.91	-25.66	0.67	0.00
C(2S) -H(2SB)	1.382	1.951	0.54	-25.17	0.31	0.01
C(2S) -H(2SC)	1.138	1.958	0.67	-25.51	0.67	0.00

Table S27: List of ring and cage critical points (RCP, CCP) found from topological analysis of experimental and theoretical models for (**3**).

	ρ (eÅ^{-3})		$\nabla^2\rho$ (eÅ^{-5})	
Bond	***Exp***	**SP**	***Exp***	**SP**
O(1')-C(1')-N(1')-H(B1)-N(2)-C(8)-N(1)-H(01)	0.036	0.029	0.40	0.45
O(1)-C(7)-N(1)-C(8)-C(9)-H(9)	0.127	0.092	1.80	1.92
O(2')-N(3)-C(12)-N(2)-H(B1)-N(1')-C(2')	0.038	0.028	0.40	0.43
O(2)-C(13)-N(3)-C(12)-C(11)-H(11)	0.115	0.085	1.50	1.63
O(3')-C(4')-N(2')-H(B2)-N(5)-C(33)-N(4)-H(04)	0.049	0.037	0.60	0.61
O(5)-C32)-N(4)-C(33)-C(34)-H(34)	0.107	0.082	1.40	1.64
O(6)-C(38)-N(6)-C(37)-C(36)-H(36)	0.131	0.094	1.80	2.00
O(1')-C(1')-N(2')-H(B2)-N(5)-C(37)-N(6)-H(06)	0.036	0.031	0.50	0.52
N(1')-C(1')-N(2')-C(4')-C(3')-C(2')	0.188	0.125	2.60	3.22
C(1)-C(2)-C(3)-C(4)-C(5)-C(6)	0.194	0.141	3.30	3.92
N(2)-C(8)-C(9)-C(10)-C(11)-C(12)	0.205	0.152	3.50	4.15
C(18)-C(19)-C(20)-C(21)-C(26)-C(27)	0.181	0.134	3.00	3.69
C(21)-C(22)-C(23)-C(24)-C(25)-C(26)	0.179	0.133	3.10	3.68
O(3')-H(31B)0C(31)-C(32)-N(4)-H(04)	0.008	0.052	0.10	0.80
N(5)-C(33)-C(24)-C(35)-C(36)-C(37)	0.049	0.153	0.60	4.15
C(25)-C(26)-C(27)-H(27)-H(8'B)-H(25)	0.030	0.023	0.30	0.29
O(2')-N(3)-C(13)-C(14)-H(14A)	0.052	0.041	0.60	0.60
O(1')-H(1)-C(1)-C(6)-C(7)-N(1)-H01)	0.053	0.045	0.70	0.73
O(1')-H(1)-C(1)-C(2)-C(38)-N(6)-H(06)	0.052	0.048	0.70	0.78
C(7')-C(8')-H(8'B)-H(27)-H(7'B)	0.030	0.029	0.30	0.39
C(7')-C(8')-H(8'B)-H(25)-H(7'A)	0.030	0.026	0.30	0.35
O(4)-C(24)-C(25)-H(25)-H(7'A)-C(7')-C(3')-C(4')-O(3')-H(31B)-C(31)-C(30)-C(29)-C(28)	0.008	0.007	0.10	0.10

O(3)-C(17)-C(16)-C(15)-C(14)-H(14A)-O(2')-C(2')-C(3')-C(7')-H(7'B)-H(27)-C(27)-C(18)	0.007	0.006	0.10	0.08
O(3)-C(17)-C(16)-C(15)-C(14)-H(14A)-O(2')-C(2')-C(3')-C(7')-C(8')-H(8'B)-H(27)-C(27)-C(18)	1.703	0.006	-9.80	0.07
O(4)-C(24)-C(25)-H(25)-H(7'A)-C(7')-C(8')-C(31)-C(3')-C(4')-C(29)-C(30)-O(3')-H(31B)-C(28)	0.008	0.006	0.10	0.07
O(6)-C(38)-C(2)-C(3)-H(3)-H(1SB)-C(1S)-O(1S)-H(OS1)	0.157	0.020	5.00	0.32
O(4)-O(3')-C(24)-C(25)-C(28)-C(29)-C(30)-C(31)-C(3')-C(4')-C97')-C(8')-H(25)-H(31B)-H(7'A)	0.008	0.006	0.10	0.08
O(3)-O(2')-C(14)-C(15)-C(16)-C(17)-C(18)-C(27)-C(2')-C(3')-C(7')-C(8')-H(14A)-H(27)-H(7'B)-H(8'B)	0.000	0.006	0.00	0.08

Hydrogen bond geometry

Table S28: Geometrical details for hydrogen bonds and short contacts found in (**1**).

Bond	**dH...A** (Å)	**dD...A**(Å)	**<(DHA)** (°)
Intramolecular			
Macrocycle – macrocycle interactions*			
C(9)-H(9)···O(1)	2.192	2.861	117.8
C(11)-H(11)···O(2)	2.149	2.830	118.4
C(34)-H(34)···O(5)	2.305	2.863	110.1
Intermolecular			
Macrocycle – ethanol interactions			
N(4)-H(04)···O(001)	1.890	2.871	163.3
O(001)-H(001)···N(5)	2.390	2.910	113.3
O(001)-H(001)···O(6)	1.814	2.757	164.0
C(30)-H(30B)···O(001)	2.452	3.272	130.8
Macrocycle – acetonitrile interactions			
C(15)-H(15B)···N(01)	2.662	3.727	165.1
C(01)-H(01A)···O(1)[a]	2.456	3.376	144.7
C(14)-H(14A)···O(1)[b]	2.525	3.251	123.1
C(14)-H(14B)···O(5)[c]	2.252	3.215	146.0
C(17)-H(17A)···O(6)[d]	2.630	3.662	157.5
C(3)-H(3)···N(01)[e]	2.503	3.307	131.1
C(36)-H(36)···N(01)[e]	2.484	3.430	145.3

N(3)-H(03)···O(5) [c]	2.002	2.955	156.7
N(6)-H(06)···N(01)[e]	2.150	3.139	166.4

Symmetry operators used to define atoms: [a]-x+2, -y, -z+1; [b]-x+1, -y, -z+1; [c]-x, -y+1, -z+1; [d] -x+1, -y+1, -z+1; [e]x-1, y, z-1

[*] Bonds located *via* topological search.

Table S29: Geometrical details for hydrogen bonds and short contacts found in (**2**).

Bond	dH...A (**Å**)	dD...A(**Å**)	<(DHA) **(°)**
Intramolecular			
N(1)-H(1)···O(2)[1]	1.904	2.842	174.5

Symmetry operators used to define atoms: [1]1/2-x,1/2-y,-z

Table S30: Geometrical details for hydrogen bonds and short contacts found in (**3**).

Bond	**dH...A (**Å**)**	**dD...A(**Å**)**	**<(DHA)** (°)
Intramolecular			
Macrocycle – macrocycle interactions*			
C(9)-H(9)···O(1)	2.074	2.764	118.8
C(11)-H(11)···O(2)	2.286	2.860	111.0
C(34)-H(34)···O(5)	2.223	2.873	116.4
C(36)-H(36)···O(6)	2.039	2.747	120.2
Macrocycle – barbital interactions			
N(1')-H(B1)···N(2)	2.055	3.054	170.0
N(2')-H(B2)···N(5)	2.013	3.012	170.5
N(4)-H(04)···O(3')	1.835	2.816	163.1
N(1)-H(01)···O(1')	2.228	3.214	165.3
N(6)-H(06)···O(1')	2.236	3.237	171.3
C(1)-H(1)···O(1')	2.224	3.010	127.7
N(3)-H(03)···O(2')	2.677	3.016	99.7
C(14)-H(14A)···O(2')	2.558	3.268	121.8
Macrocycle – ethanol interactions			
O(1S)-H(OS1)···O(6)	1.789	2.747	170.6
Intermolecular			
C(19)-H(19)···O(3')[a]	2.569	3.451	138.0
C(31)-H(31A)···O(2')[b]	2.281	3.318	158.0
N(3)-H(03)···O(1S)[a]	1.860	2.859	169.9

Symmetry operators used to define atoms: [a]x, y-1, z; [b]x+1, y, z ; [*]Bonds located *via* topological search.

Table S31: Topological analysis of hydrogen bonding in (**1**). Standard uncertainties have been omitted for clarity. They are closely scattered around 0.02 eÅ^{-3} (ρ_{bcp}) and 0.05 eÅ^{-5} ($\nabla^2\rho_{bcp}$).

	ρ	$\nabla^2\rho$	ε	$d_{H\cdots bcp}$	$d_{A\cdots bcp}$	G	V	H	E_{HB}
	/eÅ^{-3}	/eÅ^{-5}		(Å)	(Å)	/E_h eÅ^{-3}	/E_h eÅ^{-3}	/E_h eÅ^{-3}	/kJ mol^{-1}
Intramolecular									
Macrocycle – macrocycle interactions									
C(9)-H(9)···O(1)	0.123	1.66	0.07	0.936	1.314	0.10	-0.09	0.01	35.02
C(11)-H(11)···O(2)	0.134	1.80	0.05	0.919	1.300	0.11	-0.10	0.01	38.91
C(34)-H(34)···O(5)	0.120	1.55	0.18	1.023	1.337	0.10	-0.08	0.01	31.12
Intermolecular									
Macrocycle – ethanol interactions									
N(4)-H(04)···O(001)	0.108	2.20	0.43	0.755	1.198	0.12	-0.09	0.03	35.02
O(001)-H(001)···N(5)	0.100	1.23	0.22	1.049	1.415	0.07	-0.06	0.01	23.34
O(001)-H(001)···O(6)	0.113	4.16	0.47	0.682	1.210	0.22	-0.14	0.08	54.47
C(30)-H(30B)···O(001)	0.053	0.707	0.64	1.156	1.441	0.04	-0.03	0.01	11.67
Macrocycle – acetonitrile interactions									
C(15)-H(15B)···N(01)	0.027	0.521	1.21	1.037	1.717	0.03	-0.02	0.01	7.78
C(01)-H(01A)···O(1)[a]	0.028	0.48	1.81	1.186	1.554	0.02	-0.02	0.01	7.78
C(14)-H(14A)···O(1)[b]	0.060	0.75	0.49	1.192	1.448	0.04	-0.03	0.01	11.67
C(14)-H(14B)···O(5)[c]	0.094	1.44	0.19	0.912	1.356	0.08	-0.06	0.02	23.34
C(17)-H(17A)···O(6)[d]	0.030	0.51	0.10	1.088	1.569	0.03	-0.02	0.01	7.78
C(3)-H(3)···N(01)[e]	0.054	0.71	0.14	1.082	1.486	0.04	-0.03	0.01	11.67
C(36)-H(36)···N(01)[e]	0.047	0.70	0.19	1.020	1.534	0.04	-0.03	0.01	11.67
N(3)-H(03)···O(5)[c]	0.105	2.28	0.05	0.736	1.296	0.13	-0.09	0.03	35.02
N(6)-H(06)···N(01)[e]	0.072	1.75	0.08	0.775	1.395	0.09	-0.06	0.03	23.34

Table S32: Topological analysis of hydrogen bonding in (2). Standard uncertainties have been omitted for clarity. They are closely scattered around 0.02 eÅ^{-3} (ρ_{bcp}) and 0.05 eÅ^{-5} ($\nabla^2\rho_{bcp}$).

	ρ	$\nabla^2\rho$	ε	$d_{H\cdots bcp}$	$d_{A\cdots bcp}$	G	V	H	E_{HB}
	/eÅ^{-3}	/eÅ^{-5}		(Å)	(Å)	/E_h eÅ^{-3}	/E_h eÅ^{-3}	/E_h eÅ^{-3}	/kJ mol^{-1}
Intermolecular									
N(1)-H(1)···O(2)[a]	0.133	3.34	0.02	0.622	1.217	0.18	-0.13	0.05	50.58

Symmetry operators used to define atoms: [a]3/2-x, 1/2-y, -z

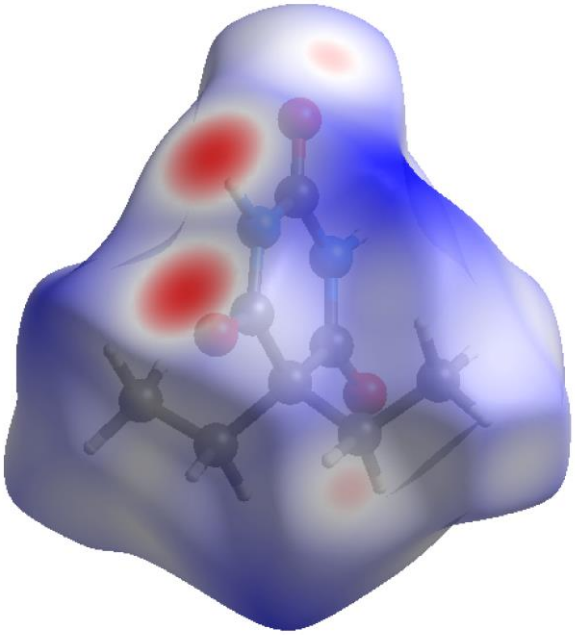
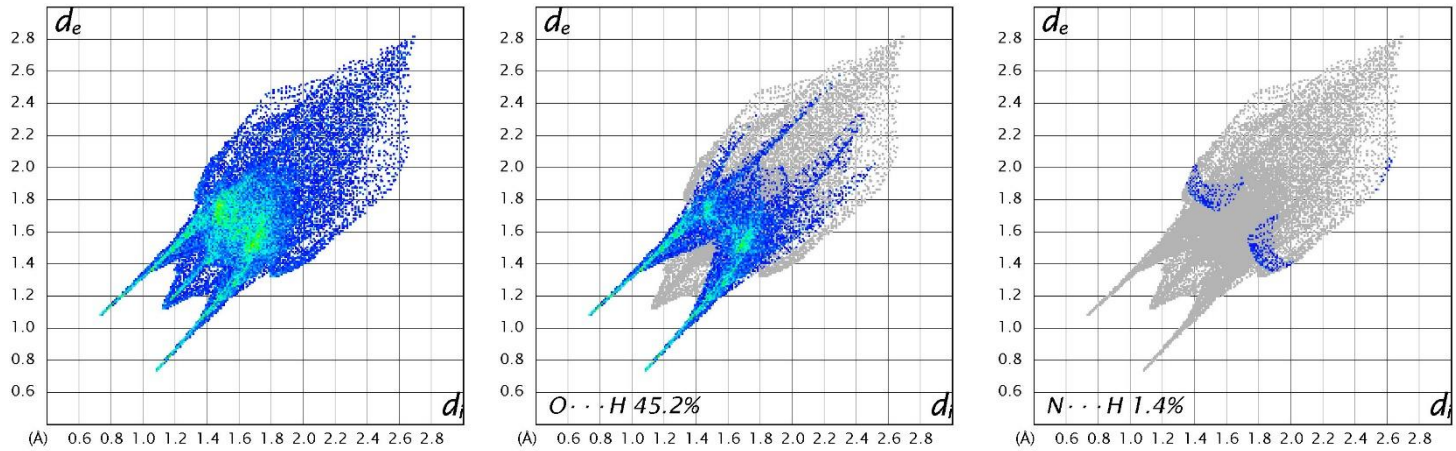
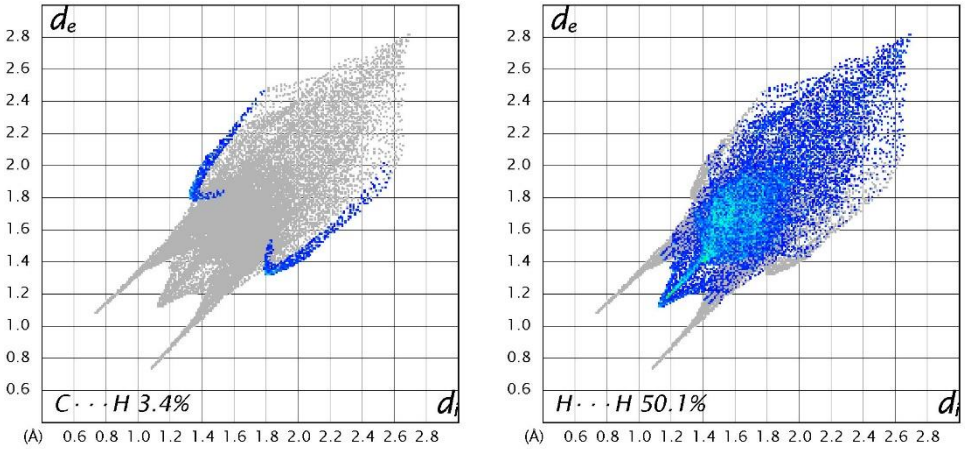


Figure S7: Hirshfeld surface for (**2**).



(a) (b) (c)



(d) (e)

Figure S8: 2-dimensional fingerprint plots of (**2**); (a) all interactions, (b) O···H interactions, (c) N···H interactions, (d) C···H interactions and (e) H···H interactions.

Table S33: Atomic charges (e) from multipole refinement for the macrocycle. Standard uncertainties have been omitted for clarity.

ATOM	**PV (*Exp*)**	**Ω (*Exp*)**	**Ω (DFT)**
O(1) (1)	6.32	-1.12	-1.20
(3)	6.47	-1.26	-1.21
O(2)	6.28	-1.00	-1.23
	6.36	-1.23	-1.22
O(3)	6.38	-1.08	-1.13
	6.36	-1.12	-1.13
O(4)	6.34	-1.04	-1.13
	6.35	-1.12	-1.13
O(5)	6.41	-1.12	-1.23
	6.32	-1.17	-1.23
O(6)	6.31	-1.01	-1.22
	6.43	-1.17	-1.22
N(1)	5.26	-1.22	-1.29
	5.54	-1.41	-1.29
N(2)	5.15	-0.86	-1.29
	5.42	-1.14	-1.29
N(3)	5.27	-1.19	-1.31
	5.54	-1.36	-1.29
N(4)	5.32	-1.22	-1.31
	5.48	-1.41	-1.33
N(5)	5.16	-0.83	-1.23
	5.43	-1.13	-1.29
N(6)	5.23	-1.15	-1.31
	5.56	-1.43	-1.29
C(1)	4.04	-0.05	-0.04
	4.16	-0.22	-0.05
C(2)	4.01	-0.02	-0.01
	4.20	-0.16	-0.01
C(3)	4.10	-0.16	-0.03
	4.22	-0.28	-0.02
C(4)	4.04	-0.08	-0.04
	4.15	-0.19	-0.03
C(5)	3.95	0.02	-0.05
	4.17	-0.22	-0.02
C(6)	3.99	-0.01	-0.01

	4.28	-0.25	-0.01
C(7)	4.05	1.06	1.52
	4.05	1.07	1.51
C(8)	3.97	0.65	1.02
	4.03	0.63	0.95
C(9)	3.87	0.10	-0.06
	4.15	-0.21	-0.03
C(10)	3.92	0.05	-0.03
	4.12	-0.22	-0.02
C(11)	3.89	0.04	-0.03
	4.17	-0.21	-0.02
C(12)	4.03	0.59	0.95
	4.10	0.55	0.97
C(13)	4.10	0.96	1.54
	4.05	1.13	1.51
C(14)	4.00	-0.07	-0.01
	4.10	-0.21	-0.02
C(15)	4.01	-0.07	0.00
	3.99	-0.09	0.02
C(16)	4.02	-0.03	0.03
	4.14	-0.25	0.02
C(17)	4.02	0.23	0.46
	4.15	0.11	0.45
C(18)	3.89	0.48	0.52
	4.13	0.28	0.52
C(19)	3.96	-0.03	-0.04
	4.11	-0.13	-0.04
C(20)	3.95	-0.02	-0.04
	4.09	-0.14	-0.04
C(21)	3.93	0.11	0.01
	4.19	-0.18	0.01
C(22)	3.90	0.08	-0.04
	3.96	0.04	-0.04
C(23)	3.99	-0.06	-0.04
	4.07	-0.12	-0.04
C(24)	3.84	0.53	0.53
	4.05	0.33	0.52
C(25)	3.95	-0.04	-0.06
	4.17	-0.21	-0.07
C(26)	3.95	0.05	0.03
	4.08	-0.07	0.02

C(27)	3.89	0.07	-0.07
	4.12	-0.18	-0.07
C(28)	3.95	0.25	0.44
	4.19	0.05	0.45
C(29)	4.03	-0.05	0.02
	4.22	-0.31	0.03
C(30)	3.82	-0.16	0.02
	4.10	-0.21	0.01
C(31)	3.49	0.10	-0.01
	4.07	-0.19	-0.02
C(32)	4.14	0.93	1.51
	4.04	1.16	1.51
C(33)	4.12	0.46	0.98
	3.95	0.72	0.98
C(34)	4.02	-0.07	-0.04
	4.22	-0.32	-0.03
C(35)	3.89	0.04	-0.03
	4.02	-0.10	-0.02
C(36)	4.05	-0.13	-0.04
	4.21	-0.27	-0.04
C(37)	3.99	0.65	0.98
	4.05	0.58	0.93
C(38)	3.92	1.18	1.53
	4.08	1.04	1.50
H(1)	0.88	0.17	0.09
	0.87	0.21	0.08
H(3)	0.89	0.18	0.10
	0.92	0.13	0.09
H(4)	0.98	0.06	0.05
	0.93	0.11	0.07
H(5)	0.87	0.17	0.07
	0.98	0.09	0.10
H(9)	0.93	0.14	0.08
	0.85	0.25	0.15
H(10)	0.97	0.04	0.06
	0.93	0.15	0.07
H(11)	0.90	0.21	0.13
	0.88	0.20	0.13
H(14A)	1.02	0.05	0.06
	0.96	0.10	0.02
H(14B)	1.04	-0.02	0.04

	0.84	0.23	0.03
H(15A)	0.90	0.14	0.01
	0.97	0.09	0.05
H(15B)	1.02	0.01	0.00
	0.85	0.21	0.02
H(16A)	1.12	-0.11	0.02
	0.83	0.23	0.03
H(16B)	1.00	0.01	0.04
	0.83	0.23	0.02
H(17A)	0.96	0.07	0.03
	0.93	0.11	0.02
H(17B)	0.91	0.11	0.03
	0.85	0.19	0.03
H(19)	1.01	0.04	0.07
	0.96	0.10	0.07
H(20)	1.01	0.04	0.05
	0.96	0.08	0.05
H(22)	0.91	0.15	0.05
	1.10	-0.07	0.05
H(23)	0.92	0.16	0.07
	0.94	0.13	0.08
H(25)	0.86	0.23	0.04
	0.88	0.18	0.04
H(27)	0.93	0.13	0.04
	0.92	0.13	0.04
H(28A)	1.03	0.02	0.01
	1.01	0.01	0.02
H(28B)	0.98	0.07	0.04
	0.78	0.29	0.03
H(29A)	0.81	0.22	0.02
	0.87	0.17	0.03
H(29B)	0.97	0.07	0.03
	0.94	0.12	0.03
H(30A)	0.89	0.26	0.05
	0.89	0.16	0.04
H(30B)	1.13	0.09	0.03
	0.99	0.07	0.01
H(31A)	1.07	0.12	0.00
	0.89	0.18	0.05
H(31B)	1.17	0.00	0.05
	0.96	0.11	0.02

H(34)	0.90	0.19	0.14
	0.77	0.34	0.14
H(35)	1.04	0.02	0.07
	0.91	0.17	0.08
H(36)	0.78	0.30	0.14
	0.86	0.23	0.15
H(01)	0.73	0.62	0.46
	0.68	0.54	0.47
H(03)	0.76	0.56	0.46
	0.62	0.57	0.44
H(04)	0.82	0.45	0.45
	0.64	0.61	0.49
H(06)	0.81	0.49	0.45
	0.69	0.54	0.47

Table S34: Atomic charges (e) from multipole refinement for barbital. Standard uncertainties have been omitted for clarity.

ATOM	**PV** (***Exp***)	Ω (***Exp***)	Ω (**DFT**)
O(1) (**2**)	5.99	-0.84	-1.14
(**3**)	6.19	-0.91	-1.24
O(2) (**2**)	6.04	-0.84	-1.12
(**3**)	6.02	-0.83	-1.21
(**3**)(**SYMM**)	6.07	-0.79	-1.22
N(1)	5.10	-1.11	-1.20
	5.18	-1.11	-1.33
	5.14	-1.11	-1.32
C(1)	3.89	1.29	1.83
	3.94	1.44	1.95
C(2)	3.99	1.11	1.38
	4.04	1.11	1.49
	4.00	0.05	0.03
C(3)	3.93	0.12	0.01
	4.24	0.81	1.48
C(4)	4.10	-0.17	0.13
	3.85	0.07	0.00
	3.88	0.10	0.01
C(5)	4.02	0.06	0.15
	3.85	0.07	0.00

	3.80	0.09	0.00
H(4A)	0.98	0.08	-0.01
	0.96	0.09	0.05
	0.91	0.14	0.04
H(4B)	0.94	0.11	-0.01
	0.94	0.13	0.05
	1.13	-0.12	0.04
H(5A)	1.12	-0.18	-0.03
	1.10	-0.11	0.01
	1.14	-0.10	0.00
H(5B)	1.08	-0.08	-0.07
	0.90	0.15	0.03
	1.07	-0.04	0.04
H(5C)	1.07	-0.12	-0.03
	1.08	-0.08	0.01
	1.16	-0.14	0.00
H(1)	0.66	0.64	0.46
	0.75	0.51	0.52
	0.69	0.59	0.53

References

1. Hoser, A. A.; Dominiak, P. M.; Wozniak, K., Towards the best model for H atoms in experimental charge-density refinement. *Acta Crystallogr., Sect. A: Found. Crystallogr.* **2009,** *65*, 300-311.
2. Madsen, A. O.; Sorensen, H. O.; Flensburg, C.; Stewart, R. F.; Larsen, S., Modeling of the nuclear parameters for H atoms in X-ray charge-density studies. *Acta Crystallogr., Sect. A: Found. Crystallogr.* **2004,** *A60*, 550-561.
3. Madsen, A. O., SHADE web server for estimation of hydrogen anisotropic displacement parameters. *J. Appl. Crystallogr.* **2006,** *39*, 757-758.
4. Nguyen, T. H.; Howard, S. T.; Hanrahan, J. R.; Groundwater, P. W.; Platts, J. A.; Hibbs, D. E., Experimental and Theoretical Charge Density Distribution in a Host-Guest System: Synthetic Terephthaloyl Receptor Complexed to Adipic Acid. *J. Phys. Chem. A* **2012,** *116*, 5618-5628.
5. Meindl, K.; Henn, J., Foundations of residual-density analysis. *Acta Crystallogr., Sect. A: Found. Crystallogr.* **2008,** *64*, 404-418.
6. Meindl, K.; Henn, J., Residual Density Analysis. In *Electron Density and Chemical Bonding II: Theoretical Charge Density Studies*, Stalke, D., Ed. Springer Berlin Heidelberg: Berlin, Heidelberg, 2012; pp 143-192.