ACTA CHEMICA SCANDINAVICA 27 (1973) 797–813

The Crystal and Molecular Structure of 4,5-Dichloro-3,6-pyridazinedione

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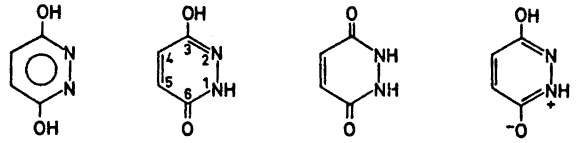
The crystal and molecular structure of 4,5-dichloro-3,6-pyridazinedione (dichloromaleic hydrazide) has been determined by X-ray methods using 2429 reflections above background level collected by counter methods. The crystals are orthorhombic, space group *Iba*2, with cell dimensions $a = 10.89_3$ Å, $b = 17.05_1$ Å, $c = 13.63_5$ Å. The structure model has been refined on the basis of a low-angle data set (1341 reflections with $\sin\theta/\lambda < 0.71$) and a high-angle data set (1317 reflections with $\sin\theta/\lambda > 0.61$). A comparison between the results of the two refinements is given. The nitrogen-nitrogen bond length is found significantly different in the two cases. Estimated standard deviations in bond lengths are about 0.004 – 0.006 Å, and in angles 0.3 – 0.4°. The molecule is found to be in the monolactim form, but the bond lengths indicate considerable resonance stabilization of the heterocycle. The hydrogen bond system is discussed.

Reactions of maleic hydrazids have been extensively studied (Ref. 1 and references therein). These compounds are interesting as starting materials for the synthesis of a number of drugs. It has also been found that maleic hydrazide is a powerful plant growth inhibitor.[2]

Maleic hydrazide has three possible tautomeric forms (I, II, and III):



From spectral evidence Druey *et al.*[3,4] concluded that the structure in the solid state corresponds to the monolactim (II). Further evidence for this structure is found in the reaction of maleic hydrazide with diazomethane,[5] in which a methoxy compound is rapidly formed, whereas the 1-methyl-3-methoxy

derivative forms more slowly. Reaction with dimethyl sulphate seems to yield 1-methyl-3-methoxy-6-pyridazone as the kinetic product, while 1,2-dimethyl-3,6-pyridazinedione is the end product.[1]

It has been pointed out that the monolactim permits resonance stabilization of the heterocycle.[5] The unshared pair of electrons on the single bonded nitrogen may be utilized to fill a molecular orbital, giving the resonance structure IV with an sp^2-hybridized N1-atom.

Both 6-amido-3-pyridazone [6,7] and 3-pyridazthione [8] are found to exist in the monolactim form in the crystal.

The main reason for the present structural work was to establish which of the tautomers exist in the solid state.

Errors in parameters connected with the asphericity of the electron density have been discussed.[9–11] Shifts in positional parameters for nitrogen atoms [9] and terminal oxygen atoms [10] may be significant. A refinement based on high-angle data would therefore be of interest.

EXPERIMENTAL

Dichloromaleic hydrazide was synthesized from dichloromaleic anhydride and hydrazine hydrochloride by the method of Mizzoni and Spoerri.[12] The product was recrystallized by slow evaporation of a 70 % ethyl alcohol solution. Rectangular, colourless plate-shaped crystals were formed.

Oscillation, Weissenberg and precession photographs indicated orthorhombic symmetry; reflections (hkl): $h+k+l$ odd, ($0kl$): k odd, and ($h0l$): h odd were systematically absent. Wilsons statistical method later applied on the intensity data indicated a non-centrosymmetric space group. It was therefore concluded that the space group was $Iba2$ with two molecules, called I and II, in the asymmetric unit.

Unit cell parameters were determined with the use of a Picker manual diffractometer using Cu$K\beta$ radiation ($\lambda = 1.3922$ Å) and a take off angle of 1.0°. 37 reflections and their Friedel equivalents were measured. The computer program used in least-squares calculations of cell parameters and programs employed in all subsequent calculations are part of an assembly of programs for CDC-3300 computer.[13]

Three-dimensional intensity data were recorded using an automatic Syntex-P$\bar{1}$ four-circle diffractometer with graphite monochromated Mo$K\alpha$ radiation. The take-off angle was 4°, and the temperature was kept constant within 1° at 18°C.

Intensity data were first recorded for reflections with 2θ less than 60°, and then for reflections with 2θ between 52° and 110°. A crystal of dimensions $0.9 \times 0.42 \times 0.14$ mm^3 was used for the data collection. The $w - 2\theta$ scanning mode was utilized with scan speed variable from 1° to 12° min^{-1} depending on the peak intensity of the reflection. Background counting time was equal to the scan time. Reflections for which the counts exceeded 10^5 cps were remeasured with reduced primary beam intensity. Intensities of six standard reflections were measured for every 50 reflections, and the data were adjusted according to the variations in the test reflection intensity.

Reflections for which the peak counts were less than 50 cps were not measured in the low-angle data set. All reflections with 2θ between 52° and 110°, which had calculated structure factors (based on the structure model found for the low-angle data set) higher than 5.0, were measured in the high-angle data set. The estimated standard deviations were taken as the square root of the total counts with a 2 % addition for adjustment uncertainty. All 1341 reflections measured in the low-angle data set had intensities higher than six times the standard deviation. 1601 reflections were measured in the high-angle data set. Of these, 1317 had intensities larger than twice the standard deviation. These were regarded as "observed" whereas the remaining reflections were excluded from further calculations.

The intensity data were corrected for Lorentz, polarization, and absorption effects.

Atomic form factors used were those of Doyle and Turner [14] for chlorine, oxygen, nitrogen, and carbon, and of Stewart *et al.*[15] for hydrogen. Anomalous scattering factors were obtained from Cromer and Liberman.[16]

CRYSTAL DATA

4,5-Dichloro-3,6-pyridazinedione (dichloromaleic hydrazide) $C_4H_2Cl_2N_2O_2$; m.p. 303 – 305°C; orthorhombic.

$a = 10.892$ (0.002) Å, $b = 17.051$ (0.003) Å, $c = 13.635$ (0.003) Å.

Figures in parentheses are estimated standard deviations. $V = 2532.3$ Å^3; $M = 181.0$, D_{obs} (flotation) $= 1.89$ g/cm^3; $Z = 16$; $D_{calc} = 1.899$ g/cm^3, $F(000) = 1472$; $\mu = 0.95$ cm^{-1}.

Absent reflections: (hkl) for $h+k+l$ odd, $(0kl)$ for k odd, $(h0l)$ for h odd. Space group $Iba2$.

STRUCTURE DETERMINATION AND REFINEMENTS

The structure was solved using the low-angle data. Positions for two of the four chlorine atoms were derived from the Patterson map. The positions for all other non-hydrogen atoms were found in a Fourier map based on these two positions. The structure model was refined to an R of 0.15. Introduction of anisotropic temperature factors for all non-hydrogen atoms and least-squares refinement lowered R to 0.043. Positions of all hydrogen atoms were localized in a difference Fourier map. Least-squares refinements of all positional parameters, anisotropic thermal parameters for non-hydrogen atoms, and isotropic thermal parameters for hydrogen yielded a conventional R of 0.032 and a weighted R_w of 0.019 (refinement A). The overdetermination ratio was 6.8.

The final parameters for non-hydrogen atoms are listed in Table 2 and for hydrogen in Table 3.

Magnitudes and directions of the principal axes of the ellipsoids of vibration are given in Table 4. The total discrepancy between the atomic vibration tensor components and those calculated from the rigid-body parameters found by analysis of the librational, translational, and screw motion of the molecules is 0.016 Å^2 for both molecules. The atomic positions were accordingly corrected for the librational motion.

At this stage the high-angle data were introduced. Refinement of positional and anisotropic thermal parameters for non-hydrogen atoms resulted in a conventional R of 0.066, and a weighted R_w of 0.025 (refinement B). The overdetermination ratio was 7.3. The final parameters are listed in Table 2. This structure model yielded a conventional R of 0.051 for the low-angle data.

A comparison of observed and calculated structure factors is given in Table 1.

Magnitudes and directions of the principal axes of the ellipsoids of vibration are given in Table 4. The total discrepancy between the atomic vibration tensor components and those calculated from the rigid-body parameters found by analysis of the librational, translational, and screw motion of the molecules is 0.019 Å^2 for molecule I and 0.014 Å^2 for molecule II. This indicates that the molecules may be regarded as rigid bodies. The atomic positions were accord-

Table 1. Observed and calculated structure factors. Data based on refinement B. The columns are h, k, l, $10|F_o|$, $|10F_c|$.

0	0	2	168	158
0	0	4	1351	1290
0	0	6	1717	1700
0	0	8	1916	1952
0	0	10	1301	1352
0	0	12	1027	1033
0	0	14	1235	1246
0	0	16	220	219
0	2	0	418	446
0	2	2	1097	1095
0	2	4	1595	1471
0	2	6	1245	1080
0	2	8	1465	1384
0	2	10	461	453
0	2	12	709	707
0	2	14	258	259
0	2	16	158	173
0	2	18	303	296
0	4	0	216	206
0	4	2	74	50
0	4	4	1997	2053
0	4	6	285	265
0	4	8	306	252
0	4	10	1877	1624
0	4	12	662	609
0	4	14	105	81
0	4	16	167	151
0	4	18	199	188
0	6	0	652	711
0	6	2	1485	1556
0	6	4	1577	1604
0	6	6	661	649
0	6	8	359	366
0	6	10	187	143
0	6	12	643	617
0	6	14	430	409
0	6	16	214	190
0	6	18	256	231
0	8	0	1027	1093
0	8	2	44	64
0	8	4	614	626
0	8	6	407	382
0	8	8	257	257
0	8	10	359	384
0	8	12	691	735
0	8	14	275	265
0	8	18	154	156
0	10	0	1084	1133
0	10	2	1299	1362
0	10	4	503	512
0	10	6	92	81
0	10	8	233	241
0	10	10	615	644
0	10	12	803	816
0	10	14	182	160
0	10	16	417	405
0	12	0	80	63
0	12	2	273	270
0	12	4	233	263
0	12	6	653	698
0	12	8	298	301
0	12	12	297	305
0	12	14	143	138
0	12	16	194	194
0	14	0	246	257
0	14	2	509	538
0	14	4	137	142
0	14	6	520	539
0	14	8	256	256
0	14	10	211	231
0	14	14	121	125
0	16	0	126	113
0	16	2	460	496
0	16	4	199	221
0	16	8	303	314
0	16	10	171	170
0	16	12	194	188
0	16	14	173	161
0	18	0	177	171
0	18	2	159	166
0	18	4	517	522
0	18	6	291	300
0	18	8	116	139
0	18	10	188	177
0	18	12	211	195
0	20	2	135	146
0	20	8	259	272
0	22	0	201	200
0	22	4	288	292
1	1	0	191	176
1	1	2	765	706
1	1	4	1226	1104
1	1	6	954	840
1	1	8	1064	944
1	1	10	262	241
1	1	12	444	441
1	1	14	288	264

1	1	18	214	209
1	2	1	870	814
1	2	3	450	426
1	2	5	450	393
1	2	7	163	158
1	2	9	264	192
1	2	11	569	557
1	2	19	191	184
1	3	0	1180	1195
1	3	2	1015	954
1	3	4	1294	1170
1	3	6	603	511
1	3	8	442	388
1	3	10	493	473
1	3	12	112	89
1	3	14	127	110
1	3	18	247	232
1	4	1	1357	1317
1	4	3	1486	1428
1	4	5	630	560
1	4	7	809	731
1	4	9	167	158
1	4	11	961	879
1	4	13	158	151
1	4	15	226	203
1	5	2	530	577
1	5	4	1413	1401
1	5	6	1094	1025
1	5	8	947	867
1	5	10	462	437
1	5	12	127	126
1	5	14	405	356
1	5	18	215	204
1	6	1	941	953
1	6	3	822	859
1	6	5	252	270
1	6	7	489	443
1	6	9	194	181
1	6	11	671	651
1	6	13	270	236
1	6	15	211	195
1	6	17	266	252
1	7	0	174	190
1	7	2	712	743
1	7	4	519	494
1	7	6	1218	1211
1	7	8	483	483
1	7	10	345	348
1	7	12	462	463
1	7	14	149	135
1	7	16	376	339
1	8	1	621	691
1	8	3	722	775
1	8	5	1035	1058
1	8	7	860	882
1	8	9	310	305
1	8	11	388	377
1	8	13	125	129
1	8	15	337	319
1	8	17	185	167
1	9	0	617	614
1	9	2	43	62
1	9	4	935	980
1	9	6	533	539
1	9	8	326	328
1	9	10	311	312
1	9	12	211	236
1	9	16	115	111
1	10	1	986	1060
1	10	3	488	532
1	10	5	291	278
1	10	7	685	695
1	10	9	112	106
1	10	11	324	348*
1	10	13	189	193
1	10	15	197	189
1	10	17	194	190
1	11	0	423	415
1	11	2	841	909
1	11	4	393	434
1	11	6	724	749
1	11	8	341	353
1	11	10	252	242
1	11	12	415	434
1	11	16	372	363
1	12	1	279	304
1	12	3	417	435
1	12	5	104	102
1	12	7	486	514
1	12	9	147	141
1	12	11	172	155
1	12	13	206	204
1	12	15	255	243
1	13	0	436	460
1	13	4	154	157
1	13	6	157	156
1	13	8	272	278

1	13	10	189	215
1	14	1	462	486
1	14	3	269	285
1	14	5	278	290
1	14	7	244	270
1	14	9	300	323
1	14	11	151	150
1	14	13	129	130
1	14	15	110	108
1	15	0	224	227
1	15	2	470	501
1	15	4	253	266
1	15	6	450	479
1	15	8	394	414
1	15	10	155	145
1	15	12	189	198
1	16	1	247	257
1	16	3	252	267
1	16	5	256	256
1	16	7	335	344
1	17	0	320	348
1	17	4	299	303
1	17	6	263	272
1	17	8	124	124
1	17	10	172	175
1	17	12	122	112
1	18	1	107	111
1	18	3	283	299
1	18	5	316	324
1	18	7	173	186
1	18	9	184	191
1	19	0	102	107
1	19	2	207	216
1	19	4	115	118
1	19	6	258	270
1	19	8	166	183
1	20	1	155	155
1	20	3	155	160
1	20	5	154	158
1	21	0	160	165
1	21	4	184	188
1	21	6	129	136
1	22	5	155	152
2	0	0	324	299
2	0	2	195	153
2	0	6	318	300
2	0	8	322	274
2	0	10	1947	1841
2	0	12	689	654
2	0	16	111	106
2	0	18	217	207
2	1	5	313	286
2	1	7	366	313
2	1	9	225	230
2	1	11	957	916
2	1	15	209	187
2	1	17	146	145
2	2	0	309	287
2	2	2	1382	1262
2	2	6	963	836
2	2	8	854	790
2	2	10	371	342
2	2	14	390	364
2	2	18	339	318
2	3	1	1120	1053
2	3	3	528	501
2	3	5	1396	1257
2	3	7	701	651
2	3	9	383	326
2	3	11	684	654
2	3	15	171	171
2	3	17	155	138
2	4	2	101	93
2	4	4	1092	998
2	4	6	910	857
2	4	8	1009	962
2	4	10	422	369
2	4	12	941	888
2	4	14	814	739
2	4	16	187	168
2	5	3	233	236
2	5	5	154	127
2	5	7	594	542
2	5	9	95	95
2	5	11	364	364
2	5	13	108	118
2	5	15	233	211
2	6	0	691	712
2	6	2	1337	1314
2	6	4	918	909
2	6	6	560	513
2	6	8	720	691
2	6	10	594	556
2	6	12	771	712
2	6	16	322	295
2	7	1	377	395
2	7	3	395	368

2	7	5	577	554
2	7	7	259	258
2	7	9	447	412
2	7	11	243	226
2	7	13	299	277
2	7	15	259	232
2	7	17	203	177
2	8	0	251	256
2	8	2	251	242
2	8	4	391	386
2	8	6	413	436
2	8	8	333	315
2	8	10	582	554
2	8	12	429	403
2	8	14	156	133
2	9	1	510	542
2	9	3	185	201
2	9	5	164	168
2	9	7	186	192
2	9	9	339	351
2	9	11	182	201
2	10	0	50	70
2	10	2	747	804
2	10	4	399	397
2	10	6	370	389
2	10	8	183	178
2	10	10	114	126
2	10	12	279	273
2	10	14	162	172
2	11	1	483	478
2	11	3	107	102
2	11	5	261	275
2	11	7	497	515
2	11	9	231	236
2	11	11	179	182
2	11	13	258	249
2	12	0	377	385
2	12	2	452	485
2	12	4	249	264
2	12	6	241	239
2	12	8	367	374
2	12	10	278	289
2	13	1	189	208
2	13	3	229	237
2	13	5	659	674
2	13	7	319	338
2	13	9	183	181
2	14	0	486	522
2	14	2	498	541
2	14	4	458	481
2	14	6	223	238
2	14	8	120	137
2	14	10	294	302
2	14	12	396	407
2	15	1	251	279
2	15	3	219	238
2	15	5	176	188
2	15	7	219	234
2	15	9	169	166
2	15	13	151	151
2	16	2	182	193
2	16	4	110	125
2	16	6	146	154
2	16	8	288	311
2	16	12	177	192
2	17	1	104	100
2	17	5	261	258
2	17	7	115	131
2	18	0	266	282
2	18	2	180	186
2	18	4	191	209
2	18	6	195	200
2	18	8	184	183
2	19	3	189	197
2	19	7	202	206
2	20	2	182	195
2	22	4	159	155
3	1	0	107	78
3	1	2	993	960
3	1	4	1792	1648
3	1	6	762	682
3	1	8	336	322
3	1	10	311	305
3	1	12	290	265
3	1	14	258	239
3	1	18	260	246
3	2	1	1278	1189
3	2	3	800	747
3	2	5	363	361
3	2	7	465	405
3	2	9	317	275
3	2	11	622	600
3	2	13	391	316
3	2	15	249	222
3	2	17	265	244
3	3	0	1008	973
3	3	2	753	725

3	3	4	323	275
3	3	6	1333	1202
3	3	8	892	811
3	3	10	152	132
3	3	12	553	553
3	3	14	297	266
3	3	16	311	281
3	4	1	1193	1148
3	4	3	377	383
3	4	5	946	872
3	4	7	783	734
3	4	9	442	399
3	4	11	412	378
3	4	13	112	115
3	4	15	259	242
3	4	17	129	124
3	5	2	487	479
3	5	4	966	943
3	5	6	333	341
3	5	8	496	483
3	5	10	725	704
3	5	12	487	473
3	5	18	221	212
3	6	1	1147	1129
3	6	3	359	348
3	6	5	124	119
3	6	7	970	936
3	6	11	173	149
3	6	13	199	190
3	6	15	202	189
3	7	0	577	514
3	7	2	1037	1063
3	7	4	924	921
3	7	6	279	269
3	7	8	293	272
3	7	10	328	293
3	7	12	359	341
3	7	16	341	313
3	7	18	158	136
3	8	1	921	948
3	8	3	906	887
3	8	5	354	349
3	8	7	452	449
3	8	9	325	305
3	8	11	704	654
3	8	13	309	277
3	8	15	403	359
3	8	17	140	133
3	9	0	692	720
3	9	2	254	267
3	9	4	307	305
3	9	6	270	276
3	9	8	393	385
3	9	10	404	405
3	9	12	313	305
3	9	14	262	259
3	10	1	247	235
3	10	3	659	673
3	10	5	334	346
3	10	7	179	188
3	10	9	388	406
3	10	11	425	435
3	11	2	343	364
3	11	4	348	358
3	11	6	747	775
3	11	8	573	596
3	11	10	169	168
3	11	12	212	213
3	11	14	166	154
3	12	1	450	493
3	12	3	475	516
3	12	5	486	500
3	12	7	529	554
3	12	9	240	236
3	12	11	251	252
3	12	13	294	290
3	12	15	161	149
3	13	0	399	413
3	13	4	335	337
3	13	6	139	142
3	13	8	127	144
3	13	12	274	286
3	13	14	118	129
3	14	1	346	377
3	14	3	462	503
3	14	5	387	400
3	14	7	212	234
3	14	9	240	239
3	14	11	200	210
3	14	13	153	145
3	15	0	173	175
3	15	2	268	302
3	15	4	273	295
3	15	6	380	408
3	15	12	196	204
3	16	1	166	173
3	16	3	283	301

3	16	5	125	144
3	16	7	131	142
3	16	9	187	198
3	17	0	209	218
3	17	4	128	145
3	17	10	150	159
3	18	1	367	382
3	18	5	187	191
3	18	7	178	179
3	18	9	165	157
3	19	0	143	157
3	19	2	166	172
3	21	0	143	144
3	21	6	185	185
4	0	2	128	98
4	0	4	1211	1117
4	0	6	675	636
4	0	8	468	452
4	0	10	373	394
4	0	12	787	803
4	0	14	505	484
4	0	16	236	225
4	0	18	233	223
4	1	1	1143	1137
4	1	3	317	309
4	1	5	1145	1080
4	1	7	1280	1203
4	1	9	335	313
4	1	11	337	312
4	1	13	179	176
4	1	15	359	352
4	1	17	161	154
4	2	2	621	614
4	2	4	1291	1217
4	2	6	713	665
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4	4	14	241	209
4	4	16	154	146
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Table 1. Continued.

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5	4	3	204	198
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6	14	6	181	187
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7	16	11	148	154
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Table 1. Continued.

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11	9	4	15[illegible]	156	7	5	16	343	344	3	4	21	174	171	0	12	14	137	138	5	21	8	144	132	9	10	17	104	115
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11	12	3	183	254	11	1	14	285	279	4	2	18	167	159	0	0	26	103	127	5	4	17	112	[illegible]	10	17	1	135	131
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Table 1. Continued.

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13	4	3	164	172	2	12	16	79	99	4	16	20	81	82	6	1	29	79	68	8	8	22	75	86	11	20	5	74	65
13	6	3	133	139	2	11	17	96	97	4	13	21	69	82	7	22	1	82	75	8	14	22	73	54	11	15	6	81	65
13	8	3	172	166	2	15	17	70	67	4	2	22	85	84	7	19	2	97	96	8	1	23	78	62	11	19	6	96	98
13	10	3	162	161	2	10	18	97	84	4	4	22	91	93	7	22	3	74	69	8	6	24	71	64	11	15	8	100	84
13	7	4	138	144	2	20	18	80	79	4	6	22	97	94	7	19	4	73	71	8	1	25	93	72	11	17	8	82	91
13	10	5	169	121	2	17	19	74	80	4	14	22	95	94	7	17	6	83	95	9	21	0	75	65	11	19	8	67	100
13	9	6	125	118	2	16	20	88	92	4	3	23	76	89	7	21	6	88	59	9	22	1	71	73	11	14	9	64	62
13	12	7	139	142	2	1	21	74	91	4	7	23	69	69	7	23	6	70	86	9	25	2	71	72	11	1	10	83	91
13	3	8	117	126	2	7	21	72	68	4	15	23	78	50	7	19	8	78	59	9	18	3	79	80	11	3	10	78	57
13	6	9	118	113	2	13	21	82	61	4	5	25	85	52	7	21	8	60	61	9	23	4	87	57	11	5	10	107	103
13	3	14	164	163	2	2	22	85	75	4	17	25	73	51	7	23	8	72	63	9	17	6	108	95	11	9	10	88	81
13	1	16	137	130	2	6	22	89	83	4	0	26	80	85	7	17	10	105	95	9	19	6	103	94	11	11	10	67	58
13	5	16	163	111	2	1	23	72	84	4	6	30	72	64	7	19	10	71	58	9	16	7	71	96	11	13	10	67	94
14	1	1	132	131	2	3	23	76	73	5	21	0	60	74	7	21	10	103	98	9	18	7	120	105	11	15	10	83	81
14	3	1	114	116	2	9	23	70	77	5	23	0	76	55	7	12	11	74	86	9	15	8	84	91	11	17	10	92	94
14	5	1	165	170	2	2	24	93	90	5	25	0	71	52	7	18	11	76	83	9	19	8	78	71	11	2	11	63	55
14	7	3	144	151	2	8	24	83	58	5	23	2	92	106	7	20	11	112	104	9	21	8	80	81	11	4	11	76	75
14	0	4	164	164	2	14	24	85	63	5	25	2	91	86	7	11	12	80	88	9	16	9	89	88	11	9	12	83	57
14	6	4	162	162	3	23	0	75	73	5	20	3	76	83	7	13	12	62	74	9	14	11	87	78	11	11	12	93	85
14	10	4	136	146	3	24	1	95	85	5	24	3	101	95	7	14	13	63	82	9	20	11	86	94	11	15	12	66	70
14	1	5	126	130	3	21	2	97	89	5	19	4	110	115	7	18	13	88	89	9	13	12	71	86	11	17	12	75	94
14	0	6	126	134	3	23	2	104	95	5	21	4	77	66	7	13	14	71	78	9	21	12	82	96	11	6	13	90	84
14	3	7	101	114	3	25	2	91	79	5	22	5	84	87	7	2	15	64	69	9	4	13	76	82	11	8	13	92	92
14	5	7	112	122	3	22	3	98	99	5	21	6	103	94	7	6	15	83	90	9	14	13	94	90	11	16	13	96	89
14	13	7	174	164	3	28	3	74	70	5	18	7	99	98	7	8	15	97	99	9	3	14	61	53	11	7	14	63	71
14	2	8	137	147	3	21	4	87	72	5	20	7	110	113	7	10	15	65	61	9	15	14	82	93	11	9	14	74	74
14	2	8	131	121	3	20	4	91	109	5	17	8	104	98	7	12	15	84	85	9	2	15	102	103	11	4	15	71	69
14	3	9	164	155	3	20	5	92	108	5	25	8	69	69	7	18	15	90	80	9	6	15	75	74	11	15	16	119	112
14	1	13	125	121	3	23	6	104	106	5	16	9	57	54	7	3	16	112	105	9	10	15	77	49	11	2	17	105	93
15	9	2	112	120	3	20	7	81	69	5	24	11	76	61	7	17	16	89	93	9	12	15	89	69	11	4	17	101	104
15	2	3	136	150	3	24	7	94	74	5	15	12	88	83	7	2	17	70	84	9	14	15	78	80	11	8	17	87	69
15	7	4	117	119	3	19	8	115	108	5	12	13	91	93	7	4	17	85	83	9	5	16	96	88	11	3	18	79	80
15	2	5	169	158	3	23	8	98	96	5	22	13	64	65	7	10	17	91	59	9	7	16	111	94	11	7	18	84	86
15	1	8	132	126	3	20	9	106	102	5	11	14	77	62	7	12	17	70	76	9	13	16	107	99	11	11	18	91	92
16	3	3	132	142	3	19	10	107	95	5	19	14	75	83	7	14	17	69	84	9	19	16	101	87	11	2	19	91	95
16	3	5	141	139	3	21	10	114	99	5	6	15	98	81	7	1	18	63	54	9	9	18	85	69	11	10	19	71	64
17	9	2	131	136	3	18	11	64	74	5	10	15	89	87	7	5	18	104	85	9	4	19	70	66	11	2	21	75	94
0	20	0	77	64	3	19	12	84	72	5	12	15	109	77	7	9	18	87	102	9	12	19	85	73	11	9	26	83	64
0	24	2	104	99	3	16	13	99	76	5	16	15	96	98	7	11	18	82	72	9	14	19	90	99	12	0	0	99	99
0	26	2	106	99	3	20	13	74	72	5	7	16	75	72	7	6	19	88	91	9	1	20	83	90	12	12	0	57	54
0	24	4	72	86	3	14	15	78	79	5	15	16	89	85	7	8	19	71	93	9	7	20	85	91	12	16	0	62	96
0	20	10	62	55	3	16	15	73	90	5	21	16	93	84	7	14	19	76	66	9	9	20	86	85	12	13	1	96	101
0	26	10	71	65	3	18	15	82	88	5	2	17	135	139	7	18	19	87	76	9	13	20	70	63	12	15	1	67	57
0	8	16	92	79	3	20	15	74	58	5	18	17	97	87	7	3	20	66	63	9	2	21	80	71	12	17	1	100	102
0	14	16	87	93	3	9	16	71	67	5	15	18	77	57	7	12	19	86	82	9	6	21	95	92	12	19	1	68	59
0	24	16	73	72	3	11	16	93	102	5	14	19	80	84	7	18	19	77	76	9	1	22	90	80	12	12	2	87	103
0	18	18	99	101	3	13	16	69	82	5	18	19	71	86	7	3	21	76	63	9	4	23	75	79	12	14	2	85	85
0	20	18	91	97	3	17	16	71	62	5	5	20	74	88	7	5	20	92	96	9	8	23	87	72	12	20	2	71	84
0	6	20	80	83	3	6	17	78	72	5	9	20	71	75	7	7	23	91	82	9	11	24	98	81	12	9	3	92	89
0	10	22	78	66	3	12	17	63	65	5	4	21	79	80	7	9	23	85	64	9	5	24	85	63	12	15	4	107	99
0	2	24	96	91	3	12	17	103	93	5	14	21	69	66	7	17	23	87	88	10	18	0	81	79	12	18	4	91	84
0	8	26	69	71	3	22	17	83	70	5	1	22	76	73	7	2	21	78	92	10	19	1	66	78	12	20	4	82	93
0	10	26	83	71	3	1	23	84	72	5	5	22	67	74	7	6	21	77	65	10	21	1	76	75	12	9	5	101	102
0	0	28	71	65	3	11	20	79	88	5	11	22	75	79	7	2	23	68	72	10	19	3	90	95	12	17	5	63	79
1	25	0	75	59	3	15	20	104	90	5	2	23	69	79	7	4	23	102	90	10	18	4	105	115	12	12	6	103	100
1	23	2	90	74	3	8	21	76	61	5	8	23	77	75	7	6	23	72	63	10	17	5	76	94	12	18	6	89	89
1	22	3	83	85	3	10	21	78	84	5	9	24	71	84	7	8	23	90	58	10	14	6	92	91	12	11	7	79	76
1	23	4	89	71	3	12	21	107	80	5	9	26	71	66	7	6	25	84	63	10	18	6	102	103	12	13	7	101	107
1	25	4	64	65	3	7	22	87	66	5	2	27	71	55	8	24	0	70	71	10	13	7	91	96	12	15	7	99	94
1	23	6	63	80	3	11	22	73	57	8	21	1	83	73	7	19	1	58	67	10	17	7	97	111	12	17	7	107	118
1	20	7	102	97	3	13	22	85	79	8	24	2	59	70	8	21	1	83	81	10	21	7	74	69	12	0	8	97	94
1	22	7	112	116	3	2	23	68	85	8	19	3	76	90	8	20	2	94	88	10	12	8	110	97	12	10	8	105	96
1	20	9	117	104	3	4	23	86	80	8	21	3	67	69	8	22	2	68	68	10	16	8	92	77	12	12	8	69	71
1	26	9	64	69	3	12	23	76	93	8	22	4	99	109	8	19	3	82	81	10	18	8	80	96	12	14	8	87	84
1	18	11	84	93	3	3	24	97	92	8	19	5	60	72	8	21	3	94	97	10	20	8	95	85	12	0	10	65	79
1	19	12	72	61	3	2	25	87	54	8	23	5	83	82	8	17	5	108	102	10	15	9	61	70	12	6	10	104	104
1	21	12	87	72	3	8	25	93	80	8	18	6	118	111	8	19	5	101	99	10	14	10	81	96	12	10	10	73	85
1	18	13	63	55	4	20	2	93	99	8	20	6	78	88	8	21	5	71	64	10	16	10	104	106	12	14	10	66	72
1	22	13	91	80	4	22	2	72	88	8	22	6	99	85	8	16	6	57	57	10	5	11	69	74	12	9	11	106	124
1	17	14	104	93	4	26	2	64	69	8	17	7	102	108	8	18	6	103	93	10	11	11	67	86	12	11	11	83	76
1	21	14	92	71	4	23	3	80	96	8	19	7	94	94	8	21	7	73	73	10	15	11	60	69	12	0	12	114	108
1	20	15	88	87	4	24	4	91	72	8	23	7	63	64	8	18	8	72	74	10	2	12	87	98	12	2	12	85	82
1	17	16	65	73	4	26	4	99	89	8	18	8	86	90	8	20	8	87	72	10	6	12	80	69	12	12	12	71	75
1	12	17	103	97	4	21	5	97	93	8	20	8	98	75	8	24	8	93	65	10	10	12	90	69	12	14	12	75	79
1	14	17	81	79	4	20	6	97	88	8	22	8	92	74	8	14	10	100	105	10	18	12	101	96	12	9	13	79	69
1	18	17	114	109	4	24	6	59	55	8	24	8	66	67	8	18	10	74	81	10	9	13	69	58	12	0	14	92	87
1	7	18	101	80	4	26	6	88	65	8	17	9	128	119	8	11	11	72	70	10	13	13	73	76	12	2	14	93	90
1	11	18	78	88	4	23	7	61	75	8	19	9	92	100	8	8	12	90	93	10	0	14	77	86	12	1	15	95	102
1	10	19	84	84	4	18	8	59	75	8	16	10	84	84	8	10	12	103	104	10	4	14	71	81	12	11	15	94	95
1	14	19	91	101	4	20	8	100	87	8	18	10	73	78	8	16	12	111	112	10	8	14	117	102	12	13	15	91	97
1	16	19	103	92	4	22	8	111	93	8	19	11	65	85	8	18	12	82	77	10	12	14	74	70	12	2	16	114	99
1	15	20	88	77	4	19	9	88	76	8	18	12	100	99	8	9	13	76	74	10	1	15	89	91	12	4	16	98	85
1	2	21	98	98	4	23	9	64	67	8	20	12	129	112	8	13	13	73	84	10	11	15	91	76	12	14	16	99	88
1	9	22	76	55	4	16	10	100	86	8	13	13	84	85	8	17	13	69	55	10	17	15	72	75	12	16	16	72	71

Table 1. Continued.

12	1	17	76	102	13	14	9	96	80	14	6	2	94	101	14	10	10	96	80	15	6	5	114	112	16	12	4	83	77
12	6	18	103	103	13	3	10	61	49	14	8	2	108	108	14	1	11	74	71	15	3	6	91	103	16	7	5	97	79
12	5	19	63	86	13	9	10	84	101	14	12	2	75	72	14	9	11	86	98	15	7	6	95	90	16	13	5	75	77
12	9	19	69	72	13	21	10	79	81	14	16	2	80	94	14	0	12	87	86	15	11	6	121	116	16	0	6	83	67
12	0	20	69	89	13	2	11	75	69	14	1	3	91	91	14	4	14	83	65	15	2	7	69	61	16	4	6	77	75
12	2	20	103	103	13	8	11	89	90	14	5	3	99	92	14	8	14	96	85	15	4	7	113	113	16	6	6	66	68
12	10	20	81	65	13	10	11	99	84	14	13	3	71	68	14	12	14	87	64	15	8	7	125	117	16	10	6	82	85
12	0	22	76	70	13	1	12	72	58	14	17	3	84	70	14	3	15	66	73	15	9	8	91	99	16	7	7	101	97
13	11	0	80	73	13	7	12	67	84	14	4	4	103	110	14	7	15	87	89	15	11	8	69	75	16	9	7	95	101
13	17	0	118	112	13	13	12	78	71	14	8	4	75	103	14	0	16	127	115	15	13	8	84	81	16	0	8	90	85
13	25	0	101	94	13	15	12	72	61	14	12	4	85	121	14	8	16	70	69	15	2	9	82	91	16	8	8	87	90
13	10	1	129	124	13	2	13	94	75	14	16	4	119	119	14	10	16	69	97	15	6	9	107	94	16	10	8	75	69
13	12	1	91	99	13	6	13	104	102	14	3	5	85	84	15	13	0	81	102	15	16	9	75	76	16	16	8	74	71
13	14	1	103	103	13	8	13	67	69	14	9	5	123	114	15	2	1	123	109	15	5	10	91	74	17	17	0	97	86
13	17	2	94	106	13	1	14	84	83	14	11	5	61	71	15	6	1	93	89	15	5	12	91	88	17	5	4	74	69
13	16	3	98	102	13	5	14	115	103	14	13	5	66	68	15	10	1	74	86	15	10	13	70	63	17	2	5	68	63
13	9	4	75	73	13	6	15	124	111	14	2	6	110	97	15	20	1	69	78	15	9	14	107	94	17	7	6	73	68
13	15	4	103	109	13	11	16	109	92	14	4	6	73	71	15	1	2	111	118	15	2	15	99	96	17	1	8	107	101
13	19	4	64	67	13	2	17	67	79	14	8	6	113	108	15	7	2	67	71	15	1	20	82	61	17	5	14	83	70
13	21	4	71	69	13	4	17	84	77	14	10	6	86	99	15	13	2	82	98	16	1	1	112	118	18	4	2	77	66
13	8	5	60	69	13	2	19	86	98	14	14	6	94	108	15	15	2	78	59	16	9	1	63	68	18	2	4	66	81
13	3	6	120	125	13	5	20	90	81	14	1	7	75	59	15	4	3	95	94	16	0	2	101	112	18	8	4	88	87
13	11	6	120	119	14	0	0	117	113	14	0	8	125	111	15	10	3	72	73	16	12	2	94	95	18	0	16	91	76
13	15	6	92	81	14	12	0	77	71	14	4	8	115	100	15	12	3	61	86	16	13	3	92	88	20	0	4	89	79
13	14	7	83	74	14	7	1	101	121	14	6	8	118	116	15	14	3	97	101	16	2	4	104	97	0	28	6	71	58
13	15	8	75	74	14	13	1	82	97	14	10	8	101	114	15	3	4	70	77	16	4	4	84	70	0	30	8	72	45
13	4	9	84	64	14	0	2	93	99	14	1	9	105	104	15	11	4	95	77	16	6	4	93	109	0	24	2	105	99
13	10	9	104	87	14	2	2	73	65	14	5	9	63	64	15	17	4	64	67	16	8	4	107	120	0	20	14	65	31

Table 2. Fractional atomic coordinates and thermal parameters with estimated standard deviations ($\times 10^5$) for non-hydrogen atoms. The first two lines are for low-angle data refinement, the next two lines for high-angle data refinement. The temperature factor is given by $\exp -(B_{11}h^2+B_{22}k^2+B_{33}l^2+B_{12}hk+B_{13}hl+B_{23}kl)$.

Atom	x	y	z	B_{11}	B_{22}	B_{33}	B_{12}	B_{13}	B_{23}
Cl2	−6506	9670	174	1163	380	284	−41	536	58
	10	5	8	9	3	4	10	11	7
	−6481	9641	176	1100	342	315	−50	560	54
	19	10	9	15	4	3	13	11	7
Cl1	1682	13579	22057	818	402	457	−439	182	−108
	9	6	8	8	3	5	9	11	80
	1707	13554	22099	726	370	493	−401	187	−112
	17	12	12	9	5	5	11	12	8
O1	−28424	−671	66	752	517	189	107	−70	−142
	23	19	19	22	11	10	27	28	20
	−28520	−729	68	688	491	204	116	−129	−135
	41	36	20	29	19	7	34	23	20
O2	−14374	6509	36370	802	416	185	−286	−170	19
	25	15	17	22	10	10	24	24	17
	−14409	6442	36463	714	393	201	−313	−166	27
	41	30	17	28	13	7	30	21	15
N1	−30296	−1402	16502	597	370	207	−74	−64	−84
	26	18	19	24	12	13	27	28	19
	−30355	−1354	16569	482	330	217	−94	−83	−34
	35	26	20	22	14	7	25	22	16
N2	−27254	151	26004	555	335	188	−9	1	6
	23	19	20	21	10	11	25	29	18
	−27283	105	25939	478	327	207	−73	2	13
	34	30	18	22	11	7	28	20	16

Table 2. Continued.

C3	−17823	4648	27410	554	253	198	59	−48	26
	28	18	21	26	10	13	26	29	19
	−17833	4637	27429	420	266	210	29	−25	5
	34	27	20	21	10	8	25	21	15
C4	−10739	7903	19510	574	238	247	23	10	17
	28	18	23	24	9	15	26	31	20
	−10688	7909	19448	544	208	237	15	−8	−27
	38	22	22	24	16	8	24	23	15
C5	−14212	6238	10200	663	277	205	110	144	30
	31	20	22	28	12	14	28	30	20
	−14258	6257	10132	532	283	209	51	124	−21
	41	28	20	26	12	7	28	22	16
C6	−24710	1198	8416	628	336	208	151	−61	−35
	29	21	23	24	13	15	30	36	22
	−24802	1197	8430	525	325	204	73	−39	−7
	44	32	21	24	13	8	27	23	17
Cl12	26860	23483	35416	850	414	493	501	−91	−37
	9	6	9	8	3	5	9	11	8
	26865	23467	35404	786	384	505	460	−120	−60
	18	12	13	10	5	5	12	13	9
Cl22	20381	28360	57575	1317	453	319	164	−619	103
	10	6	8	11	4	5	11	13	8
	20303	28334	57595	1216	404	340	151	−600	94
	22	13	10	16	5	3	15	13	7
O12	−674	39422	57973	970	397	223	−178	234	−70
	28	16	20	24	16	11	27	30	19
	−596	39394	57968	900	371	213	−109	165	−90
	50	31	20	35	15	7	35	29	17
O22	11757	31391	21444	765	371	198	258	102	−53
	22	14	17	22	9	10	24	26	17
	11750	31391	21425	723	328	218	205	78	−60
	40	27	18	28	12	7	30	23	15
N12	−3111	39941	41531	582	303	241	2	144	−60
	25	16	20	24	10	14	25	27	18
	−3127	39887	41500	497	248	253	55	103	−68
	34	25	21	21	10	8	23	22	16
N22	−481	38159	31931	607	292	195	20	6	−28
	23	17	20	21	9	12	26	26	19
	−558	38139	32038	449	217	228	43	−16	−39
	35	24	18	22	9	8	24	21	15
C32	8375	33283	30563	522	254	197	−77	−10	−22
	27	18	21	26	10	13	26	30	19
	8444	33307	30483	472	224	213	10	−4	3
	36	24	20	22	9	8	23	22	15
C42	15355	29766	38380	622	268	273	60	−66	−18
	29	19	23	26	11	16	28	33	21
	15342	29817	38373	560	243	249	136	−85	−34
	41	26	23	26	10	9	27	26	17

Table 2. Continued.

C52	12560	31811	47700	715	294	241	−103	−196	88
	31	20	24	30	11	15	31	34	22
	12434	31773	47727	719	255	236	109	−183	12
	52	29	23	35	11	9	35	28	18
C62	2615	37187	49558	660	304	224	−184	−8	−41
	30	21	25	29	11	15	28	36	21
	2569	37166	49567	610	272	209	−94	−1	−51
	44	28	23	26	12	8	28	25	17

Table 3. Fractional atomic coordinates ($\times 10^4$) and isotropic thermal parameters with estimated standard deviation for hydrogen atoms.

Atom	x	y	z	B
HN1	−3703	−522	1436	6.32
	40	27	41	1.37
HO2	−1859	346	4044	4.82
	37	25	37	1.13
HN12	−1006	4377	4302	3.07
	29	19	31	.79
HO22	635	3362	1747	4.30
	36	23	34	1.04

Table 4. R.m.s. amplitudes of vibration $(\overline{u^2})^{\frac{1}{2}}$ (Å) and B-values (Å^2) along the principal axes of vibration given by the components of a unit vector **e** in fractional coordinates ($\times 10^3$). Results from the low-angle data refinement are given in parentheses.

Atom	$(\overline{u^2})^{\frac{1}{2}}$	B	e_x	e_y	e_z
Cl12	.274(.284)	5.94(6.36)	57(58)	44(45)	−14(−8)
	.216(.215)	3.67(3.64)	6(0)	12(8)	72(72)
	.172(.178)	2.34(2.51)	72(71)	−36(−36)	5(6)
Cl22	.290(.299)	6.62(7.07)	83(84)	11(13)	−28(−24)
	.246(.259)	4.77(5.30)	−8(−13)	56(56)	19(17)
	.143(.139)	1.62(1.52)	37(34)	−12(−10)	65(66)
O12	.247(.262)	4.83(5.41)	63(65)	−41(−39)	14(14)
	.222(.224)	3.89(3.98)	66(61)	41(43)	2(7)
	.136(.138)	1.46(1.50)	−14(−19)	7(3)	72(71)
O22	.236(.251)	4.41(4.99)	56(53)	46(48)	−2(0)
	.193(.197)	2.93(3.05)	71(73)	−34(−32)	19(17)
	.139(.132)	1.52(1.37)	−17(−17)	11(8)	71(71)
N12	.195(.213)	3.00(3.57)	19(7)	56(57)	−16(−12)
	.178(.192)	2.50(2.91)	81(86)	−5(8)	34(23)
	.143(.142)	1.62(1.60)	−40(−31)	17(8)	63(68)

Table 4. Continued.

N22	.198(.208)	3.10(3.42)	15(12)	57(58)	−10(−4)
	.163(.192)	2.11(2.87)	−90(−91)	10(−8)	3(5)
	.145(.135)	1.67(1.44)	5(−1)	7(4)	73(73)
C32	.182(.200)	2.61(3.17)	9(53)	58(47)	1(2)
	.168(.180)	2.24(2.56)	−91(−75)	6(33)	2(−5)
	.142(.136)	1.58(1.45)	2(3)	−1(4)	73(73)
C42	.205(.204)	3.31(3.30)	59(54)	43(46)	14(−10)
	.169(.189)	2.24(2.81)	−66(−72)	39(36)	14(9)
	.150(.159)	1.78(1.99)	24(17)	1(2)	71(72)
C52	.218(.225)	2.76(4.01)	80(63)	25(38)	−17(−22)
	.189(.196)	2.81(3.03)	−35(−61)	52(43)	18(5)
	.141(.141)	1.57(1.57)	28(24)	−7(−9)	69(69)
C62	.208(.227)	3.42(4.06)	50(54)	−49(−47)	8(4)
	.184(.183)	2.67(2.64)	77(76)	31(34)	−8(−9)
	.139(.144)	1.52(1.64)	4(6)	9(7)	72(72)
Cl2	.275(.279)	5.98(6.15)	83(85)	−2(−1)	30(26)
	.226(.237)	4.02(4.45)	−2(−1)	58(58)	9(7)
	.140(.136)	1.54(1.45)	38(33)	7(6)	−66(−67)
Cl1	.269(.279)	5.70(6.15)	53(55)	−43(−44)	25(18)
	.208(.202)	3.41(3.22)	−11(−2)	20(18)	68(69)
	.171(.181)	2.31(2.58)	74(73)	34(34)	−8(−12)
O1	.273(.280)	5.90(6.17)	17(15)	57(57)	−11(−10)
	.202(.211)	3.23(3.51)	−89(90)	12(10)	10(3)
	.132(.129)	1.37(1.30)	16(6)	7(7)	72(72)
O2	.260(.265)	5.32(5.53)	−49(−49)	49(49)	7(6)
	.187(.202)	2.77(3.22)	74(75)	31(32)	−19(−14)
	.131(.127)	1.36(1.27)	25(19)	4(2)	71(71)
N1	.223(.236)	3.91(4.39)	19(14)	−57(−57)	3(9)
	.171(.189)	2.32(2.83)	84(89)	10(8)	−27(−13)
	.138(.135)	1.50(1.44)	32(15)	7(8)	68(71)
N2	.221(.222)	3.85(3.90)	16(−2)	58(58)	2(0)
	.168(.183)	2.22(2.63)	90(91)	10(1)	−1(0)
	.139(.133)	1.54(1.40)	0(0)	2(0)	73(73)
C3	.198(.197)	3.10(3.06)	9(41)	58(52)	1(2)
	.159(.180)	2.00(2.55)	−90(−81)	6(25)	13(12)
	.140(.135)	1.55(1.44)	16(12)	−2(−6)	72(73)
C4	.182(.190)	2.61(2.84)	86(54)	19(46)	−6(6)
	.175(.184)	2.42(2.66)	31(35)	−54(−74)	12(2)
	.148(.152)	1.74(1.83)	2(2)	11(5)	72(73)
C5	.206(.215)	3.34(3.66)	20(64)	57(40)	−1(13)
	.182(.189)	2.62(2.81)	84(62)	−12(−42)	25(10)
	.134(.134)	1.42(1.42)	−30(−21)	5(1)	69(71)
C6	.220(.230)	3.83(4.19)	18(39)	57(53)	−2(−6)
	.176(.186)	2.45(2.72)	−89(−82)	12(25)	8(5)
	.138(.139)	1.50(1.52)	11(10)	0(2)	73(72)

ingly corrected for the librational motion. The eigenvalues of **T** are 0.19, 0.17, and 0.14 Å,^{2} and 0.19, 0.18, and 0.14 Å^2 for molecules I and II, respectively. The r.m.s. librational amplitudes are 4.5, 3.7, and 2.5°, and 4.0, 3.2, and 2.8°, respectively. The major axes of libration are nearly parallel to a line through O1 – O2 and a line through O12 – O22 for the two molecules.

For both data sets it was found that effects caused by secondary extinction and anomalous scattering were insignificant.

Standard deviations were calculated from the correlation matrix ignoring standard deviations in cell parameters.

Differences between the results from the two refinements are significant. The thermal parameters are generally smaller in refinement B (see Table 4). This is in agreement with what is usually found.[11]

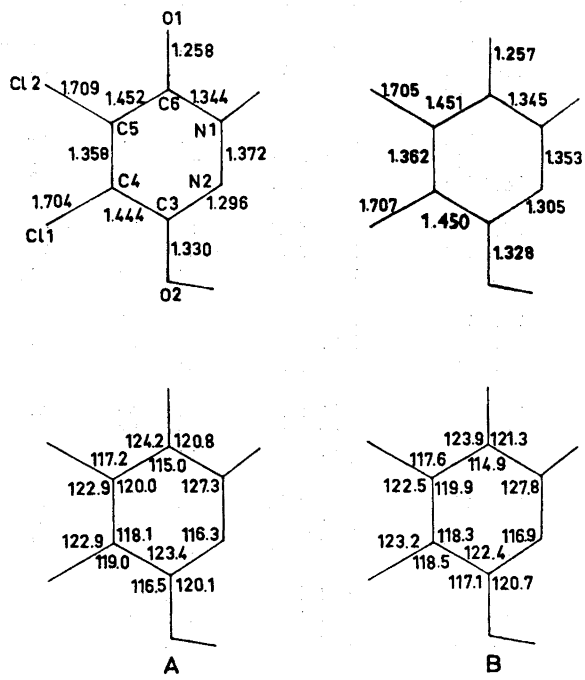

Fig. 1. Mean bond lengths (Å) (corrected for thermal vibration effects), and mean bond angles (°). A, results from low-angle data. B, results from high-angle data.

Mean bond lengths found in the two refinements are listed in Fig. 1. The O2 – C3 bond length, which was found significantly different in the two molecules after refinement A, is found less than one σ different after refinement B. The nitrogen bond length is found 0.019 Å shorter. In molecule II the nitrogen atoms are moved in towards the center of the ring in refinement B as compared with refinement A. This was also found in *s*-triazine.[17] But in molecule I the nitrogen atoms have only moved towards each other. This

is not as expected.[9] Further it was found that the thermal motion in the molecular plane had increased for some of the atoms. It has been noted earlier that parameter shifts attributed to valence electron influence is not a function of the electron density only, but rather of the total crystallographic-experimental environment.[18]

In the discussion only results from the refinement based on high-angle data will be used.

DISCUSSION

Bond lengths and bond angles are listed in Tables 5 and 6 and also in Fig. 1, where the numbering of the atoms is indicated.

The monolactim nature of the molecule is revealed not only by the different carbon-nitrogen and carbon-oxygen bond lengths, but also by the location of the hydrogen atoms on O2 and N1.

Table 5. Bond lengths (Å). Estimated standard deviations in parentheses. Results from the low-angle data refinement are given on the second line for each bond. The first three columns are for molecule I, the next three for molecule II.

	Bond length	E.s.d. ($\times 10^4$)	Corrected bond length	Bond length	E.s.d. ($\times 10^4$)	Corrected bond length
Cl1 – C4	1.697	(37)	1.702	1.706	(48)	1.711
	1.699	(32)	1.705	1.698	(33)	1.703
Cl2 – C5	1.701	(46)	1.706	1.700	(40)	1.704
	1.708	(31)	1.713	1.699	(32)	1.704
O1 – C6	1.254	(40)	1.256	1.255	(41)	1.257
	1.250	(41)	1.252	1.261	(44)	1.263
O2 – C3	1.323	(36)	1.326	1.327	(38)	1.330
	1.317	(36)	1.320	1.336	(35)	1.339
N1 – N2	1.344	(36)	1.348	1.353	(38)	1.357
	1.363	(37)	1.367	1.374	(37)	1.377
N2 – C3	1.303	(54)	1.307	1.298	(53)	1.302
	1.296	(43)	1.301	1.278	(42)	1.291
C3 – C4	1.449	(48)	1.454	1.441	(49)	1.445
	1.437	(42)	1.442	1.440	(42)	1.445
C4 – C5	1.358	(44)	1.363	1.356	(48)	1.360
	1.357	(44)	1.359	1.352	(45)	1.357
C5 – C6	1.455	(74)	1.460	1.436	(76)	1.441
	1.451	(57)	1.456	1.442	(59)	1.447
N1 – C6	1.337	(47)	1.341	1.345	(42)	1.349
	1.335	(42)	1.339	1.344	(43)	1.349
O2 – HO2	0.87			0.88		
	0.89			0.89		
N1 – HN1	1.03			1.03		
	1.02			1.02		

Table 5. Continued.

Hydrogen bond lengths	
O2 – O1 $(x, -y, \frac{1}{2}+z)$	2.598 2.611
N1 – O12 $(x-\frac{1}{2}, y-\frac{1}{2}, z-\frac{1}{2})$	2.953 2.954
O22 – O12 $(-x, y, -\frac{1}{2}+z)$	2.589 2.590
N12 – O1 $(-\frac{1}{2}-x, \frac{1}{2}-y, \frac{1}{2}+z)$	2.963 2.957

Table 6. Bond angles (°). Estimated standard deviations in parentheses. Results from low-angle data refinement are given on the second line. The first columns are for molecule I, the last columns for molecule II.

Atom	Bond angle	E.s.d.	Bond angle	E.s.d.
Cl1 – C4 – C5	123.0	(.31)	123.4	(.31)
	122.2	(.25)	123.5	(.26)
Cl1 – C4 – C3	119.1	(.25)	117.9	(.27)
	119.6	(.23)	118.4	(.24)
Cl2 – C5 – C6	117.8	(.24)	117.4	(.29)
	117.1	(.23)	117.2	(.26)
Cl2 – C5 – C4	122.2	(.37)	122.8	(.39)
	122.8	(.27)	122.9	(.29)
O1 – C6 – C5	123.7	(.40)	124.0	(.40)
	124.0	(.30)	124.4	(.33)
O1 – C6 – N1	121.6	(.49)	121.0	(.45)
	121.4	(.31)	120.2	(.31)
O2 – C3 – N2	120.4	(.31)	120.9	(.31)
	120.4	(.28)	119.8	(.27)
O2 – C3 – C4	117.4	(.36)	116.8	(.38)
	116.7	(.28)	116.3	(.27)
N1 – N2 – C3	117.0	(.30)	116.8	(.31)
	116.6	(.27)	115.9	(.27)
N2 – C3 – C4	122.4	(.27)	122.3	(.28)
	122.9	(.27)	123.9	(.27)
C3 – C4 – C5	117.9	(.37)	118.6	(.29)
	118.1	(.29)	118.0	(.30)

Table 6. Continued.

C4–C5–C6	119.9	(.32)	119.8	(.33)
	120.1	(.28)	119.9	(.30)
C5–C6–N1	114.7	(.30)	115.0	(.29)
	114.7	(.28)	115.3	(.29)
C6–N1–N2	128.1	(.39)	127.5	(.35)
	127.6	(.28)	127.0	(.27)

The mean nitrogen-nitrogen bond length of 1.353 Å is significantly longer and the mean C4–C5 bond length of 1.362 Å significantly shorter than the corresponding bond lengths of 1.330 Å and 1.395 Å, respectively, found in pyridazine.[19] The nitrogen-nitrogen bond and both of the nitrogen-carbon bonds (mean lengths 1.305 Å and 1.345 Å) are significantly shorter than in diacetylhydrazine.[20] The nitrogen-nitrogen bond length is also shorter than those reported for phenylhydrazine [21] and phenylhydrazine hydrochloride.[22]

The double bonds are more localized in dichloromaleic hydrazine than in pyridazine, but resonance stabilization of the heterocycle is still evident.

On the basis of these results it may be concluded that both the nitrogen atoms in the ring are sp^2-hybridized.

The heterocycle is planar, the atoms being displaced from a least-squares plane through the six ring atoms by less than 0.006 Å for both molecules. The oxygen and the chlorine atoms deviate significantly from the plane (see Table 7).

Table 7. Deviations from a least-squares plane through the six ring atoms. The first column is for molecule I, the second for molecule II. (Molecule I: Eqn.: $(-0.0547\,X + 0.0471\,Y - 0.0003\,Z)\,R - 1.771 = 0$. (Molecule II: Eqn.: $(0.0591\,X + 0.0448\,Y - 0.0027\,Z)\,R - 4.777 = 0$.)

Atom	Deviation (Å)	Deviation (Å)
Cl1	−0.030	−0.010
Cl2	−0.038	0.055
O1	−0.021	0.030
O2	0.024	0.033
N1	0.002	−0.002
N2	−0.003	−0.003
C3	0.005	0.006
C4	−0.006	−0.004
C5	0.005	0.000
C6	−0.004	0.003

The chlorine-carbon bond lengths are as expected for bonds between chlorine and an sp^2-hydridized carbon atom. The distortion of the external angles from 120° at C4 and C5 is possibly caused by repulsion between the chlorine atoms in the molecule.

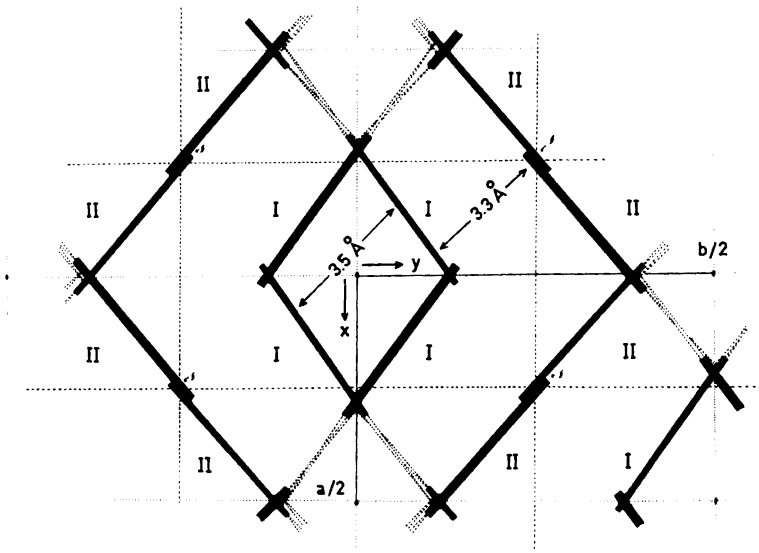

Fig. 2. The crystal structure as seen along the *c*-axis. The figure indicates the numbering of the molecules and the mean distances between the molecular planes. Hydrogen bonds are indicated by dotted lines.

The molecular arrangement in the crystal is visualized in Figs. 2 and 3 and may be described as layers parallel to (110). Within these layers the molecules are hydrogen bonded from O2(O22) to O1(O12).

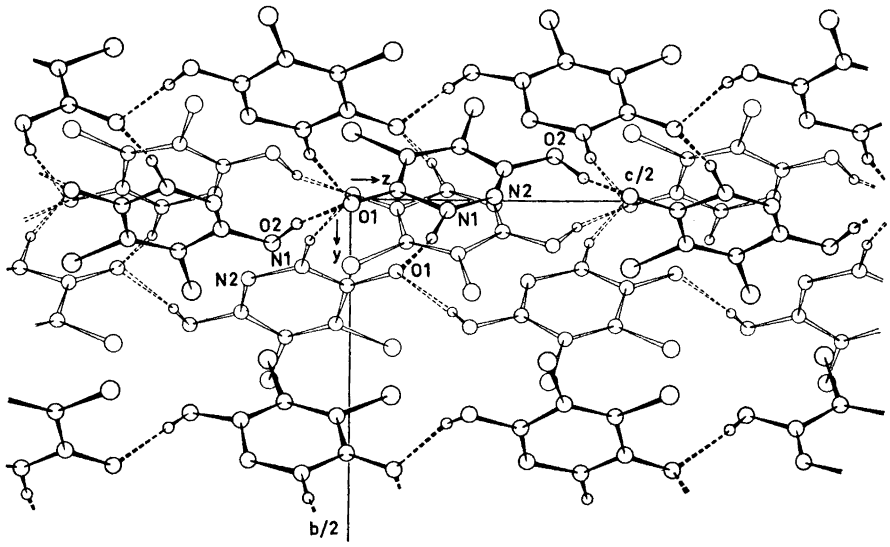

Fig. 3. The crystal structure as seen along the *a*-axis.

Each molecule of type I (II) is bonded to two molecules of type II (I) by hydrogen bonds parallel to (420) from N1 to O12 and from N12 to O1. The N1 – O12 hydrogen bond is close to the plane through molecule I and the N12 – O1 hydrogen bond to the plane through molecule II.

Between the layers the bonding seems to be dominated by van der Waals forces with many short contacts (3.2 – 3.6 Å) (see Fig. 3).

Acknowledgement. The author thanks Professor H. Hope for helpful discussions.

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Received September 11, 1972.