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Crystal and Molecular Structure of 3-Hydroxybiuret Potassium Salt (2:1), $KH(C_2H_4N_3O_3)_2$

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The structure of the 3-hydroxybiuret potassium salt (2:1) ($C_2H_4KN_3O_3$, $C_2H_5N_3O_3$) has been determined, using three-dimensional diffractometer-collected X-ray data. The salt crystallizes in the space group $P2_1/c$ with 4 formula units $C_4H_9KN_6O_6$ in the unit cell with the dimensions $a = 3.868(2)$, $b = 20.38(1)$, $c = 12.400(7)$ Å, $\beta = 90.38(7)°$. The structure was solved by the heavy-atom method and refined by full-matrix least-squares calculations to a conventional R of 0.032.

The crystals are built up of K^+ ions and $H(C_2H_4N_3O_3)_2^-$ ions. The two 3-hydroxybiuret residues are joined by a very short hydrogen bond (2.442(3) Å) between the hydroxyl oxygen atoms. In addition the anions are connected by several NH···O hydrogen bonds. Each potassium ion is coordinated to eight oxygen atoms. The K···O distances lie in the range 2.663–3.026 Å.

The structure determination of 3-hydroxybiuret, $H_2N-(C=O)-N(OH)-(C=O)-NH_2$, was undertaken as part of an X-ray study of organic hydroxylamine derivatives, which are inhibitors of DNA synthesis in several cell systems. 3-Hydroxybiuret (3-HB) and 1-hydroxybiuret (1-HB) were by Gale *et al.*[1] shown to inhibit DNA synthesis in an *in vitro* Ehrlich ascites tumor test system. The 3-HB used in these experiments was prepared by the method described by Exner,[2] but following the same method the present author obtained a compound with a different melting point (138–139 °C decomp.; Exner: 133 °C decomp.). The elementary analysis suggested the compound to be a 3-HB potassium salt (2:1), and this assumption was verified by the present structure determination. The *CA* name of the compound for the present collective-index period (1972–76) is 2-hydroxyimidodicarbonic diamide potassium salt (2:1). By treatment of the potassium salt with a strongly acidic ion-exchange resin a compound was obtained which is presumed to be 3-HB, but the melting point of 158–160 °C decomp. does still not agree with that reported by Exner. X-Ray structure determination of this compound and of 1-HB are in progress.

EXPERIMENTAL

The potassium acid salt of 3-HB was synthesized from hydroxyurea and potassium cyanate following the method described by Exner [2] for preparation of 3-HB. The yield was 48 % of a crystalline crude product. Recrystallization by precipitation with absolute ethanol from a saturated aqueous solution of the crude product yielded thin, colourless needles, m.p. 138–139 °C (decomp.). (Found: C 17.35; H 3.34; N 30.16. Calc. for $C_4H_9KN_6O_6$: C 17.40; H 3.28; N 30.20).

Table 1. Crystal data for the 3-HB potassium salt (2:1).

Mol. formula	$C_4H_9KN_6O_6$
Mol. weight	276.3
Melting point	138–139 °C (decomp.)
Space group	$P2_1/c$
a	3.868(2) Å
b	20.38(1) Å
c	12.400(7) Å
β	90.38(7)°
V	977.5 Å^3
Z	4
D_x	1.87 g/cm^3
D_m	1.85 g/cm^3
μMo$K\alpha$	5.78 cm^{-1}
Crystal size	0.10 × 0.12 × 0.50 mm^3
Rot. axis	a

Crystals suitable for X-ray work were obtained by diffusion at room temperature of absolute ethanol into a saturated solution of the salt in aqueous ethanol. Some crystal data of the compound are given in Table 1. The density was measured by flotation in a mixture of carbon tetrachloride and methyl iodide. The melting points were determined with a hot stage microscope (Mikroskop Heiztisch Ernst Leitz G.m.b.H., Wetzlar). The microanalysis was performed by Preben Hansen, Microanalytical Laboratory, University of Copenhagen.

The lattice parameters were calculated from a series of diffractometer-measured θ-values. The intensity data were collected with a NONIUS 3-circle automatic diffractometer by the moving crystal-stationary detector technique, using graphite monochromatized Mo$K\alpha$-radiation ($\lambda = 0.71069$ Å). The scan speed was 0.6°/min and each reflexion was scanned over a range of 1.2°. The background was measured on each side of the reflexion for half the scanning time. The background count was in the data reduction set equal to twice the lowest count obtained in order to avoid weak satellite peaks, observed on one side of the strong reflexions. Intensities of reflexions were measured in the range $2.5° < \theta < 25°$, and the intensity of a reference reflexion was measured for every 25 reflexions. Thus 1663 independent reflexions were measured. A reflexion was considered unobserved and was omitted when the intensity observed was less than 2.5 times its estimated standard deviation. The number of observed reflexions was reduced to 1446. These data were corrected for Lorentz and polarization effects, but no corrections for absorption or extinction were made.

STRUCTURE DETERMINATION

The structure was solved from the three-dimensional Patterson map by the heavy-atom method. The electron density map, based on the position of the potassium ion only, revealed the positions of all 16 non-hydrogen atoms of the two 3-HB residues in the asymmetric unit. The conventional R-value was 0.31 at this stage. Three cycles of full-matrix least-squares refinement, in which positional as well as individual atomic, isotropic thermal parameters were varied, reduced the R-value to 0.074.

The difference Fourier map, calculated at this stage of the refinement, revealed the 8 hydrogen atoms of the NH_2-groups unambiguously (*cf.* Fig. 1). The 9th hydrogen atom is situated between the hydroxyl oxygen atoms O(3) and O(3′), apparently either symmetrically or statistically disordered.

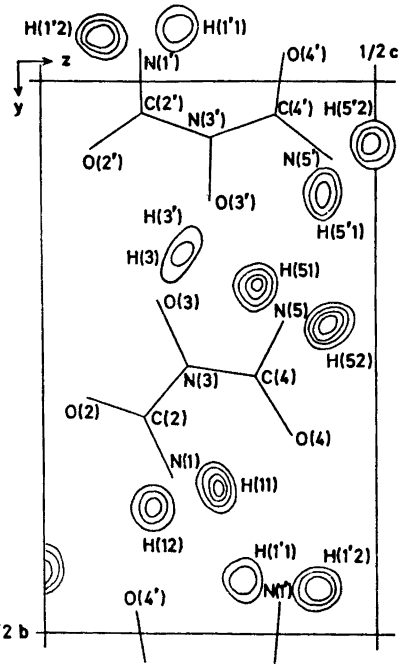

Fig. 1. Projection along the a-axis of sections of the difference Fourier map. Contours are at intervals of about 0.1 e Å^{-3}.

In the following refinement this hydrogen atom was treated as 2 half hydrogen atoms, H(3) and H(3′), *i.e.* each with a population parameter of 0.5 and situated in either end of the elongated peak, about 0.8 Å apart.

Two cycles of anisotropic least-squares refinement with all H-atom parameters fixed, reduced the R-value to 0.042. In the remaining two cycles of refinement the positional parameters of the H-atoms were also varied, but the isotropic thermal parameters of 2.0 Å^2 were still fixed. The final R-value was 0.032. Refinement of the population parameters of H(3) and H(3′) was also carried out, but the shifts were not significant, and these parameters were therefore fixed on 0.5.

In another attempt of refinement the hydrogen atom H(3) was included with a position midway between the oxygen atoms O(3) and O(3′) and allowed to vibrate isotropically. Only these four parameters of H(3) and the scale factor were refined, and at convergence ($R = 0.031$) the hydrogen atom had moved toward the oxygen atom O(3′). The dimensions corre-

Table 2. Observed and calculated structure factors. The columns are l, $10 \times F_o$ and $10 \times F_c$.

	0,0,L		8	318	-317	3	67	-60	-10	290	283	13	102	-109		1,9,L			
			9	28	22	4	180	-190	-12	272	-276	-13	52	56					
2	412	-448	10	81	76	5	126	-131	-12	209	205	14	107	114	0	336	-337		
4	294	-286	11	208	-206	6	110	-109	14	100	91	-14	113	-112	1	155	-136		
6	439	457	12	132	-133	7	74	-79	-14	145	140				-1	353	342		
8	128	139	13	30	24	8	192	-189					1,5,L		2	450	-443		
10	592	-598	14	116	-98	9	36	33		1,1,L					-2	410	-411		
12	44	35				10	170	-169				0	1350	1376	3	366	364		
14	123	126		0,8,L		11	36	15	0	541	520	1	112	108	4	118	-125		
									1	713	717	-1	164	161	-4	180	170		
	0,1,L		0	422	-416		0,16,L		-1	524	-520	2	314	307	-5	170	161		
			1	203	181				-2	117	-120	-2	239	229	6	103	96		
2	316	-315	2	436	-425	0	266	-262	-2	1409	-1479	3	226	-219	-6	253	-266		
3	300	303	3	208	-217	1	125	121	-3	330	-327	-3	194	188	7	53	52		
4	139	-136	4	337	334	2	253	-247	4	571	-576	4	612	595	-7	277	-278		
5	165	159	5	519	-514	3	107	-109	-4	426	-428	-4	528	521	8	59	-58		
6	814	-812	6	129	121	4	30	-22	5	33	13	5	167	-160	9	152	150		
7	97	96	7	420	-414	5	111	113	-5	117	119	-5	283	275	-9	320	-325		
8	216	-225	8	56	57	6	327	-328	6	39	37	6	31	-28	10	64	-66		
10	107	-105	9	146	-141	7	157	-150	-6	210	-215	-6	107	113	-10	100	90		
12	99	-93	10	205	209	8	158	-154	7	213	-207	-7	136	-134	11	320	323		
14	168	-163	11	187	-179	9	132	135	-8	283	-279	8	65	-64	-11	222	-230		
			12	97	-86	10	115	117	9	213	-213	-8	77	-77	12	124	128		
	0,2,L		13	164	-163	11	68	-72	-10	99	-105	9	120	-122	-12	121	-111		
									11	337	-336	-9	155	-151	13	65	66		
1	329	-334		0,9,L			0,17,L		-11	431	436	10	141	-144					
2	50	-45							13	118	-114	11	107	-116		1,10,L			
3	110	-119	1	769	-783	1	27	6	-13	138	136	-11	109	104					
4	200	-206	2	115	-109	2	79	-78	14	56	59	12	47	-45	0	45	35		
5	852	-855	3	195	-207	3	208	-210	-14	137	140	-12	128	128	1	164	176		
6	361	-359	4	123	-117	4	337	335				13	90	93	-1	37	38		
7	137	-143	6	166	-164	5	148	-149		1,2,L					2	125	127		
8	201	188	7	135	-144	6	156	155					1,6,L		-2	93	-87		
9	328	-331	8	82	70	7	53	-45	0	464	448				3	32	29		
10	217	-215	10	335	-339	8	83	80	1	254	-274	0	239	224	-3	305	303		
11	63	61	11	190	189	9	63	-71	-1	298	-293	1	414	407	-4	49	-40		
12	153	162	12	120	-120	10	112	112	2	212	206	-1	586	576	5	31	-24		
13	268	-269							-2	188	187	2	389	-378	-5	240	253		
14	115	106		0,10,L			0,18,L		3	889	-905	-2	127	113	6	283	291		
									-3	601	-598	3	179	-181	-6	56	-47		
	0,3,L		0	438	-439	0	105	-97	4	113	110	-3	61	61	7	75	-80		
			1	236	230	1	211	211	-4	589	-579	4	328	-332	-7	90	96		
1	493	-490	2	278	-292	2	190	186	-5	631	-627	-4	112	118	8	334	339		
2	73	-71	3	244	242	3	46	-38	6	160	160	5	116	-119	-8	390	-394		
3	398	-425	4	587	-576	4	342	336	-6	119	112	-5	211	217	-9	118	-124		
4	85	79	5	324	323	5	68	-69	7	309	-310	6	335	-334	10	179	169		
5	233	-240	7	302	317	6	321	-317	-7	155	156	-6	82	87	-11	73	70		
6	186	179	8	83	84	7	86	86	8	120	117	7	98	107	12	145	156		
7	413	-420	9	166	-168	8	279	271	-8	117	-113	-7	132	133	-12	247	-245		
8	97	-89	10	172	-162				9	99	94	-8	257	253	-13	68	-70		
9	172	-167	12	72	64		0,19,L		-9	282	-287	9	207	-213					
10	63	-59							10	82	-86	-9	52	50		1,11,L			
11	42	22		0,11,L		1	116	114	-10	180	-177	10	518	-525					
12	134	-141				2	145	152	-11	41	38	-10	211	214	0	140	136		
13	113	112	1	105	99	3	240	237	12	47	48	11	120	118	1	166	-177		
			2	53	-41	4	168	-176	-12	189	-194	-11	101	-104	-1	93	89		
	0,4,L		3	273	-271	5	170	166	13	143	147	12	69	72	2	307	312		
			4	183	184	6	72	69	-13	45	-37	-12	136	131	-2	156	159		
0	577	609	5	42	-37	7	44	-37	14	107	102				3	57	-56		
1	605	-633	6	271	272	8	89	87	-14	134	135		1,7,L		-3	89	87		
2	445	-473	7	34	-2	9	86	84							4	130	132		
3	141	-137	8	35	36	10	91	28		1,3,L		0	286	270	-4	47	-32		
4	952	-983	9	34	34							1	394	-382	5	89	-88		
5	464	-441	10	292	285		0,20,L		0	118	108	-1	139	-138	-5	115	-112		
6	235	235	11	86	86				1	429	436	2	227	-233	6	413	421		
7	357	363	12	188	188	0	115	111	-1	748	-750	-2	233	-242	-6	526	527		
8	73	-69	13	31	-26	1	216	-223	2	27	14	3	544	-529	7	342	355		
9	184	186				2	34	9	-2	698	702	-3	205	203	-7	62	-59		
10	229	-227		0,12,L		4	342	343	3	760	757	4	363	-363	8	155	202		
11	342	342				5	154	-146	-3	172	-175	-4	531	-534	-8	162	164		
12	32	-21	0	304	-287	6	39	-26	4	206	-204	5	537	-528	9	66	-68		
13	216	215	2	322	323	7	34	-39	-4	99	83	-5	597	594	-9	145	153		
14	235	233	3	609	596	8	36	25	5	129	140	6	67	-70	10	57	-57		
			4	348	340				-5	161	-166	7	202	-211	11	56	66		
	0,5,L		5	95	94		0,21,L		6	373	-357	-7	238	242	-11	154	-160		
			7	42	-31				-6	467	-462	8	147	146	12	139	-145		
1	25	-13	8	32	-30	1	103	97	7	206	198	-8	46	43	-12	137	132		
2	230	251	9	325	322	2	251	-256	-7	95	93	9	36	-31					
3	154	-146	10	271	274	3	45	-36	8	164	-166	-9	230	239		1,12,L			
4	70	-60	11	78	69	5	87	-90	-8	30	7	10	200	-199					
5	199	200	12	129	-125	6	79	-79	9	564	564	-10	259	-268	0	29	19		
6	320	331				7	45	53	-9	373	-376	11	55	59	1	546	546		
7	100	96		0,13,L					10	177	-188	-11	44	36	-1	40	46		
8	410	413					0,22,L		-10	198	-194	12	105	108	2	213	-219		
10	114	118	1	146	153				11	132	139	-12	124	127	-2	150	156		
11	166	168	2	163	158	0	325	-323	-11	139	-140	13	152	-156	3	67	67		
12	302	304	3	425	430	1	34	26	12	211	215	-13	110	108	-3	93	99		
13	69	-75	4	52	56	2	136	-137	13	77	76				4	30	21		
14	161	152	5	190	186	3	36	-31	14	139	136		1,8,L		-4	205	219		
			6	48	42	4	39	-36	-14	46	30				5	111	112		
	0,6,L		7	132	133	5	235	-236				0	44	-35	-5	167	176		
			8	65	60	6	90	-84		1,4,L			222	-223	6	164	-167		
0	392	396	9	35	30							-1	666	-664	-6	91	-102		
1	93	-99	10	169	-170		0,23,L		0	984	981	2	143	134	7	76	76		
2	383	399	11	45	45				-1	661	-642	-2	241	235	-7	298	299		
3	149	155				1	179	-177	2	280	-272	3	165	152	8	325	-337		
4	312	-301		0,14,L		2	48	51	-2	724	-713	-3	817	-805	-8	150	151		
5	165	167				3	100	-107	3	765	768	4	127	-123	9	165	176		
6	793	781	0	31	26				-3	529	518	-4	226	-212	10	69	-73		
8	335	331	1	270	263		0,24,L		4	77	-61	5	463	-474	-10	45	37		
9	228	235	2	487	484				-4	250	-243	-5	96	-94	11	90	-97		
10	39	-30	3	191	-170	0	203	-205	5	83	84	6	107	98	12	64	-64		
11	195	195	4	139	143				-5	263	266	7	90	-77	-12	97	100		
12	128	-122	5	64	-69		1,0,L		6	527	523	-7	294	-292					
14	58	-56	6	39	-35				-6	44	-60	8	135	-139		1,13,L			
			7	137	-132	0	123	-117	7	44	-40	-8	138	136					
	0,7,L		8	274	-284	-2	273	-265	-7	64	-60	9	248	260	0	82	-70		
			9	202	-200	-2	170	169	8	313	314	-9	186	-190	1	360	-348		
1	602	614	10	74	78	4	1360	-1390	-8	50	49	10	103	111	-1	137	138		
2	116	-123	11	245	-243	-4	495	-474	9	194	201	-10	275	-267	2	325	-324		
3	639	647				6	419	402	10	121	141	11	78	-83	-2	65	61		
4	141	-148		0,15,L		-6	670	646	-10	348	-348	12	48	-45	3	91	82		
5	69	76				8	160	-166	11	201	-199	-12	50	55	4	139	-138		
6	156	-151	1	174	-175	-8	485	463	-11	198	194	13	52	-52					
7	139	143	2	146	-144	10	339	-344	12	112	107	-13	176	179					

Table 2. Continued.

	1,13,L		-7	112	113	-2	106	-101	-8	313	-307	-4	182	176	-7	92	89
			8	134	-136	3	362	357	9	30	35	6	148	137	8	138	-139
-4	115	-120	10	51	99	-3	376	-373	-9	166	-167	6	341	-338	-8	142	143
5	32	17				-4	179	160	10	58	57	-6	124	119			
-5	81	20		1,19,L		5	271	271	-10	38	26	7	64	-61		2,18,L	
6	164	165				-5	135	-129	11	365	-370	-7	103	-106			
-6	131	135	-1	75	-79	6	207	210	-11	390	381	8	164	-162	1	106	-101
7	463	-476	2	131	130	-6	227	224	12	154	153	-8	36	33	-1	66	63
-7	136	137	-2	202	207	7	562	560	-12	110	-108	9	107	-106	2	56	-54
9	216	-222	3	78	-81	-7	313	-311				10	270	-258	-2	118	-108
-9	240	248	-3	180	187	8	84	79		2,7,L		-10	155	156	3	109	-110
10	42	56	4	121	119	-8	203	213							-3	73	68
-10	74	79	-4	42	29	9	60	63	0	45	44		2,12,L		-4	69	-71
11	51	-53	5	69	-71	-9	105	-102	-1	169	-177				5	79	-81
-11	157	158	6	79	-79	10	126	127	2	91	86	0	384	-388	-6	38	-21
12	49	-45	-6	74	-73	-10	191	184	-2	170	170	1	54	-48	-7	76	74
-12	154	-150	7	218	-220	11	41	28	3	154	-141	-1	35	-3			
			-7	135	130	-11	75	-74	-3	436	-430	2	206	-204		2,19,L	
	1,14,L		-8	47	-55	12	90	-85	4	101	94	-2	79	-72			
						-12	89	-92	-4	168	-168	3	186	-187	0	55	-58
0	182	-184		1,20,L		13	88	85	5	146	-136	-3	68	-66	1	232	-234
1	159	-169				-13	71	-66	-5	424	-422	-4	219	-207	-1	152	-146
-1	477	-481	0	244	-250				6	312	311	5	596	-591	3	160	-155
2	62	-72	1	65	-62		2,3,L		-6	85	-79	-5	467	461	-3	164	-165
-2	145	-147	-1	170	-170				-7	43	-51	-6	79	-76	-4	47	29
3	79	-81	2	165	170	0	153	-141	8	155	150	-5	40	-30	5	58	-54
-3	254	-259	3	134	-136	1	764	738	-8	143	-142	9	35	-25	-5	137	-137
4	72	75	-3	46	48	-1	581	571	9	56	-56	10	91	64			
-4	168	160	4	54	-54	2	43	43	-9	257	-254	11	138	-121		2,20,L	
5	328	-337	-4	189	192	3	76	75	10	36	-38	-11	60	57			
-5	43	46	5	36	-22	-3	85	-86	-10	78	79				0	135	-132
6	359	-369	-5	132	-128	4	292	288	11	57	-44		2,13,L		-2	115	-114
-6	106	110	6	149	-156	-4	274	-272	-11	66	-59				3	208	202
7	73	83	7	125	-125	5	32	15	12	82	74	0	104	103	-3	71	-73
-7	177	-177	-7	90	-85	-5	514	506	-12	143	-149	1	353	-342	4	144	-140
8	124	132				6	90	82				2	65	-65	-4	89	-92
-8	149	152		1,21,L		-6	49	34		2,8,L		3	254	-257	5	171	167
9	56	52				7	166	158				-3	261	-258	-5	156	-156
-9	98	-102	0	110	-106	-7	323	324	0	435	420	-4	57	53			
10	42	-53	1	51	-47	8	204	208	1	569	556	-5	368	-371		2,21,L	
-10	49	55	2	181	-182	9	92	-90	-1	383	-365	6	185	186			
			-2	112	-117	10	34	-23	2	101	-102	-6	203	-210	1	62	-56
	1,15,L		3	217	-213	-10	60	63	-2	42	44	7	104	-107	-1	66	73
			-3	48	-45	11	110	-106	3	246	242	-7	41	34	2	134	133
0	526	-533	-4	89	-82	-12	86	-82	-3	275	267	-8	87	87			
1	237	243	5	100	96	13	202	-199	-4	127	-124	-9	85	-81		3,0,L	
-1	49	-46	-5	147	-139	-13	88	87	5	47	-46	10	59	-64			
-2	193	-200	-6	197	-196				6	220	-216	-10	67	-60	0	48	-46
-3	100	-103					2,4,L		-6	65	-67				2	314	313
4	138	-139		1,22,L					7	161	163		2,14,L		-2	203	204
-4	226	-230				0	126	123	-8	38	43				4	272	274
5	299	295	-1	71	-72	1	80	75	9	416	407	8	39	32	-4	54	46
-5	232	-221	-2	84	83	-1	450	-432	-9	300	-288	1	34	22	6	111	-106
6	60	57	3	84	-73	2	537	527	10	49	-49	-1	137	129	8	394	393
-6	130	-137	-3	119	-113	-2	754	734	-10	202	-196	2	55	-50	-8	134	-136
7	166	169	4	223	221	3	307	301	11	65	71	-2	264	-261	10	229	226
-7	131	-136	-4	41	-49	-3	38	-33	-11	153	-145	3	230	231	-10	150	-155
8	48	-49	-5	138	-139	4	73	-75	12	61	65	-3	62	-59			
-8	168	-171				-4	202	-201	-12	104	96	4	168	-169		3,1,L	
9	87	-91		1,23,L		5	352	-349				-4	227	-219			
-9	47	52				-5	94	91		2,9,L		5	64	65	0	105	-109
-10	46	38	0	60	65	6	102	97				-5	76	71	-1	125	118
-11	59	-54	2	70	63	-6	325	321	0	39	42	6	143	145	2	298	309
			-2	74	73	7	466	-469	1	207	209	-6	65	59	-2	154	148
	1,16,L		3	47	47	-7	83	83	-1	123	115	7	104	105	3	120	123
						8	46	45	2	189	182	-7	85	-78	-3	83	77
0	97	-102		2,0,L		-8	119	120	3	95	91	8	40	-36	4	97	98
1	257	-264				-9	78	74	-3	338	334	-8	246	242	-4	238	234
-1	184	183	0	467	-455	10	133	-135	4	204	203	-9	173	-169	5	71	66
2	224	231	2	375	-366	11	193	-194	-4	130	-125	10	45	29	-5	125	124
-2	33	32	-2	551	-547	-11	178	181	5	183	179				6	69	69
3	70	67	4	120	-118	12	166	-165	-5	137	130		2,15,L		-6	238	239
-3	171	-168	-4	452	-440	-13	203	202	6	197	190				7	94	96
4	273	289	6	101	104				7	149	-145	0	104	105	-7	56	-59
-4	99	100	-6	269	-266		2,5,L		-7	180	180	2	72	65	8	62	61
5	33	-31	8	160	160				8	48	45	3	163	157	-8	62	65
-5	105	107	-8	76	-68	1	299	-277	-8	120	-116	-3	255	257	9	107	107
6	197	199	10	86	-73	-1	496	-499	9	93	90	4	236	228	-9	82	-82
-6	351	-358	-10	89	-89	-2	176	-170	-9	109	108	-4	75	77	10	77	-80
7	47	-51	12	45	29	3	244	234	10	196	197	5	61	-64	-10	143	145
-8	219	-224	-12	33	-23	-3	151	152	-10	185	-180	-5	96	93	11	199	205
10	183	183				4	498	-489	11	68	-65	6	84	-84	-11	175	-177
				2,1,L		-4	170	166	-11	96	94	7	65	72			
	1,17,L					5	110	-106				8	168	164		3,2,L	
			0	320	306	-5	206	200		2,10,L		-8	193	-192			
0	366	372	1	328	-320	6	130	-127				9	51	-49	0	153	152
1	170	-166	-1	475	-458	-6	215	214	0	641	612	-9	143	133	1	168	161
-1	114	119	2	86	74	7	88	-89	1	34	22				-1	200	202
2	74	76	-2	85	75	-7	264	-264	-1	141	-137		2,16,L		2	195	-192
-2	261	267	3	63	56	8	404	-397	2	201	202				3	81	75
3	132	129	-3	146	-140	-8	156	161	-2	163	163	0	113	-109	-3	349	354
-4	133	134	4	97	82	9	152	139	3	46	-39	1	277	278	4	155	-155
5	331	334	-4	34	29	10	115	-108	-3	203	-195	-1	232	-232	5	62	61
-5	369	-380	5	325	-306	-10	169	167	4	212	214	2	101	101	-5	300	303
6	77	82	-5	145	133	11	81	84	-4	442	431	-2	229	233	6	91	93
-6	101	105	6	379	372	-11	167	165	5	337	-336	4	135	137	-6	102	-112
8	81	-81	-6	49	-35	12	141	-142	-5	370	359	-4	218	217	7	99	-96
-8	47	37	-7	201	-199	-12	210	212	6	70	-72	5	110	-107	-7	151	156
9	89	89	8	65	57				-6	63	-63	6	153	150	8	216	-217
10	55	-42	-8	213	-215		2,6,L		-7	60	50	-6	237	232	-8	38	35
			9	163	156				8	54	58	7	133	131	-9	74	72
	1,18,L		-9	142	-139	0	113	-110	-8	182	162	-8	132	132	10	147	-143
			10	211	207	1	481	461	9	88	90	-9	72	-68	11	123	-116
0	171	-172	-10	147	-146	-1	294	-284	-9	84	-90				-11	123	127
1	321	332	12	146	138	2	288	-279	10	268	-274		2,17,L				
-1	127	127	-12	100	-97	-2	313	-302	-10	42	-37					3,3,L	
-2	58	58	13	53	61	3	265	-253				0	50	-50			
3	201	213	-13	47	48	-3	426	-415		2,11,L		1	41	-45	0	71	-71
4	65	65				4	245	-241				-1	301	303	1	208	-209
-4	99	-107		2,2,L		-4	58	-102	1	230	225	2	199	-203	-1	72	-74
5	39	-43				5	52	51	-1	37	-40	-3	103	102	-2	40	39
-5	201	202	0	30	35	-5	32	9	2	254	-246	4	138	-134	3	343	-359
6	36	-39	1	507	500	-6	387	-378	3	57	60	5	244	243	-3	91	-90
-6	136	140	-1	33	34	-7	158	169	-3	217	217	-5	58	57	-4	143	142
7	188	200	2	213	-207	8	152	-154	4	249	-245	7	43	-36			

Table 2. Continued.

	3,3,L		-6	93	-99		3,10,L		-7	98	-93	-6	75	75	2	149	150
			7	108	99										-2	137	141
5	325	-323	-7	224	-227	0	302	-298		3,14,L			4,1,L		-3	41	42
-5	122	124	-8	51	-47	1	165	167							4	264	252
6	102	103	10	163	165	-1	44	-56	0	103	98	0	207	-210	-4	153	164
7	132	-130	-10	131	-132	-2	66	-80	1	277	288	1	108	112	5	63	-58
-7	153	152				3	88	-93	-1	169	168	-1	68	62	-5	49	50
8	65	-66		3,7,L		-3	195	-205	2	41	19	2	142	-146	-6	110	-116
-8	121	119				4	326	-327	-3	41	34	-3	135	140			
9	119	-123	0	43	-46	5	103	-100	5	37	20	4	48	43		4,6,L	
-9	70	72	1	325	326	6	57	-55	-5	139	144	-4	72	64			
10	45	48	2	110	112	-6	132	131	6	89	83	5	69	67	0	106	105
-10	135	134	-2	40	22	8	239	-232	-6	64	-62	6	399	-407	1	68	66
-11	147	146	3	96	98	-8	65	64	-7	209	212	7	110	-119	-2	174	172
			-3	178	-182	9	160	157				-7	150	155	-3	50	51
	3,4,L		4	108	112	-9	164	-172		3,15,L					5	79	75
			-4	399	403								4,2,L		6	72	74
-1	295	-285	5	243	238		3,11,L		0	143	138				-6	251	261
2	316	-322	-5	58	56				1	111	-110	1	196	-204			
-2	146	-149	6	44	-34	0	164	-165	2	126	129	-1	124	-128		4,7,L	
3	75	-70	7	198	193	1	56	-54	3	178	-176	2	80	-85			
-3	362	-368	-7	75	-77	-1	66	-69	4	112	106	3	129	-135	0	225	-235
4	101	-96	8	34	-26	-2	295	-287	-4	194	192	-3	58	-54	1	67	67
-4	83	85	9	131	134	-3	65	-65	5	64	-60	4	64	-57	-1	166	152
5	236	-235	-9	138	-144	4	59	-54	-5	71	72	-4	68	-67	-2	52	-59
6	318	-313	10	44	-32	5	86	-90	6	36	-25	5	251	-262	3	51	57
7	174	173	-10	200	205	-5	66	69	-6	72	69	7	77	-76	-3	68	70
-8	152	157				6	105	-108							-4	175	169
9	186	187		3,8,L		-6	271	-273		3,16,L			4,3,L		-5	180	185
-9	170	-171				7	65	-64									
10	131	-131	1	231	223	-7	72	78	0	77	-76	0	66	63		4,8,L	
-10	97	93	-1	105	100	8	45	35	1	70	76	1	58	-59			
			-2	162	166	-8	291	-286	2	129	-127	-1	131	-136	1	65	-66
	3,5,L		3	280	282				3	60	58	2	78	-82	-1	162	-160
			4	55	-48		3,12,L		4	48	-50	-2	92	-95	2	42	-30
0	492	-480	-4	272	-281				-4	36	38	3	101	-98	-2	132	-131
2	162	-160	5	152	-150	0	166	169	5	100	-104	-3	166	-175	3	166	-163
-2	256	-254	-5	424	435	1	111	114	-5	58	58	4	56	-59	-4	141	149
3	253	256	6	170	166	-1	463	-471				-4	76	79			
-3	119	117	-6	109	108	2	72	78		3,17,L		5	100	108		4,9,L	
4	62	56	7	113	111	-2	72	75				-5	195	-203			
-4	334	-335	-7	104	110	4	198	201	0	122	-120	-6	70	-72	0	78	-84
5	142	138	9	43	35	-4	99	-108	1	99	-99	-7	167	-173	-1	139	-140
-5	177	-175	-9	64	69	5	116	-120	-1	91	-95				2	183	-187
-6	71	-68	-10	87	77	-5	144	-142	2	70	-73		4,4,L		-2	43	-38
7	121	-119				6	169	166	-2	130	-133				-3	48	-44
8	100	101		3,9,L		-7	45	41	3	45	-54	0	78	-81			
-8	177	-178				-7	173	-178	-3	65	57	1	61	64		4,10,L	
10	219	219	0	48	43	8	50	46	4	69	-63	-1	170	176			
			-1	95	-97	-8	105	-100	-4	75	-68	-2	90	-92	0	97	-100
	3,6,L		2	112	112							-3	69	71	1	73	70
			-2	329	331		3,13,L			3,18,L		4	75	-73	-1	80	72
0	81	70	4	121	116							-4	196	-198	2	85	-138
1	128	-117	-4	141	135	1	170	167	0	121	125	5	81	87	-2	85	-89
-1	75	-76	5	68	-73	2	34	-31	1	130	-125	-5	85	92	3	63	70
2	260	257	-5	174	-172	-2	257	254	-1	124	-131	6	78	80	-4	242	-246
3	36	21	6	110	-112	3	96	93				-6	105	-120			
-3	104	-107	7	172	-177	-3	115	118		4,0,L						4,11,L	
4	242	237	8	115	-114	4	81	84					4,5,L				
-4	60	63	-8	83	79	-4	37	36	0	396	423				0	83	-91
5	274	-267	9	205	-207	5	160	155	2	38	45	0	105	107	2	244	257
-5	156	158	-9	144	146	6	107	-103	-2	117	113	1	68	-64	-2	87	94
6	221	222				7	189	192	-4	148	148	-1	43	31			

sponding to this refinement are also given in the description of the structure.

The above refinements were based on F, minimizing the function $\sum w(|F_o|-|F_c|)^2$. The weights for all observed reflexions were unity. The weight analysis, *i.e.* the plot of the $\langle w(|F_o|-|F_c|)^2\rangle$ values as a function of F_o and $\sin\theta$ for groups of reflexions, indicated that this choice was satisfactory. The atomic scattering factors for K^+ and neutral O, N, C, and H, respectively, were taken from *International Tables for X-Ray Crystallography*.[3] Observed and calculated structure factors are listed in Table 2. Table 3 lists the fractional coordinates of the atoms, and Table 4 the thermal parameters of the non-hydrogen atoms.

The input tapes for the diffractometer were produced on a GIER computer using a program written by Sørensen.[4] The rest of the calculations were performed on the IBM 360/75 computer at NEUCC, Lundtofte, Denmark, using a local version of the N.R.C. 2A Picker data reduction program,[5] and *The X-Ray System*.[6] The drawings were prepared by *ORTEP*.[7]

DISCUSSION

The structure of the 3-HB potassium salt (2:1) is built up of K^+ ions and $H(C_2H_4N_3O_3)_2^-$ ions. As illustrated in Fig. 2 the potassium ions form slightly puckered layers, which are perpendicular to the c-axis and nearly at $z=0$ and $\frac{1}{2}$. The 3-HB anions are situated between these layers. The anion is composed of the two crystallographically nonequivalent 3-HB residues, which are joined by a short hydrogen bond between the hydroxyl oxygen atoms, O(3) and O(3′), and are linked to the metal ions by ionic interactions. In addition the anions are linked to a three-dimensional network by several NH···O hydrogen bonds. The lengths of these bonds are given in Fig. 2, and the angles concerning the hydrogen bonding system are given in Table 7.

Table 3. Fractional atomic coordinates.

	x	*y*	*z*
K^+	0.2825(1)	0.1554(1)	0.0242(1)
N(1)	0.4725(7)	0.3586(1)	0.1933(2)
C(2)	0.5465(7)	0.3006(1)	0.1491(2)
O(2)	0.4649(6)	0.2852(1)	0.0571(1)
N(3)	0.7314(6)	0.2564(1)	0.2146(1)
O(3)	0.8180(5)	0.1970(1)	0.1667(1)
C(4)	0.8479(7)	0.2660(1)	0.3193(2)
O(4)	0.7810(5)	0.3168(1)	0.3709(1)
N(5)	1.0356(6)	0.2167(1)	0.3594(2)
N(1′)	0.9760(7)	−0.0274(1)	0.1482(2)
C(2′)	0.8047(6)	0.0298(1)	0.1502(2)
O(2′)	0.7428(5)	0.0618(1)	0.0683(1)
N(3′)	0.6879(5)	0.0520(1)	0.2504(1)
O(3′)	0.4831(5)	0.1083(1)	0.2475(1)
C(4′)	0.7963(7)	0.0300(1)	0.3509(2)
O(4′)	0.9568(5)	−0.0224(1)	0.3610(1)
N(5′)	0.7118(7)	0.0680(1)	0.4339(2)
H(11)	0.517(10)	0.366(1)	0.262(3)
H(12)	0.345(10)	0.386(1)	0.158(3)
H(3)	0.717(22)	0.166(4)	0.198(6)
H(51)	1.078(10)	0.182(2)	0.317(3)
H(52)	1.113(09)	0.217(1)	0.430(3)
H(1′1)	1.036(10)	−0.044(1)	0.208(3)
H(1′2)	1.063(10)	−0.038(1)	0.087(3)
H(3′)	0.574(24)	0.140(4)	0.219(7)
H(5′1)	0.604(10)	0.107(2)	0.420(3)
H(5′2)	0.795(10)	0.057(1)	0.497(3)

The 3-HB anions. Fig. 3 shows the geometry of the anion together with the bond lengths and angles. No significant differences in the dimensions of the two non-equivalent 3-HB residues were found, and in addition the bond lengths and angles are in agreement with literature values for comparable molecules, *e.g.* biuret,[8-9] and hydroxyurea.[10]

An analysis of planarity of the 3-HB residues was carried out by calculation of the best plane through the non-hydrogen atoms of each residue and in addition through the N, C, and O atoms of each half of the residues (*cf.* Table 5). The analysis clearly shows that the atoms defining plane 1 is more coplanar than the atoms defining plane 2. In residue 1 there is a twist of only 3.4° between the two $H_2N-(C=O)-N-$ moieties; the corresponding angle of residue 2 is 13.2°. Both residues have adapted the conformation that gives rise to an *intra*molecular NH···O hydrogen bond (*cf.* Fig. 2). The same conformation was found in the biuret molecule [8] and the perdeuterated biuret molecule,[9] where the length of the *intra*molecular hydrogen bond was found to be 2.72 Å and the twisting angle 6.3°.

The environment of the potassium ion. The potassium ion has eight oxygen neighbours belonging to six different 3-HB residues, with K−O distances in the range 2.663−3.026 Å (*cf.* Fig. 4). These values are in good agreement

Table 4. Thermal parameters for the non-hydrogen atoms. $T_i = \exp[-1/4\ (B_{11}h^2a^{*2} + \cdots + 2B_{23}klb^*c^*)]$.

	B_{11}	B_{22}	B_{33}	B_{12}	B_{13}	B_{23}
K^+	1.62(02)	2.12(02)	1.81(03)	0.19(02)	−0.07(02)	0.09(02)
N(1)	3.56(11)	1.95(09)	2.56(06)	0.65(10)	−0.44(10)	−0.14(09)
C(2)	1.71(11)	1.79(10)	1.76(11)	−0.22(06)	0.12(09)	0.09(09)
O(2)	3.75(11)	2.71(10)	1.91(09)	0.21(08)	−0.84(08)	−0.10(07)
N(3)	1.95(10)	1.51(09)	1.48(09)	0.13(08)	−0.03(07)	−0.35(07)
O(3)	2.55(09)	1.77(08)	1.86(08)	0.44(07)	0.28(07)	−0.38(07)
C(4)	1.58(11)	2.00(12)	1.72(11)	−0.59(10)	0.20(09)	−0.01(07)
O(4)	2.81(09)	2.15(09)	1.81(08)	−0.08(07)	−0.13(07)	−0.60(07)
N(5)	2.63(12)	2.37(11)	1.95(10)	0.25(09)	−0.47(09)	−0.02(09)
N(1′)	3.28(09)	2.01(11)	1.89(10)	0.89(10)	0.27(09)	0.08(09)
C(2′)	1.60(11)	1.69(11)	1.88(11)	−0.18(09)	−0.07(09)	−0.17(09)
O(2′)	2.89(09)	2.07(08)	1.52(08)	0.50(07)	0.02(07)	0.27(06)
N(3′)	1.82(10)	1.58(09)	1.69(09)	0.33(08)	0.05(08)	0.22(07)
O(3′)	2.15(09)	1.94(09)	2.15(09)	0.70(07)	0.29(07)	0.13(07)
C(4′)	1.77(11)	1.80(12)	1.86(11)	−0.30(09)	0.00(08)	0.13(09)
O(4′)	3.17(10)	1.98(09)	2.03(08)	0.44(08)	−0.29(07)	0.20(07)
N(5′)	3.44(13)	2.62(12)	1.72(10)	0.68(10)	−0.04(09)	−0.10(09)

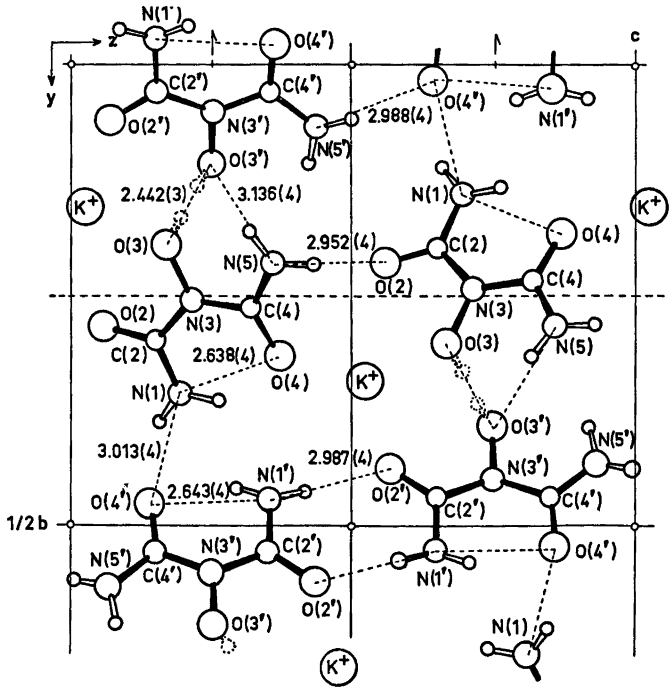

Fig. 2. The structure viewed along the *a*-axis.

Table 5. Least-squares planes. The equations of the planes can be expressed as $Px+Qy+Rz=S$ in direct space.

	Plane 1	Plane 2	Plane 3	Plane 4	Plane 5	Plane 6
P	3.304	3.253	3.343	3.227	3.326	3.337
Q	8.054	11.019	7.584	8.706	10.059	10.099
R	−4.258	0.083	−4.270	−4.391	1.546	−1.314
S	3.591	2.939	3.472	3.650	3.201	2.494

Distances in Å of the atoms from the planes.

Plane 1		Plane 2		Plane 3		Plane 4		Plane 5		Plane 6	
N(1)	0.03	N(1′)	−0.05	N(1)	0.001	N(3)	0.000	N(1′)	−0.003	N(3′)	−0.002
C(2)	0.00	C(2′)	0.02	C(2)	−0.003	C(4)	0.000	C(2′)	0.008	C(4′)	0.005
O(2)	0.00	O(2′)	0.16	O(2)	0.001	O(4)	0.000	O(2′)	−0.003	O(4′)	−0.002
N(3)	−0.02	N(3′)	−0.11	N(3)	0.001	N(5)	0.000	N(3′)	−0.002	N(5′)	−0.002
O(3)	−0.01	O(3′)	−0.15								
C(4)	−0.01	C(4′)	0.01								
O(4)	−0.04	O(4′)	−0.04								
N(5)	0.05	N(5′)	0.16								

Additional atoms:				Angles between normals to planes:	
H(11)	−0.04	H(1′1)	−0.04	1:2	21.9°
H(12)	−0.01	H(1′2)	0.10	3:4	3.6°
H(51)	0.09	H(5′1)	0.25	5:6	13.2°
H(52)	0.01	H(5′2)	0.32		
H(3)	−0.72	H(3′)	0.50		

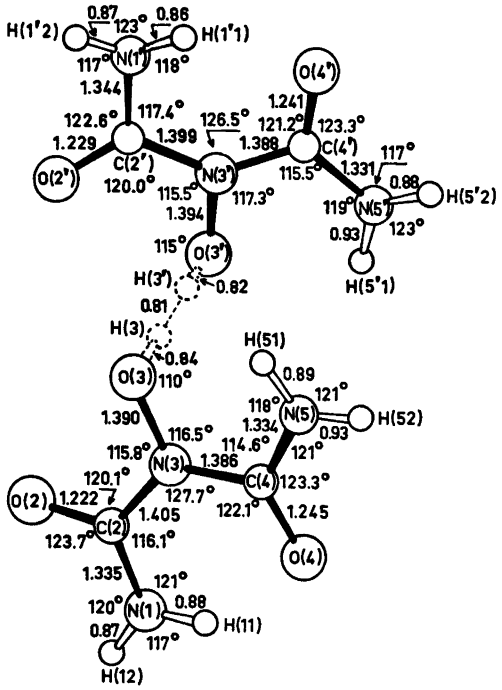

Fig. 3. Bond lengths and valency angles. The standard deviations on bonds between non-hydrogen atoms are 0.003 – 0.004 Å, and on the angles 0.2 – 0.3°. The standard deviations on bonds to hydrogen atoms are 0.03 – 0.04 Å and on angles including hydrogen atoms 2 – 6°.

with those earlier reported for eight-coordinated potassium,[3] *e.g.* in the structure of potassium hydrogen diformate.[11] As in this structure there is no regular geometry of the arrangement (*cf.* Table 6). It seems likely that the reason for this is the intensive *inter*molecular hydrogen bonding in the structure.

Two of the coordinating oxygen atoms, O(2) and O(3), belong to residue 1, and other two, O(2′) and O(3′), belong to residue 2 (*cf.* Fig. 4), but the planes through the chelate ligands do not pass through the central potassium ion. The displacements of K^+ from the planes defined by O(2) – (C2) – N(3) – O(3) and O(2′) – C(2′) – N(3′) – O(3′) are −1.51 and −0.47 Å, respectively. A similar situation was found in the potassium acid salts of isonitrosoacetophenone,[12] and the cyclic hydroxamic acid 5-bromo-3-hydroxy-6-methyl-uracil,[13] and is not a matter for surprise considering the electrostatic nature of the bonding. Nevertheless, the chelate rings in the structures of the potassium acid salts of carboxylic acids are often found to be planar.[11]

The number of ionic contacts to potassium is different for the two 3-HB residues: residue 1 has five and residue 2 has three contacts to K^+. This may mean that there is a tendency towards less ionization in residue 2 than in residue 1.

The distance between the layers of the potassium ions is about 6 Å ($\sim \frac{1}{2}c$). The nearest potassium-potassium contact is 3.868 Å ($=a$).

The short hydrogen bond. In most structures with very short hydrogen bonds, *e.g.* the acid salts of carboxylic acids, the short hydrogen bond has to be symmetric, because the acidic residues are crystallographically symmetry related. In the present structure the hydrogen bond, which links the 3-HB residues to a "dimer", has no crystallographical symmetry.

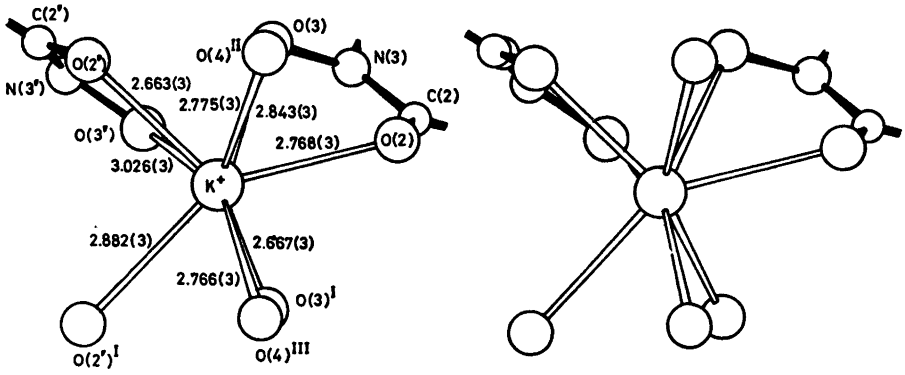

Fig. 4. Stereoscopic view of the potassium ion with its eight oxygen neighbours. In addition the chelate ligands of each 3-HB residue are indicated.

Table 6. The 28 independent O – K – O angles (°). The standard deviations are 0.05 – 0.07°.

O(2) – K$^+$ – O(3)	55.9	O(2′) – K$^+$ – O(3)^{I}	123.0
O(2) – K$^+$ – O(2′)	119.0	O(2′) – K$^+$ – O(2′)^{I}	88.4
O(2) – K$^+$ – O(3′)	96.0	O(2′) – K$^+$ – O(4)^{II}	79.6
O(2) – K$^+$ – O(3)^{I}	76.7	O(2′) – K$^+$ – O(4)^{III}	138.8
O(2) – K$^+$ – O(2′)^{I}	142.1	O(3′) – K$^+$ – O(3)^{I}	70.4
O(2) – K$^+$ – O(4)^{II}	74.3	O(3′) – K$^+$ – O(2′)^{I}	78.4
O(2) – K$^+$ – O(4)^{III}	94.8	O(3′) – K$^+$ – O(4)^{II}	121.1
O(3) – K$^+$ – O(2′)	66.6	O(3′) – K$^+$ – O(4)^{III}	150.2
O(3) – K$^+$ – O(3′)	49.1	O(3)^{I} – K$^+$ – O(2′)^{I}	66.0
O(3) – K$^+$ – O(3)^{I}	89.1	O(3)^{I} – K$^+$ – O(4)^{II}	149.7
O(3) – K$^+$ – O(2′)^{I}	127.2	O(3)^{I} – K$^+$ – O(4)^{III}	85.3
O(3) – K$^+$ – O(4)^{II}	81.9	O(2′)^{I} – K$^+$ – O(4)^{II}	140.5
O(3) – K$^+$ – O(4)^{III}	150.6	O(2′)^{I} – K$^+$ – O(4)^{III}	76.1
O(2′) – K$^+$ – O(3′)	54.4	O(4)^{II} – K$^+$ – O(4)^{III}	88.6

Roman numerals as superscripts denote the following equivalent positions relative to the reference molecule of x, y, z: I $x-1, y, z$; II $x, -y+\frac{1}{2}, z-\frac{1}{2}$; III $x-1, -y+\frac{1}{2}, z-\frac{1}{2}$.

*Table 7. Intera*tomic angles (°) concerning the hydrogen bonding system.

N(1) – H(11) – O(4)	131(3)
N(1) – H(12) – O(4′)^{IV}	156(3)
O(3) – H(3) – O(3′)	172(9)
O(3) – H(3′) – O(3′)	170(9)
N(5) – H(51) – O(3′)^{V}	148(3)
N(5) – H(52) – O(2)^{VI}	158(3)
N(1′) – H(1′1) – O(4′)	134(4)
N(1′) – H(1′2) – O(2′)^{VII}	176(4)
N(5′) – H(5′2) – O(4′)^{VIII}	172(4)

Roman numerals as superscripts denote the following equivalent positions relative to the reference molecule of x, y, z: IV $-x+1, y+\frac{1}{2}, -z+\frac{1}{2}$; V $x+1, y, z$; VI $x+1, -y+\frac{1}{2}, z+\frac{1}{2}$; VII $-x+2, -y, -z$; VIII $-x+2, -y, -z+1$.

But the short O···O distance of 2.442(3) Å, the favourable N – O···O angles of 109.8(3)° and 108.5(2)°, respectively, and the almost identical geometry of the 3-HB-residues being connected, are consistent with the existence of a virtually symmetrical hydrogen bond.[14–16] Symmetry due to statistical disorder is also possible. The rather elongated peak between the oxygen atoms of the difference map could perhaps be interpreted as an unresolved peak due to two half H-atoms about 0.4 Å on either side of the midpoint. The final refinement ($\sim R = 0.032$) was based on this assumption.

But the X-ray data were also consistent with an asymmetric hydrogen bond. Refinement of the positional parameters of the hydrogen atom, with a start position midway between the oxygen atoms, led to the following, perhaps most reliable, arrangement of the system: O(3) – H(3) = 1.31(4) Å, O(3′) – H(3) = 1.13(4) Å and the angle O(3) – H(3) – O(3′) = 179(3)°. In fact the O(3′) – H(3) bond has just the length corresponding to an O···O distance of 2.44 Å, using the curve, given by Lundgren,[16] of O – H distance as a function of O···O distance.

The conclusion must be that it is not possible from the X-ray data to establish whether this hydrogen bond is symmetric or slightly asymmetric.

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