Supplementary materials

New series of 3,5-diamino-1,2,4-triazolium(1+) inorganic salts and their potential in crystal engineering of novel NLO materials

Irena Matulková[a,b], Jaroslav Cihelka[b], Michaela Pojarová[b], Karla Fejfarová[b], Michal Dušek[b], Přemysl Vaněk[b], Jan Kroupa[b], Radmila Krupková[b], Jan Fábry[b], Ivan Němec[a]

[a]*Faculty of Science, Department of Inorganic Chemistry, Charles University in Prague, Hlavova 8, 128 40 Prague 2, Czech Republic*
[b]*Institute of Physics ASCR, v.v.i., Na Slovance 2, 182 21 Prague 8, Czech Republic*

* Corresponding author. Phone: +420 221 951 247; Fax: +420 221 951 253

E-mail address: irena.mat@atlas.cz

TABLES

Table 1S. Experimental powder data for **dat$_2$SeO$_4$2H$_2$O.**

2 Theta (°)	d (Å)	Intensity (a.u.)	2 Theta (°)	d (Å)	Intensity (a.u.)
10.05	8.80	1316.61	26.90	3.31	1735.16
13.96	6.34	20172.35	27.67	3.22	4260.94
15.57	5.69	2751.54	28.09	3.17	19840.11
15.75	5.62	2465.01	28.17	3.16	15016.86
16.29	5.44	6655.41	28.27	3.15	10287.42
18.53	4.79	2430.93	29.37	3.04	2980.08
18.81	4.71	3194.73	29.66	3.01	1581.87
19.05	4.66	3182.11	29.88	2.99	1204.31
19.46	4.56	1442.06	30.60	2.92	1392.48
20.23	4.39	4846.15	30.81	2.90	1648.96
21.03	4.22	1556.26	31.50	2.84	2370.27
21.60	4.11	2118.96	33.70	2.66	1222.57
22.94	3.87	873.44	34.28	2.61	1254.37
23.32	3.81	893.88	35.89	2.50	930.67
23.94	3.71	2599.96	37.33	2.41	1182.47
26.07	3.42	1289.66	57.21	1.61	993.15

Table 2S. Experimental powder data for **dat$_2$SO$_4$2H$_2$O.**

2 Theta (°)	d (Å)	Intensity (a.u.)	2 Theta (°)	d (Å)	Intensity (a.u.)
14.17	6.25	20100.00	27.94	3.19	3361.87
15.97	5.55	2949.50	28.45	3.13	43118.56
16.58	5.34	16007.78	29.80	3.00	3037.67
19.02	4.66	2259.90	31.05	2.88	1685.16
19.43	4.56	2426.42	31.17	2.87	1957.88
20.80	4.27	7939.40	32.09	2.79	3106.73
21.24	4.18	1567.67	33.98	2.64	1959.80
21.80	4.07	1708.62	42.19	2.14	1143.10
23.44	3.79	2044.32			

Table 3S. Experimental powder data for **datClO$_4$**.

2 Theta (°)	d (Å)	Intensity (a.u.)	2 Theta (°)	d (Å)	Intensity (a.u.)
9.86	3.72	4582.41	27.00	3.30	3042.63
17.58	5.04	7136.91	27.07	3.29	1805.62
19.74	4.49	13407.70	27.83	3.20	45409.19
20.27	4.38	3083.65	27.91	3.19	20140.00
21.72	4.09	3786.32	28.73	3.10	2898.17
22.88	3.88	2520.75	29.76	3.00	1373.69
23.71	3.75	6617.72	40.88	2.21	1259.92
24.04	3.70	2712.90	40.99	2.20	791.10
26.75	3.33	4347.39			

Table 4S. Experimental powder data for **datNO$_3$**.

2 Theta (°)	d (Å)	Intensity (a.u.)
23.88	3.72	2963.98
24.15	5.04	11499.83
24.50	4.49	8386.61
28.43	4.38	325675.75
28.51	4.09	148280.00

Table 5S. Experimental powder data for **dat$_2$Cl$_2$H$_2$O**.

2 Theta (°)	d (Å)	Intensity (a.u.)	2 Theta (°)	d (Å)	Intensity (a.u.)
6.53	13.53	501.10	35.17	2.55	631.14
10.66	8.29	528.54	35.50	2.53	445.52
11.41	7.75	816.15	35.70	2.51	1145.77
12.42	7.12	580.15	36.25	2.48	337.06
12.98	6.81	729.84	36.73	2.45	506.42
13.58	6.52	552.63	37.09	2.42	997.44
14.29	6.19	1540.22	37.19	2.42	874.00
14.85	5.96	508.98	38.22	2.35	794.10
15.09	5.87	1439.04	38.32	2.35	488.95
16.31	5.43	484.80	38.66	2.33	381.22
17.94	4.94	1240.86	39.15	2.30	326.45
18.18	4.88	409.20	39.49	2.28	275.06
18.90	4.69	693.65	39.94	2.26	456.94
19.22	4.62	1777.69	41.41	2.18	308.12
20.19	4.39	1288.91	41.68	2.17	387.44
20.60	4.31	1163.30	42.47	2.13	462.54
20.92	4.24	935.85	43.00	2.10	247.53
21.46	4.14	336.12	44.15	2.05	344.32
22.71	3.91	249.45	44.27	2.04	270.10
23.17	3.84	1685.67	44.98	2.01	248.57
23.73	3.75	750.25	45.75	1.98	257.84
24.09	3.69	1026.43	46.34	1.96	473.23
24.24	3.67	1421.11	46.45	1.95	302.93
25.14	3.54	4594.71	46.71	1.94	228.07
25.58	3.48	2751.32	48.41	1.88	244.70
26.06	3.42	4858.17	48.78	1.87	301.28
27.55	3.24	2880.86	48.89	1.86	265.30
28.22	3.16	389.08	50.98	1.79	261.66
29.16	3.06	4975.37	51.50	1.77	351.00
29.50	3.03	1703.54	52.15	1.75	304.64
29.71	3.00	1171.96	52.69	1.74	284.21
30.79	2.90	1398.99	53.14	1.72	302.19
31.44	2.84	761.33	53.27	1.72	233.78
31.52	2.84	852.71	53.71	1.71	206.66
32.38	2.76	1284.26	55.29	1.66	279.75
32.68	2.74	500.52	55.43	1.66	216.56
32.88	2.72	342.46	56.72	1.62	290.77
33.40	2.68	542.60	57.01	1.61	302.00
34.08	2.63	535.76	57.15	1.61	265.88
34.25	2.62	1147.42	58.76	1.57	454.78
34.55	2.59	440.26	58.92	1.57	383.99
34.97	2.56	382.15			

Table 6S. Experimental powder data for **datH$_2$PO$_3$**.

2 Theta (°)	d (Å)	Intensity (a.u.)	2 Theta (°)	d (Å)	Intensity (a.u.)
18.83	4.71	3875.83	25.91	3.44	1842.00
19.06	4.65	9195.69	29.63	3.01	9555.79
21.52	4.13	26799.41	29.70	3.01	4935.00
22.88	3.88	3671.35	31.05	2.88	3878.24
24.31	3.66	140585.20	31.13	2.87	1986.68
24.38	3.65	65822.20	32.49	2.75	3686.88
25.53	3.49	3890.10	32.58	2.75	2000.00

Table 7S. Experimental powder data for **datH$_2$PO$_4$**.

2 Theta (°)	d (Å)	Intensity (a.u.)	2 Theta (°)	d (Å)	Intensity (a.u.)
9.63	9.17	2441.12	29.90	2.99	1148.00
14.02	6.31	3377.22	30.19	2.96	2354.63
16.30	5.43	7762.22	31.10	2.87	1189.14
17.60	5.04	2859.25	32.85	2.72	2158.05
19.23	4.61	8841.91	32.94	2.72	2175.09
19.53	4.54	968.43	33.05	2.71	1265.24
20.06	4.42	6034.53	35.99	2.49	1354.80
21.42	4.14	1141.05	36.48	2.46	963.00
23.32	3.81	9861.90	38.91	2.31	1736.92
24.76	3.59	5130.52	39.97	2.25	1053.37
25.70	3.46	828.42	40.30	2.24	911.75
26.42	3.37	3337.84	46.09	1.97	1197.44
26.87	3.32	16973.42	50.08	1.82	889.74
27.49	3.24	2930.35	51.11	1.79	842.08
28.15	3.17	23993.14	54.52	1.68	1156.17
29.84	2.99	1556.88			

Table 8S. Selected bond lengths (Å) and angles (°) for **$dat_2SeO_4 2H_2O$.**

Bond/Angle	Value	Angle	Value
Se1-O1	1.645(1)	O2-Se1-O4	108.29(7)
Se1-O2	1.639(1)	O3-Se1-O4	108.88(6)
Se1-O3	1.637(1)	N1-N2-C3	103.2(1)
Se1-O4	1.641(1)	N1-C5-N4	106.9(2)
O1W-H1W	0.82(2)	N1-C5-N6	126.7(2)
O1W-H2W	0.82(1)	N2-N1-C5	111.4(2)
O2W-H3W	0.81(2)	N2-C3-N4	112.0(2)
O2W-H4W	0.81(2)	N2-C3-N7	126.9(2)
N1-C5	1.321(2)	N4-C3-N7	121.2(2)
N2-N1	1.410(2)	N4-C5-N6	126.4(2)
N2-C3	1.304(3)	N8-N9-C10	103.1(1)
N4-C3	1.382(2)	N8-C12-N11	106.6(2)
N4-C5	1.350(2)	N8-C12-N13	127.1(2)
N6-C5	1.328(2)	N9-N8-C12	111.7(2)
N6-H6A	0.87(2)	N9-C10-N11	111.8(2)
N6-H6B	0.87(1)	N9-C10-N14	126.7(2)
N7-C3	1.339(2)	N11-C10-N14	121.5(2)
N7-H7A	0.87(2)	N11-C12-N13	126.3(2)
N7-H7B	0.86(1)	C3-N7-H7B	123(2)
N8-C12	1.321(2)	C3-N7-H7A	118(1)
N9-N8	1.410(2)	C5-N4-C3	106.6(2)
N9-C10	1.308(3)	C5-N6-H6A	120(1)
N11-C12	1.351(2)	C5-N6-H6B	122(2)
N11-C10	1.383(2)	C10-N14-H14A	119(1)
N13-C12	1.329(2)	C10-N14-H14B	120(2)
N13-H13A	0.87(2)	C12-N11-C10	106.9(2)
N13-H13B	0.88(1)	C12-N13-H13A	121(1)
N14-C10	1.344(2)	C12-N13-H13B	118(2)
N14-H14A	0.87(2)	H6A-N6-H6B	114(2)
N14-H14B	0.86(1)	H7B-N7-H7A	119(2)
O1-Se1-O2	109.46(6)	H13A-N13-H13B	120(2)
O1-Se1-O3	110.05(7)	H14A-N14-H14B	119(2)
O1-Se1-O4	108.95(7)	H1W-O1W-H2W	100(3)
O2-Se1-O3	111.16(7)	H3W-O2W-H4W	102(2)

Hydrogen bonds

D-H…A	d (D-H)	d (A…H)	d (D…A)	<(DHA)
O1W-H1W…O3[a]	0.82(2)	1.96(2)	2.777(2)	175(3)
O1W-H2W…O2W	0.82(1)	2.09(1)	2.880(2)	163(3)
O2W-H3W…O1	0.82(2)	2.02(2)	2.748(2)	148(2)
O2W-H4W…O4[c]	0.81(2)	2.08(2)	2.849(2)	157(2)
N1-H1A…O3	0.87	1.88	2.734(2)	165
N4-H4A…O2[b]	0.87	1.90	2.698(2)	151
N6-H6A…N2[d]	0.87(2)	2.18(2)	3.028(2)	167(2)
N6-H6B…Se1[d]	0.87(1)	2.92(2)	3.650(2)	142(2)
N6-H6B…O1[d]	0.87(1)	2.06(1)	2.926(2)	175(2)
N7-H7A…O1W[e]	0.88(2)	2.29(2)	3.040(2)	144(2)
N7-H7B…O2[f]	0.86(1)	2.02(1)	2.879(2)	177(2)
N8-H8A…O2W[g]	0.87	1.90	2.734(2)	160
N11-H11A…O4	0.87	1.93	2.720(2)	150
N13-H13A…N9[d]	0.87(2)	2.21(2)	3.073(2)	171(2)
N13-H13B…O1W[g]	0.88(1)	2.07(1)	2.908(2)	161(2)
N14-H14A…O1	0.87(2)	2.14(2)	3.000(2)	177(2)
N14-H14B…O4[a]	0.86(1)	2.05(1)	2.914(2)	174(2)

Note.
Equivalent positions: [a] -1+x, y, z, -1/2+z; [b] 2-x, -y, 1-z, 1/4+z, -1/2-z; [c] x, 1+y, z; [d] 1+x, y, z; [e] 1-x, 1-y, 1-z; [f] 1-x, -y, 1-z; [g] 1-x, 1-y, 2-z.
Abbreviations: A, acceptor; D, donor.

Table 9S. Selected bond lengths (Å) and angles (°) for **$dat_2SO_4 2H_2O$.**

Bond/Angle	Value	Angle	Value
S1-O1	1.476(1)	N1-N2-C3	103.3(1)
S1-O2	1.472(1)	N1-C5-N4	106.4(2)
S1-O3	1.474(1)	N1-C5-N6	127.3(2)
S1-O4	1.479(1)	N2-N1-C5	111.4(1)
O1W-H1W	0.87(3)	N2-C3-N4	111.8(2)
O1W-H2W	0.85(3)	N2-C3-N7	126.7(2)
O2W-H3W	0.78(3)	N2-N1-H1A	120.4(2)
O2W-H4W	0.84(2)	N4-C3-N7	121.6(2)
N1-H1A	0.84(2)	N4-C5-N6	126.3(2)
N1-C5	1.322(2)	N8-N9-C10	103.3(2)
N2-N1	1.410(2)	N8-C12-N11	105.7(2)
N2-C3	1.305(2)	N8-C12-N13	127.6(2)
N4-H4A	0.87(2)	N9-N8-H8A	118(1)
N4-C3	1.375(2)	N9-N8-C12	112.1(1)
N4-C5	1.354(2)	N9-C10-N11	111.5(2)
N6-H6A	0.91(2)	N9-C10-N14	127.1(2)
N6-H6B	0.82(2)	N11-C10-N14	121.4(2)
N6-C5	1.322(3)	N11-C12-N13	126.7(2)
N7-H7A	0.87(2)	C3-N4-H4A	122(2)
N7-H7B	0.88(3)	C3-N7-H7B	118(1)
N7-C3	1.335(3)	C3-N7-H7A	121(2)
N8-H8A	0.87(2)	C5-N4-C3	107.0(1)
N8-C12	1.325(2)	C5-N1-H1A	128(2)
N9-N8	1.405(2)	C5-N4-H4A	128(2)
N9-C10	1.306(2)	C5-N6-H6A	122(2)
N11-C12	1.355(2)	C5-N6-H6B	120(1)
N11-C10	1.381(2)	C10-N11-H11A	123(2)
N11-H11A	0.86(2)	C10-N14-H14A	123(2)
N13-C12	1.318(3)	C10-N14-H14B	120(2)
N13-H13A	0.81(2)	C12-N11-C10	107.4(1)
N13-H13B	0.90(2)	C12-N8-H8A	129(1)
N14-C10	1.333(3)	C12-N11-H11A	129(2)
N14-H14A	0.86(3)	C12-N13-H13A	126(2)
N14-H14B	0.90(2)	C12-N13-H13B	119(1)
O1-S1-O2	110.07(7)	H6A-N6-H6B	116(2)
O1-S1-O3	109.28(8)	H7B-N7-H7A	121(2)
O1-S1-O4	109.11(7)	H13A-N13-H13B	113(2)
O2-S1-O3	110.38(7)	H14A-N14-H14B	116(2)
O2-S1-O4	109.06(7)	H1W-O1W-H2W	99(2)
O3-S1-O4	108.91(7)	H3W-O2W-H4W	100(2)

Hydrogen bonds

D-H…A	d (D-H)	d (A…H)	d (D…A)	<(DHA)
O1W-H1W…O2	0.87(2)	1.91(2)	2.774(2)	174(2)
O1W-H2W…O2W[a]	0.85(2)	2.03(2)	2.871(2)	167(2)
O2W-H3W…O1	0.78(2)	2.01(2)	2.767(2)	163(2)
O2W-H4W…O4[c]	0.84(2)	2.10(3)	2.866(2)	152(2)
N1-H1A…O2	0.84(2)	1.92(2)	2.750(2)	170(2)
N4-H4A…O3[b]	0.87(2)	1.86(2)	2.712(2)	166(2)
N6-H6A…N2[a]	0.82(2)	2.18(2)	2.997(2)	172(2)
N6-H6B…O1[a]	0.91(2)	2.01(2)	2.911(2)	169(2)
N7-H7A…O1W[d]	0.87(2)	2.28(2)	3.014(2)	142(2)
N7-H7B…O3[e]	0.88(2)	2.00(2)	2.878(2)	171(2)
N8-H8A…O2W[a]	0.87(2)	1.90(2)	2.756(2)	169(2)
N11-H11A…O4[f]	0.86(2)	1.90(2)	2.728(2)	162(2)
N13-H13A…N9[g]	0.81(2)	2.23(2)	3.039(2)	179(2)
N13-H13B…O1W	0.90(2)	2.01(2)	2.875(2)	161(2)
N14-H14A…O1[f]	0.91(2)	2.07(2)	2.962(2)	168(2)
N14-H14B…O4[h]	0.86(2)	2.05(2)	2.912(2)	175(2)

Note.
Equivalent positions: [a] -1+x, y, z, -1/2+z; [b] 2-x, -y, 1-z; [c] x, -1+y, z; [d] -x, -y, -z; [e] 1-x, 1-y, -z; [f] 1-x, 1-y, 1-z; [g] 1+x, y, z; [h] -x, 1-y, 1-z.
Abbreviations: A, acceptor; D, donor.

Table 10S. Selected bond lengths (Å) and angles (°) for **datClO$_4$**.

Bond/Angle	Value	Angle	Value
Cl1-O1	1.442(2)	O3-Cl1-O2	109.1(1)
Cl1-O2	1.444(2)	O4-Cl1-O1	109.87(9)
Cl1-O3	1.439(2)	O4-Cl1-O2	109.65(9)
Cl1-O4	1.431(2)	O4-Cl1-O3	110.11(9)
N1-N2	1.397(2)	N1-C5-N4	106.0(2)
N1-C5	1.317(3)	N1-C5-N6	127.9(2)
N2-C3	1.307(3)	N2-C3-N4	111.4(2)
N4-C3	1.373(3)	N2-C3-N7	125.7(2)
N4-C5	1.346(3)	N6-C5-N4	126.1(2)
C3-N7	1.344(3)	N7-C3-N4	122.8(2)
C5-N6	1.323(3)	C3-N2-N1	103.2(2)
O1-Cl1-O2	108.75(9)	C5-N1-N2	112.1(2)
O3-Cl1-O1	109.35(9)	C5-N4-C3	107.4(2)

Hydrogen bonds

D-H…A	d (D-H)	d (A…H)	d (D…A)	<(DHA)
N1-H1A…O4[a]	0.79(3)	2.36(3)	3.008(3)	139(2)
N1-H1A…O3[b]	0.79(3)	2.46(3)	3.004(2)	128(2)
N4-H4A…O2	0.78(3)	2.12(2)	2.896(2)	173(3)
N6-H6A…O1	0.82(3)	2.23(3)	3.003(3)	156(3)
N6-H6A…O1[c]	0.82(3)	2.47(3)	2.988(3)	122(2)
N6-H6B…O3[d]	0.78(3)	2.33(3)	2.968(3)	140(3)
N7-H7A…O2[e]	0.81(3)	2.46(3)	3.085(3)	136(2)
N7-H7B…N2[f]	0.79(3)	2.22(3)	2.979(3)	161(3)

Note.
Equivalent positions: [a] -1+x, -1+y, z; [b] -x, 1-y, 2-z, -1/2-z; [c] 1-x, 1-y, 2-z; [d] x, -1+y, z; [d] x, -1+y, z; [d] x, -1+y, z; [e] -x, 2-y, 1-z; [f] -1-x, 1-y, 1-z.
Abbreviations: A, acceptor; D, donor.

Table 11S. Selected bond lengths (Å) and angles (°) for **datNO$_3$**.

Bond/Angle	Value	Angle	Value
O1-N3	1.243(2)	O1-N3-O3	120.1(1)
O2-N3	1.258(1)	O3-N3-O2	119.1(1)
O3-N3	1.256(1)	N1-C5-N4	106.6(1)
N1-C5	1.320(2)	N1-C5-N6	128.5(1)
N2-N1	1.402(2)	N2-C3-N4	111.3(1)
N2-C3	1.306(2)	N2-C3-N7	125.9(1)
N4-C3	1.379(2)	N6-C5-N4	124.8(1)
N4-C5	1.349(2)	N7-C3-N4	122.7(1)
N6-C5	1.325(2)	C3-N2-N1	103.6(1)
N7-C3	1.339(2)	C5-N1-N2	111.5(1)
O1-N3-O2	120.8(1)	C5-N4-C3	107.0(1)

Hydrogen bonds

D-H…A	d (D-H)	d (A…H)	d (D…A)	<(DHA)
N1-H1A…O1[a]	0.88(2)	2.03(2)	2.913(2)	176(2)
N4-H4A…O2[b]	0.92(2)	1.91(2)	2.828(2)	177(1)
N6-H6A…O3[b]	0.89(2)	2.10(2)	2.950(2)	161(2)
N6-H6B…O3[c]	0.86(2)	2.06(2)	2.899(2)	166(2)
N7-H7A…N2[b]	0.89(2)	2.09(2)	2.916(2)	156(1)
N7-H7B…O2	0.88(2)	2.06(2)	2.932(2)	176(2)

Note.
Equivalent positions: [a] -x, -1/2+y, -1/2-z; [b] -x, 1/2+y, -1/2-z; [c] -1+x, y, -1+z.
Abbreviations: A, acceptor; D, donor.

Table 12S. Selected bond lengths (Å) and angles (°) for **dat$_2$Cl$_2$H$_2$O**.

Bond/Angle	Value	Angle	Value
N1-N2	1.397(2)	N2-C3-N7	126.1(2)
N9-N8	1.399(2)	N6-C5-N1	126.5(2)
C3-N2	1.300(2)	N6-C5-N4	126.8(2)
C3-N4	1.385(2)	N7-C3-N4	122.0(2)
C3-N7	1.356(2)	N8-C12-N11	106.8(1)
C5-N1	1.321(2)	N8-C12-N13	127.6(2)
C5-N4	1.350(2)	N9-C10-N11	111.3(1)
C5-N6	1.320(2)	N9-C10-N14	125.6(1)
C10-N9	1.313(2)	N13-C12-N11	125.6(2)
C10-N11	1.371(2)	N14-C10-N11	123.2(1)
C10-N14	1.339(2)	C3-N2-N1	103.6(1)
C12-N8	1.315(2)	C5-N1-N2	111.6(1)
C12-N11	1.348(2)	C5-N4-C3	106.3(1)
C12-N13	1.330(2)	C10-N9-N8	103.5(1)
N1-C5-N4	106.7(1)	C12-N8-N9	111.4(1)
N2-C3-N4	111.8(1)	C12-N11-C10	107.0(1)

Hydrogen bonds

D-H…A	d (D-H)	d (A…H)	d (D…A)	<(DHA)
O1W-H1W…Cl1[a]	0.74	2.40	3.126(1)	166
O1W-H2W…Cl2[b]	0.82	2.34	3.148(1)	170
N1-H1A…O1W	0.80	2.06	2.803(2)	153
N4-H4A…Cl1[c]	0.89	2.27	3.154(1)	175
N6-H6A…Cl2[d]	0.87	2.48	3.239(1)	147
N6-H6B…Cl2[e]	0.83	2.42	3.244(2)	169
N7-H7B…N2[a]	0.86	2.22	3.068(2)	169
N8-H8A…O1W[f]	0.80	2.17	2.856(2)	143
N11-H11A…N9[g]	0.78	2.13	2.834(2)	151
N13-H13A…Cl1[g]	0.86	2.68	3.241(1)	124
N13-H13B…Cl2	0.82	2.38	3.153(2)	158
N14-H14A…Cl2[h]	0.86	2.46	3.318(2)	174
N14-H14B…Cl1[c]	0.83	2.47	3.294(2)	174

Note.
Equivalent positions: [a] 1-x, 1-y, 1-z; [b] 1/2-x, -1/2+y, 3/2-z, -*z*; [c] 1+x, y, z, 1-*z*; [d] 3/2+x, 3/2-y, 1/2+z; [e] 3/2-x, -1/2+y, 3/2-z; [e] 3/2-x, -1/2+y, 3/2-z; [f] 3/2-x, 1/2+y, 3/2-z; [g] -1/2+x, 3/2-y, -1/2+z; [h] 1/2+x, 3/2-y, -1/2+z.
Abbreviations: A, acceptor; D, donor.

Table 13S. Selected bond lengths (Å) and angles (°) for **datH$_2$PO$_3$**.

Bond/Angle	Value	Angle	Value
P1-H1	1.29	O1-P1-O3	114.76(7)
N1-N2	1.397(2)	O2-P1-H1	100.0
O1-P1	1.502(1)	O3-P1-H1	109.0
O2-P1	1.576(1)	O3-P1-O2	109.75(7)
O3-P1	1.510(1)	N1-C5-N4	106.8(2)
C3-N2	1.306(2)	N2-C3-N7	125.6(2)
C3-N4	1.378(2)	N2-C3-N4	111.6(2)
C3-N7	1.352(2)	N6-C5-N1	127.3(2)
C5-N1	1.324(2)	N6-C5-N4	126.0(2)
C5-N4	1.351(2)	N7-C3-N4	122.8(2)
C5-N6	1.321(2)	C3-N2-N1	103.8(1)
P1-O2-H2	109.3	C5-N1-N2	111.3(2)
O1-P1-H1	110.7	C5-N4-C3	106.6(1)
O1-P1-O2	111.69(7)		

Hydrogen bonds

D-H…A	d (D-H)	d (A…H)	d (D…A)	<(DHA)
O2-H2…O3[a]	0.87	1.71	2.575(2)	174
N1-H1A…O1	0.81	1.92	2.710(2)	165
N4-H4A…O1[b]	0.86	1.91	2.759(2)	169
N6-H6A…N2[b]	0.86	2.05	2.906(2)	177
N6-H6B…O2[c]	0.86	2.17	3.001(2)	164
N7-H7A…O3[b]	0.90	1.99	2.894(2)	175
N7-H7B…O3[d]	0.83	2.15	2.938(2)	159

Note.
Equivalent positions: [a] x, 1/2-y, -1/2+z; [b] 1-x, 1/2+y, 3/2-z, -1/2-z; [c] 2-x, 1-y, 2-z; [d] -1+x, 1/2-y, -1/2+z.
Abbreviations: A, acceptor; D, donor.

Table 14S. Selected bond lengths (Å) and angles (°) for **datH$_2$PO$_4$**.

Bond/Angle	Value	Angle	Value
P1-O1	1.561(1)	O2-P1-O4	111.74(6)
P1-O2	1.571(1)	O3-P1-O4	114.81(6)
P1-O3	1.505(1)	P1-O1-H1	116(1)
P1-O4	1.516(1)	P1-O2-H2	117(1)
O1-H1	0.83(2)	N1-N2-C3	103.7(1)
O2-H2	0.82(1)	N1-C5-N6	128.3(1)
N1-H1A	0.87(1)	N2-N1-H1A	120(1)
N1-C5	1.326(2)	N2-N1-C5	111.3(1)
N2-N1	1.407(2)	N2-C3-N7	127.4(2)
N2-C3	1.299(2)	N4-C3-N2	111.5(1)
N4-H4A	0.86(2)	N4-C3-N7	121.1(2)
N4-C3	1.382(2)	N4-C5-N1	106.4(1)
N4-C5	1.348(2)	N4-C5-N6	125.3(2)
N6-H6A	0.88(2)	C3-N4-H4A	125(1)
N6-H6B	0.88(1)	C3-N4-C5	107.1(1)
N6-C5	1.324(2)	C3-N7-H7A	119(1)
N7-H7A	0.88(2)	C3-N7-H7B	113(2)
N7-H7B	0.88(1)	C5-N1-H1A	128(1)
N7-C3	1.342(2)	C5-N4-H4A	128(1)
O1-P1-O2	106.92(7)	C5-N6-H6A	117(1)
O1-P1-O3	112.27(6)	C5-N6-H6B	115(2)
O1-P1-O4	104.95(6)	H6B-N6-H6A	128(2)
O2-P1-O3	105.99(6)	H7B-N7-H7A	111(2)

Hydrogen bonds

D-H…A	d (D-H)	d (A…H)	d (D…A)	<(DHA)
O1-H1…O3[a]	0.83(2)	1.70(2)	2.527(2)	178(2)
O2-H2…O4[c]	0.82(1)	1.76(1)	2.585(2)	177(2)
N1-H1A…O3[b]	0.88(1)	1.94(2)	2.756(2)	155(2)
N4-H4A…O4[d]	0.86(2)	1.94(2)	2.766(2)	162(2)
N6-H6A…N2[d]	0.88(1)	2.16(1)	3.026(2)	167(2)
N6-H6B…O2[e]	0.88(1)	2.18(2)	2.987(2)	153(2)
N7-H7A…O1[d]	0.88(2)	2.09(2)	2.939(2)	164(2)
N7-H7B…O4	0.88(1)	2.03(1)	2.893(2)	167(2)

Note.
Equivalent positions: [a] -x, 1/2-y, -1/2+z; [b] 1/4+x, 1/4-y, 1/4+z, -1/2-z; [c] 1/2-x, 1/2-y, z; [d] x, y, 1+z; [e] 1/4-x, -1/4+y, 3/4+z.
Abbreviations: A, acceptor; D, donor.

Table 15S. Comparison of geomeric parameters (distances from the triazole ring plane; angles between the plane of triazole ring and plane of NH_2 groups).

	Deviation of fitted atoms of NH_2 groups (Å)			C-N bond (Å)	inter-planar angles (°)
	N - atom	HXA - atom	HXB - atom		
dat*	0.0924	-0.0860	-0.2901	1.376(0)	37.77(2)
	-0.0682	0.1851	0.1088	1.353(0)	27.33(2)
datNO$_3$	-0.0663	-0.0007	0.0904	1.325(2)	15.37(2)
	-0.0430	0.1755	0.0654	1.339(2)	21.00(1)
datClO$_4$	0.0144	-0.0490	-0.0785	1.323(3)	11.42(1)
	-0.0665	0.1535	0.0565	1.344(3)	25.22(1)
datH$_2$PO$_3$	-0.0055	-0.0132	0.0027	1.320(2)	0.61(6)
	-0.0376	0.1180	0.2470	1.352(2)	31.60(4)
datH$_2$PO$_4$	-0.0121	0.0768	-0.1394	1.324(2)	8.41(0)
	0.0642	-0.2689	-0.1853	1.342(2)	36.08(0)
dat$_2$Cl$_2$H$_2$O	0.0003	0.0034	-0.1434	1.320(2)	11.23(1)
	-0.0999	0.3415	-0.0394	1.356(2)	38.40(0)
	0.0495	-0.1507	-0.0926	1.330(2)	24.11(1)
	0.0351	-0.0911	-0.0545	1.339(2)	14.82(1)
dat$_2$SO$_4$2H$_2$O	-0.0159	0.0831	0.1249	1.322(3)	14.98(0)
	0.0151	-0.0407	0.1377	1.335(3)	8.00(0)
	-0.0248	0.0602	0.0813	1.318(3)	11.72(0)
	0.0521	-0.1418	0.1197	1.333(3)	12.40(0)
dat$_2$SeO$_4$2H$_2$O	0.0344	-0.1632	-0.0817	1.328(2)	19.65(0)
	-0.0047	0.1027	-0.1576	1.339(2)	10.50(0)
	-0.0279	0.0133	0.1103	1.329(2)	12.33(0)
	0.0706	-0.1925	0.2142	1.344(2)	17.73(1)

Note: Symbols: N-atom (N6, N7, or N13, N14), HXA-atom (H6A, H7A, or H13A, H14A), and HXB-atom (H6B, H7B, or H13B, H14B). The asterisk indicate the 3,5-diamino-1,2,4-triazole from the Crystal Structural Database (database code CSD – **Damtrz20**).

Table 16S: Calculated (scaled) fundamental frequencies (cm^{-1}) of **dat**(1+) cation

B3LYP 6-31+G S.F.[a] = 0.960	Relative intensities[b] IR	Raman	Assignment
183	0	0	τ NH_2, γ rg
211	1	0	τ NH_2, γ rg, ω NH_2
255	0	0	τ NH_2, γ NH, γ rg
306	1	2	ρ NH_2
354	1	0	γ rg, γ NH
466	0	2	ρ NH_2
557	20	0	ω NH_2, γ NH
570	0	0	ω NH_2, γ NH, δ rg
585	7	0	γ NH, δ rg
620	29	0	γ NH, γ rg, ω NH_2
640	0	14	ν rg, ρ NH_2
645	40	1	γ NH, ω NH_2, γ rg
705	18	0	δ CNC(rg), γ rg
771	1	5	δ CNN(rg), γ NH, γ rg
928	1	5	ν NN(rg), ρ NH_2, δ NH, δ rg
963	0	0	ρ NH_2, ν rg
1032	0	9	ρ NH_2, ν NC(rg), δ rg
1050	1	7	ρ NH_2, ν NC(rg), δ rg
1103	1	14	ρ NH_2, ν NN(rg), δ rg
1306	0	5	δ NH, ν CN, ν CN(rg), δ rg
1340	3	4	δ NH, ρ NH_2, ν CN(rg), δ rg
1385	0	8	ν rg, δ NH, ρ NH_2
1489	3	2	ν rg, ρ NH_2, δ NH
1615	2	2	δ NH_2, δ rg
1641	1	1	δ NH_2, δ NH, δ rg
1651	100	3	δ NH_2, ν CN, δ rg, δ NH
1679	20	18	δ NH_2, ν CN, δ rg, δ NH
3461	25	59	ν NH
3472	14	100	ν NH
3509	14	44	ν NH
3539	16	57	ν NH
3571	11	29	ν NH
3592	10	27	ν NH

[a] Precomputed vibrational scaling factor[40].

[b] The intensities of the calculated IR and Raman bands are presented on a relative scale from 0 to 100.

Table 17S. FTIR and Raman spectra of **$dat_2SeO_4 2H_2O$**.

FTIR cm^{-1}	Raman cm^{-1}	Assignment
3470 sh		ν OH(…O)
3310 mb	3317 mb	ν OH(…O), ν NH(…O)
3260 m	3258 m	ν NH(…O)
3191 m	3201 m	ν NH(…N), ν NH(…O)
3130 m	3130 m	ν NH(…N), ν NH(…O)
2916 m		ν NH(…O)
2797 m		ν NH(…O)
2660 m		ν NH(…O)
1676 s	1692 s	δ NH_2, ν CN, δ rg, δ NH
1622 m	1627 m	δ NH_2, δ NH, δ rg
1537 m	1543 m	ν rg, ρ NH_2, δ NH
1464 vw	1471 m	ν rg, δ NH, ρ NH_2
	1411 sh	ν rg, δ NH, ρ NH_2
1378 w	1388 m	ν rg, δ NH, ρ NH_2
1353 sh	1349 w	δ NH, ρ NH_2, ν CN(rg), δ rg
1157 w	1155 m	ρ NH_2, ν NN(rg), δ rg
1110 wb		ρ NH_2, ν NN(rg), δ rg
1066 vw	1073 s	ρ NH_2, ν NC(rg), δ rg
1046 w	1052 sh	ρ NH_2, ν NC(rg), δ rg
1015 m	1019 m	ρ NH_2, ν rg
	890 w	ν NN (rg), ρ NH_2, δ NH, δ rg
865 s	864 m	ν_3 SeO_4^{2-}
	836 vs	ν_1 SeO_4^{2-}
797 m	798 s	δ CNN(rg), γ NH, γ rg, γ NH(…N)
724 m	735 vw	δ CNC(rg), γ rg, γ OH(…O)
662 m	665 s	γ NH, ω NH_2, γ rg, γ OH(…O)
560 wb		ω NH_2, γ NH
519 w	518 m	?
	437 w	?
421 m	421 w	ν_4 SeO_4^{2-}
	405 w	?
	361 w	γ rg, γ NH
	333 wb	ν_2 SeO_4^{2-}
	251 w	?
	236 sh	?
	161 m	τ NH_2, γ rg
	141 m	External modes
	120 m	External modes

Note: Abbreviations and symbols: vs, very strong; s, strong; m, medium; w, weak; b, broad; sh, shoulder; ν, stretching; δ, deformation or in-plane bending; γ, out-of-plane bending; ρ, rocking; ω, wagging; τ, torsion; $_s$, symmetric; $_{as}$, antisymmetric.

Table 18S. FTIR and Raman spectra of **$dat_2SO_4 2H_2O$**.

FTIR cm^{-1}	Raman cm^{-1}	Assignment
	3320 mb	ν OH(…O)
3304 m		ν OH(…O)
3263 m	3266 m	ν NH(…O)
3201 m	3212 m	ν NH(…O)
3122 m	3129 m	ν NH(…N), ν NH(…O)
	2927 vw	ν NH(…O)
2803 m	2827 vw	ν NH(…O)
2731 m		ν NH(…O)
2666 m	2674 vw	ν NH(…O)
1692 s	1694 m	δ NH_2, ν CN, δ rg, δ NH
1678 s		δ NH_2, ν CN, δ rg, δ NH
1653 sh		δ NH_2, ν CN, δ rg, δ NH, δ H_2O
1630 m	1630 m	δ NH_2, δ NH, δ rg
1620 m		δ NH_2, δ NH, δ rg
1540 m	1541 w	ν rg, ρ NH_2, δ NH
1466 vw	1467 w	ν rg, δ NH, ρ NH_2
1382 m	1384 w	ν rg, δ NH, ρ NH_2
	1353 vw	δ NH, ρ NH_2, ν CN(rg), δ rg
1157 sh	1159 m	ρ NH_2, ν NN(rg), δ rg
1102 s		ν_3 SO_4^{2-}
	1071 s	ρ NH_2, ν NC(rg), δ rg
1053 m	1050 sh	ρ NH_2, ν NC(rg), δ rg
1012 m	1017 s	ρ NH_2, ν rg
	981 vs	ν_1 SO_4^{2-}
	820 wb	δ CNN(rg), γ NH, γ rg
799 mb	798 m	δ CNN(rg), γ NH, γ rg, γ NH(…N)
774 sh		δ CNN(rg), γ NH, γ rg
721 m	735 vw	δ CNC(rg), γ rg, γ OH(…O)
664 w	667 m	γ NH, ω NH_2, γ rg, γ OH(…O)
613 m	605 w	ν_4 SO_4^{2-}, γ NH, γ rg, ω NH_2
514 m	518 w	ρ NH_2
	469 m	ν_2 SO_4^{2-}, ρ NH_2
	375 m	γ rg, γ NH
	164 s	τ NH_2, γ rg
	147 s	External modes
	130 s	External modes
	102 s	External modes

Table 19S. FTIR and Raman spectra of **datClO$_4$**.

FTIR cm^{-1}	Raman cm^{-1}	Assignment
3472 m		ν NH(…O)
3458 m	3462 w	ν NH(…O)
3410 m	3417 w	ν NH(…O)
3368 m	3371 w	ν NH(…O)
3340 sh		ν NH(…O)
3278 m		ν NH(…O)
3220 m	3222 wb	ν NH(…O)
3174 m	3177 wb	ν NH(…N), ν NH(…O)
3006 w		ν NH(…N), ν NH(…O)
2908 w		?
2828 w		?
2752 w		?
1700 m	1701 m	δ NH$_2$, ν CN, δ rg, δ NH
1681 mb	1667 m	δ NH$_2$, ν CN, δ rg, δ NH
1645 m		δ NH$_2$, δ NH, δ rg
1602 w	1602 w	δ NH$_2$, δ rg
	1586 w	δ NH$_2$, δ rg
1542 m	1546 w	ν rg, ρ NH$_2$, δ NH
1458 m	1461 m	ν rg, δ NH, ρ NH$_2$
1367 w		δ NH, ν CN, ν CN(rg), δ rg
1345 sh	1345 m	δ NH, ν CN, ν CN(rg), δ rg
	1172 m	ρ NH$_2$, ν NN(rg), δ rg
1157 sh		ρ NH$_2$, ν NN(rg), δ rg
1127 sh	1122 sh	ν$_3$ ClO$_4^-$
1105 sh	1110 m	ν$_3$ ClO$_4^-$
1087 s		ν$_3$ ClO$_4^-$, ρ NH$_2$, ν CN(rg), δ rg
	1073 m	ρ NH$_2$, ν NC(rg), δ rg
1052 m		ρ NH$_2$, ν NC(rg), δ rg
1013 m	1018 m	ρ NH$_2$, ν rg
934 w	935 vs	ν$_1$ ClO$_4^-$
	908 w	ν NN (rg), ρ NH$_2$, δ NH, δ rg
	799 w	δ CNN(rg), γ NH, γ rg
	743 w	δ CNC(rg), γ rg
722 w		δ CNC(rg), γ rg, γ NH(…N)
672 w	669 m	γ NH, ω NH$_2$, γ rg
631 sh	632 m	ν$_4$ ClO$_4^-$, γ NH, ρ rg, δ CN
625 m	627 m	ν$_4$ ClO$_4^-$
573 w		ω NH$_2$, γ NH, δ rg
500 w	493 w	ν$_2$ ClO$_4^-$
471 w	470 w	ν$_2$ ClO$_4^-$, ρ NH$_2$
458 vw	455 w	ρ NH$_2$
	344 w	γ rg, γ NH
	232 vw	τ NH$_2$, γ rg, ω NH$_2$
	151 m	External modes
	126 s	External modes
	98 sh	External modes
	84 s	External modes
	70 s	External modes

Table 20S. FTIR and Raman spectra of **datNO$_3$**.

FTIR cm^{-1}	Raman cm^{-1}	Assignment
3360 s	3360 w	ν NH(…O)
3268 s	3277 w	ν NH(…O)
3189 s	3193 w	ν NH(…O)
3065 sh		ν NH(…O)
2964 w		ν NH(…O)
2812 w		ν NH(…N)
2680 w		ν NH(…N)
2438 w		?
2139 w		?
1777 w		$\nu_1 + \nu_4$ NO_3^-
1767 w		?
1717 m		?
1698 s	1707 m	δ NH_2, ν CN, δ rg, δ NH
1695 s		δ NH_2, ν CN, δ rg, δ NH
1687 s		δ NH_2, ν CN, δ rg, δ NH
1684 s	1677 sh	δ NH_2, ν CN, δ rg, δ NH
	1668 m	δ NH_2, ν CN, δ rg, δ NH
1659 s	1651 w	δ NH_2, ν CN, δ rg, δ NH
1616 vw	1618 w	δ NH_2, δ rg
1540 m	1545 w	ν rg, ρ NH_2, δ NH
	1468 m	ν rg, ρ NH_2, δ NH
1393 s	1394 w	ν_3 NO_3^-
1375 sh	1381 w	ν rg, δ NH, ρ NH_2
1355 sh	1357 vw	δ NH, ν CN, ν CN(rg), δ rg
1338 s		ν_3 NO_3^-
1153 m	1155 s	ρ NH_2, ν NN(rg), δ rg
1072 sh	1074 vs	ρ NH_2, ν NC(rg), δ rg
1067 m		ρ NH_2, ν NC(rg), δ rg
1053 m	1055 vs	ν_1 NO_3^-
1012 mb	1015 m	ρ NH_2, ν rg
817 m	820 vw	ν_2 NO_3^-
804 vw	807 m	δ CNN(rg), γ NH, γ rg
790 mb	794 w	δ CNN(rg), γ NH, γ rg, γ NH(…N)
724 m	730 m	ν_4 NO_3^-, γ CNC(rg), γ rg
716 m	717 vw	ν_4 NO_3^-, δ CNC(rg), γ rg
692 mb		γ NH, ω NH_2, γ rg
666 vw	671 m	ν rg, ρ NH_2
585 mb		ω NH_2, γ NH, δ rg
520 sh	515 w	ρ NH_2
	366 m	γ rg, γ NH
	242 vw	τ NH_2, γ NH, γ rg
	228 vw	τ NH_2, γ rg, ω NH_2
	191 vw	τ NH_2, γ rg
	147 vs	External modes
	133 sh	External modes
	105 vs	External modes
	80 vs	External modes
	71 s	External modes

Table 21S. FTIR and Raman spectra of **$dat_2Cl_2H_2O$**.

FTIR cm^{-1}	Raman cm^{-1}	Assignment
	3419 w	ν NH
3338 mb	3318 wb	ν NH
	3236 wb	ν NH
3161 mb	3172 wb	ν NH(…N), ν NH(…O)
2949 s		ν NH(…O)
2914 s		ν NH(…O)
2870 s		ν NH(…O)
2781 s		?
2724 s		?
2658 s		?
2132 s		?
1695 m	1688 m	δ NH_2, ν CN, δ rg, δ NH
1659 m	1658 m	δ NH_2, ν CN, δ rg, δ NH, δ H_2O
1598 m	1609 w	δ NH_2, δ NH, δ rg
	1591 w	δ NH_2, δ rg
	1568 m	ν rg, ρ NH_2, δ NH
1536 m	1541 w	ν rg, ρ NH_2, δ NH
1455 m	1455 w	ν rg, δ NH, ρ NH_2
1353 s		δ NH, ρ NH_2, ν CN(rg), δ rg
1338 m	1332 m	δ NH, ρ NH_2, ν CN(rg), δ rg
1302 m	1305 m	δ NH, ν CN, ν CN(rg), δ rg
1162 m	1164 m	ρ NH_2, ν NN(rg), δ rg
1063 m	1070 s	ρ NH_2, ν NC(rg), δ rg
1013 vs	1014 s	ρ NH_2, ν rg
928 w		ν NN (rg), ρ NH_2, δ NH, δ rg
799 s	796 m	δ CNN(rg), γ NH, γ rg, γ NH(…N)
717 m	723 w	δ CNC(rg), γ rg
653 m	664 m	γ NH, ω NH_2, γ rg
607 mb		γ NH, γ rg, ω NH_2
546 m		ω NH_2, γ NH
489 mb	498 m	ρ NH_2
	353 m	γ rg, γ NH
	340 sh	ρ NH_2
	202 w	τ NH_2, γ rg, ω NH_2
	172 sh	τ NH_2, γ rg
	147 s	External modes
	115 vs	External modes
	102 vs	External modes

Table 22S. FTIR and Raman spectra of **datH$_2$PO$_3$**.

FTIR cm^{-1}	Raman cm^{-1}	Assignment
3364 m		ν NH(…O)
	3317 w	ν NH(…O)
	3266 w	ν NH(…O)
	3194 m	ν NH(…O)
3174 m	3077 m	ν NH(…O)
3078 m		ν NH(…O)
2816 m		ν NH(…O), ν NH(…N)
2757 mb		ν NH(…O)
2701 m		ν NH(…O), ν (OH…O)
2418 m	2421 vs	ν PH
1709 sh		
1696 s	1701 m	δ NH_2, ν CN, δ rg, δ NH
1663 s	1662 vw	δ NH_2, ν CN, δ rg, δ NH
1629 w	1634 m	δ NH_2, δ NH, δ rg
1541 w	1549 vw	ν rg, ρ NH_2, δ NH
1476 vw	1482 m	ν rg, δ NH, ρ NH_2
1398 w	1402 m	ν rg, δ NH, ρ NH_2
1373 w		ν rg, δ NH, ρ NH_2
1231 mb		δ POH
1185 sh	1187 m	ρ NH_2, ν NN(rg), δ rg
1145 s		ν_a PO_2, ρ NH_2, ν NN(rg), δ rg
1118 sh		ρ NH_2, ν NN(rg), δ rg
1084 m		ν_s PO_2
1053 m	1055 vs	γ PH, ρ NH_2, ν CN(rg), δ rg
1028 m	1033 vs	δ PH, ρ NH_2, ν CN(rg), δ rg
	1019 vs	ρ NH_2, ν CN(rg), δ rg
993 m	994 m	ρ NH_2, ν rg
927 m		ν PO_H, ν NN (rg), ρ NH_2, δ NH, δ rg, γ OH(…O)
886 sh		?
866 sh		?
842 mb		γ NH(…N)
809 w	809 m	δ CNN(rg), γ NH, γ rg
796 w		δ CNN(rg), γ NH, γ rg
724 m	738 w	δ CNC(rg), γ rg
669 w	671 m	γ NH, ω NH_2, γ rg
635 wb		γ NH, γ rg, ω NH_2
	583 w	γ NH, δ rg
562 m		ρ PO_2
532 sh	536 m	ω NH_2, γ NH
522 m	523 sh	δ PO_2, ω NH_2, γ NH
	465 w	ρ NH_2
453 m	456 w	δ PO_H
433 sh	423 w	?
	371 m	γ rg, γ NH
	238 vw	τ NH_2, γ rg, ω NH_2
	157 m	τ NH_2, γ rg
	132 vs	External modes
	86 vs	External modes
	69 s	External modes

Table 23S. FTIR and Raman spectra of **datH$_2$PO$_4$**.

FTIR cm^{-1}	Raman cm^{-1}	Assignment
3405 m	3407 wb	ν NH(…O)
3367 m	3361 w	ν NH(…O)
	3319 w	ν NH(…O)
3269 m	3270 w	ν NH(…O)
3224 m	3231 w	ν NH(…N), ν NH(…O)
	3161 m	ν NH(…N), ν NH(…O)
2725 m		ν OH(…O), ν NH(…O)
2656 m		ν OH(…O), ν NH(…O)
2356 mb		ν OH(…O)
1680 vs	1687 m	δ NH$_2$, ν CN, δ rg, δ NH
	1632 w	δ NH$_2$, δ NH, δ rg
	1621 sh	δ NH$_2$, δ NH, δ rg
1529 m	1532 w	ν rg, ρ NH$_2$, δ NH
1459 vw	1457 m	ν rg, δ NH, ρ NH$_2$
	1403 vw	δ NH, ρ NH$_2$, ν CN(rg), δ rg
1362 m	1363 w	δ NH, ρ NH$_2$, ν CN(rg), δ rg
	1342 w	δ NH, ν CN, ν CN(rg), δ rg
1262 sh		?
1237 m		δ POH
1165 sh	1167 w	ρ NH$_2$, ν NN(rg), δ rg
1126 s	1120 vw	ν$_3$ PO$_4$
1087 s		ν$_3$ PO$_4$
1055 m	1055 s	ρ NH$_2$, ν NC(rg), δ rg, γ OH(…O)
1013 m	1014 s	ν CN, γ ring, γ OH(…O)
1001 m	995 s	ν CN, γ ring, γ OH(…O)
970 s		ν$_3$ PO$_4$
	912 s	ν$_1$ PO$_4$
883 m	888 w	?
831 mb		γ NH(…N)
796 m	796 m	δ CNN(rg), γ NH, γ rg
721 m		δ CNC(rg), γ rg
	704 wb	δ CNC(rg), γ rg
670 w	668 m	γ NH, ω NH$_2$, γ rg
637 wb		γ NH, γ rg, ω NH$_2$
552 m	547 w	ν$_4$ PO$_4$, ω NH$_2$, γ NH
529 sh	523 sh	ν$_4$ PO$_4$, ω NH$_2$, γ NH
509 m	510 m	ν$_4$ PO$_4$
496 sh		ρ NH$_2$
480 m	475 w	ρ NH$_2$
	442 w	?
	404 w	?
	371 m	ν$_2$ PO$_4$, γ rg, γ NH
	360 w	ν$_2$ PO$_4$, γ rg, γ NH
	351 w	External modes
	234 vw	External modes
	150 m	External modes
	126 vs	External modes
	107 vs	External modes

Table 24S: Correlation diagram of XO_4^{2-} (X= S or Se) internal modes in **$dat_2SO_4 2H_2O$** and **$dat_2SeO_4 2H_2O$** crystals.

Free ion modes	Degrees of freedom	Free ion XO_4^{2-} T_d	Site symmetry C_1	Factor group C_i	Vibrational modes
ν_1	2	A_1	A	A_g (Ra)	$\nu_1, 2\nu_2, 3\nu_3, 3\nu_4$
ν_2	2	E			
ν_3	2	F_2		A_u (IR)	$\nu_1, 2\nu_2, 3\nu_3, 3\nu_4$
ν_4	2	F_2			

Table 25S: Correlation diagram of ClO_4^- internal modes in **datClO$_4$** crystals.

Free ion modes	Degrees of freedom	Free ion ClO_4^- T_d	Site symmetry C_1	Factor grup C_i	Vibrational modes
ν_1	2	A_1	A	A_g (Ra)	$\nu_1, 2\nu_2, 3\nu_3, 3\nu_4$
ν_2	2	E			
ν_3	2	F_2		A_u (IR)	$\nu_1, 2\nu_2, 3\nu_3, 3\nu_4$
ν_4	2	F_2			

Table 26S: Correlation diagram of NO_3^- internal modes in **datNO$_3$** crystals.

Free ion modes	Degrees of freedom	Free ion NO_3^- D_{3h}	Site symmetry C_1	Factor grup C_{2h}	Vibrational modes
ν_1	4	A_1'	A	A_g (Ra)	$\nu_1, \nu_2, 2\nu_3, 2\nu_4$
ν_2	4	A_2''		A_u (IR)	$\nu_1, \nu_2, 2\nu_3, 2\nu_4$
ν_3	4	E′		B_g (Ra)	$\nu_1, \nu_2, 2\nu_3, 2\nu_4$
ν_4	4	E′		B_u (IR)	$\nu_1, \nu_2, 2\nu_3, 2\nu_4$

Table 27S: Correlation diagram of $H_2PO_3^-$ internal modes in **datH_2PO_3** crystals.

Free ion modes of HPO_3^{2-}	Degrees of freedom	Free ion HPO_3^{2-} C_{3v}	Free ion $H_2PO_3^-$ C_s	Site symmetry C_1	Factor group C_s	Vibrational modes
ν_1 (ν PH)	4	A_1	A´	A	A´ (IR, Ra)	$4\nu_1$ (ν PH)
ν_2 (δ PH)	4	E				$4\nu_2$ (δ PH)
ν_3´ (ν_s PO_2)	4	A_1				$4\nu_2$ (γ PH)
ν_3´´ (ν_{as} PO_2)	4	E				$4\nu_3$´(ν PO_H)
ν_4´ (δ_s PO_3)	4	A_1	A´´		A´´ (IR, Ra)	$4\nu_3$´´(ν_s PO_2)
ν_4´´ (δ_{as} PO_3)	4	E				$4\nu_3$´´(ν_{as} PO_2)
						$4\nu_4$´(δ PO_H)
						$4\nu_4$´´ (δ PO_2)
						$4\nu_4$´´ (ρ PO_2)

Table 28S: Correlation diagram of $H_2PO_4^-$ internal modes in **datH_2PO_4** crystals.

Free ion modes of PO_4^{3-}	Degrees of freedom	Free ion PO_4^{3-} T_d	Free ion $H_2PO_4^-$ C_{2v}	Site symmetry C_1	Factor group C_{2v}	Vibrational modes
ν_1	16	A_1	A_1	A	A_1 (IR, Ra)	$16\nu_1$ (ν_s $P(O_H)_2$)
ν_2	16	E	A_2		A_2 (Ra)	$16\nu_2$ (δ_s $P(O_H)_2$)
						$16\nu_2$ (τ $P(O_H)_2$)
ν_3	16	F_2	B_1		B_1 (IR, Ra)	$16\nu_3$ (ν_s PO_2)
						$16\nu_3$ (ν_{as} PO_2)
						$16\nu_3$ (ν_{as} $P(O_H)_2$)
ν_4	16	F_2	B_2		B_2 (IR, Ra)	$16\nu_4$ (δ_s PO_2)
						$16\nu_4$ (ρ PO_2)
						$16\nu_4$ (ω PO_2)

FIGURES

Figure 1S: Atom numbering of the isomorphous crystals of **$dat_2SO_4 2H_2O$/$dat_2SeO_4 2H_2O$.** The dashed lines indicate the hydrogen bonds.

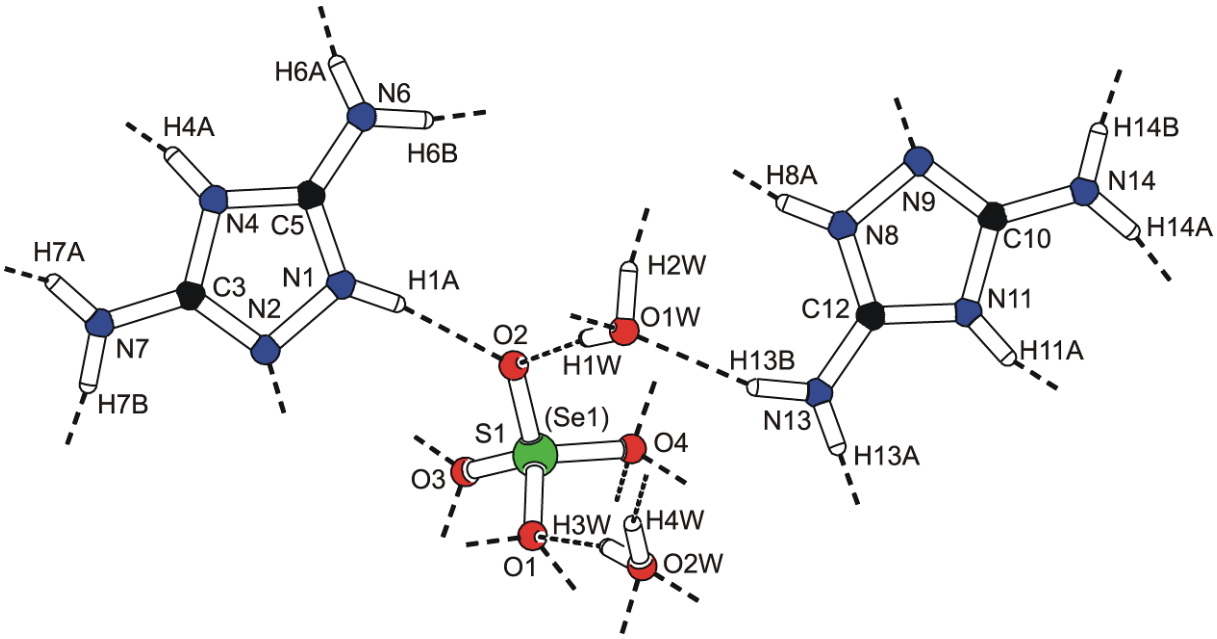

Figure 2S: Two structurally independent cationic chains in the crystals of **$dat_2SO_4 2H_2O$/$dat_2SeO_4 2H_2O$** (view along [010] direction). The dashed lines indicate the hydrogen bonds.

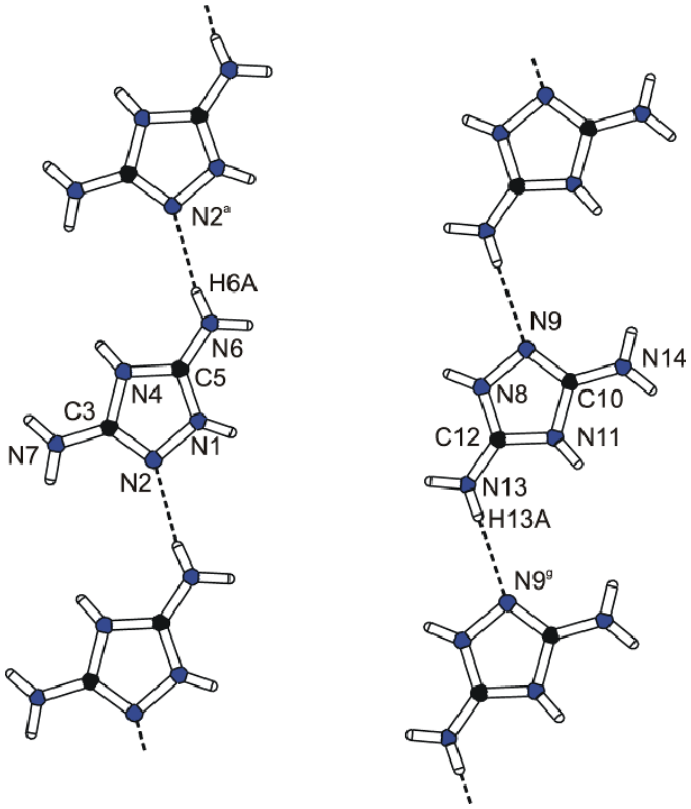

Figure 3S: Atom numbering of **datClO$_4$**. The dashed lines indicate the hydrogen bonds.

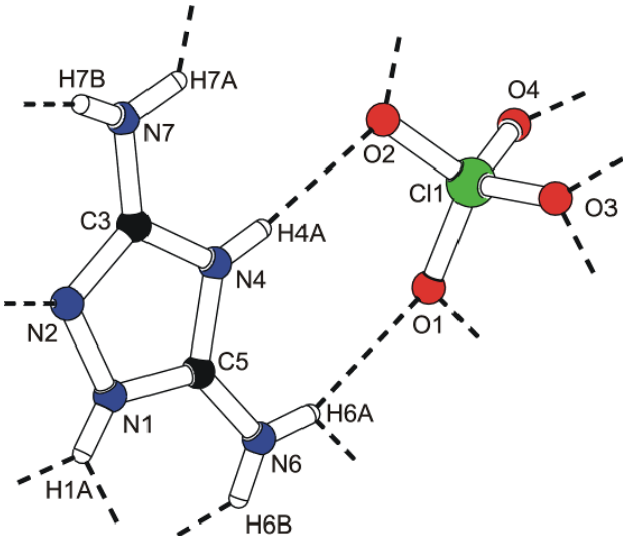

Figure 4S: The hydrogen bonds (dashed lines) system in the layers of **datClO$_4$** crystal structure.

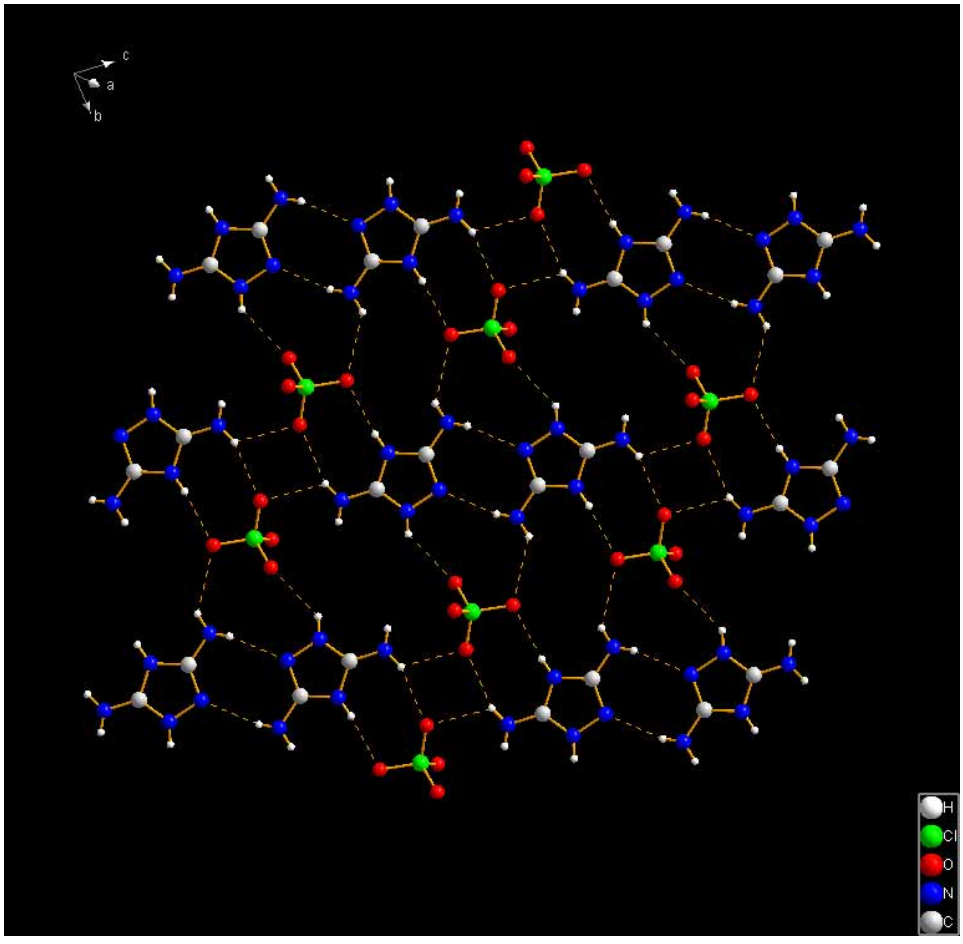

Figure 5S: Atom numbering of **datNO$_3$**. The dashed lines indicate the hydrogen bonds.

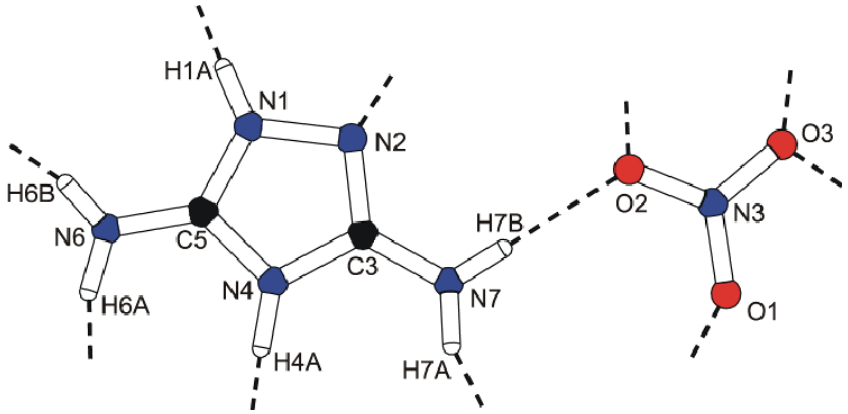

Figure 6S: The hydrogen bonds (dashed lines) system in the layers of **datNO$_3$** crystal structure.

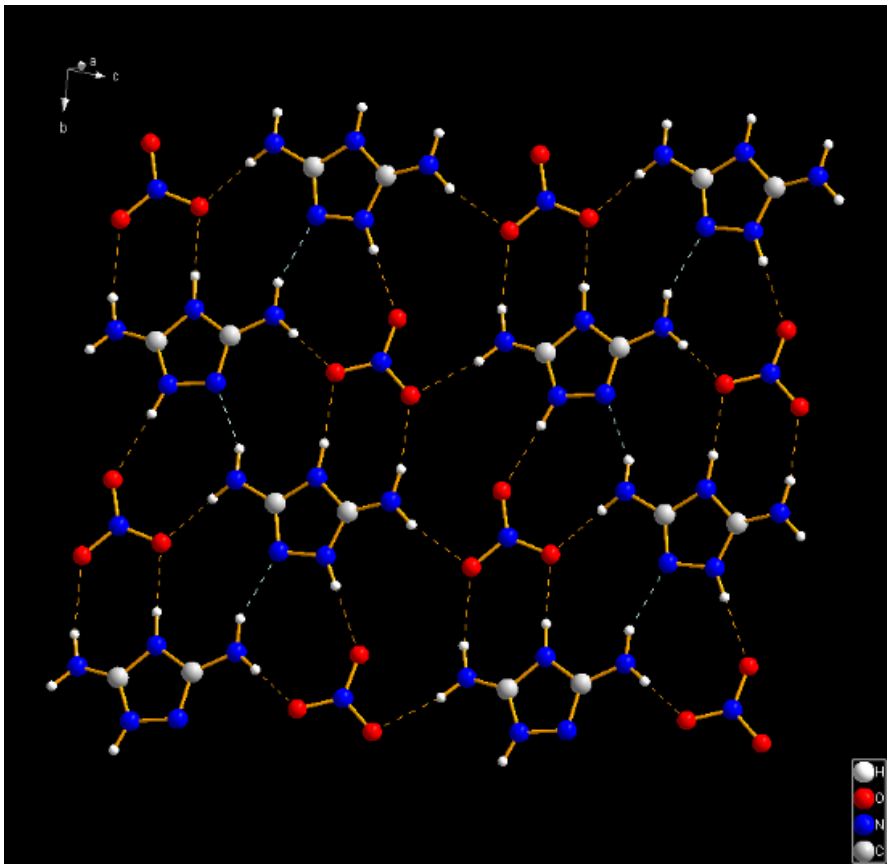

Figure 7S: Atom numbering of **dat$_2$Cl$_2$H$_2$O**. The dashed lines indicate the hydrogen bonds.

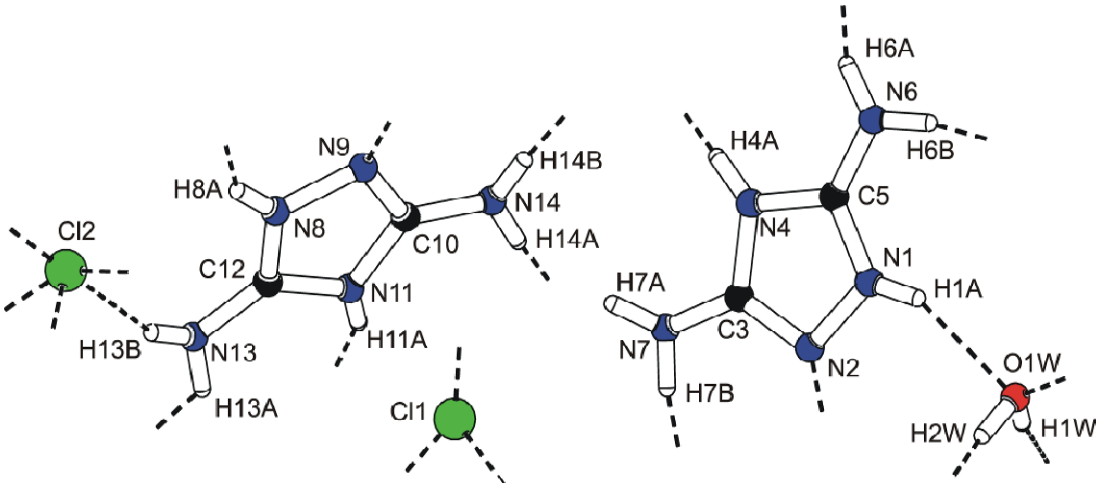

Figure 8S: Packing scheme of the cations in the crystal of **dat$_2$Cl$_2$H$_2$O** (view along [100] direction). The dashed lines indicate the hydrogen bonds.

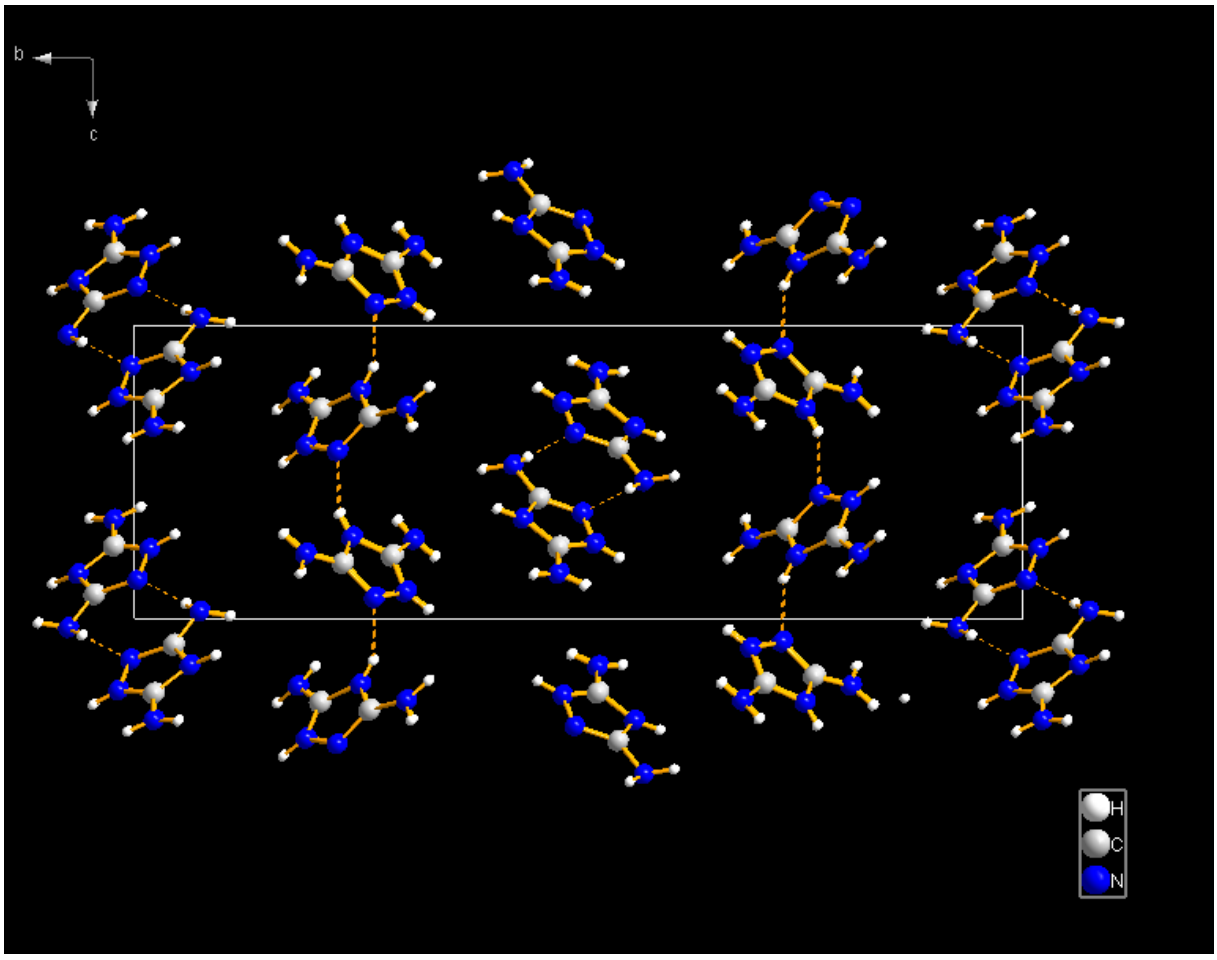

Figure 9S: Atom numbering of **datH$_2$PO$_3$**. The dashed lines indicate the hydrogen bonds.

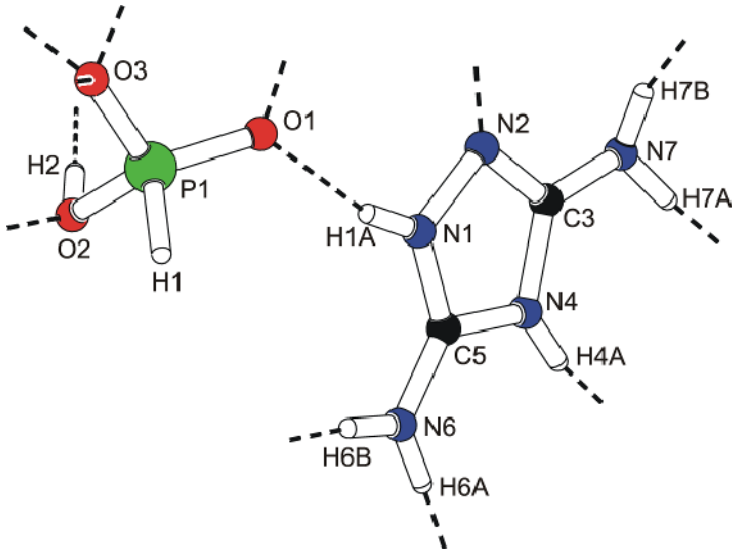

Figure 10S: Atom numbering of **datH$_2$PO$_4$**. The dashed lines indicate the hydrogen bonds.