Exploring temperature dependent proton migration behaviour across short strong hydrogen bonds engineered in organic acid molecular crystals using single crystal synchrotron X-ray diffraction (SCSXRD)

L. K. Saunders,[a] H. Nowell,[a] L. E. Hatcher,[b] H. J. Shepherd,[c] S. J. Teat,[d] D. R. Allan, [d] P. R. Raithby[b] and C. C. Wilson[b]

a Diamond Light Source, Harwell Science and Innovation Campus, Didcot OX11 0DE, U.K.

b Department of Chemistry, University of Bath, Bath, BA2 7AY, U.K.

c School of Physical Sciences, Ingram Building, University of Kent, Canterbury, CT2 7NH, U.K.

d Advanced Light Source, 1 Cyclotron Road, Lawrence Berkeley National Laboratory, Berkeley, CA 94720-8229, U.S.A

E-mail: lucy.saunders@diamond.ac.uk

SUPPLEMENTARY INFORMATION

Contents

Table S1	Crystallographic data for **1** (*N,N*-dimethylurea phthalic acid 2:1) over 100 to 300 K temperature range collected on beamline 11.3.1 at the Advanced Light Source, U.S.A.
Table S2	Crystallographic data for **2** (*N,N*-dimethylurea 2,4-dinitrobenzoic acid 1:1) over 100 to 350 K temperature range collected on beamline I19 at Diamond Light Source, U.K.
Table S3	Crystallographic data for **3** (*N,N*-dimethylurea 3,5-dinitrobenzoic acid 1:1) over 100 to 300 K temperature range collected on beamline 11.3.1 at the Advanced Light Source, U.S.A.
Table S4	Crystallographic data for **4** (*N,N*-dimethylurea 3,5-dinitrobenzoic acid) over 100 to 300 K temperature range collected on beamline I19 at Diamond Light Source, U.K.
Table S5	Crystallographic data for **5** (isonicotinamide phthalic acid 2:1) over 100 to 350 K temperature range collected on beamline 11.3.1 at the Advanced Light Source, U.S.A.
Table S6	Crystallographic data for **6** (isonicotinamide 2,4-dinitrobenzoic acid 1:1) over 100 to 350 K temperature range collected on beamline I19 at Diamond Light Source, U.K.
Table S7	Crystallographic data for **7** (isonicotinamide 2,4-dinitrobenzoic acid 2:2) over 100 to 350 K temperature range collected on beamline 11.3.1 at the Advanced Light Source, U.S.A.
Table S8	Crystallographic data for **8** (isonicotinamide 3,5-dinitrobenzoic acid 1:1) over 100 to 350 K temperature range collected on beamline I19 at Diamond Light Source, U.K.
Table S9	Selected structural parameters in **5 – 8**: <CONC is the torsion angle between the acid co-former and isonicotinamide (IN) molecule; <OCCC is the torsion angle of the amide group to *N*-heterocyclic ring in the IN molecule.
Table S10	The $O1^{+}$—H1···$O2^{-}$ SSHB parameters in **2** over the 100 to 350 K temperature range.
Table S11	The $N3^{+}$—H3···$O3^{-}$ SSHB parameters in **5** between 100 and 350 K.
Table S12	Bond distances of the groups forming the $O1^{+}$—H1···$O2^{-}$ SSHB over the temperature range in **2**.

Table S13	Bond distances of the groups forming the $N3^{+}$—H3···$O3^{-}$ SSHB over the temperature range in **5**.
Table S14	The $N1^{+}$—H2···$O5^{-}$ SSHB parameters in **7** between 100 and 350 K.
Table S15	The $N1^{+}$—H1···$O1^{-}$ SSHB parameters over the 100 to 350 K temperature range in **8**.
Table S16	Bond distances of the groups forming the $N1^{+}$—H2···$O5^{-}$ SSHB over the temperature range in **7**.
Table S17	Bond distances of the groups forming the $N1^{+}$—H1···$O1^{-}$ SSHB over the temperature range in **8**.
Table S18	The O1—H1···O2 SSHB parameters in **3** over the 100 to 350 K temperature range.
Table S19	The O—H···O SSHB parameters in **1** over the 100 to 300 K temperature range.
Table S20	The O—H···O SSHB parameters in **4** over the 100 to 300 K temperature range.
Table S21	The $N1^{+}$—H1···$O1^{-}$ SSHB parameters over the 100 to 350 K temperature range in **6**.
Fig. S1	The D—H and H···A distances over the 100 to 350 K temperature range in **1**, **3**, **4** and **6**.
Fig. S2	Fourier difference electron density maps showing the electron density associated with the H-atom in the O1—H1···O2 SSHB in **3** over the 100 to 300 K temperature range.
Fig. S3	Fourier difference electron density maps showing the electron density associated with the H-atom in the O3—H1···O6 and O2—H2···O5 SSHBs in **1** over the 100 to 300 K temperature range.
Fig. S4	Fourier difference electron density maps showing the electron density associated with the H-atom in the O1—H1···O7 and O8—H13···O14 SSHB in **4** over the 100 to 300 K temperature range.
Fig. S5	Fourier difference electron density maps showing the electron density associated with the H-atom in the $N1^{+}$—H1···$O1^{-}$ SSHB in **6** over the 100 to 300 K temperature range.

Table S1 Crystallographic data for **1** (*N,N*-dimethylurea phthalic acid 2:1) over 100 to 300 K temperature range collected on beamline 11.3.1 at the Advanced Light Source, U.S.A.

Formula	$C_{14}H_{22}N_4O_6$	$C_{14}H_{22}N_4O_6$	$C_{14}H_{22}N_4O_6$
Mol. W ($gmol^{-1}$)	342.35	342.35	342.35
T (K)	100	200	300
Space group	$P2_1/c$	$P2_1/c$	$P2_1/c$
a (Å)	7.8576(5)	7.9905(5)	8.1487(4)
b (Å)	6.4443(4)	6.4455(4)	6.4317(3)
c (Å)	32.5997(18)	32.7612(18)	32.9567(15)
α (°)	90	90	90
β (°)	94.616(3)	95.375(3)	96.369(3)
γ (°)	90	90	90
Volume ($Å^3$)	1645.39(17)	1679.87(17)	1716.60(14)
Z	4	4	4
ρ_{calc} ($Mg\ m^{-3}$)	1.382	1.354	1.325
μ (mm^{-1})	0.096	0.094	0.092
Θ range (°)	2.734-30.650	2.723-30.646	2.712-30.646
Reflections collected	22440	22109	23830
Independent reflections	3940	4026	4102
R_{int}	0.0700	0.0783	0.0726
Completeness (%)	99.9	100.0	100.0
SHELXL			
Parameters	305	305	272
GooF	1.023	1.018	1.009
Final R indices [I>2sigma(I)] (R_1, wR2)	0.0451, 0.1037	0.0447, 0.1059	0.0516, 0.1228
Final R indices (all data) (R_1, wR2)	0.0671, 0.1124	0.0795, 0.1188	0.0959, 0.1439
Δρ (max, min) ($e\ Å^{-3}$)	0.305, -0.275	0.276, -0.294	0.213, -0.205

Table S2 Crystallographic data for **2** (*N*,*N*-dimethylurea 2,4-dinitrobenzoic acid 1:1) over 100 to 350 K temperature range collected on beamline I19 at Diamond Light Source, U.K.

Formula	$C_{10}H_{12}N_4O_7$	$C_{10}H_{12}N_4O_7$	$C_{10}H_{12}N_4O_7$	$C_{10}H_{12}N_4O_7$	$C_{10}H_{12}N_4O_7$	$C_{10}H_{12}N_4O_7$
Mol. W ($gmol^{-1}$)	300.24	300.24	300.24	300.24	300.24	300.24
T (K)	100	150	200	250	300	350
Space group	Pbca	Pbca	Pbca	Pbca	Pbca	Pbca
a (Å)	22.95610(12)	22.97700(7)	23.02270(8)	23.08380(10)	23.1543(2)	23.2312(3)
b (Å)	19.16910(11)	19.20650(6)	19.24130(7)	19.28500(10)	19.32370(17)	19.3458(3)
c (Å)	5.80590(3)	5.83570(2)	5.87020(2)	5.90715(3)	5.95150(6)	6.00460(10)
α (°)	90	90	90	90	90	90
β (°)	90	90	90	90	90	90
γ (°)	90	90	90	90	90	90
Volume ($Å^3$)	2554.87(2)	2575.340(14)	2600.421(17)	2629.69(2)	2662.86(4)	2698.62(7)
Z	8	8	8	8	8	8
ρ_{calc} ($Mg\ m^{-3}$)	1.561	1.549	1.534	1.517	1.498	1.478
μ (mm^{-1})	0.084	0.083	0.082	0.082	0.081	0.079
Θ range (°)	2.004-27.739	1.718-27.740	1.998-27.740	1.710-27.740	1.988-27.740	1.982-27.739
Reflections collected	36127	37203	37690	37981	38298	38400
Independent reflections	3283	3315	3350	3383	3428	3474
R_{int}	0.0446	0.0619	0.0525	0.0404	0.0400	0.0397
Completeness (%)	99.8	100.0	99.8	99.7	99.8	99.7
SHELXL						
Parameters	238	238	238	227	227	216
GooF	1.061	1.051	1.068	1.109	1.121	0.979
Final R indices [I>2sigma(I)] (R_1, wR2)	0.0306, 0.0833	0.0341, 0.0953	0.0354, 0.0990	0.0380, 0.1086	0.0408, 0.1308	0.0443, 0.1315
Final R indices (all data) (R_1, wR2)	0.0376, 0.0872	0.0388, 0.0988	0.0417, 0.1035	0.0494, 0.1146	0.0614, 0.1423	0.0904, 0.1517
Δρ (max, min) ($e\ Å^{-3}$)	0.3, -0.238	0.257, -0.232	0.218, -0.224	0.222, -0.189	0.184, -0.183	0.157, -0.179
HAR						
Chi2 final	1.870031	1.936550	2.674991	3.975747	9.109069	9.267979
Max. shift (esd)	0.004216	0.005028	0.03588	0.009017	0.006246	0.008051
Final R indices [I>2sigma(I)] (R_1, wR2)	0.0173, 0.0169	0.026, 0.0225	0.0231, 0.0232	0.0192, 0.0019	0.0366, 0.0332	0.0379, 0.0305
Δρ (max, min) ($e\ Å^{-3}$)	0.0747, -0.0616	0.0841, -0.091	0.0933, -0.0938	0.0636, -0.0654	0.1155, -0.1094	0.0828, -0.0816

Table S3 Crystallographic data for **3** (*N,N*-dimethylurea 3,5-dinitrobenzoic acid 1:1) over 100 to 300 K temperature range collected on beamline 11.3.1 at the Advanced Light Source, U.S.A.

Formula	$C_{10}H_{12}N_4O_7$	$C_{10}H_{12}N_4O_7$	$C_{10}H_{12}N_4O_7$	$C_{10}H_{12}N_4O_7$	$C_{10}H_{12}N_4O_7$
Mol. W ($gmol^{-1}$)	300.24	300.24	300.24	300.24	300.24
T (K)	100	150	200	250	300
Space group	$P2_1$	$P2_1$	$P2_1$	$P2_1$	$P2_1$
a (Å)	7.4233(4)	7.4488(4)	7.4809(4)	7.5150(4)	7.5476(4)
b (Å)	11.3405(5)	11.3580(5)	11.3782(6)	11.4026(6)	11.4257(6)
c (Å)	7.8840(4)	7.8908(4)	7.8924(4)	7.8998(5)	7.9214(5)
α (°)	90	90	90	90	90
β (°)	103.648(3)	103.062(3)	102.334(3)	101.509(4)	100.763(4)
γ (°)	90	90	90	90	90
Volume ($Å^3$)	644.96(6)	650.31(6)	656.29(6)	663.33(7)	671.10(7)
Z	2	2	2	2	2
ρ_{calc} ($Mg\ m^{-3}$)	1.546	1.533	1.519	1.503	1.486
μ (mm^{-1})	0.183	0.182	0.180	0.178	0.176
Θ range (°)	3.311-36.180	3.302-36.176	3.292-36.185	3.279-36.164	3.262-36.177
Reflections collected	10290	10340	10496	10617	10837
Independent reflections	3155	3166	3191	3231	3282
R_{int}	0.0287	0.0260	0.0264	0.0258	0.0279
Completeness (%)	100.0	99.9	100.0	100.0	100.0
SHELXL					
Parameters	216	216	216	216	216
Flack parameter	-0.2(2)	-0.1(2)	-0.2(3)	-0.5(3)	-0.1(3)
GooF	1.075	1.065	1.047	1.039	0.983
Final R indices [I>2sigma(I)] (R_1, wR2)	0.0376, 0.0913	0.0386, 0.0918	0.0422, 0.0950	0.0439, 0.0952	0.0419, 0.0914
Final R indices (all data) (R_1, wR2)	0.0454, 0.0952	0.0489, 0.0964	0.0616, 0.1040	0.0736, 0.1078	0.0874, 0.1082
Δρ (max, min) ($e\ Å^{-3}$)	0.259, -0.285	0.226, -0.220	0.194, -0.193	0.159, -0.201	0.137, -0.165

Table S4 Crystallographic data for **4** (*N,N*-dimethylurea 3,5-dinitrobenzoic acid 2:2) over 100 to 300 K temperature range collected on beamline I19 at Diamond Light Source, U.K.

Formula	$C_{20}H_{24}N_8O_{14}$	$C_{20}H_{24}N_8O_{14}$	$C_{20}H_{24}N_8O_{14}$
Mol. W ($gmol^{-1}$)	600.47	600.47	600.47
T (K)	100	200	300
Space group	C2/c	C2/c	C2/c
a (Å)	28.3637(2)	28.48730(16)	28.61360(19)
b (Å)	6.10113(4)	6.09630(4)	6.10500(4)
c (Å)	29.8215(2)	30.29390(19)	30.8489(2)
α (°)	90	90	90
β (°)	92.5650(10)	92.3750(5)	92.1270(6)
γ (°)	90	90	90
Volume ($Å^3$)	5155.46(6)	5256.54(5)	5385.16(6)
Z	8	8	8
ρ_{calc} ($Mg\ m^{-3}$)	1.547	1.518	1.481
μ (mm^{-1})	0.083	0.082	0.080
Θ range (°)	1.325-27.338	1.304-27.337	1.280-27.399
Reflections collected	36276	38334	39335
Independent reflections	6377	6495	6662
R_{int}	0.0453	0.0510	0.0459
Completeness (%)	99.7	99.3	99.4
SHELXL			
Parameters	464	431	415
GooF	1.062	1.079	1.083
Final R indices [I>2sigma(I)] (R_1, wR2)	0.0356, 0.1002	0.0425, 0.01261	0.0492, 0.1564
Final R indices (all data) (R_1, wR2)	0.0396, 0.1031	0.0473, 0.1304	0.0630, 0.1677
Δρ (max, min) ($e\ Å^{-3}$)	0.309, -0.258	0.264, -0.209	0.200, -0.191

Table S5 Crystallographic data for **5** (isonicotinamide phthalic acid 2:1) over 100 to 350 K temperature range collected on beamline 11.3.1 at the Advanced Light Source, U.S.A.

Formula	$C_{20}H_{18}N_4O_6$	$C_{20}H_{18}N_4O_6$	$C_{20}H_{18}N_4O_6$	$C_{20}H_{18}N_4O_6$	$C_{20}H_{18}N_4O_6$	$C_{20}H_{18}N_4O_6$
Mol. W ($gmol^{-1}$)	410.38	410.38	410.38	410.38	410.38	410.38
T (K)	100	150	200	250	300	350
Space group	$P2_1/n$	$P2_1/n$	$P2_1/n$	$P2_1/n$	$P2_1/n$	$P2_1/n$
a (Å)	7.0539(3)	7.0883(3)	7.1282(3)	7.1747(3)	7.2267(3)	7.2897(3)
b (Å)	14.3314(6)	14.3309(6)	14.3290(6)	14.3223(6)	14.3176(6)	14.3165(6)
c (Å)	18.3058(8)	18.3184(7)	18.3340(7)	18.3518(7)	18.3746(7)	18.4105(7)
α (°)	90	90	90	90	90	90
β (°)	92.9626(19)	92.8585(18)	92.7370(18)	92.6162(19)	92.5051(19)	92.410(2)
γ (°)	90	90	90	90	90	90
Volume ($Å^3$)	1848.10(14)	1858.50(13)	1870.50(13)	1883.83(13)	1899.38(13))	1919.67(13)
Z	4	4	4	4	4	4
ρ_{calc} ($g\ cm^{-3}$)	1.475	1.467	1.457	1.447	1.435	1.420
μ (mm^{-1})	0.147	0.146	0.145	0.144	0.143	0.141
Θ range (°)	2.250-41.364	2.249-41.365	2.249-41.363	2.248-41.368	2.248-41.366	2.246-41.361
Reflections collected	33784	34116	34324	34716	35041	35430
Independent reflections	6362	6415	6455	6517	6548	6608
R_{int}	0.0406	0.0362	0.0389	0.0372	0.0374	0.0415
Completeness (%)	100.0	100.0	100.0	100.0	100.0	100.0
SHELXL						
Parameters	343	343	343	343	343	343
GooF	1.044	1.043	1.039	1.039	1.030	1.024
Final R indices [I>2sigma(I)] (R_1, wR2)	0.0387, 0.1041	0.0391, 0.1082	0.0412, 0.1125	0.0444, 0.1181	0.0472, 0.1205	0.0533, 0.1299
Final R indices (all data) (R_1, wR2)	0.0482, 0.1115	0.0487, 0.1163	0.0535, 0.1223	0.0605, 0.1308	0.0713, 0.1355	0.0922, 0.1502
Δρ (max, min) ($e\ Å^{-3}$)	0.538, -0.257	0.433, -0.220	0.386, -0.215	0.341, -0.212	0.272, -0.207	0.244, -0.199
HAR						
Chi2 final	1.975890	2.209121	2.392734	2.649042	2.634677	3.243190
Max. shift (esd)	0.008125	0.009183	0.007896	0.007837	0.006606	0.005517
Final R indices [I>2sigma(I)] (R_1, wR2)	0.0221, 0.0194	0.0220, 0.0176	0.0253, 0.0196	0.0289, 0.0194	0.0337, 0.0196	0.0436, 0.0230
Δρ (max, min) ($e\ Å^{-3}$)	0.1169, -0.1341	0.1045, -0.1145	0.1262, -0.1337	0.1068, -0.1539	0.1178, -0.1587	0.1629, -0.1547

Table S6 Crystallographic data for **6** (isonicotinamide 2,4-dinitrobenzoic acid 1:1) over 100 to 350 K temperature range collected on beamline I19 at Diamond Light Source, U.K.

Formula	$C_{13}H_{10}N_4O_7$	$C_{13}H_{10}N_4O_7$	$C_{13}H_{10}N_4O_7$
Mol. W ($gmol^{-1}$)	334.25	334.25	334.25
T (K)	100	200	300
Space group	$P2_1/c$	$P2_1/c$	$P2_1/c$
a (Å)	11.03460(7)	11.11940(5)	11.22350(8)
b (Å)	11.48110(8)	11.56830(5)	11.66850(7)
c (Å)	11.85960(8)	11.89730(6)	11.95710(8)
α (°)	90	90	90
β (°)	107.6910(6)	108.2370(4)	108.9360(6)
γ (°)	90	90	90
Volume ($Å^3$)	1431.431(17)	1453.509(12)	1481.174(17)
Z	4	4	4
ρ_{calc} ($Mg\ m^{-3}$)	1.551	1.527	1.499
μ (mm^{-1})	0.079	0.078	0.077
Θ range (°)	2.451-36.017	2.442-36.135	1.859-36.080
Reflections collected	30463	31718	32404
Independent reflections	6889	7061	7216
R_{int}	0.0424	0.0550	0.0559
Completeness (%)	99.5	99.5	99.5
SHELXL			
Parameters	257	257	257
GooF	1.117	1.070	1.047
Final R indices [I>2sigma(I)] (R_1, wR2)	0.410, 0.1257	0.0476, 0.1417	0.0539, 0.1681
Final R indices (all data) (R_1, wR2)	0.0501, 0.1309	0.0596, 0.1502	0.0826, 0.1857
Δρ (max, min) ($e\ Å^{-3}$)	0.580, -0.259	0.465, -0.221	0.344, -0.221

Table S7 Crystallographic data for **7** (isonicotinamide 2,4-dinitrobenzoic acid 2:2) over 100 to 350 K temperature range collected on beamline 11.3.1 at the Advanced Light Source, U.S.A.

Formula	$C_{26}H_{20}N_8O_{14}$	$C_{26}H_{20}N_8O_{14}$	$C_{26}H_{20}N_8O_{14}$	$C_{26}H_{20}N_8O_{14}$	$C_{26}H_{20}N_8O_{14}$	$C_{26}H_{20}N_8O_{14}$
Mol. W ($gmol^{-1}$)	668.5	668.5	668.5	668.5	668.5	668.5
T (K)	100	150	200	250	300	350
Space group	P-1	P-1	P-1	P-1	P-1	P-1
a (Å)	9.6450(4)	9.6632(4)	9.6926(4)	9.7313(4)	9.7819(4)	9.8448(6
b (Å)	10.2722(4)	10.3059(4)	10.3369(4)	10.3587(4)	10.3742(4)	10.3745(6)
c (Å)	15.2241(6)	15.2750(6)	15.3321(6)	15.3935(6)	15.4651(7)	15.5573(9)
α (°)	79.3772(18)	79.0739(18)	78.854(2)	78.736(2)	78.774(2)	79.023(4)
β (°)	72.0170(17)	71.8454(18)	71.624(2)	71.341(2)	71.008(2)	70.556(3)
γ (°)	77.3694(18)	77.2714(19)	77.257(2)	77.307(2)	77.449(2)	77.704(4)
Volume ($Å^3$)	1388.80(10)	1397.91(10)	1409.14(10)	1421.05(10)	1435.37(11)	1451.58(15)
Z	2	2	2	2	2	2
ρ_{calc} ($Mg\ m^{-3}$)	1.599	1.588	1.576	1.562	1.547	1.529
μ (mm^{-1})	0.181	0.180	0.178	0.177	0.175	0.173
Θ range (°)	2.552-35.633	2.546-35.634	2.540-35.633	2.535-35.633	2.529-35.632	2.525-35.631
Reflections collected	21882	22262	22472	22770	23053	23376
Independent	6582	6629	6692	6755	6832	6895
R_{int}	0.0439	0.0458	0.0355	0.0309	0.0300	0.0373
Completeness (%)	99.9	99.8	99.8	99.9	99.9	99.9
SHELXL						
Parameters	513	513	513	513	513	513
GooF	1.029	1.034	1.016	1.036	1.023	1.010
Final R indices [I>2sigma(I)] (R_1, wR2)	0.0364, 0.0980	0.0405, 0.1035	0.0408, 0.1050	0.0446, 0.1129	0.0478, 0.1208	0.0553, 0.1322
Final R indices (all data) (R_1, wR2)	0.0433, 0.1033	0.0517, 0.1106	0.0546, 0.1140	0.0681, 0.1257	0.0821, 0.1392	0.1214, 0.1639
Δρ (max, min) ($e\ Å^{-3}$)	0.359, -0.294	0.362, -0.278	0.257, -0.280	0.211, -0.258	0.211, -0.214	0.182, -0.215
HAR						
Chi2 final	1.384186	1.401731	1.971126	2.404034	3.167608	3.934570
Max. shift (esd)	0.004283	0.003376	0.003874	0.002829	0.001562	0.005373
Final R indices [I>2sigma(I)] (R_1, wR2)	0.0241, 0.0251	0.0286, 0.0275	0.0282, 0.0241	0.0329, 0.0229	0.0373, 0.0239	0.0498, 0.0279
Δρ (max, min) ($e\ Å^{-3}$)	0.1759, -0.2009	0.1881, -0.2216	0.2130, -0.2501	0.2016, -0.2232	0.2117, -0.2220	0.2194, -0.2659

Table S8 Crystallographic data for **8** (isonicotinamide 3,5-dinitrobenzoic acid 1:1) over 100 to 350 K temperature range collected on beamline I19 at Diamond Light Source, U.K.

Formula	$C_{13}H_{10}N_4O_7$	$C_{13}H_{10}N_4O_7$	$C_{13}H_{10}N_4O_7$	$C_{13}H_{10}N_4O_7$
Mol. W ($gmol^{-1}$)	334.25	334.25	334.25	334.25
T (K)	100	200	300	300
Space group	P-1	P-1	P-1	P-1
a (Å)	7.24910(3)	7.33360(4)	7.44850(5)	7.52240(7)
b (Å)	9.00520(4)	9.01180(4)	9.01200(5)	9.00890(6)
c (Å)	10.79980(4)	10.81310(5)	10.83100(6)	10.84300(8)
α (°)	87.8510(4)	87.9490(4)	88.0350(5)	88.0720(7)
β (°)	76.1640(3)	76.4520(4)	76.7370(5)	76.8850(7)
γ (°)	86.9090(3)	87.1840(4)	87.5550(5)	87.7960(7)
Volume ($Å^3$)	683.334(5)	693.685(6)	706.780(7)	714.884(10)
Z	2	2	2	2
ρ_{calc} ($Mg\ m^{-3}$)	1.624	1.600	1.571	1.553
μ (mm^{-1})	0.083	0.082	0.080	0.080
Θ range (°)	1.883-36.085	1.878-36.015	1.873-36.113	1.870-36.157
Reflections collected	15124	23674	24234	24632
Independent reflections	6229	6677	6827	6918
R_{int}	0.0237	0.0292	0.0288	0.0268
Completeness (%)	98.9	99.8	99.8	99.9
Parameters	257	257	257	257
SHELXL				
GooF	1.035	1.031	1.054	1.056
Final R indices [I>2sigma(I)] (R_1, wR2)	0.0360, 0.1040	0.0434, 0.1246	0.0509, 0.1491	0.0544, 0.1639
Final R indices (all data) (R_1, wR2)	0.0390, 0.1080	0.0522, 0.1351	0.0720, 0.1686	0.0872, 0.1935
Δρ (max, min) ($e\ Å^{-3}$)	0.58, -0.37	0.52, -0.303	0.391, -0.241	0.316, -0.230

Table S9 Selected structural parameters in **5** – **8**: <CONC is the torsion angle between the acid co-former and isonicotinamide (IN) molecule; <OCCC is the torsion angle of the amide group to *N*-heterocyclic ring in the IN molecule.

System	Interaction	dD…A acid:IN (Å)	dD…A IN:IN (Å)	<CONC (°)	<OCCC (°)
5	N3—H3···O3	2.5499(10)	2.992(1)	-85.41(9)	25.8(1)
	O1—H1···N1	2.6955(10)	2.834(1)	-4.06(8)	-1.1(1)
6	N1—H1···O1	2.6347(12)	2.8414(8)	28.59(8)	10.4(1)
7	N2—H1···O9	2.6347(12)	2.949(1)	0.5(1)	-18.0(2)
	N1—H2···O5	2.5542(12)	2.864(1)	-5.4(1)	18.5(2)
8	N1—H1···O1	2.5350(13)	2.9337(13)	29.3(2)	-24.5(2)

Table S10 The $O1^{+}$—H1···$O2^{-}$ SSHB parameters in **2** over the 100 to 350 K temperature range.

	dD—H (Å)		dH···A (Å)		dD···A (Å)		<DHA (°)	
T (K)	SHELXL	HAR	SHELXL	HAR	SHELXL	HAR	SHELXL	HAR
100	1.10(2)	1.147(8)	1.36(2)	1.306(8)	2.4435(10)	2.4418(6)	168.6(18)	169.1(9)
150	1.10(2)	1.133 (10)	1.36(2)	1.321(9)	2.4448(11)	2.4444(6)	169(2)	170(1)
200	1.17(2)	1.149(8)	1.29(2)	1.306(8)	2.4477(11)	2.4452(7)	169(2)	170(1)
250	1.21(3)	1.19(3)	1.26(2)	1.28(3)	2.4496(13)	2.446(2)	164(2)	154(4)
300	1.21(3)	1.28(4)	1.26(3)	1.20(4)	2.4502(14)	2.450(1)	166(2)	164(3)
350	1.34(3)	1.39(4)	1.15(3)	1.10(4)	2.4543(17)	2.452(2)	162(3)	159(3)

Table S11 The $N3^{+}$—H3···$O3^{-}$ SSHB parameters in **5** between 100 and 350 K.

	dD—H (Å)		dH···A (Å)		dD···A (Å)		<DHA (°)	
T (K)	SHELXL	HAR	SHELXL	HAR	SHELXL	HAR	SHELXL	HAR
100	1.102(18)	1.141(8)	1.449(18)	1.411(8)	2.5499(10)	2.5513(5)	176.7(16)	175.9(7)
150	1.115(18)	1.144(8)	1.435(18)	1.407(8)	2.5490(10)	2.5504(5)	176.9(16)	175.9(7)
200	1.115(19)	1.136(8)	1.436(19)	1.415(8)	2.5495(10)	2.5497(5)	176.6(17)	176.2(7)
250	1.14(2)	1.148(9)	1.41(2)	1.40(1)	2.5490(11)	2.5493(6)	176.0(18)	175.8(8)
300	1.17(2)	1.147(10)	1.38(2)	1.40(1)	2.5501(12)	2.5494(7)	175.3(18)	176.6(9)
350	1.19(2)	1.178(14(	1.37(2)	1.38(1)	2.5536(14)	2.5524(9)	175(2)	176(1)

Table S12 Bond distances of the groups forming the $O1^{+}$—H1···$O2^{-}$ SSHB over the temperature range in **2**.

T (K)	dC1=$O1_{DMU}$ (Å)	dC3—$O2^{-}_{2,4\text{-DNBA}}$ (Å)	dC4=$O3_{2,4\text{-DNBA}}$ (Å)
100	1.2940(12)	1.2723(12)	1.2359(12)
150	1.2934(13)	1.2707(13)	1.2346(12)
200	1.2904(13)	1.2670(13)	1.2346(13)
250	1.2878(15)	1.2648(15)	1.2323(14)
300	1.2832(17)	1.2611(17)	1.2300(16)
350	1.277(2)	1.260(2)	1.2250(19)

Table S13 Bond distances of the groups forming the $N3^{+}$—H3···$O3^{-}$ SSHB over the temperature range in **5**.

T (K)	dC12—N3 (Å)	dC13—N3 (Å)	<C13N1C12 (°)	dC2—$O3^{-}$ (Å)	dC2=O4 (Å)
100	1.3429(11)	1.3438(12)	121.06(8)	1.2913(11)	1.2391(11)
150	1.3420(12)	1.3418(13)	120.99(8)	1.2886(11)	1.2381(11)
200	1.3386(13)	1.3390(14)	120.89(9)	1.2867(13)	1.2365(12)
250	1.3375(15)	1.3362(16)	120.7(1)	1.2864(14)	1.2341(13)
300	1.3334(16)	1.3329(18)	120.5(1)	1.2847(16)	1.2313(15)
350	1.3299(18)	1.329(2)	120.2(1)	1.2843(18)	1.2281(17)

Table S14 The $N1^{+}$—H2···$O5^{-}$ SSHB parameters in **7** between 100 and 350 K.

	dD—H (Å)		dH···A (Å)		dD···A (Å)		<DHA (°)	
T (K)	SHELXL	HAR	SHELXL	HAR	SHELXL	HAR	SHELXL	HAR
100	1.05(2)	1.205(15)	1.50(2)	1.35(1)	2.5542(12)	2.5534(8)	178(2)	175(1)
150	1.08(2)	1.210(17)	1.48(2)	1.35(1)	2.5553(13)	2.5542(9)	177(2)	172(1)
200	1.07(2)	1.207(17)	1.49(2)	1.36(1)	2.5596(14)	2.5579(9)	176(2)	172(1)
250	1.08(3)	1.23(2)	1.48(3)	1.34(2)	2.5616(15)	2.560(1)	176(2)	171(2)
300	1.10(3)	1.24(2)	1.47(3)	1.33(2)	2.5664(17)	2.564(2)	174(2)	169(2)
350	1.12(4)	1.26(4)	1.45(4)	1.31(3)	2.570(2)	2.567(2)	175(3)	173(3)

Table S15 The $N1^{+}$—H1···$O1^{-}$ SSHB parameters over the 100 to 350 K temperature range in **8**.

T (K)	dD—H (Å)	dH···A (Å)	dD···A (Å)	<DHA (°)
100	1.023(14)	1.543(14)	2.5368(7)	162.3(13)
200	1.034(16)	1.524(17)	2.5354(9)	164.4(16)
300	1.07(2)	1.49(2)	2.5326(11)	164.4(19)
350	1.09(2)	1.48(2)	2.5310(13)	160(2)

Table S16 Bond distances of the groups forming the $N1^{+}$—H2···$O5^{-}$ SSHB over the temperature range in **7**.

T (K)	dC19—N1 (Å)	dC14—N1 (Å)	<CNC (°)	dC—O^{-} (Å)	dC=O (Å)
100	1.3377(15)	1.3352(15)	120.7(1)	1.269(1)	1.240(1)
150	1.3353(17)	1.3316(18)	120.3(1)	1.266(2)	1.238(1)
200	1.3285(18)	1.3265(18)	120.5(1)	1.2641(16)	1.2377(16)
250	1.323(2)	1.325(2)	120.0(2)	1.2628(18)	1.2356(17)
300	1.321(2)	1.315(2)	119.9(2)	1.259(2)	1.233(2)
350	1.313(3)	1.309(3)	119.8(2)	1.260(3)	1.230(3)

Table S17 Bond distances of the groups forming the $N1^{+}$—H1···$O1^{-}$ SSHB over the temperature range in **8**.

T (K)	dC12—N1 (Å)	dC8—N1 (Å)	<C12N1C8 (°)	dC1—$O1^{-}$ (Å)	dC1=O2 (Å)
100	1.3382(8)	1.3408(8)	121.50(6)	1.2674(8)	1.2409(8)
200	1.3340(10)	1.3374(10)	121.46(7)	1.2638(10)	1.2342(10)
300	1.3306(14)	1.3302(14)	121.2 (1)	1.2588(13)	1.2242(14)
350	1.3256(16)	1.3268(17)	120.9(1)	1.2560(15)	1.2184(16)

Table S18 The O1—H1···O2 SSHB parameters in **3** over the 100 to 350 K temperature range.

T (K)	dD—H (Å)	dH···A (Å)	dD···A (Å)	<DHA (°)
100	1.22(7)	1.26(7)	2.469(3)	172(6)
150	1.23(6)	1.24(6)	2.466(3)	176(6)
200	1.05(7)	1.43(7)	2.470(3)	175(6)
250	1.21(7)	1.27(7)	2.472(3)	168(6)
300	1.15(7)	1.34(7)	2.468(4)	162(6)

Table S19 The O—H···O SSHB parameters in **1** over the 100 to 300 K temperature range.

T (K)	D—H···A	dD—H (Å)	dH···A (Å)	dD···A (Å)	<DHA (°)
100	O3—H1···O6	1.09(3)	1.44(3)	2.4925(16)	160(3)
	O2—H2···O5	1.01(3)	1.46(3)	2.4520(16)	167(2)
200	O3—H1···O6	1.11(3)	1.42(3)	2.4984(16)	162(2)
	O2—H2···O5	1.06(3)	1.41(3)	2.4606(16)	168(3)
300	O3—H1···O6	1.11(3)	1.42(3)	2.4954(19)	159(2)
	O2—H2···O5	1.05(3)	1.45(3)	2.472(2)	164(2)

Table S20 The O—H···O SSHB parameters in **4** over the 100 to 300 K temperature range.

T (K)	D—H···A	dD—H (Å)	dH···A (Å)	dD···A (Å)	<DHA (°)
100	O8—H13···O14	1.03(2)	1.45(2)	2.4640(11)	167(2)
	O1—H1···O7	1.06(2)	1.39(2)	2.4445(12)	170(2)
200	O8—H13···O14	1.00(2)	1.48(2)	2.4687(13)	172(2)
	O1—H1···O7	1.04(2)	1.42(2)	2.4500(1)	167(2)
300	O8—H13···O14	0.94(3)	1.55(3)	2.4832(16)	174(3)
	O1—H1···O7	0.97(3)	1.51(3)	2.4596(18)	166(2)

Table S21 The $N1^{+}$—H1···$O1^{-}$ SSHB parameters over the 100 to 350 K temperature range in **6**.

T (K)	dD—H (Å)	dH···A (Å)	dD···A (Å)	<DHA (°)
100	1.046(18)	1.572(18)	2.6000(8)	166.2(15)
200	1.02(2)	1.59(2)	2.5968(10)	168.9(17)
300	0.99(2)	1.63(2)	2.5941(12)	163.2(18)

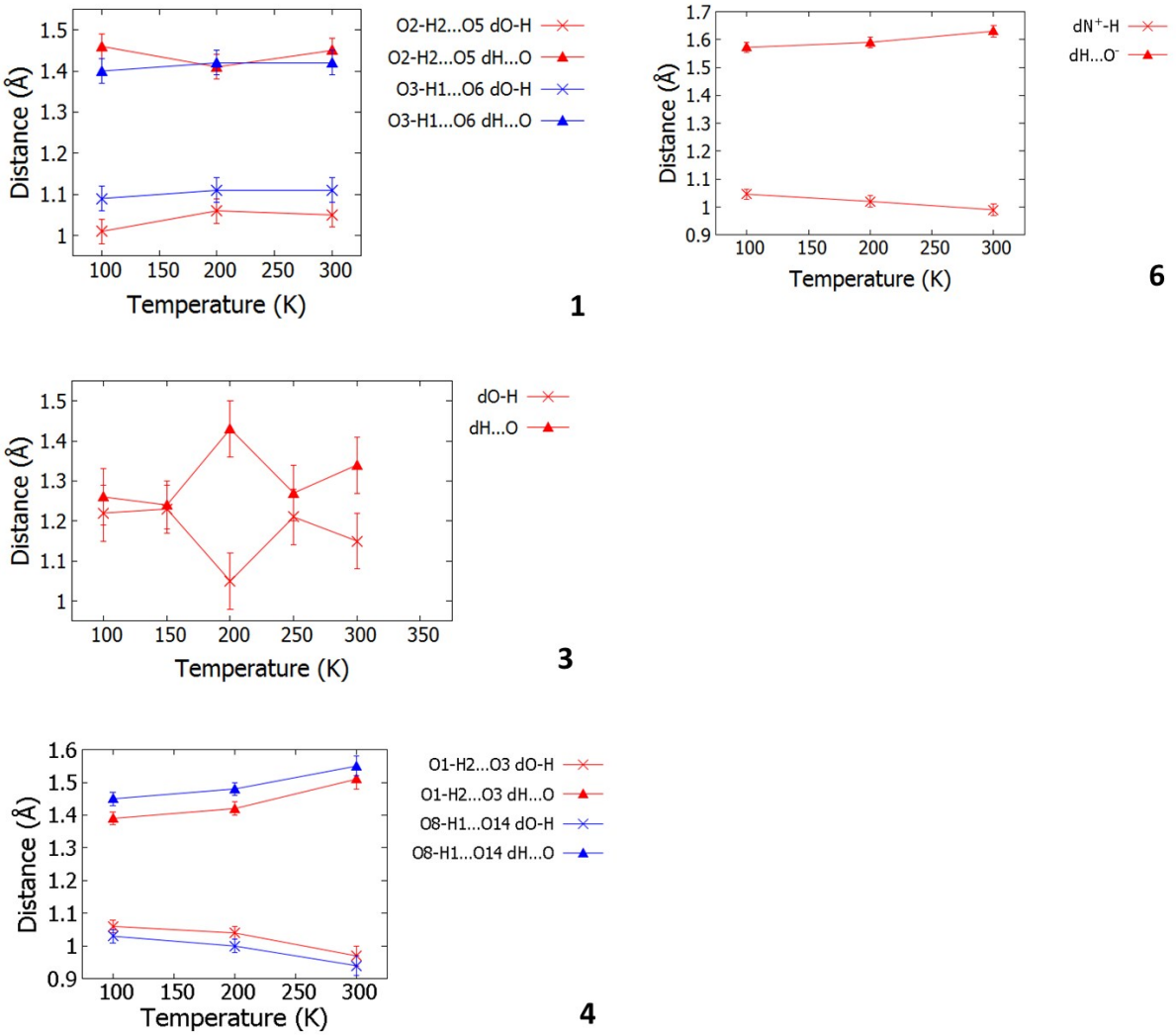

Fig. S1 The D—H and H···A distances over the 100 to 350 K temperature range in **1**, **3**, **4** and **6**.

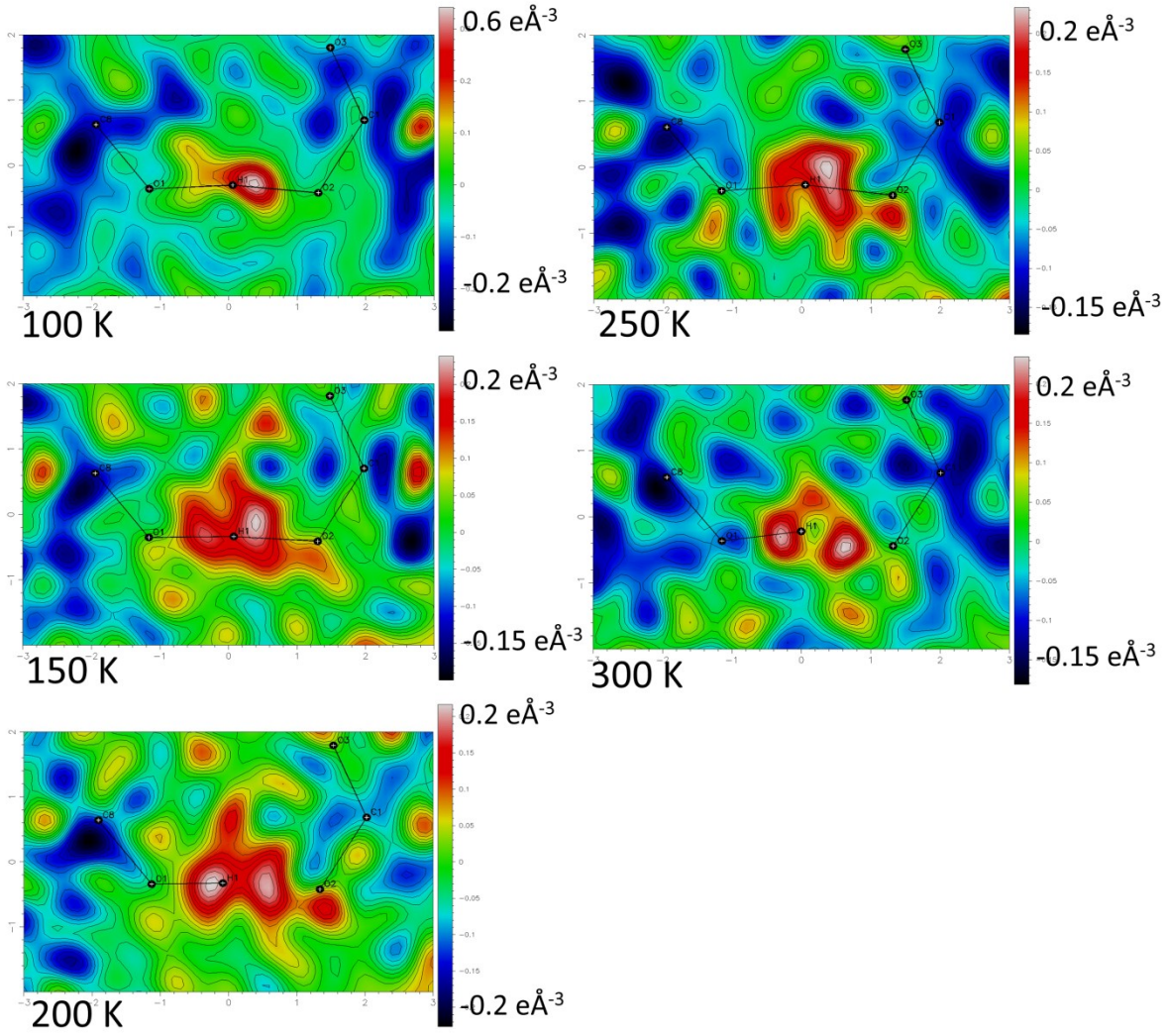

Fig. S2 Fourier difference electron density maps showing the electron density associated with the H-atom in the O1—H1···O2 SSHB in **3** over the 100 to 300 K temperature range.

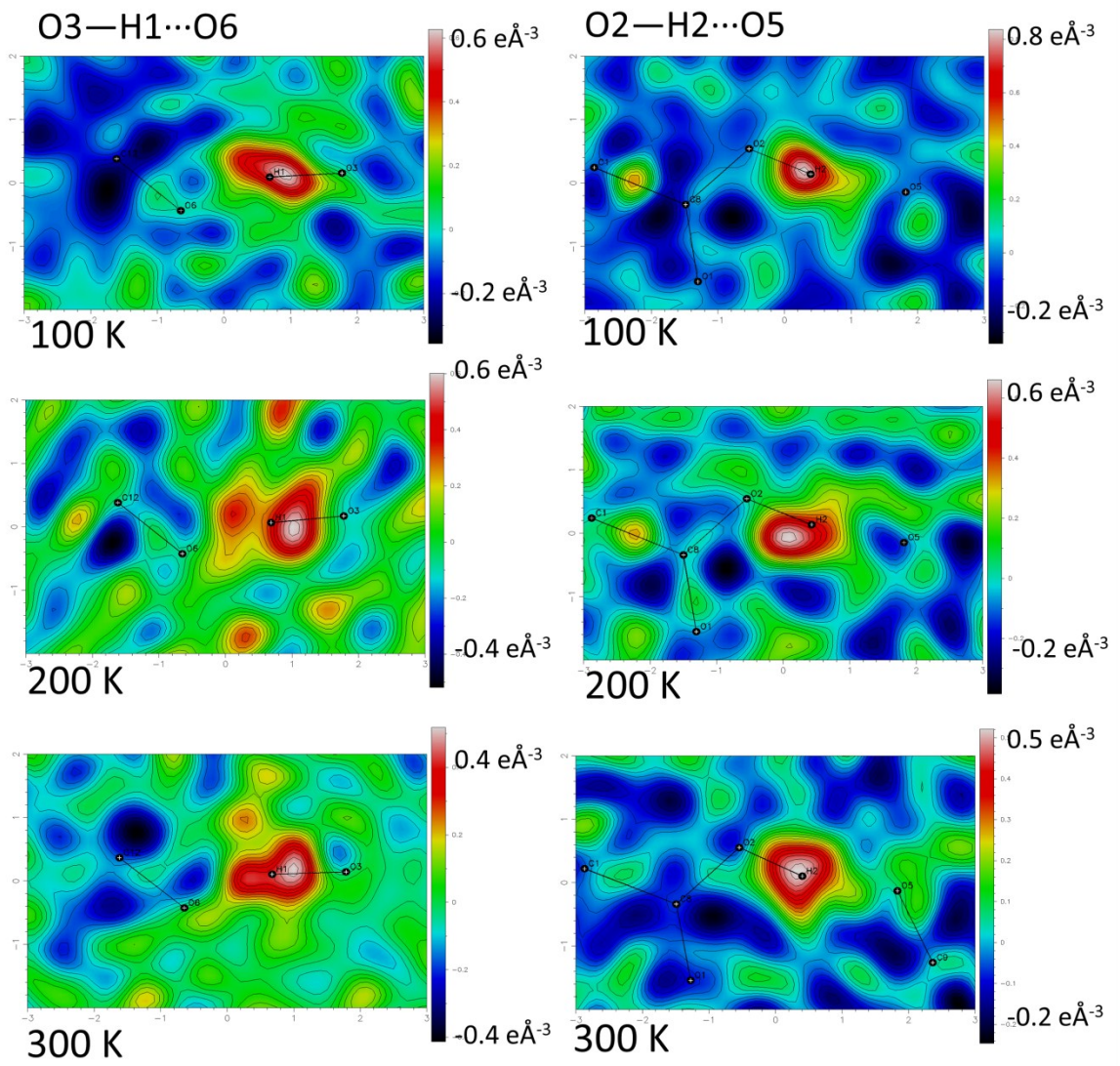

Fig. S3 Fourier difference electron density maps showing the electron density associated with the H-atom in the O3—H1···O6 and O2—H2···O5 SSHBs in **1** over the 100 to 300 K temperature range.

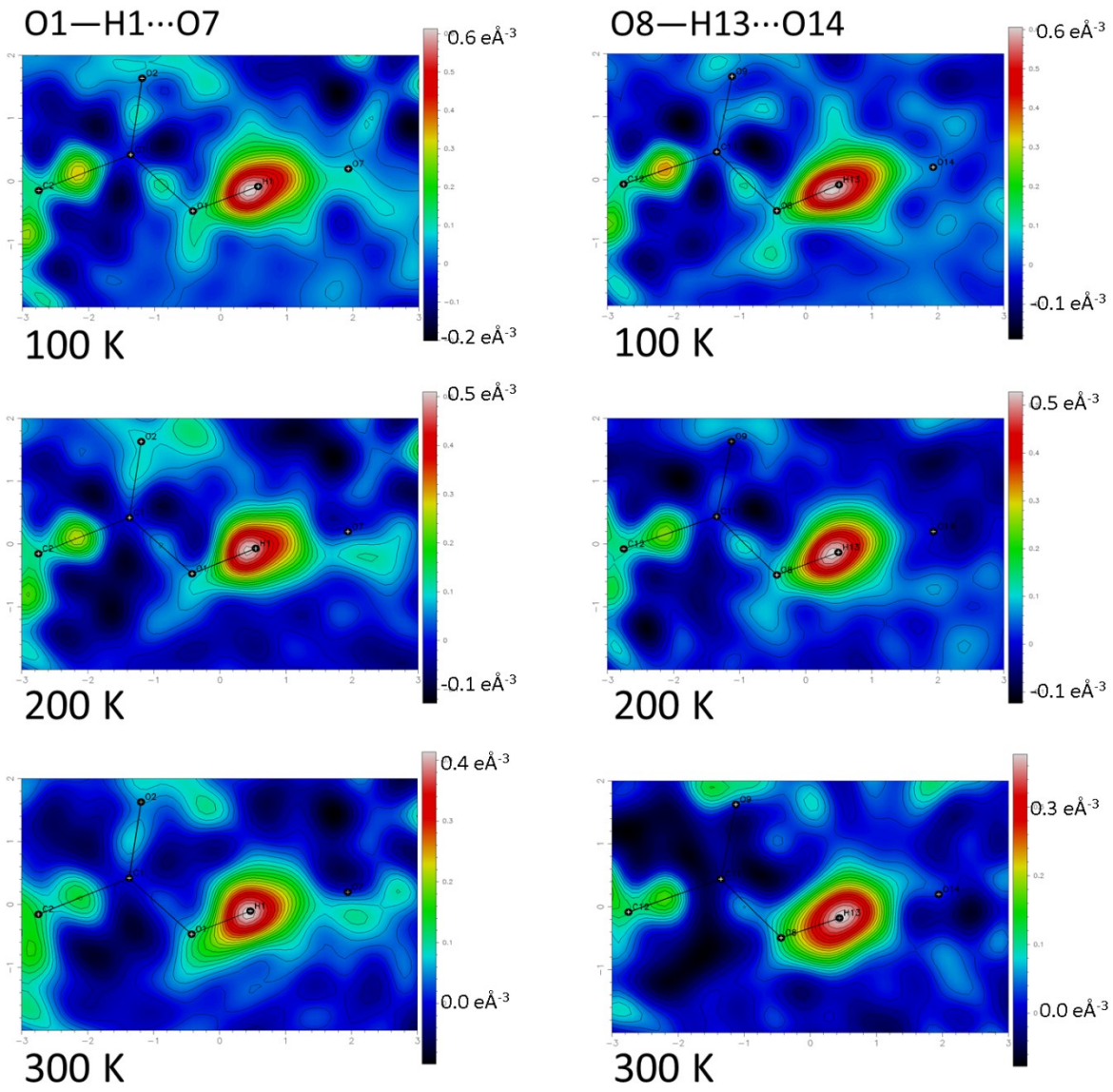

Fig. S4 Fourier difference electron density maps showing the electron density associated with the H-atom in the O1—H1···O7 and O8—H13···O14 SSHB in **4** over the 100 to 300 K temperature range.

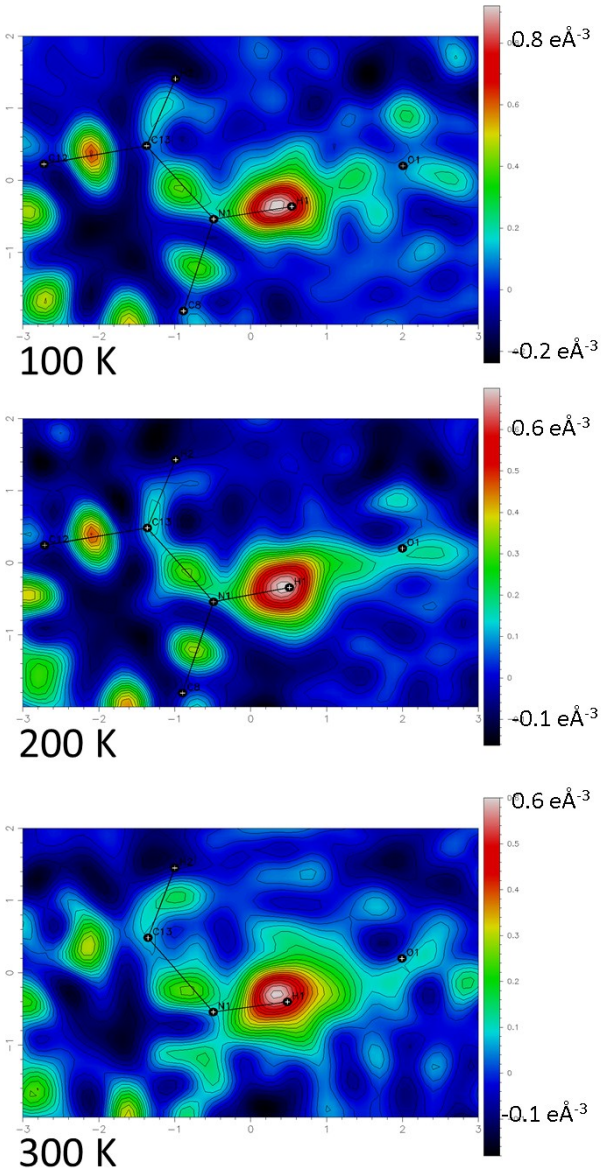

Fig. S5 Fourier difference electron density maps showing the electron density associated with the H-atom in the $N1^{+}$—H1···$O1^{-}$ SSHB in **6** over the 100 to 300 K temperature range.