Exploring why monoclinic paracetamol has poor tableting properties†

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Data collection, integration and reduction

Single-crystal X-ray diffraction experiments were carried out in the Faculty of Pharmacy at the University of Sydney using an Agilent SuperNova™ X-ray diffractometer with an X-ray wavelength of 0.7107 Å (Mo*K*α) at 150K. Crystals of (**1**), (**2**) and (**3**) with dimensions (0.1 x 0.25 x 0.15) mm, (0.1 x 0.2 x 0.5) mm and (0.3 x 0.25 x 0.3) mm, respectively, were mounted on thin glass fibres with Paratone-N oil used as both adhesive and cryoprotectant. Data were collected for all crystals using 1° ω-scans maintaining the crystal-to-detector distance at 5.3 cm for (**1**), (**2**) and (**3**). For (**1**), reciprocal space coverage was achieved during the data collection by positioning the detector arm at two different angles in 2θ, at 41.6° and 90.5° with exposure times of 20 and 60 seconds. For (**2**), reciprocal space coverage was achieved by positioning the detector arm at two angles in 2θ, at 0° and 49° with exposure times of 50 and 150 seconds. For (**3**), reciprocal space coverage was covered in 2θ at 28° and 66° with exposure times of 25 and 50 seconds. A total of 6680, 1276 and 4618 frames were collected for (**1**), (**2**) and (**3**) respectively.

Integration and reduction of the collected data were performed with the CrysAlis^{Pro} software package[1]. All crystals were cooled to 150 K with an Oxford Cryosystems COBRA cooler. The unit cell parameters for (**1**) were refined from 100164 reflections in the monoclinic space group $P2_1/n$ with $Z = 4$, $F(000) = 320$ and $\mu = 0.096$ mm^{-1}. The unit cell parameters for (**2**) were refined from 19186 reflections in the triclinic space group $P\bar{1}$ with $Z = 2$, $F(000) = 328$ and $\mu = 0.078$ mm^{-1}. The unit cell parameters for co-crystal (**3**) were refined from 170540 reflections in the monoclinic space group $P2_1/c$ with $Z=4$, $F(000) = 1316$ and $\mu = 0.102$ mm^{-1}. Refer to Table 1 for selected crystallographic information from the independent atom model (IAM) and multipole (*EXP*) refinements.

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

Atom	U11	U22	U33	U12	U13	U23
H02	0.030239	0.051887	0.047266	-0.019786	-0.004603	0.018488
H03	0.025852	0.059896	0.040552	-0.013989	-0.011945	0.017423
H05	0.027912	0.044326	0.038833	-0.016919	-0.000219	0.006340
H06	0.025217	0.061884	0.037042	-0.011820	-0.010200	0.011908
H08A	0.064924	0.033707	0.050582	0.001713	0.007159	0.009195
H08B	0.029242	0.068955	0.050665	-0.003159	0.000839	0.022690
H08C	0.051442	0.063808	0.033235	0.009050	0.017402	0.008143
H01A	0.020788	0.051058	0.037994	-0.000332	0.000381	0.014486
H01	0.025077	0.044602	0.039273	-0.004550	0.001690	0.014276

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

Atom	U11	U22	U33	U12	U13	U23
H1	0.061420	0.042469	0.036631	0.018625	-0.010644	-0.011173
H2	0.076599	0.054484	0.034468	0.005943	-0.023107	-0.010937
H3	0.061508	0.053499	0.050724	0.030360	-0.010496	-0.004375
H4	0.065371	0.049773	0.033141	0.019011	-0.008841	-0.012604
H7	0.055019	0.058459	0.032963	0.023242	-0.000593	-0.007081
H8	0.075704	0.075651	0.024158	0.017663	-0.007689	-0.003800
H9	0.056043	0.075282	0.046676	0.036926	-0.006717	-0.000921
H10	0.052655	0.063622	0.027766	0.021221	-0.000756	-0.005328
H1'	0.061573	0.034158	0.034716	0.005255	0.001880	0.005600
H2'	0.062824	0.035360	0.053843	0.014145	0.002112	-0.007541
H3'	0.070664	0.060329	0.028152	-0.001969	0.007398	0.003044
H4'	0.056567	0.036420	0.035059	0.007052	-0.003366	0.005018
H7'	0.042521	0.056316	0.033969	0.008437	-0.014012	-0.006875
H8'	0.036420	0.062947	0.055749	0.017283	-0.008129	-0.015667
H9'	0.062359	0.065878	0.026433	0.007981	-0.007105	-0.008657
H10'	0.043301	0.048281	0.033934	0.013283	-0.009898	-0.002065

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

Atom	U11	U22	U33	U12	U13	U23
H02	0.046872	0.044426	0.039226	0.007098	-0.018231	-0.007667
H03	0.047518	0.033075	0.045141	0.002674	-0.012889	-0.014763
H05	0.046772	0.046080	0.038779	0.006677	-0.017665	-0.012264
H06	0.063235	0.036800	0.050913	0.009094	-0.016042	-0.019830
H08A	0.046458	0.061184	0.064981	0.014085	0.012852	-0.002816
H08B	0.050506	0.040069	0.072446	0.003859	-0.012522	-0.009218
H08C	0.068371	0.061278	0.044524	0.023762	-0.014651	-0.000153
H01	0.039364	0.029407	0.046963	-0.000019	-0.008289	-0.008551
H01A	0.053575	0.038758	0.036390	0.009522	-0.009163	-0.001235
H02'	0.036700	0.051793	0.047999	0.011082	-0.019226	-0.007413
H03'	0.042518	0.037642	0.050596	0.019073	-0.011464	-0.003304
H05'	0.034525	0.041152	0.047225	0.010293	-0.017518	-0.005740
H06'	0.044158	0.035478	0.064293	0.018041	-0.015605	-0.011529
H08D	0.083212	0.054886	0.044166	0.005530	-0.006215	-0.014690
H08E	0.063384	0.039967	0.069835	0.015015	-0.017433	-0.008879
H08F	0.055171	0.049898	0.084740	-0.012842	0.005247	-0.016781
H01'	0.046493	0.029205	0.044152	0.011382	-0.008636	0.001306
H01B	0.037219	0.046579	0.050147	0.004790	-0.013516	-0.011761
H1	0.042069	0.062216	0.041749	-0.017021	0.015164	-0.012486
H2	0.046662	0.073450	0.056529	-0.030075	0.008559	-0.018836
H3	0.056472	0.059985	0.041771	-0.007449	0.016204	-0.018121
H4	0.036409	0.053536	0.047798	-0.011509	0.013220	-0.012374
H7	0.033033	0.049756	0.054292	0.008129	0.003343	-0.016669
H8	0.029653	0.061274	0.058397	-0.002611	-0.004756	-0.017856
H9	0.042928	0.050511	0.048670	0.009493	0.005658	-0.018007
H10	0.027351	0.056000	0.048635	0.002740	0.000423	-0.012826
H1'	0.043858	0.054539	0.051809	0.015508	0.017056	0.012259
H2'	0.046473	0.053827	0.067152	0.020985	-0.000278	0.011962
H3'	0.073740	0.067158	0.048776	0.002865	0.020511	0.021150
H4'	0.047575	0.065558	0.061767	0.016611	0.024697	0.018432
H7'	0.032802	0.072699	0.077326	0.008005	0.015557	0.030026
H8'	0.041487	0.081570	0.096456	0.025818	0.003208	0.034131

H9'	0.068434	0.058520	0.058894	0.009422	0.025531	0.025337
H10'	0.033985	0.055300	0.062752	0.007991	0.014990	0.017631

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(01)	6344(1)	1702(1)	6299(1)	17(1)
C(02)	4632(1)	1721(1)	5551(1)	21(1)
C(03)	4410(1)	2648(1)	4602(1)	20(1)
C(04)	5886(1)	3566(1)	4385(1)	15(1)
C(05)	7584(1)	3568(1)	5147(1)	18(1)
C(06)	7807(1)	2636(1)	6098(1)	19(1)
C(07)	6790(1)	5121(1)	2807(1)	18(1)
C(08)	5929(1)	5979(1)	1763(1)	26(1)
N(01)	5524(1)	4472(1)	3400(1)	17(1)
O(01)	6646(1)	788(1)	7229(1)	24(1)
O(02)	8535(1)	5038(1)	3088(1)	26(1)

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

C(01)-O(01)	1.3667(3)
C(01)-C(06)	1.3936(3)
C(01)-C(02)	1.3950(2)
C(02)-C(03)	1.3900(3)

C(03)-C(04)	1.3964(2)
C(04)-C(05)	1.3961(2)
C(04)-N(01)	1.4150(2)
C(05)-C(06)	1.3937(3)
C(07)-O(02)	1.2395(3)
C(07)-N(01)	1.3446(2)
C(07)-C(08)	1.5085(3)

O(01)-C(01)-C(06)	118.368(16)
O(01)-C(01)-C(02)	122.109(17)
C(06)-C(01)-C(02)	119.523(17)
C(03)-C(02)-C(01)	119.940(17)
C(02)-C(03)-C(04)	120.680(16)
C(05)-C(04)-C(03)	119.344(17)
C(05)-C(04)-N(01)	124.067(15)
C(03)-C(04)-N(01)	116.571(15)
C(06)-C(05)-C(04)	119.914(16)
C(01)-C(06)-C(05)	120.574(16)
O(02)-C(07)-N(01)	123.30(2)
O(02)-C(07)-C(08)	121.74(2)
N(01)-C(07)-C(08)	114.957(18)
C(07)-N(01)-C(04)	128.261(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(01)	14(1)	20(1)	18(1)	1(1)	1(1)	-1(1)
C(02)	15(1)	24(1)	22(1)	5(1)	-2(1)	-5(1)
C(03)	14(1)	24(1)	20(1)	4(1)	-2(1)	-5(1)
C(04)	13(1)	17(1)	16(1)	-1(1)	2(1)	-1(1)
C(05)	13(1)	20(1)	19(1)	0(1)	0(1)	-3(1)
C(06)	13(1)	23(1)	19(1)	1(1)	-1(1)	-2(1)
C(07)	17(1)	19(1)	18(1)	-1(1)	5(1)	-1(1)
C(08)	28(1)	27(1)	25(1)	7(1)	7(1)	2(1)
N(01)	13(1)	20(1)	18(1)	2(1)	2(1)	-1(1)
O(01)	16(1)	30(1)	25(1)	10(1)	0(1)	-1(1)
O(02)	16(1)	39(1)	24(1)	1(1)	6(1)	-4(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(02)	3453(7)	1039(7)	5708(7)	32(2)
H(03)	3090(7)	2645(10)	4014(6)	46(2)
H(05)	8722(7)	4295(7)	5000(7)	33(2)
H(06)	9148(6)	2681(9)	6664(6)	33(2)
H(08A)	6041(14)	7109(2)	1903(10)	70(3)
H(08B)	4452(4)	5827(13)	1513(9)	68(3)
H(08C)	6660(13)	5798(13)	1041(6)	70(3)
H(01A)	4160(4)	4540(10)	3024(7)	42(2)
H(01)	5416(5)	499(9)	7419(7)	42(2)

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

O(01)-C(01)-C(02)-C(03)	-178.55(2)
C(06)-C(01)-C(02)-C(03)	1.24(3)
C(01)-C(02)-C(03)-C(04)	0.00(4)
C(02)-C(03)-C(04)-C(05)	-1.38(3)
C(02)-C(03)-C(04)-N(01)	-179.89(2)
C(03)-C(04)-C(05)-C(06)	1.50(3)
N(01)-C(04)-C(05)-C(06)	179.891(18)

O(01)-C(01)-C(06)-C(05)	178.68(2)
C(02)-C(01)-C(06)-C(05)	-1.12(3)
C(04)-C(05)-C(06)-C(01)	-0.26(3)
O(02)-C(07)-N(01)-C(04)	-2.45(3)
C(08)-C(07)-N(01)-C(04)	177.92(2)
C(05)-C(04)-N(01)-C(07)	22.69(3)
C(03)-C(04)-N(01)-C(07)	-158.87(2)

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

D-H	d(D-H)	d(H..A)	<DHA	d(D..A)	A
C03-H03	1.083	2.624	135.28	3.478	O01 [x-1/2, -y+1/2, z-1/2]
C05-H05	1.082	2.315	112.92	2.912	O02
N01-H01A	1.009	1.919	166.91	2.910	O01 [x-1/2, -y+1/2, z-1/2]
O01-H01	0.966	1.706	166.05	2.654	O02 [x-1/2, -y+1/2, z+1/2]

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)
N(1)	200(1)	11439(1)	1502(1)	31(1)
N(2)	5042(1)	7370(1)	6114(1)	34(1)
C(1)	2309(1)	9388(1)	2136(1)	24(1)
C(2)	1283(1)	10164(1)	1283(1)	28(1)
C(3)	160(1)	11984(1)	2609(1)	30(1)
C(4)	1126(1)	11291(1)	3528(1)	26(1)
C(5)	2222(1)	9934(1)	3303(1)	20(1)
C(6)	3221(1)	9083(1)	4268(1)	20(1)
C(7)	2859(1)	9338(1)	5501(1)	30(1)
C(8)	3787(1)	8460(1)	6375(1)	35(1)
C(9)	5396(1)	7147(1)	4928(1)	37(1)
C(10)	4539(1)	7954(1)	3987(1)	31(1)
N(1')	5595(1)	6786(1)	11103(1)	32(1)
N(2')	10471(1)	2879(1)	6384(1)	32(1)
C(1')	6658(1)	6720(1)	9012(1)	25(1)
C(2')	5738(1)	7448(1)	9964(1)	30(1)
C(3')	6403(1)	5339(1)	11310(1)	31(1)
C(4')	7383(1)	4519(1)	10428(1)	26(1)
C(5')	7520(1)	5216(1)	9245(1)	21(1)
C(6')	8539(1)	4399(1)	8264(1)	21(1)

C(7')	9949(1)	3399(1)	8509(1)	25(1)
C(8')	10864(1)	2685(1)	7546(1)	31(1)
C(9')	9102(1)	3827(1)	6161(1)	30(1)
C(10')	8112(1)	4611(1)	7054(1)	25(1)

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

N(1)-C(2)	1.3384(7)
N(1)-C(3)	1.3389(7)
N(2)-C(8)	1.3325(7)
N(2)-C(9)	1.3367(7)
C(1)-C(2)	1.3879(6)
C(1)-C(5)	1.3974(6)
C(1)-H(1)	1.08299(11)
C(2)-H(2)	1.08299(10)
C(3)-C(4)	1.3884(6)
C(3)-H(3)	1.08300(10)
C(4)-C(5)	1.3967(5)
C(4)-H(4)	1.08300(10)
C(5)-C(6)	1.4820(5)
C(6)-C(7)	1.3924(6)
C(6)-C(10)	1.3939(6)
C(7)-C(8)	1.3861(6)
C(7)-H(7)	1.08299(11)
C(8)-H(8)	1.08300(10)

C(9)-C(10)	1.3851(6)
C(9)-H(9)	1.08300(11)
C(10)-H(10)	1.08299(11)
N(1')-C(3')	1.3347(7)
N(1')-C(2')	1.3391(7)
N(2')-C(8')	1.3365(7)
N(2')-C(9')	1.3401(7)
C(1')-C(2')	1.3850(6)
C(1')-C(5')	1.3960(5)
C(1')-H(1')	1.08299(11)
C(2')-H(2')	1.08299(11)
C(3')-C(4')	1.3884(6)
C(3')-H(3')	1.08299(10)
C(4')-C(5')	1.3941(6)
C(4')-H(4')	1.08300(10)
C(5')-C(6')	1.4796(5)
C(6')-C(7')	1.3931(5)
C(6')-C(10')	1.3961(6)
C(7')-C(8')	1.3884(6)
C(7')-H(7')	1.08299(11)
C(8')-H(8')	1.08299(10)
C(9')-C(10')	1.3864(6)
C(9')-H(9')	1.08299(10)
C(10')-H(10')	1.08299(11)

C(2)-N(1)-C(3)	116.31(4)
C(8)-N(2)-C(9)	116.15(4)
C(2)-C(1)-C(5)	119.39(4)
C(2)-C(1)-H(1)	119.7(4)
C(5)-C(1)-H(1)	120.9(4)
N(1)-C(2)-C(1)	123.90(4)
N(1)-C(2)-H(2)	117.0(4)
C(1)-C(2)-H(2)	119.1(4)
N(1)-C(3)-C(4)	124.27(4)
N(1)-C(3)-H(3)	116.8(5)
C(4)-C(3)-H(3)	118.9(5)
C(3)-C(4)-C(5)	119.01(4)
C(3)-C(4)-H(4)	120.6(4)
C(5)-C(4)-H(4)	120.4(4)
C(4)-C(5)-C(1)	117.06(3)
C(4)-C(5)-C(6)	121.69(4)
C(1)-C(5)-C(6)	121.23(3)
C(7)-C(6)-C(10)	116.82(4)
C(7)-C(6)-C(5)	121.60(3)
C(10)-C(6)-C(5)	121.56(4)
C(8)-C(7)-C(6)	119.45(4)
C(8)-C(7)-H(7)	119.7(4)
C(6)-C(7)-H(7)	120.8(4)
N(2)-C(8)-C(7)	124.11(4)
N(2)-C(8)-H(8)	114.7(5)

C(7)-C(8)-H(8)	121.2(5)
N(2)-C(9)-C(10)	124.20(4)
N(2)-C(9)-H(9)	116.6(5)
C(10)-C(9)-H(9)	119.2(5)
C(9)-C(10)-C(6)	119.27(4)
C(9)-C(10)-H(10)	121.6(5)
C(6)-C(10)-H(10)	119.1(5)
C(3')-N(1')-C(2')	116.76(4)
C(8')-N(2')-C(9')	116.59(4)
C(2')-C(1')-C(5')	118.76(4)
C(2')-C(1')-H(1')	117.4(4)
C(5')-C(1')-H(1')	123.8(4)
N(1')-C(2')-C(1')	124.02(4)
N(1')-C(2')-H(2')	116.0(5)
C(1')-C(2')-H(2')	119.9(5)
N(1')-C(3')-C(4')	123.79(4)
N(1')-C(3')-H(3')	118.6(4)
C(4')-C(3')-H(3')	117.5(4)
C(3')-C(4')-C(5')	118.97(4)
C(3')-C(4')-H(4')	120.4(5)
C(5')-C(4')-H(4')	120.6(5)
C(4')-C(5')-C(1')	117.68(4)
C(4')-C(5')-C(6')	121.51(3)
C(1')-C(5')-C(6')	120.81(3)
C(7')-C(6')-C(10')	117.75(4)

C(7')-C(6')-C(5')	121.56(3)
C(10')-C(6')-C(5')	120.69(3)
C(8')-C(7')-C(6')	118.81(4)
C(8')-C(7')-H(7')	119.8(4)
C(6')-C(7')-H(7')	121.4(4)
N(2')-C(8')-C(7')	124.05(4)
N(2')-C(8')-H(8')	115.9(5)
C(7')-C(8')-H(8')	120.0(5)
N(2')-C(9')-C(10')	123.92(4)
N(2')-C(9')-H(9')	117.7(4)
C(10')-C(9')-H(9')	118.3(4)
C(9')-C(10')-C(6')	118.87(4)
C(9')-C(10')-H(10')	120.5(4)
C(6')-C(10')-H(10')	120.7(4)

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
N(1)	29(1)	33(1)	27(1)	3(1)	-10(1)	3(1)
N(2)	34(1)	38(1)	24(1)	4(1)	-8(1)	7(1)
C(1)	27(1)	22(1)	20(1)	-2(1)	-6(1)	1(1)
C(2)	33(1)	29(1)	23(1)	0(1)	-10(1)	0(1)

C(3)	27(1)	31(1)	28(1)	2(1)	-5(1)	8(1)
C(4)	26(1)	27(1)	22(1)	-1(1)	-3(1)	6(1)
C(5)	20(1)	21(1)	18(1)	0(1)	-3(1)	0(1)
C(6)	20(1)	21(1)	18(1)	0(1)	-3(1)	0(1)
C(7)	33(1)	33(1)	18(1)	-2(1)	-3(1)	8(1)
C(8)	40(1)	41(1)	19(1)	1(1)	-6(1)	9(1)
C(9)	32(1)	43(1)	26(1)	2(1)	-6(1)	14(1)
C(10)	28(1)	37(1)	21(1)	-1(1)	-3(1)	12(1)
N(1')	32(1)	36(1)	26(1)	-9(1)	3(1)	0(1)
N(2')	30(1)	37(1)	27(1)	-9(1)	2(1)	3(1)
C(1')	29(1)	21(1)	23(1)	-2(1)	0(1)	2(1)
C(2')	32(1)	26(1)	30(1)	-7(1)	1(1)	3(1)
C(3')	34(1)	35(1)	21(1)	-3(1)	2(1)	-1(1)
C(4')	30(1)	26(1)	20(1)	-1(1)	-2(1)	-1(1)
C(5')	22(1)	20(1)	19(1)	-3(1)	-3(1)	-1(1)
C(6')	22(1)	19(1)	19(1)	-2(1)	-3(1)	0(1)
C(7')	23(1)	27(1)	24(1)	-5(1)	-5(1)	2(1)
C(8')	24(1)	34(1)	31(1)	-8(1)	-2(1)	5(1)
C(9')	34(1)	32(1)	20(1)	-4(1)	0(1)	2(1)
C(10')	28(1)	25(1)	19(1)	-2(1)	-3(1)	3(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)	3148(5)	8355(4)	1894(7)	42(2)
H(2)	1318(11)	9703(6)	394(2)	44(2)
H(3)	-659(6)	13060(4)	2772(8)	49(3)
H(4)	1024(12)	11785(6)	4409(2)	46(2)
H(7)	1868(4)	10217(5)	5785(7)	46(3)
H(8)	3542(13)	8636(11)	7340(2)	56(3)
H(9)	6410(4)	6258(5)	4709(8)	63(3)
H(10)	4859(12)	7718(10)	3038(2)	59(3)
H(1')	6698(11)	7379(6)	8133(2)	41(2)
H(2')	5095(7)	8631(3)	9811(9)	50(3)
H(3')	6260(12)	4744(6)	12207(3)	54(3)
H(4')	8016(7)	3348(3)	10655(8)	48(3)
H(7')	10346(7)	3184(12)	9428(2)	44(2)
H(8')	11976(4)	1922(6)	7713(8)	59(3)
H(9')	8722(7)	3933(13)	5238(2)	51(3)
H(10')	7019(4)	5362(6)	6817(7)	46(3)

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

C(3)-N(1)-C(2)-C(1)	1.46(8)
C(5)-C(1)-C(2)-N(1)	0.67(8)
C(2)-N(1)-C(3)-C(4)	-1.80(8)
N(1)-C(3)-C(4)-C(5)	-0.01(8)
C(3)-C(4)-C(5)-C(1)	2.14(7)
C(3)-C(4)-C(5)-C(6)	-176.19(4)
C(2)-C(1)-C(5)-C(4)	-2.45(7)
C(2)-C(1)-C(5)-C(6)	175.89(4)
C(4)-C(5)-C(6)-C(7)	16.62(7)
C(1)-C(5)-C(6)-C(7)	-161.64(5)
C(4)-C(5)-C(6)-C(10)	-165.39(5)
C(1)-C(5)-C(6)-C(10)	16.35(7)
C(10)-C(6)-C(7)-C(8)	-1.15(8)
C(5)-C(6)-C(7)-C(8)	176.93(5)
C(9)-N(2)-C(8)-C(7)	0.25(11)
C(6)-C(7)-C(8)-N(2)	0.71(10)
C(8)-N(2)-C(9)-C(10)	-0.75(11)
N(2)-C(9)-C(10)-C(6)	0.28(11)
C(7)-C(6)-C(10)-C(9)	0.69(8)
C(5)-C(6)-C(10)-C(9)	-177.39(5)
C(3')-N(1')-C(2')-C(1')	0.12(9)
C(5')-C(1')-C(2')-N(1')	-1.22(8)
C(2')-N(1')-C(3')-C(4')	1.24(9)

N(1')-C(3')-C(4')-C(5')	-1.43(8)
C(3')-C(4')-C(5')-C(1')	0.24(7)
C(3')-C(4')-C(5')-C(6')	-179.70(4)
C(2')-C(1')-C(5')-C(4')	0.98(7)
C(2')-C(1')-C(5')-C(6')	-179.08(4)
C(4')-C(5')-C(6')-C(7')	-34.64(6)
C(1')-C(5')-C(6')-C(7')	145.41(5)
C(4')-C(5')-C(6')-C(10')	145.40(5)
C(1')-C(5')-C(6')-C(10')	-34.54(6)
C(10')-C(6')-C(7')-C(8')	0.90(7)
C(5')-C(6')-C(7')-C(8')	-179.05(4)
C(9')-N(2')-C(8')-C(7')	-0.06(9)
C(6')-C(7')-C(8')-N(2')	-0.86(8)
C(8')-N(2')-C(9')-C(10')	0.94(9)
N(2')-C(9')-C(10')-C(6')	-0.87(8)
C(7')-C(6')-C(10')-C(9')	-0.10(7)
C(5')-C(6')-C(10')-C(9')	179.85(4)

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

D-H	d(D-H)	d(H..A)	<DHA	d(D..A)	A
C1-H1	1.083	2.438	158.59	3.469	N1' [x, y, z-1]
C4-H4	1.083	2.420	173.31	3.497	N2' [x-1, y+1, z]
C7-H7	1.083	2.522	156.33	3.540	N2' [x-1, y+1, z]
C10-H10	1.083	2.338	170.79	3.411	N1' [x, y, z-1]
C4'-H4'	1.083	2.462	154.04	3.468	N1 [x+1, y-1, z+1]
C7'-H7'	1.083	2.640	144.61	3.578	N1 [x+1, y-1, z+1]
C10'-H10'	1.083	2.336	166.79	3.399	N2

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)
C(1)	2887(1)	2851(1)	-64(1)	27(1)
C(2)	2464(1)	2510(1)	-981(1)	31(1)
C(3)	4174(1)	2598(1)	-1840(1)	29(1)
C(4)	4676(1)	2946(1)	-960(1)	24(1)
C(5)	4022(1)	3080(1)	-44(1)	20(1)
C(6)	4493(1)	3454(1)	910(1)	19(1)
C(7)	5704(1)	3480(1)	1305(1)	24(1)
C(8)	6086(1)	3851(1)	2183(1)	26(1)
C(9)	4197(1)	4155(1)	2322(1)	24(1)

C(10)	3729(1)	3796(1)	1455(1)	22(1)
N(1)	3081(1)	2379(1)	-1867(1)	31(1)
N(2)	5359(1)	4193(1)	2685(1)	24(1)
C(01)	3938(1)	6582(1)	3423(1)	22(1)
C(02)	3810(1)	6103(1)	2744(1)	22(1)
C(03)	4540(1)	5649(1)	3027(1)	21(1)
C(04)	5404(1)	5660(1)	3986(1)	19(1)
C(05)	5515(1)	6138(1)	4675(1)	23(1)
C(06)	4785(1)	6592(1)	4389(1)	24(1)
C(07)	7144(1)	5128(1)	4895(1)	23(1)
C(08)	7740(1)	4571(1)	4876(1)	32(1)
N(01)	6120(1)	5180(1)	4193(1)	21(1)
O(01)	3266(1)	7046(1)	3176(1)	31(1)
O(02)	7587(1)	5511(1)	5511(1)	34(1)
C(1')	7024(1)	9699(1)	6123(1)	24(1)
C(2')	6642(1)	10083(1)	5269(1)	28(1)
C(3')	8231(1)	9916(1)	4272(1)	31(1)
C(4')	8689(1)	9525(1)	5080(1)	26(1)
C(5')	8084(1)	9411(1)	6040(1)	21(1)
C(6')	8556(1)	9008(1)	6942(1)	22(1)
C(7')	9752(1)	8856(1)	7077(1)	37(1)
C(8')	10143(1)	8476(1)	7932(1)	42(1)
C(9')	8297(1)	8393(1)	8525(1)	35(1)
C(10')	7820(1)	8765(1)	7687(1)	28(1)
N(1')	7221(1)	10198(1)	4354(1)	32(1)

N(2')	9444(1)	8243(1)	8658(1)	35(1)
C(01')	9902(1)	5874(1)	7470(1)	22(1)
C(02')	9181(1)	6345(1)	7398(1)	22(1)
C(03')	9469(1)	6792(1)	8134(1)	21(1)
C(04')	10468(1)	6779(1)	8960(1)	19(1)
C(05')	11182(1)	6304(1)	9040(1)	21(1)
C(06')	10897(1)	5861(1)	8292(1)	23(1)
C(07')	11532(1)	7334(1)	10556(1)	23(1)
C(08')	11673(1)	7919(1)	11008(1)	34(1)
N(01')	10713(1)	7263(1)	9636(1)	22(1)
O(01')	9681(1)	5421(1)	6784(1)	31(1)
O(02')	12160(1)	6952(1)	11005(1)	33(1)

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

C(1)-C(2)	1.3864(8)
C(1)-C(5)	1.3946(7)
C(1)-H(1)	1.0821(10)
C(2)-N(1)	1.3375(9)
C(2)-H(2)	1.0826(10)
C(3)-N(1)	1.3413(8)
C(3)-C(4)	1.3920(7)
C(3)-H(3)	1.0823(10)
C(4)-C(5)	1.3944(7)

C(4)-H(4)	1.0822(10)
C(5)-C(6)	1.4781(6)
C(6)-C(10)	1.3933(7)
C(6)-C(7)	1.3969(7)
C(7)-C(8)	1.3879(7)
C(7)-H(7)	1.0820(10)
C(8)-N(2)	1.3409(8)
C(8)-H(8)	1.0829(10)
C(9)-N(2)	1.3387(7)
C(9)-C(10)	1.3859(7)
C(9)-H(9)	1.0823(10)
C(10)-H(10)	1.0821(10)
C(01)-O(01)	1.3652(6)
C(01)-C(06)	1.3912(7)
C(01)-C(02)	1.3945(7)
C(02)-C(03)	1.3887(7)
C(02)-H(02)	1.0825(10)
C(03)-C(04)	1.3968(6)
C(03)-H(03)	1.0821(10)
C(04)-C(05)	1.3989(6)
C(04)-N(01)	1.4185(6)
C(05)-C(06)	1.3892(7)
C(05)-H(05)	1.0823(10)
C(06)-H(06)	1.0826(10)
C(07)-O(02)	1.2392(6)

C(07)-N(01)	1.3461(6)
C(07)-C(08)	1.5045(8)
C(08)-H(08A)	1.0584(10)
C(08)-H(08B)	1.0587(10)
C(08)-H(08C)	1.0583(10)
N(01)-H(01)	1.0081(10)
O(01)-H(01A)	0.9671(10)
C(1')-C(2')	1.3890(7)
C(1')-C(5')	1.3983(7)
C(1')-H(1')	1.0826(10)
C(2')-N(1')	1.3335(10)
C(2')-H(2')	1.0823(10)
C(3')-N(1')	1.3412(9)
C(3')-C(4')	1.3884(8)
C(3')-H(3')	1.0823(10)
C(4')-C(5')	1.3936(8)
C(4')-H(4')	1.0824(10)
C(5')-C(6')	1.4814(7)
C(6')-C(10')	1.3900(8)
C(6')-C(7')	1.3942(8)
C(7')-C(8')	1.3853(9)
C(7')-H(7')	1.0830(10)
C(8')-N(2')	1.3366(11)
C(8')-H(8')	1.0828(10)
C(9')-N(2')	1.3384(9)

C(9')-C(10')	1.3871(8)
C(9')-H(9')	1.0826(10)
C(10')-H(10')	1.0829(10)
C(01')-O(01')	1.3585(6)
C(01')-C(06')	1.3927(6)
C(01')-C(02')	1.3957(7)
C(02')-C(03')	1.3893(7)
C(02')-H(02')	1.0827(10)
C(03')-C(04')	1.3985(6)
C(03')-H(03')	1.0826(10)
C(04')-C(05')	1.3976(6)
C(04')-N(01')	1.4169(6)
C(05')-C(06')	1.3916(7)
C(05')-H(05')	1.0822(10)
C(06')-H(06')	1.0819(10)
C(07')-O(02')	1.2409(6)
C(07')-N(01')	1.3439(6)
C(07')-C(08')	1.5071(8)
C(08')-H(08D)	1.0588(10)
C(08')-H(08E)	1.0587(10)
C(08')-H(08F)	1.0585(10)
N(01')-H(01')	1.0090(10)
O(01')-H(01B)	0.9671(10)

C(2)-C(1)-C(5)	119.22(5)

C(2)-C(1)-H(1)	120.6(6)
C(5)-C(1)-H(1)	119.9(6)
N(1)-C(2)-C(1)	124.16(5)
N(1)-C(2)-H(2)	117.1(7)
C(1)-C(2)-H(2)	118.7(7)
N(1)-C(3)-C(4)	123.83(6)
N(1)-C(3)-H(3)	114.1(6)
C(4)-C(3)-H(3)	121.8(6)
C(3)-C(4)-C(5)	119.17(5)
C(3)-C(4)-H(4)	119.9(6)
C(5)-C(4)-H(4)	120.9(6)
C(4)-C(5)-C(1)	117.26(4)
C(4)-C(5)-C(6)	122.05(4)
C(1)-C(5)-C(6)	120.68(5)
C(10)-C(6)-C(7)	117.30(4)
C(10)-C(6)-C(5)	120.51(4)
C(7)-C(6)-C(5)	122.19(4)
C(8)-C(7)-C(6)	119.04(5)
C(8)-C(7)-H(7)	120.7(6)
C(6)-C(7)-H(7)	120.3(6)
N(2)-C(8)-C(7)	123.82(5)
N(2)-C(8)-H(8)	115.4(5)
C(7)-C(8)-H(8)	120.6(6)
N(2)-C(9)-C(10)	123.67(5)
N(2)-C(9)-H(9)	117.6(6)

C(10)-C(9)-H(9)	118.7(6)
C(9)-C(10)-C(6)	119.42(4)
C(9)-C(10)-H(10)	118.3(6)
C(6)-C(10)-H(10)	122.1(6)
C(2)-N(1)-C(3)	116.36(5)
C(9)-N(2)-C(8)	116.70(4)
O(01)-C(01)-C(06)	118.23(4)
O(01)-C(01)-C(02)	122.65(4)
C(06)-C(01)-C(02)	119.12(4)
C(03)-C(02)-C(01)	119.80(4)
C(03)-C(02)-H(02)	122.0(5)
C(01)-C(02)-H(02)	118.2(5)
C(02)-C(03)-C(04)	121.32(4)
C(02)-C(03)-H(03)	120.2(5)
C(04)-C(03)-H(03)	118.4(5)
C(03)-C(04)-C(05)	118.61(4)
C(03)-C(04)-N(01)	117.16(4)
C(05)-C(04)-N(01)	124.23(4)
C(06)-C(05)-C(04)	119.97(4)
C(06)-C(05)-H(05)	121.9(5)
C(04)-C(05)-H(05)	118.1(5)
C(05)-C(06)-C(01)	121.16(4)
C(05)-C(06)-H(06)	120.5(6)
C(01)-C(06)-H(06)	118.3(6)
O(02)-C(07)-N(01)	123.07(5)

O(02)-C(07)-C(08)	121.34(5)
N(01)-C(07)-C(08)	115.59(5)
C(07)-C(08)-H(08A)	110.9(7)
C(07)-C(08)-H(08B)	112.7(8)
H(08A)-C(08)-H(08B)	106.4(11)
C(07)-C(08)-H(08C)	111.2(8)
H(08A)-C(08)-H(08C)	104.0(10)
H(08B)-C(08)-H(08C)	111.3(11)
C(07)-N(01)-C(04)	127.96(4)
C(07)-N(01)-H(01)	114.9(5)
C(04)-N(01)-H(01)	117.0(5)
C(01)-O(01)-H(01A)	102.1(8)
C(2')-C(1')-C(5')	119.09(5)
C(2')-C(1')-H(1')	117.6(6)
C(5')-C(1')-H(1')	123.3(6)
N(1')-C(2')-C(1')	124.19(5)
N(1')-C(2')-H(2')	116.9(6)
C(1')-C(2')-H(2')	118.7(6)
N(1')-C(3')-C(4')	123.79(6)
N(1')-C(3')-H(3')	116.9(6)
C(4')-C(3')-H(3')	119.3(6)
C(3')-C(4')-C(5')	119.41(6)
C(3')-C(4')-H(4')	117.9(6)
C(5')-C(4')-H(4')	122.7(6)
C(4')-C(5')-C(1')	117.06(5)

C(4')-C(5')-C(6')	121.36(5)
C(1')-C(5')-C(6')	121.57(5)
C(10')-C(6')-C(7')	117.24(5)
C(10')-C(6')-C(5')	121.24(5)
C(7')-C(6')-C(5')	121.51(5)
C(8')-C(7')-C(6')	119.03(6)
C(8')-C(7')-H(7')	118.1(7)
C(6')-C(7')-H(7')	122.8(7)
N(2')-C(8')-C(7')	124.12(6)
N(2')-C(8')-H(8')	114.2(6)
C(7')-C(8')-H(8')	121.6(6)
N(2')-C(9')-C(10')	123.62(6)
N(2')-C(9')-H(9')	120.1(7)
C(10')-C(9')-H(9')	116.2(7)
C(9')-C(10')-C(6')	119.49(6)
C(9')-C(10')-H(10')	120.0(6)
C(6')-C(10')-H(10')	120.5(6)
C(2')-N(1')-C(3')	116.45(5)
C(8')-N(2')-C(9')	116.49(5)
O(01')-C(01')-C(06')	117.68(4)
O(01')-C(01')-C(02')	123.44(4)
C(06')-C(01')-C(02')	118.88(4)
C(03')-C(02')-C(01')	119.85(4)
C(03')-C(02')-H(02')	122.0(5)
C(01')-C(02')-H(02')	118.2(5)

C(02')-C(03')-C(04')	121.32(4)
C(02')-C(03')-H(03')	122.1(5)
C(04')-C(03')-H(03')	116.6(5)
C(05')-C(04')-C(03')	118.79(4)
C(05')-C(04')-N(01')	124.15(4)
C(03')-C(04')-N(01')	117.01(4)
C(06')-C(05')-C(04')	119.67(4)
C(06')-C(05')-H(05')	123.2(5)
C(04')-C(05')-H(05')	117.1(6)
C(05')-C(06')-C(01')	121.49(4)
C(05')-C(06')-H(06')	123.2(5)
C(01')-C(06')-H(06')	115.3(5)
O(02')-C(07')-N(01')	123.48(5)
O(02')-C(07')-C(08')	120.96(5)
N(01')-C(07')-C(08')	115.54(5)
C(07')-C(08')-H(08D)	110.7(7)
C(07')-C(08')-H(08E)	113.1(7)
H(08D)-C(08')-H(08E)	110.0(10)
C(07')-C(08')-H(08F)	108.9(9)
H(08D)-C(08')-H(08F)	110.8(11)
H(08E)-C(08')-H(08F)	103.0(11)
C(07')-N(01')-C(04')	128.21(4)
C(07')-N(01')-H(01')	116.9(7)
C(04')-N(01')-H(01')	114.9(7)
C(01')-O(01')-H(01B)	111.0(7)

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
C(1)	27(1)	30(1)	25(1)	-5(1)	5(1)	-9(1)
C(2)	32(1)	33(1)	28(1)	-6(1)	3(1)	-13(1)
C(3)	35(1)	28(1)	24(1)	-7(1)	4(1)	-4(1)
C(4)	27(1)	23(1)	23(1)	-3(1)	5(1)	-3(1)
C(5)	24(1)	17(1)	19(1)	-1(1)	2(1)	-3(1)
C(6)	22(1)	16(1)	19(1)	0(1)	2(1)	-1(1)
C(7)	21(1)	22(1)	29(1)	-5(1)	2(1)	0(1)
C(8)	23(1)	25(1)	29(1)	-4(1)	-1(1)	-2(1)
C(9)	27(1)	22(1)	23(1)	-4(1)	2(1)	1(1)
C(10)	21(1)	22(1)	23(1)	-3(1)	1(1)	1(1)
N(1)	37(1)	31(1)	25(1)	-7(1)	1(1)	-8(1)
N(2)	28(1)	22(1)	23(1)	-3(1)	0(1)	-2(1)
C(01)	24(1)	20(1)	20(1)	-1(1)	-2(1)	0(1)
C(02)	23(1)	21(1)	20(1)	-2(1)	-4(1)	-1(1)
C(03)	21(1)	20(1)	19(1)	-3(1)	-3(1)	-2(1)
C(04)	19(1)	19(1)	17(1)	-1(1)	0(1)	-2(1)
C(05)	27(1)	23(1)	17(1)	-3(1)	-4(1)	-1(1)
C(06)	31(1)	21(1)	20(1)	-4(1)	-5(1)	0(1)
C(07)	21(1)	27(1)	20(1)	-2(1)	-2(1)	1(1)
C(08)	30(1)	31(1)	33(1)	-3(1)	-4(1)	8(1)

N(01)	21(1)	21(1)	21(1)	-2(1)	-3(1)	0(1)
O(01)	36(1)	23(1)	30(1)	-4(1)	-8(1)	7(1)
O(02)	28(1)	36(1)	36(1)	-13(1)	-13(1)	4(1)
C(1')	21(1)	23(1)	27(1)	2(1)	-1(1)	1(1)
C(2')	26(1)	23(1)	33(1)	1(1)	-9(1)	1(1)
C(3')	38(1)	30(1)	26(1)	7(1)	1(1)	-6(1)
C(4')	26(1)	27(1)	26(1)	5(1)	4(1)	-1(1)
C(5')	20(1)	19(1)	23(1)	2(1)	1(1)	-1(1)
C(6')	22(1)	19(1)	24(1)	2(1)	2(1)	2(1)
C(7')	23(1)	44(1)	44(1)	18(1)	5(1)	8(1)
C(8')	31(1)	44(1)	50(1)	17(1)	0(1)	13(1)
C(9')	42(1)	29(1)	34(1)	12(1)	9(1)	9(1)
C(10')	29(1)	26(1)	31(1)	8(1)	8(1)	6(1)
N(1')	38(1)	26(1)	29(1)	6(1)	-10(1)	-4(1)
N(2')	43(1)	26(1)	34(1)	8(1)	1(1)	10(1)
C(01')	18(1)	23(1)	23(1)	-1(1)	-2(1)	2(1)
C(02')	17(1)	25(1)	22(1)	1(1)	-4(1)	4(1)
C(03')	18(1)	23(1)	22(1)	2(1)	-2(1)	6(1)
C(04')	17(1)	19(1)	19(1)	2(1)	-1(1)	3(1)
C(05')	16(1)	20(1)	25(1)	0(1)	-4(1)	3(1)
C(06')	18(1)	21(1)	28(1)	-2(1)	-4(1)	4(1)
C(07')	23(1)	23(1)	23(1)	-1(1)	-2(1)	3(1)
C(08')	39(1)	26(1)	35(1)	-8(1)	-4(1)	2(1)
N(01')	22(1)	19(1)	22(1)	0(1)	-3(1)	4(1)
O(01')	27(1)	29(1)	35(1)	-10(1)	-9(1)	4(1)

O(02')	34(1)	29(1)	32(1)	-4(1)	-14(1)	9(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)	2383(9)	2908(5)	671(6)	56(3)
H(1')	6484(8)	9644(5)	6833(7)	54(3)
H(02')	8396(5)	6346(4)	6773(6)	35(2)
H(2')	5867(6)	10333(4)	5380(9)	42(3)
H(2)	1586(5)	2330(5)	-985(12)	62(4)
H(02)	3120(6)	6096(4)	2019(6)	35(2)
H(03)	4477(9)	5284(2)	2480(7)	33(2)
H(3)	4610(8)	2514(4)	-2609(5)	43(3)
H(03')	8945(7)	7168(2)	8091(9)	34(2)
H(3')	8683(9)	9992(5)	3509(6)	48(3)
H(4)	5570(4)	3102(4)	-985(9)	39(3)
H(4')	9510(5)	9319(4)	4930(10)	50(3)
H(05)	6199(6)	6145(4)	5405(6)	37(2)
H(05')	11928(6)	6300(4)	9710(6)	38(3)
H(06')	11430(7)	5488(2)	8291(9)	35(2)
H(06)	4888(10)	6969(3)	4896(8)	46(3)
H(7)	6333(7)	3219(4)	918(9)	45(3)

H(7')	10384(8)	9002(5)	6506(9)	68(4)
H(8)	7025(2)	3898(4)	2470(9)	37(2)
H(8')	11047(4)	8320(4)	8024(10)	49(3)
H(08A)	8483(7)	4587(6)	4394(9)	65(4)
H(08B)	7170(10)	4259(4)	4485(12)	73(4)
H(08C)	8109(12)	4452(6)	5720(5)	70(4)
H(08D)	11681(11)	7927(5)	11924(2)	62(4)
H(08E)	11014(8)	8192(4)	10620(10)	61(3)
H(08F)	12470(7)	8087(6)	10753(13)	84(5)
H(9)	3594(7)	4416(4)	2744(8)	44(3)
H(9')	7676(9)	8205(5)	9059(9)	65(4)
H(10')	6876(2)	8854(4)	7597(9)	43(3)
H(10)	2772(1)	3770(4)	1269(10)	43(3)
H(01')	10213(9)	7596(3)	9374(11)	56(3)
H(01)	5870(8)	4838(2)	3726(7)	32(2)
H(01A)	2921(11)	6975(6)	2388(4)	67(4)
H(01B)	8944(6)	5462(5)	6283(8)	54(3)

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

C(5)-C(1)-C(2)-N(1)	0.27(10)
N(1)-C(3)-C(4)-C(5)	0.00(9)
C(3)-C(4)-C(5)-C(1)	0.29(8)
C(3)-C(4)-C(5)-C(6)	-179.06(5)
C(2)-C(1)-C(5)-C(4)	-0.42(8)
C(2)-C(1)-C(5)-C(6)	178.94(5)
C(4)-C(5)-C(6)-C(10)	147.72(5)
C(1)-C(5)-C(6)-C(10)	-31.61(7)
C(4)-C(5)-C(6)-C(7)	-32.40(7)
C(1)-C(5)-C(6)-C(7)	148.27(6)
C(10)-C(6)-C(7)-C(8)	-1.81(8)
C(5)-C(6)-C(7)-C(8)	178.31(5)
C(6)-C(7)-C(8)-N(2)	-0.21(9)
N(2)-C(9)-C(10)-C(6)	-0.45(9)
C(7)-C(6)-C(10)-C(9)	2.12(8)
C(5)-C(6)-C(10)-C(9)	-178.00(5)
C(1)-C(2)-N(1)-C(3)	0.02(10)
C(4)-C(3)-N(1)-C(2)	-0.16(10)
C(10)-C(9)-N(2)-C(8)	-1.56(8)
C(7)-C(8)-N(2)-C(9)	1.89(9)
O(01)-C(01)-C(02)-C(03)	178.55(5)
C(06)-C(01)-C(02)-C(03)	-1.25(8)
C(01)-C(02)-C(03)-C(04)	0.24(8)

C(02)-C(03)-C(04)-C(05)	0.84(8)
C(02)-C(03)-C(04)-N(01)	-178.97(5)
C(03)-C(04)-C(05)-C(06)	-0.92(8)
N(01)-C(04)-C(05)-C(06)	178.88(5)
C(04)-C(05)-C(06)-C(01)	-0.08(9)
O(01)-C(01)-C(06)-C(05)	-178.63(6)
C(02)-C(01)-C(06)-C(05)	1.17(9)
O(02)-C(07)-N(01)-C(04)	2.88(10)
C(08)-C(07)-N(01)-C(04)	-176.37(5)
C(03)-C(04)-N(01)-C(07)	165.31(5)
C(05)-C(04)-N(01)-C(07)	-14.49(9)
C(5')-C(1')-C(2')-N(1')	0.26(8)
N(1')-C(3')-C(4')-C(5')	-0.14(9)
C(3')-C(4')-C(5')-C(1')	0.82(8)
C(3')-C(4')-C(5')-C(6')	-178.23(5)
C(2')-C(1')-C(5')-C(4')	-0.87(7)
C(2')-C(1')-C(5')-C(6')	178.17(5)
C(4')-C(5')-C(6')-C(10')	-159.87(6)
C(1')-C(5')-C(6')-C(10')	21.12(8)
C(4')-C(5')-C(6')-C(7')	19.88(8)
C(1')-C(5')-C(6')-C(7')	-159.12(6)
C(10')-C(6')-C(7')-C(8')	0.04(11)
C(5')-C(6')-C(7')-C(8')	-179.73(7)
C(6')-C(7')-C(8')-N(2')	-0.59(13)
N(2')-C(9')-C(10')-C(6')	-0.82(11)

C(7')-C(6')-C(10')-C(9')	0.61(9)
C(5')-C(6')-C(10')-C(9')	-179.62(6)
C(1')-C(2')-N(1')-C(3')	0.43(8)
C(4')-C(3')-N(1')-C(2')	-0.49(9)
C(7')-C(8')-N(2')-C(9')	0.43(13)
C(10')-C(9')-N(2')-C(8')	0.29(11)
O(01')-C(01')-C(02')-C(03')	179.90(5)
C(06')-C(01')-C(02')-C(03')	0.58(8)
C(01')-C(02')-C(03')-C(04')	-0.57(8)
C(02')-C(03')-C(04')-C(05')	-0.14(8)
C(02')-C(03')-C(04')-N(01')	177.44(5)
C(03')-C(04')-C(05')-C(06')	0.84(8)
N(01')-C(04')-C(05')-C(06')	-176.57(5)
C(04')-C(05')-C(06')-C(01')	-0.84(9)
O(01')-C(01')-C(06')-C(05')	-179.23(5)
C(02')-C(01')-C(06')-C(05')	0.12(9)
O(02')-C(07')-N(01')-C(04')	-5.31(10)
C(08')-C(07')-N(01')-C(04')	173.26(6)
C(05')-C(04')-N(01')-C(07')	-10.28(9)
C(03')-C(04')-N(01')-C(07')	172.27(5)

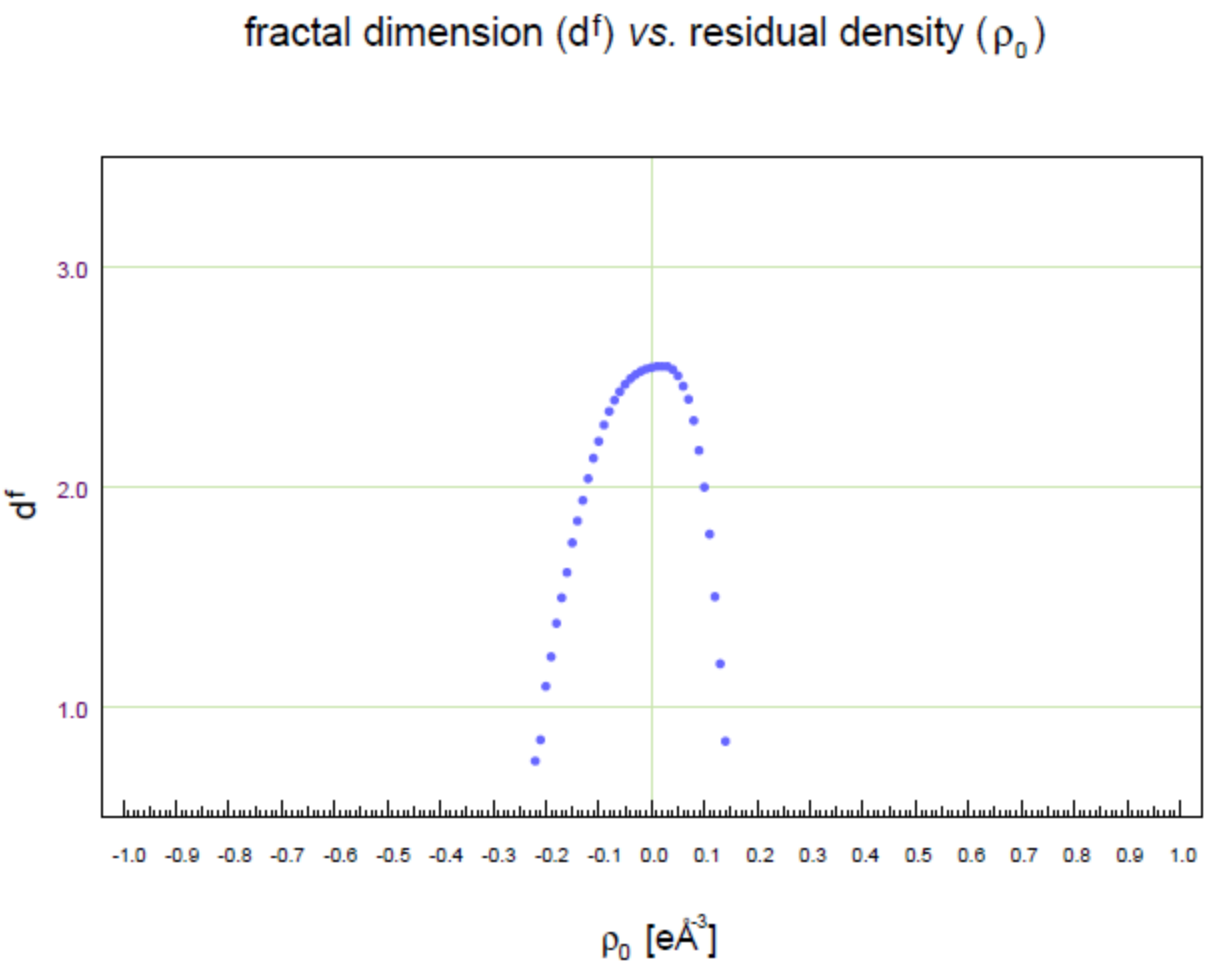
Table S21: Hydrogen bonds for (**3).** [Å and °].

D-H	d(D-H)	d(H..A)	<DHA	d(D..A)	A
C02'-H02'	1.083	2.594	126.07	3.348	O02
C02-H02	1.082	2.687	121.10	3.376	N1' [-x+1, y-1/2, -z+1/2]
C02-H02	1.082	2.558	125.20	3.303	O02' [x-1, y, z-1]
C03'-H03'	1.083	2.609	129.21	3.399	N1 [-x+1, y+1/2, -z+1/2]
C3'-H3'	1.082	2.595	147.65	3.557	O01' [x, -y+3/2, z-1/2]
C4-H4	1.082	2.573	156.60	3.592	O02' [-x+2, -y+1, -z+1]
C05-H05	1.082	2.185	118.75	2.867	O02
C05'-H05'	1.082	2.169	120.96	2.879	O02'
C8'-H8'	1.083	2.648	178.00	3.731	O01 [x+1, -y+3/2, z+1/2]
C08-H08A	1.058	2.603	177.44	3.661	O01' [-x+2, -y+1, -z+1]
C9-H9	1.082	2.544	143.92	3.477	O02 [-x+1, -y+1, -z+1]
N01'-H01'	1.009	1.930	169.70	2.928	N2'
N01-H01	1.008	2.013	175.63	3.019	N2
O01-H01A	0.967	1.733	169.91	2.691	O02' [x-1, y, z-1]
O01'-H01B	0.967	1.694	174.56	2.658	O02

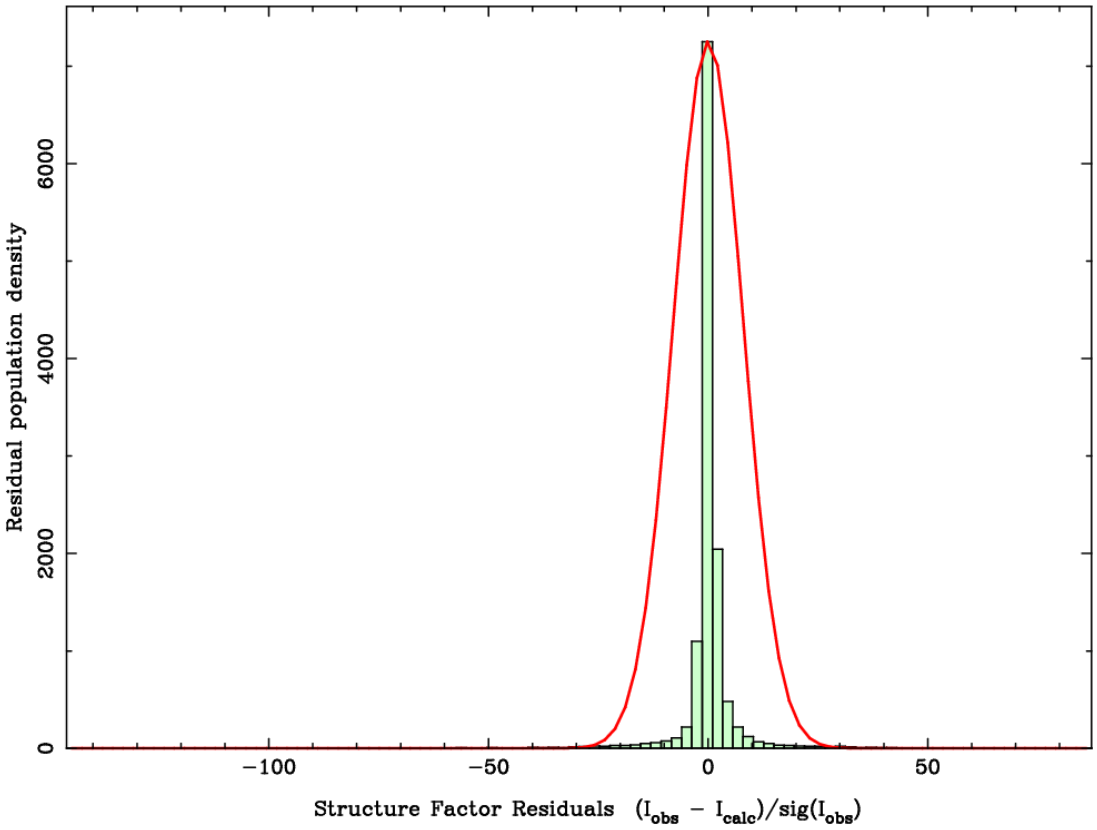
Residual density analysis

Residual density analysis

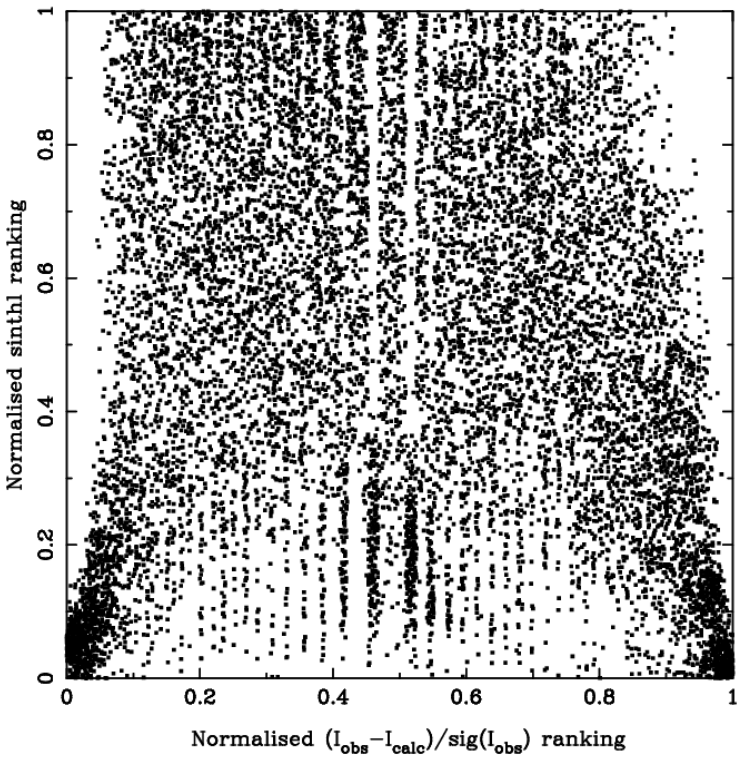
The residual density analysis introduced by Meindl *et al.*[2, 3] was also performed on the data for (**1**), (**2**) and (**3**). The tests were performed to ensure that the residual density in both the IAM and multipolar models both conform to a gaussian distribution allowing it to be attributed to experimental “noise” and ensure critical data has not been overlooked in the refinement process. The parabolic curves in the fractal diagram and residual density histogram and even spread of data points in the structure factor residuals *vs*. standard uncertainties graph for all models are in accordance with what is recommended by Meindl *et al*. to classify the residual density as noise.



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

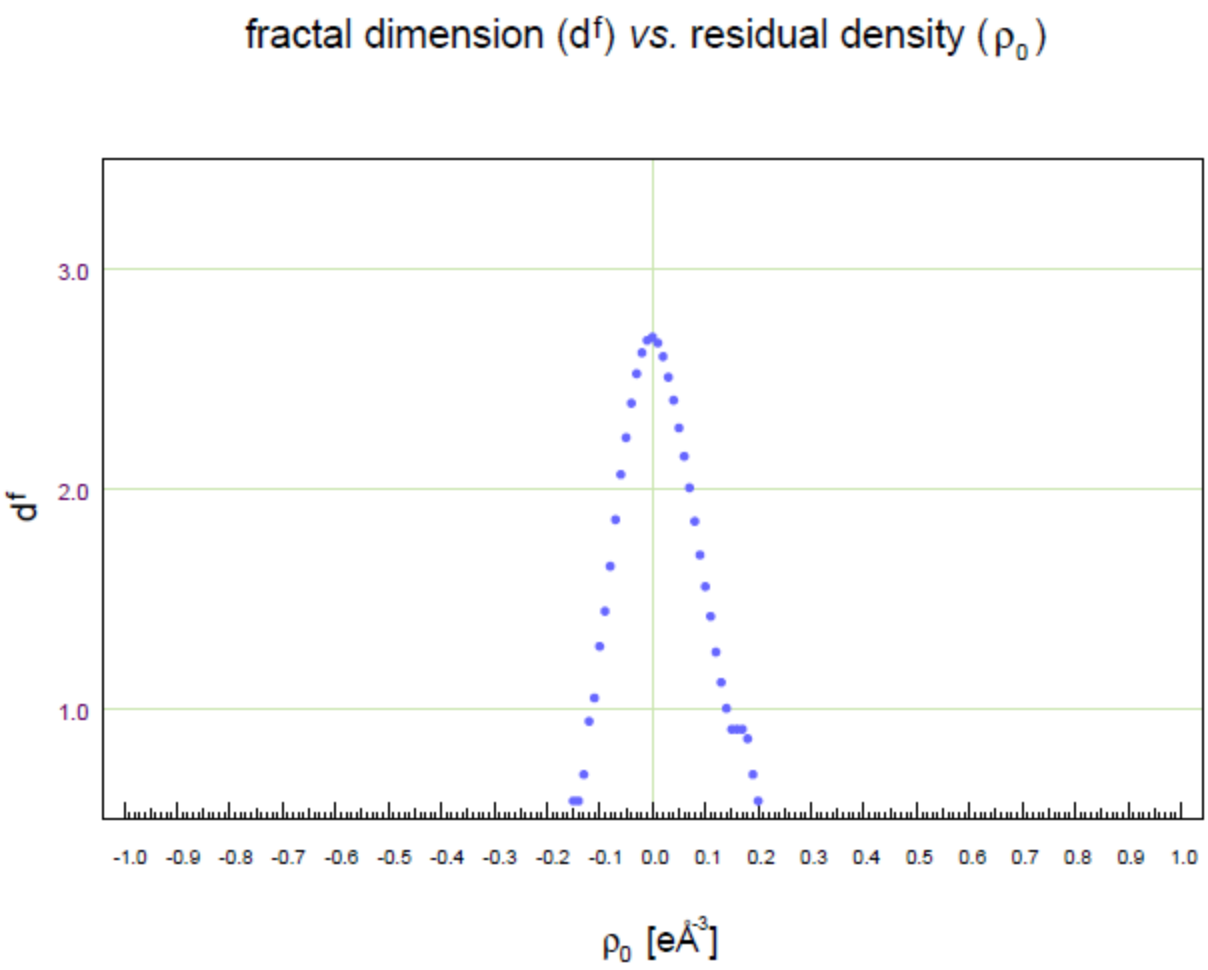


(b) histogram showing residual density of multipole refinement for (1)

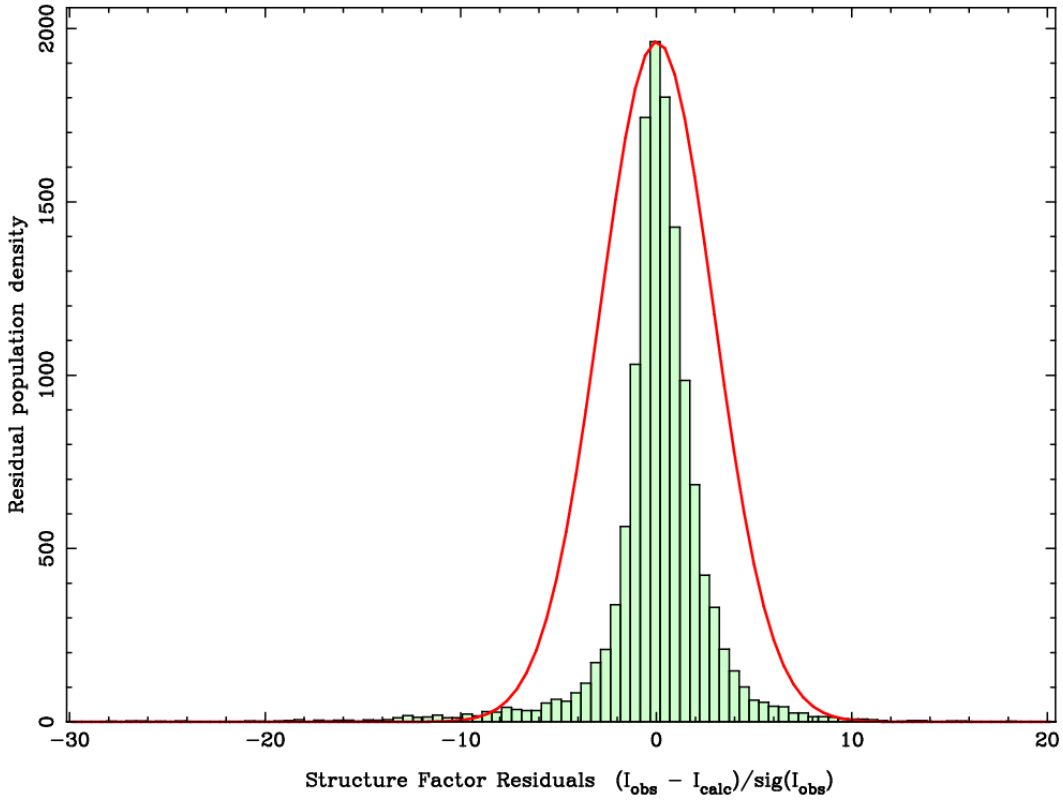


(c) structure factor residuals vs. standard uncertainties for (1)

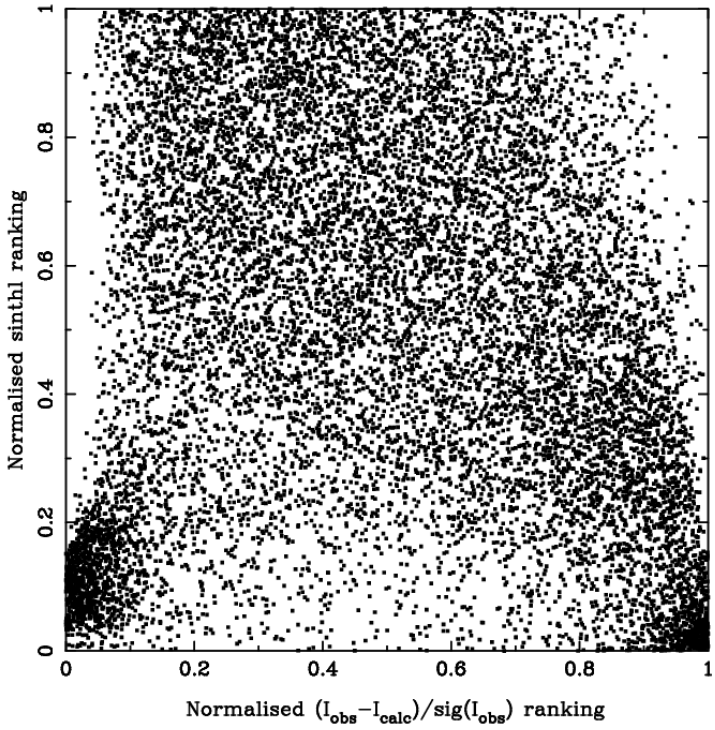
Figure S1: Results of the residual density analysis on the data sets for (**1**)



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

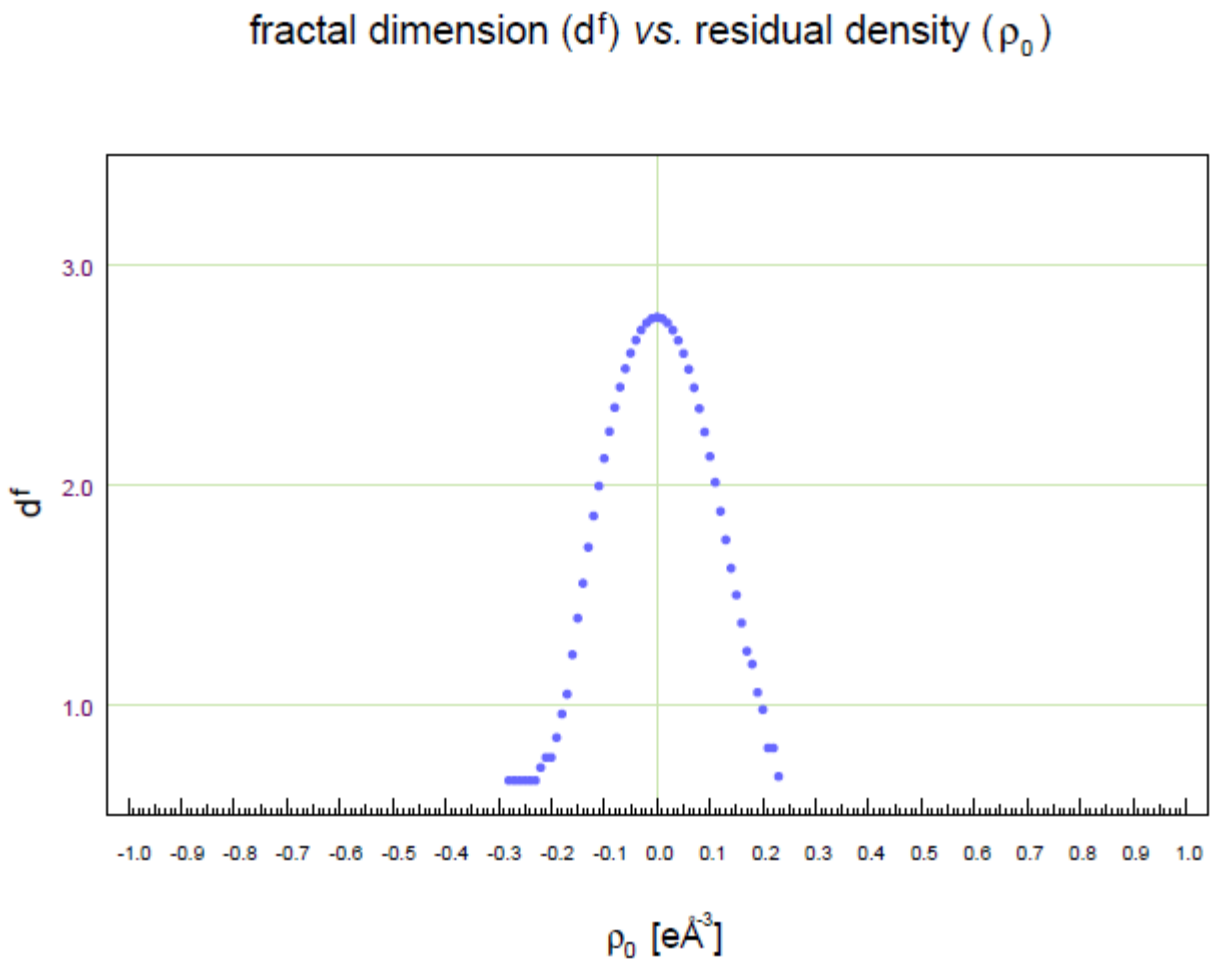


(b) histogram showing residual density of multipole refinement for (**2**)

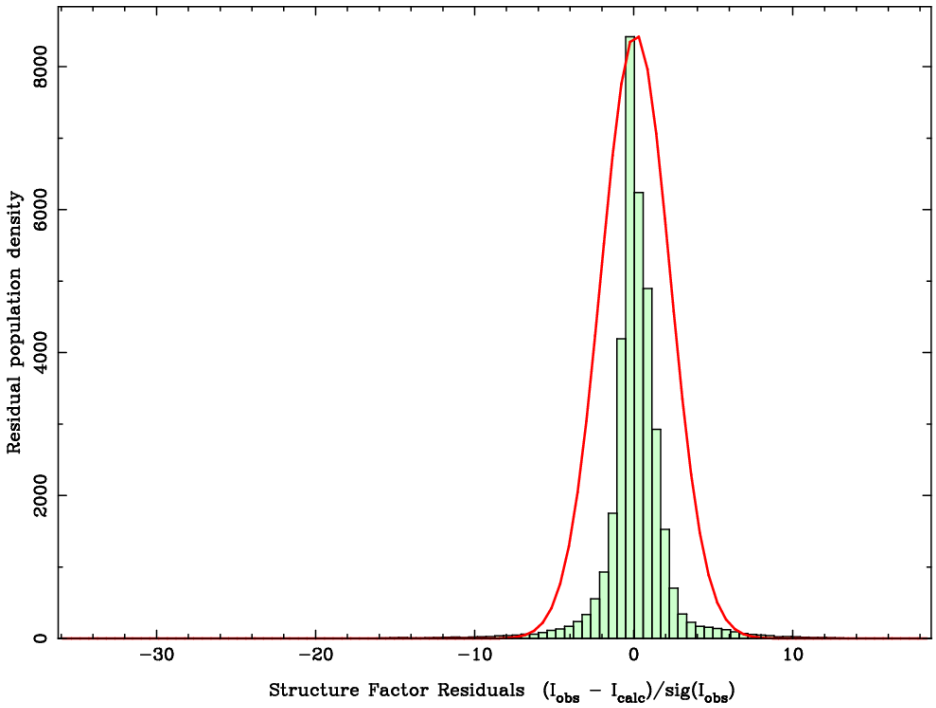


(c) structure factor residuals vs. standard uncertainties for (**2**)

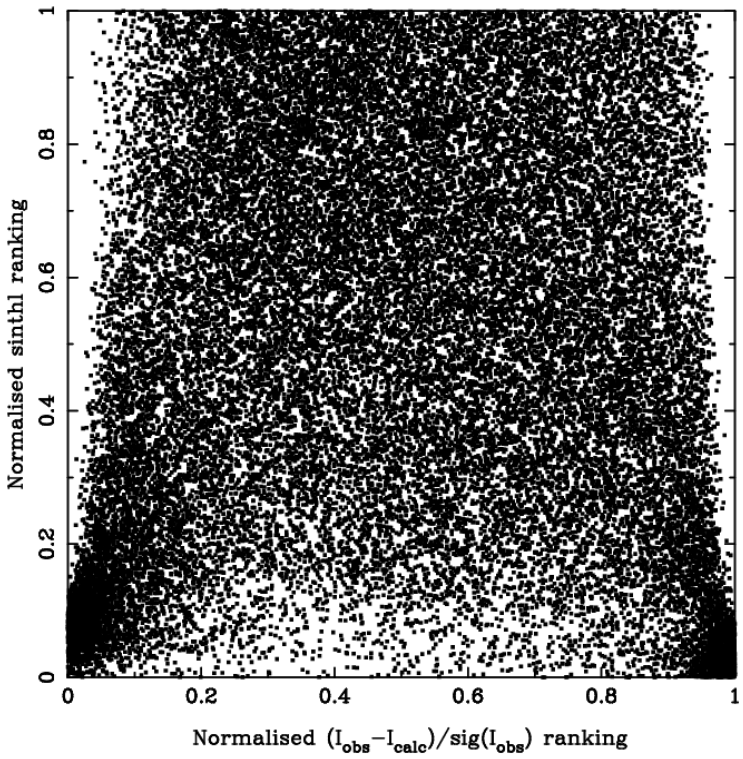
Figure S2: Results of the residual density analysis on the data sets for (**2**)



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



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



(c) structure factor residuals vs. standard uncertainties for (3)

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

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***	**Bond**	***Exp***	**SP**
O(01) -C(01)	2.006	1.918	-17.04	-9.06	0.14	0.02
O(01) -H(01)	2.043	2.435	-30.10	-60.11	0.04	0.02
O(02) -C(07)	2.813	2.653	-35.63	-11.34	0.16	0.08
N(01) -C(04)	1.956	1.894	-15.01	-18.36	0.19	0.07
N(01) -C(07)	2.374	2.202	-25.73	-21.03	0.27	0.10
N(01) -H(01A)	2.072	2.304	-23.66	-39.74	0.05	0.06
C(01) -C(02)	2.112	2.094	-18.77	-20.98	0.26	0.25
C(01) -C(06)	2.160	2.109	-20.13	-21.45	0.29	0.24
C(02) -C(03)	2.129	2.083	-19.38	-20.64	0.23	0.23
C(02) -H(02)	1.851	1.884	-16.85	-23.01	0.09	0.02
C(03) -C(04)	2.135	2.086	-19.35	-20.73	0.27	0.25
C(03) -H(03)	1.800	1.890	-17.00	-23.17	0.08	0.02
C(04) -C(05)	2.099	2.076	-18.28	-20.70	0.25	0.23
C(05) -C(06)	2.109	2.073	-18.51	-20.50	0.23	0.22
C(05) -H(05)	1.827	1.924	-17.27	-24.19	0.06	0.02
C(06) -H(06)	1.827	1.899	-16.42	-23.47	0.09	0.02
C(07) -C(08)	1.765	1.734	-13.00	-15.33	0.16	0.07
C(08) -H(08A)	1.614	1.963	-9.79	-24.60	0.38	0.02
C(08) -H(08B)	1.747	1.972	-12.79	-24.89	0.47	0.02
C(08) -H(08C)	1.809	1.975	-14.41	-25.06	0.36	0.03
O(02) - H(05)	0.107	0.102	1.36	1.31	0.22	0.29

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**	**Bond**	***Exp***
RCP				
-C(01) - C(02) - C(03) - C(04) - C(05) - C(06)-	0.152	0.144	3.10	3.77
-O(02) - C(07) - N(01) - C(04) - C(05) - H(05)-	0.098	0.080	1.40	1.48

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***	**Bond**	***Exp***	**SP**
N(1) -C(2)	2.351	2.297	-27.18	-24.03	0.20	0.10
N(1) -C(3)	2.247	2.299	-26.92	-24.03	0.22	0.10
N(2) -C(8)	2.288	2.306	-27.86	-24.02	0.20	0.10
N(2) -C(9)	2.304	2.318	-24.75	-23.71	0.32	0.10
N(1') -C(2')	2.242	2.290	-26.66	-23.90	0.27	0.11
N(1') -C(3')	2.383	2.303	-27.54	-23.82	0.16	0.11
N(2') -C(8')	2.297	2.312	-28.07	-23.97	0.24	0.11
N(2') -C(9')	2.301	2.291	-27.71	-23.73	0.18	0.10
C(1) -C(2)	2.149	2.123	-22.11	-21.59	0.25	0.22
C(1) -C(5)	2.083	2.082	-19.52	-20.82	0.23	0.19
C(1) -H(1)	1.714	1.902	-16.67	-23.46	0.05	0.02
C(2) -H(2)	1.816	1.939	-19.24	-24.54	0.15	0.03
C(3) -C(4)	2.129	2.126	-22.06	-21.66	0.23	0.22
C(3) -H(3)	1.780	1.939	-17.80	-24.55	0.10	0.03
C(4) -C(5)	2.064	2.079	-19.71	-20.74	0.27	0.19
C(4) -H(4)	1.749	1.902	-18.00	-23.45	0.09	0.02
C(5) -C(6)	1.729	1.796	-13.15	-16.39	0.15	0.06
C(6) -C(7)	2.093	2.097	-19.61	-21.17	0.22	0.19
C(6) -C(10)	2.087	2.100	-19.74	-21.21	0.25	0.19
C(7) -C(8)	2.149	2.133	-22.00	-21.79	0.26	0.22
C(7) -H(7)	1.722	1.904	-16.76	-23.51	0.08	0.02
C(8) -H(8)	1.840	1.937	-20.47	-24.46	0.10	0.03
C(9) -C(10)	2.121	2.129	-22.59	-21.74	0.29	0.22
C(9) -H(9)	1.866	1.940	-22.23	-24.55	0.06	0.03
C(10) -H(10)	1.809	1.904	-19.43	-23.51	0.08	0.02
C(1') -C(2')	2.164	2.134	-22.99	-21.84	0.30	0.21
C(1') -C(5')	2.040	2.083	-19.79	-20.86	0.27	0.19
C(1') -H(1')	1.786	1.921	-18.31	-24.04	0.08	0.02
C(2') -H(2')	1.841	1.932	-19.08	-24.34	0.12	0.03
C(3') -C(4')	2.171	2.123	-22.92	-21.63	0.28	0.22
C(3') -H(3')	1.795	1.937	-18.99	-24.47	0.10	0.03
C(4') -C(5')	2.072	2.093	-19.94	-21.04	0.28	0.20
C(4') -H(4')	1.754	1.900	-17.51	-23.43	0.09	0.02
C(5') -C(6')	1.787	1.809	-14.76	-16.69	0.15	0.06

C(6') -C(7')	2.088	2.090	-20.23	-20.98	0.25	0.20
C(6') -C(10')	2.048	2.080	-19.89	-20.79	0.24	0.19
C(7') -C(8')	2.109	2.126	-21.49	-21.69	0.28	0.22
C(7') -H(7')	1.743	1.900	-17.06	-23.44	0.07	0.02
C(8') -H(8')	1.823	1.936	-19.12	-24.44	0.09	0.03
C(9') -C(10')	2.120	2.141	-21.91	-21.99	0.28	0.22
C(9') -H(9')	1.780	1.935	-17.98	-24.41	0.11	0.03
C(10') -H(10')	1.767	1.910	-16.63	-23.74	0.09	0.02
N(2) - H(1')	0.045	0.092	0.53	1.03	0.47	0.06
N(2) - H(10')	0.070	0.048	1.25	0.50	0.20	0.12

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**	**Bond**	***Exp***
RCP				
-N(1') - C(3') - C(4') - C(5') - C(1') - C(2')-	0.220	0.164	3.50	4.24
-N(2') - C(9') - C(10') - C(6') - C(7') - C(8')-	0.226	0.164	3.50	4.24
-N(2) - H(10') - C(10') - C(6') - C(5') - C(1') - H(1')-		0.039		0.46
-N(2) - C(8) - H(8) - H(2') - C(2') - C(1') - H(1')-		0.020		0.27
-N(2) - C(9) - C(10) - C(6) - C(7) - C(8)-	0.228	0.167	3.60	4.27
-N(1) - C(3) - C(4) - C(5) - C(1) - C(2)-	0.232	0.163	3.50	4.21
-C(1) - C(5) - C(6) - C(10) - H(10) - H(1)-		0.080		1.45
-C(4) - C(5) - C(6) - C(7) - H(7) - H(4)-		0.084		1.55

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***	**Bond**	***Exp***	**SP**
N(1) -C(2)	2.351	2.297	-27.18	-24.03	0.20	0.10
N(1) -C(3)	2.247	2.299	-26.92	-24.03	0.22	0.10
N(2) -C(8)	2.288	2.306	-27.86	-24.02	0.20	0.10
N(2) -C(9)	2.304	2.318	-24.75	-23.71	0.32	0.10
N(1') -C(2')	2.242	2.290	-26.66	-23.90	0.27	0.11
N(1') -C(3')	2.383	2.303	-27.54	-23.82	0.16	0.11
N(2') -C(8')	2.297	2.312	-28.07	-23.97	0.24	0.11
N(2') -C(9')	2.301	2.291	-27.71	-23.73	0.18	0.10
C(1) -C(2)	2.149	2.123	-22.11	-21.59	0.25	0.22
C(1) -C(5)	2.083	2.082	-19.52	-20.82	0.23	0.19
C(1) -H(1)	1.714	1.902	-16.67	-23.46	0.05	0.02
C(2) -H(2)	1.816	1.939	-19.24	-24.54	0.15	0.03
C(3) -C(4)	2.129	2.126	-22.06	-21.66	0.23	0.22
C(3) -H(3)	1.780	1.939	-17.80	-24.55	0.10	0.03
C(4) -C(5)	2.064	2.079	-19.71	-20.74	0.27	0.19
C(4) -H(4)	1.749	1.902	-18.00	-23.45	0.09	0.02
C(5) -C(6)	1.729	1.796	-13.15	-16.39	0.15	0.06
C(6) -C(7)	2.093	2.097	-19.61	-21.17	0.22	0.19
C(6) -C(10)	2.087	2.100	-19.74	-21.21	0.25	0.19
C(7) -C(8)	2.149	2.133	-22.00	-21.79	0.26	0.22
C(7) -H(7)	1.722	1.904	-16.76	-23.51	0.08	0.02
C(8) -H(8)	1.840	1.937	-20.47	-24.46	0.10	0.03
C(9) -C(10)	2.121	2.129	-22.59	-21.74	0.29	0.22
C(9) -H(9)	1.866	1.940	-22.23	-24.55	0.06	0.03
C(10) -H(10)	1.809	1.904	-19.43	-23.51	0.08	0.02
C(1') -C(2')	2.164	2.134	-22.99	-21.84	0.30	0.21
C(1') -C(5')	2.040	2.083	-19.79	-20.86	0.27	0.19
C(1') -H(1')	1.786	1.921	-18.31	-24.04	0.08	0.02
C(2') -H(2')	1.841	1.932	-19.08	-24.34	0.12	0.03
C(3') -C(4')	2.171	2.123	-22.92	-21.63	0.28	0.22
C(3') -H(3')	1.795	1.937	-18.99	-24.47	0.10	0.03
C(4') -C(5')	2.072	2.093	-19.94	-21.04	0.28	0.20
C(4') -H(4')	1.754	1.900	-17.51	-23.43	0.09	0.02
C(5') -C(6')	1.787	1.809	-14.76	-16.69	0.15	0.06

C(6') -C(7')	2.088	2.090	-20.23	-20.98	0.25	0.20
C(6') -C(10')	2.048	2.080	-19.89	-20.79	0.24	0.19
C(7') -C(8')	2.109	2.126	-21.49	-21.69	0.28	0.22
C(7') -H(7')	1.743	1.900	-17.06	-23.44	0.07	0.02
C(8') -H(8')	1.823	1.936	-19.12	-24.44	0.09	0.03
C(9') -C(10')	2.120	2.141	-21.91	-21.99	0.28	0.22
C(9') -H(9')	1.780	1.935	-17.98	-24.41	0.11	0.03
C(10') - H(10')	1.767	1.910	-16.63	-23.74	0.09	0.02
N(2) - H(1')	0.045	0.092	0.53	1.03	0.47	0.06
N(2) - H(10')	0.070	0.048	1.25	0.50	0.20	0.12

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**	**Bond**	***Exp***
RCP			**RCP**	
-N(1') - C(3') - C(4') - C(5') - C(1') - C(2')-	0.220	0.164	3.50	4.24
-N(2') - C(9') - C(10') - C(6') - C(7') - C(8')-	0.226	0.164	3.50	4.24
-N(2) - H(10') - C(10') - C(6') - C(5') - C(1') - H(1')-		0.039		0.46
-N(2) - C(8) - H(8) - H(2') - C(2') - C(1') - H(1')-		0.020		0.27
-N(2) - C(9) - C(10) - C(6) - C(7) - C(8)-	0.228	0.167	3.60	4.27
-N(1) - C(3) - C(4) - C(5) - C(1) - C(2)-	0.232	0.163	3.50	4.21
-C(1) - C(5) - C(6) - C(10) - H(10) - H(1)-		0.080		1.45
-C(4) - C(5) - C(6) - C(7) - H(7) - H(4)-		0.084		1.55

Hydrogen bond geometry

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

Bond	**H...A**	**D...A**	**<(DHA)**
Intramolecular			
C(07)-O(02)···H(05)	2.315	2.912	112.9
Intermolecular			
C(03)-H(03)···O(01)[a]	2.624	2.478	135.3
N(01)-H(01A)···O(01)[a]	1.919	2.91	166.9
O(01)-H(01)···O(02)[b]	1.706	2.654	166.1

Symmetry operators: [a] *x-0.5, -y+0.5,*z *-0.5;* [b] *x*-0.50, *-y+0.5, z+0.5*

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

Bond	H...A	D...A	<(DHA)
Intramolecular			
C(1')-H(1')···N(2)	2.741	3.541	130.5
C(10')-H(10')···N(2)	2.336	3.399	166.8
H(4) - H(7)			
Intermolecular			
C(1)-H(1)···N(1')[a]	2.438	3.469	158.6
C(4)-H(4)···N(2')[b]	2.420	3.497	173.3
C(7)-H(7)···N(2')[b]	2.522	3.540	156.3
C(10)-H(10)···N(1')[a]	2.338	3.411	170.8
C(4')-H(4')···N(1)[c]	2.462	3.468	154.0
C(7')-H(7')···N(1)[c]	2.640	3.578	144.6

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

Bond	**H...A**	**D...A**	**<(DHA)**
Intramolecular			
C(05')-H(05')···O(02')	2.169	2.879	121.0
C(05)-H(05)···O(02)	2.185	2.867	118.8
N(01)-H(01)···N(2)	2.013	3.019	175.6
C(08)-H(08D)···N(2')	2.728	3.588	138.3
N(01')-H(01')···N(2')	1.93	2.928	169.7
Intermolecular			
C(02)-H(02)···N(1')	2.687	3.376	121.1
C(03')-H(03')···N(1)	2.609	3.399	129.2
C(3')-H(3')···O(01')	2.595	3.557	147.7
C(4)-H(4)···O(02')	2.573	3.592	156.6
C(8')-H(8')···O(01)	2.648	3.731	178.0
C(08)-H(08A)···O(01')	2.603	3.661	177.4
C(9)-H(9)···O(02)	2.544	3.477	143.9
H(01A)···O(02')	1.733	2.691	169.9
Short Contacts			
H(9)···H(03)	0.041	0.553	0.91

Symmetry operators: [a] -x+1, y-1/2, -z+1/2; [b] -x+1, y+1/2, -z+1/2; [c] x, -y+3/2, z-1/2; [d] -x+2, -y+1, -z+1; [e] x+1, -y+3/2, z+1/2; [f] -x+2, -y+1, -z+1; [g] -x+1, -y+1, -z+1; [h] x-1, y, z-1

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

Bond	ρ (eÅ^{-3})	$\nabla^2\rho$ (eÅ^{-5})	ε	$d_{H\cdots bcp}$ (Å)	$d_{A\cdots bcp}$ (Å)	G/E$_h$ (eÅ^{-3})	V/E_h (eÅ^{-3})	H/E_h (eÅ^{-3})	G/r	E_{HB} (kJ mol^{-1})
Intramolecular										
C(05)-H(05)···O(02)	0.107	1.4	0.22	1.044	1.358	0.08	-0.07	0.01	0.78	27.46
Intermolecular										
C(03)-H(03)···O(01)[a]	0.143	2.8	0.07	1.247	2.715	0.16	-0.13	0.03	1.14	50.19
N(01)-H(01A)···O(01)[a]	0.143	2.8	0.07	0.691	1.247	0.16	-0.13	0.03	1.14	50.19

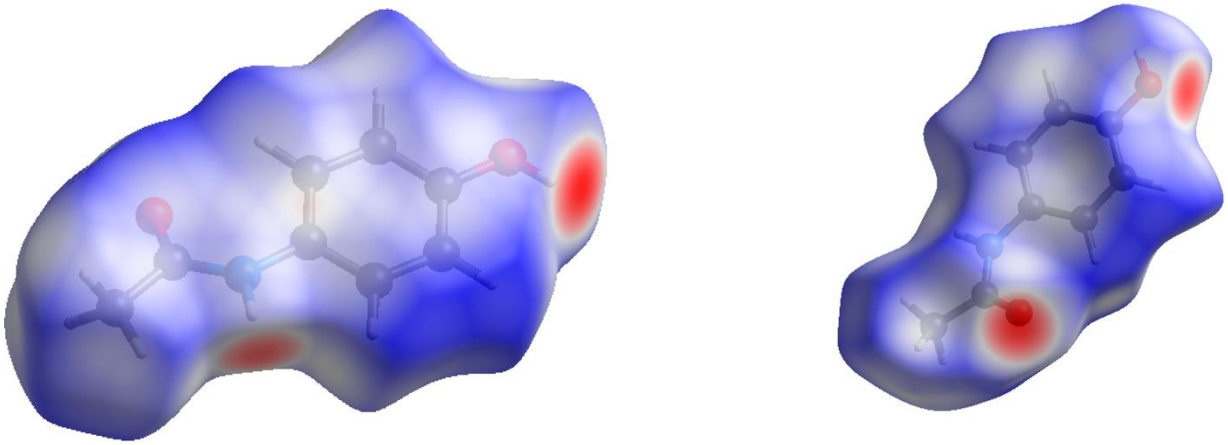
O(01)-H(01)···O(02)[b]	0.216	4.3	0.02	0.579	1.134	0.26	-0.23	0.04	1.22	87.8

Symmetry operators: [a] *x-0.5, -y+0.5,*z *-0.5*; [b] *x*-0.50, *-y+0.5, z+0.5*

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

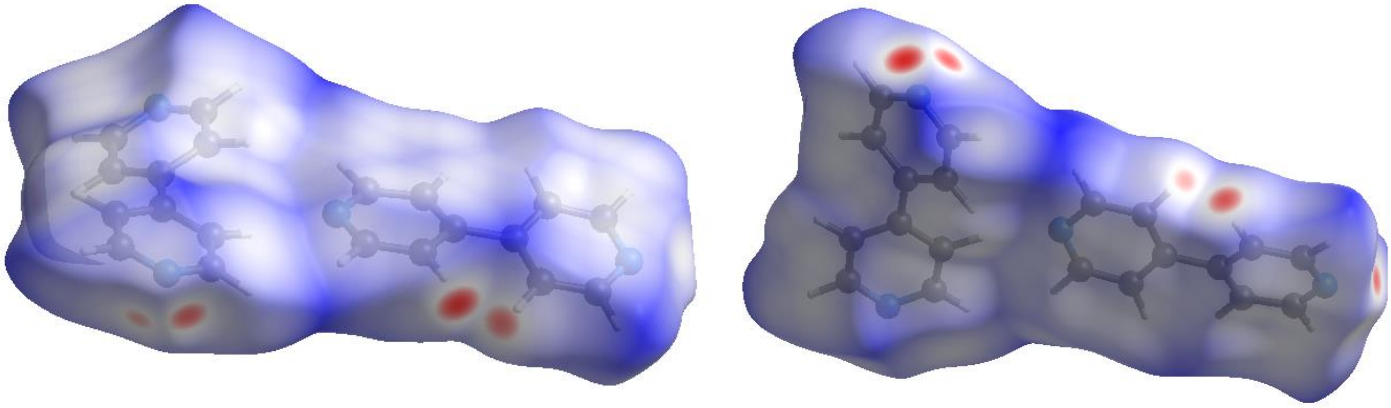
Bond	$\boldsymbol{\rho}$ (eÅ^{-3})	$\boldsymbol{\nabla^2\rho}$ (eÅ^{-5})	$\boldsymbol{\varepsilon}$	$\boldsymbol{d_{H\cdots bcp}}$ (Å)	$\boldsymbol{d_{A\cdots bcp}}$ (Å)	$\boldsymbol{G/E_h}$ (eÅ^{-3})	$\boldsymbol{V/E_h}$ (eÅ^{-3})	$\boldsymbol{H/E_h}$ (eÅ^{-3})	$\boldsymbol{G/r}$	$\boldsymbol{E_{HB}}$ (kJ mol^{-1})
Intermolecular										
C(1')-H(1')···N(2)	0.045	0.5	0.47	1.173	1.633	0.03	-0.02	0.01	0.65	8.36
C(10')-H(10')···N(2)	0.07	1.3	0.2	0.865	1.482	0.07	-0.05	0.02	0.97	18.8
C(1)-H(1)···N(1')[a]	0.051	0.9	0.17	0.922	1.538	0.05	-0.03	0.02	0.94	12.65
C(4)-H(4)···N(2')[b]	0.035	1.0	0.11	0.863	1.566	0.05	-0.03	0.02	1.43	11.49
C(7)-H(7)···N(2')[b]	0.041	0.7	0.24	0.989	1.578	0.04	-0.02	0.01	0.88	9.28
C(10)-H(10)···N(1')[a]	0.05	1.3	0.19	0.835	1.514	0.06	-0.04	0.02	1.3	15.82
C(4')-H(4')···N(1)[c]	0.048	0.8	0.29	0.964	1.541	0.04	-0.03	0.01	0.89	11.29
C(7')-H(7')···N(1)[c]	0.042	0.6	0.25	1.076	1.607	0.03	-0.02	0.01	0.73	8.32
Short Contacts										
H(4)···H(7)	0.029	0.5	0	1.074	1.251	0.03	-0.02	0.01	0.96	6.68

Symmetry operators: [a]x, y, z-1; [b]x-1, y+1, z; [c]x+1, y-1, z+1



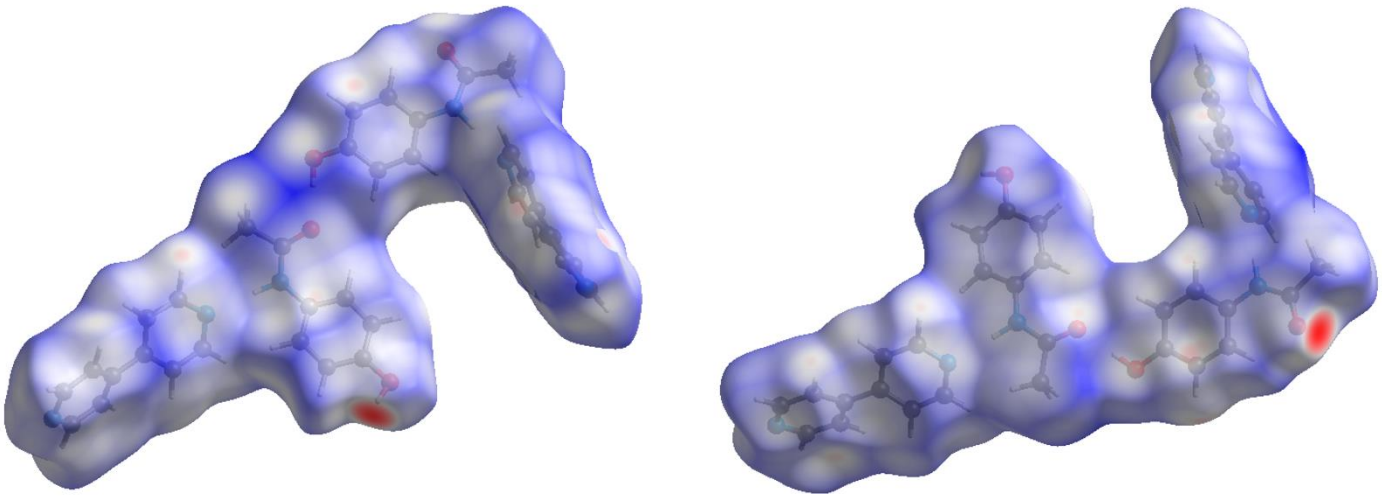
(a) (b)

Figure S4: Hirshfeld surfaces for (**1**).



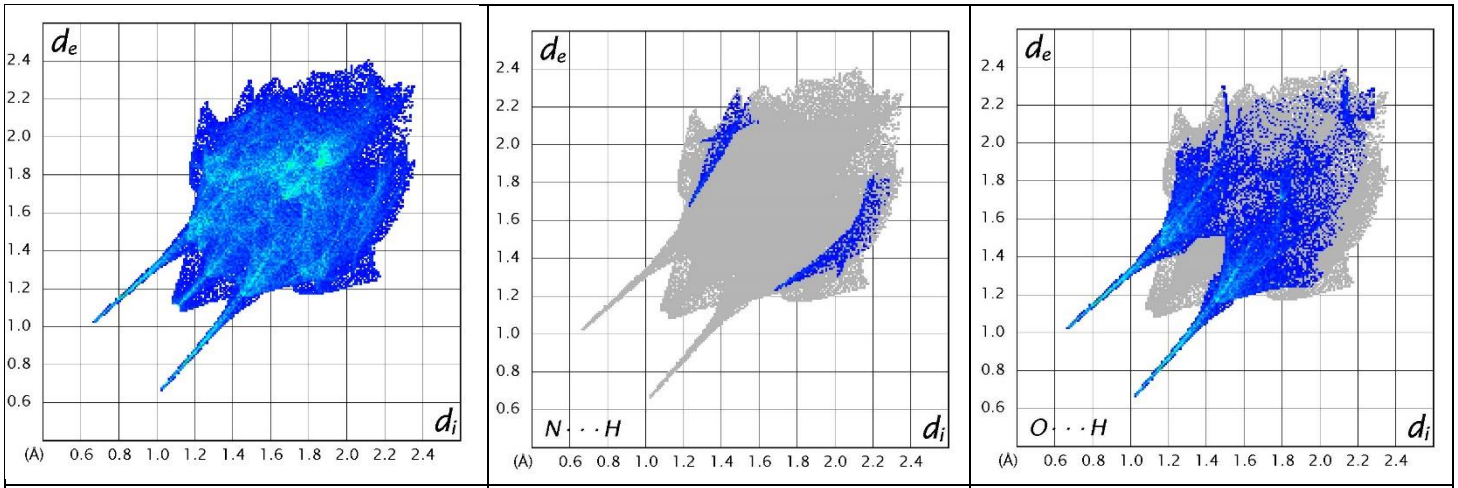
(c) (d)

Figure S5: Hirshfeld surfaces for (**2**).



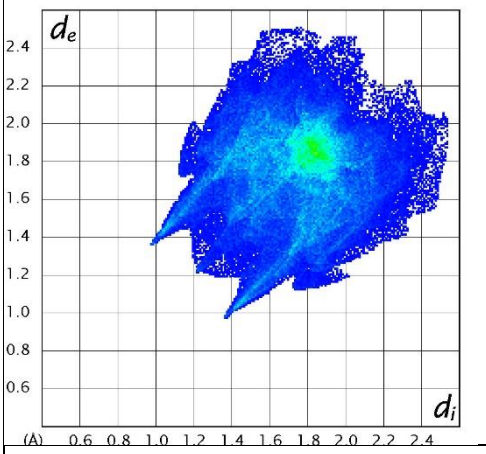
(e) (f)

Figure S6: Hirshfeld surfaces for (**3**).

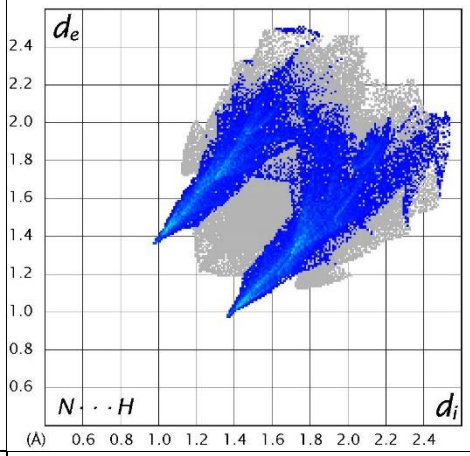


(a) (b) (c)

Figure 11: 2-dimensional fingerprint plots of (a) all interactions in (**1**), (b)N···H interactions in (**1**) and (c) O···H interactions in (**1**)



(d)



(e)

Figure 11: 2-dimensional fingerprint plots of (a) all interaction in (**2**) and (b) N···H interactions in (**2**).

Table S33: Atomic charges (e) from multipole refinement (**1**). Standard uncertainties have been omitted for clarity.

Name	Ω (***Exp***)	Ω (**DFT**)
O(01)	-1.19	-1.11
O(02)	-1.09	-1.14
N(01)	-1.21	-1.20
C(01)	0.40	0.50
C(02)	0.07	-0.02
C(03)	0.03	-0.01
C(04)	0.26	0.34
C(05)	-0.05	0.01
C(06)	0.03	0.00
C(07)	1.14	1.42
C(08)	-0.13	0.03
H(01)	0.67	0.57
H(01A)	0.48	0.38
H(02)	-0.01	0.02
H(03)	0.09	0.01

H(05)	0.15	0.10
H(06)	-0.01	0.05
H(08A)	0.16	0.03
H(08B)	0.12	-0.02
H(08C)	0.14	0.05

Table S34: Atomic charges (e) from multipole refinement (**2**). Standard uncertainties have been omitted for clarity.

Name	Ω (***Exp***)	Ω (**DFT**)
N(1')	-0.74	-1.12
N(2')	-0.77	-1.12
N(1)	-0.80	-1.10
N(2)	-0.81	-1.14
C(1)	-0.11	-0.03
C(2)	0.27	0.51
C(3)	0.40	0.51
C(4)	-0.04	-0.03
C(5)	-0.08	0.00
C(6)	0.04	0.00
C(7)	-0.06	-0.03
C(8)	0.44	0.51
C(9)	0.51	0.52
C(10)	-0.15	-0.03
H(1)	0.17	0.04
H(2)	-0.05	0.05
H(3)	-0.01	0.05
H(4)	0.21	0.03
H(7)	0.15	0.04
H(8)	-0.06	0.04
H(9)	-0.16	0.04
H(10)	0.15	0.04
C(1')	-0.05	-0.05
C(2')	0.38	0.50
C(3')	0.29	0.51
C(4')	-0.07	-0.04
C(5')	-0.01	-0.01
C(6')	-0.04	0.00

C(7')	-0.09	-0.04
C(8')	0.32	0.52
C(9')	0.30	0.50
C(10')	-0.01	-0.03
H(1')	0.14	0.09
H(2')	-0.04	0.03
H(3')	0.07	0.04
H(4')	0.12	0.04
H(7')	0.16	0.04
H(8')	0.00	0.04
H(9')	0.02	0.03
H(10')	0.06	0.06

Table S35: Atomic charges (e) from multipole refinement (**3**). Standard uncertainties have been omitted for clarity.

Name	Ω (***Exp***)	Ω (**DFT**)
N(1')	-0.74	-1.12
N(2')	-0.77	-1.12
N(1)	-0.80	-1.10
N(2)	-0.81	-1.14
C(1)	-0.11	-0.03
C(2)	0.27	0.51
C(3)	0.40	0.51
C(4)	-0.04	-0.03
C(5)	-0.08	0.00
C(6)	0.04	0.00
C(7)	-0.06	-0.03
C(8)	0.44	0.51
C(9)	0.51	0.52
C(10)	-0.15	-0.03
H(1)	0.17	0.04
H(2)	-0.05	0.05
H(3)	-0.01	0.05
H(4)	0.21	0.03
H(7)	0.15	0.04

H(8)	-0.06	0.04
H(9)	-0.16	0.04
H(10)	0.15	0.04
C(1')	-0.05	-0.05
C(2')	0.38	0.50
C(3')	0.29	0.51
C(4')	-0.07	-0.04
C(5')	-0.01	-0.01
C(6')	-0.04	0.00
C(7')	-0.09	-0.04
C(8')	0.32	0.52
C(9')	0.30	0.50
C(10')	-0.01	-0.03
H(1')	0.14	0.09
H(2')	-0.04	0.03
H(3')	0.07	0.04
H(4')	0.12	0.04
H(7')	0.16	0.04
H(8')	0.00	0.04
H(9')	0.02	0.03
H(10')	0.06	0.06

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