Table 4 (*cont.*)

C. Ba–O polyhedron

Interatomic distances (Å)	
Ba–O(1*a*+*c*)	2·85 (1)
Ba–O(3*c*)	3·02 (1)
Ba–O(1*a*)	2·88 (1)
Ba–O(3)	2·78 (1)
Ba–O(4)	2·85 (1)
Ba–O(2*c*)	2·81 (1)
Ba–O(2*a*+*c*)	2·88 (1)
Ba–O(4*c*)	2·92 (1)
Ba–O(5*b*)	3·22 (1)

D. Hydrogen bond

O(4)–O(3*c*)	2·86 (1) Å

A more satisfactory distribution might be obtained by assigning some arbitrary small value to the bonds between barium and bridging oxygen atoms; this would tend to equalize the electrostatic bond strengths of the Al–O bonds and to reduce slightly those of the remaining Ba–OH bonds.

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The Crystal and Molecular Structure of a Hydrazone Derivative of 2,2′-Di-(1,4-naphthoquinone)

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The title compound, $C_{21}H_{12}N_2O_3$, crystallizes in the orthorhombic space group *Pbca* with unit-cell dimensions of $a=14·45$, $b=8·22$ and $c=25·58$ Å, and with eight molecules per unit cell. Three-dimensional X-ray diffraction data were recorded on equi-inclination Weissenberg films using Cu $K\alpha$ radiation, and the 2196 intensities were visually estimated. The structure was solved with the symbolic addition method and has been refined with full-matrix least-squares to an R index of 0·10; anisotropic temperature factors were used for the carbon, nitrogen and oxygen atoms and the hydrogen atoms were included with isotropic temperature factors. The molecule is reasonably planar and structural requirements have produced two close intramolecular contacts, $O\cdots H$ and $CH_3\cdots O$. Bond distances in the heterocyclic portion of the molecule show the effects of π-electron delocalization.

Introduction

The reaction of 1,4-naphthoquinone (I) with hydrazine hydrate does not occur in the usual way to yield the familar ketonic hydrazone derivative. Hand & Cohen (1967) have reported that the naphthoquinone 'hydrazone' and the compound isolated from the reaction of hydrazine hydrate with 2,2′-di-(1,4-naphthoquinone) (II) are identical, and they have shown that the material is the cinnoline derivative (III*a*). Our investigation of the structure of (III) was stimulated by some recent

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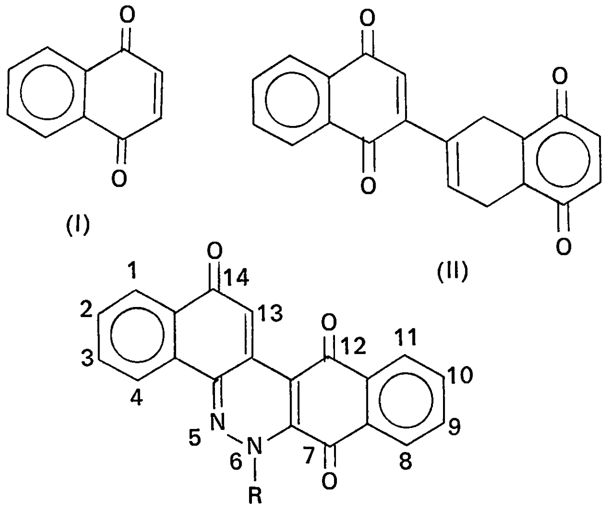
work on the crystal structure of compound (II) (Ammon, Sundaralingam & Stewart, 1969). The 'hydrazone' must be held in a rigid, planar configuration by the heterocyclic ring and the nonbonded interactions caused by such an arrangement should give rise to some interesting structural features. A determination of the crystal and molecular structure of the of the *N*-methyl derivative (III*b*), 6-methylbenzo[*g*]naphtho[1,2-*c*]-cinnoline-7,12,14-trione, is reported in this paper.



(I)

(II)

(III)

(III*a*) R=H; (III*b*) R=CH_3

Experimental

A generous sample of (III*b*) was obtained from Dr T. Cohen. The compound crystallized from anisole as blue-black, rectangular prisms. The crystals appeared to be constructed of small, but clearly visible, blocks and were somewhat fragile. Although recrystallization from anisole and other organic solvents was tried, in addition to high vacuum sublimation, we were not able to improve the crystal quality or grow larger specimens.

Cell constants were measured from rotation and Weissenberg photographs (Cu $K\alpha$, $\lambda = 1\cdot5418$ Å, calibrated with NaCl, $a = 5\cdot6396$ Å) of a sample mounted parallel to the long prism axis (*b*). The compound crystallizes in the orthorhombic system and unit cell parameters are

$$a = 14\cdot45_1,\ b = 8\cdot22_1,\ c = 25\cdot58_1\ \text{Å}.$$

For $C_{21}H_{12}N_2O_3$, mol. wt. 328·3 and $Z = 8$, $\varrho_{\text{X-ray}} = 1\cdot487$ g.cm^{-3}; $\varrho_{\text{meas}} = 1\cdot485$ g.cm^{-3}.* Systematic absences, $0kl$ absent for k odd, $h0l$ absent for l odd and $hk0$ absent for h odd, were consistent with space group *Pbca*. None of the specimens examined proved to be suitable for diffractometry: the crystals suffered from a substantial mosaic spread (peak widths of 5–6° in 2θ were observed at low 2θ) and all of the specimens were multiple crystals.

Intensity data subsequently were recorded using Ni-filtered Cu $K\alpha$ radiation on equi-inclination Weissenberg photographs. The $h0l-h7l$ levels were recorded with a crystal that was 0·1 mm square by 0·2 mm long, the $0kl-2kl$ levels were recorded with a crystal that was *ca.* 0·08 mm square by 0·1 mm long, and the intensities were visually estimated by comparison with a calibrated film scale. Although the crystal splitting was quite evident in certain regions of the *a*-axis photographs, we were able to estimate intensities in one octant where the spots were not noticeably split. Extended spot shape corrections (Lonsdale, 1964; the crystal to source distance was measured in the way described by Singh & Ramaseshan, 1967) were applied and the data were reduced using a set of IBM 1130 programs (Ammon, 1967). The data from the *a* and *b* axes were scaled together using the 'shortest path' method of Fox & Holmes (1966). Of the 2196 data (*ca.* 3473 data within the Cu sphere) measured out to a sin θ of *ca.* 0·86, 1263 (58%) were observed; the unobserved data were coded as such and given an intensity equal to the smallest observable intensity on the appropriate photographic level.

Table 1. *Starting set of reflections for construction of the phase pyramids*

Reflection	*E*	Phase	
9,0,14	3·123	+	origin
0,2,29	3·120	+	
11,3,18	3·524	+	
0,0,26	3·750	*a*	arbitrary
14,0, 0	3·340	*b*	
0,2,20	3·167	*c*	
12,3,19	4·279	*d*	
0,6,13	2·939	*e*	
0,4,16	2·896	*f*	

Table 2. *Summary criteria for the two best phase solutions*

Criterion	No. 6	No. 16
No. of + signs	93	97
No. of − signs	101	100
No. undetermined	14	11
No. correct Σ_2 triples	1998	2092
No. incorrect Σ_2 triples	274	174

The data were scaled and normalized structure-factor amplitudes ($|E|$) were computed using the *X-ray 67* (1967) subprogram *DATFIX*. The structure was solved by the symbolic addition procedure (Karle & Karle, 1966), using the *PHASER* system of programs (Ammon, 1964) modified to run on an IBM 360/40 computer. We first used the 155 $|E|$'s$\geq 2\cdot0$ (1128 $\sum_2$ triples) and the 'symbolic' phase pyramid generated by *PHASER*. The starting set of reflections necessary for the phase pyramid construction was readily narrowed

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* This density was measured by Dr H. Berman, who also made an initial determination of the cell parameters and space group (see Hand & Cohen, 1967).

Table 3. *Structure factor list*

The data are listed in blocks of constant h and k. Within each block the three columns are l, $10F_o$ and $10F_c$. An 'L' designates the unobserved data; an 'E' designates those reflections which were not included in the final cycles of least-squares refinement.

```
 L    FO     FC

 H =  0, K =  0
 2   432    426
 4   490    486
 6   184    187
 8   908E -1073
10   639   -692
12   251   -256
14    78L    58
16   488E   636
18   124    147
20   177   -208
22   148    135
24   149    129
26   521E  -671
28    93L    40
30   100    110

 H =  1, K =  0
 2  1475E  2217
 4   222   -137
 6   831   -865
 8   592    544
10    77L    13
12   291   -272
14   174    159
16   327   -303
18   115L     4
20   251    242
22   148L   -41
24   950E  1008
26   162    159
28   117    101

 H =  2, K =  0
 0  1814E -1879
 2    68    -66
 4  1017  -1037
 6   669    665
 8    66L   -36
10   771   -752
12   395   -366
14   792   -729
16   351    322
18   110L    68
20   166   -153
22   321    299
24   123L   -52

 H =  3, K =  0
 2   813    798
 4   164    149
 6   233    207
 8   333    302
10   321   -227
12   409   -386
14   724   -665
16    96L    67
18   149    137

 H =  4, K =  0
 0   200    183
 2   213   -179
 4    59L    66
 6   397    361
 8   314   -319
10   422    418
12    83L     2
14   349   -353

 H =  5, K =  0
 2   308   -266
 4   127   -121
 6   100    -85
 8   146   -101
10    81L   -56
12   307    266
14   192   -140

 H =  6, K =  0
 0   663   -631
 2    70L   -11
 4   106   -134
 6   253    237
 8   446   -434
10    86L    23
12   190    165
14    99L   -28
16   105L    70
18   137    106
20   113L    56
22   113L    45
24   108L   -55
26   154    163

 H =  7, K =  0
 2   852E  -933
 4   239    255
 6   369    329
 8   210   -169
10    92L   -55
12    97L    22
14   103L    54
16   109L   -52
18   113L   -52
20   114L   -72
22   112L    53
24   404   -385

 H =  8, K =  0
 0  1973E  2173
 2   231    193
 4   135   -117
 6    90L    50
 8   478   -498
10    98L    46
12   103L    15
14   412    356
16   111L     0
18   276    235
20   113L   -11
22   109L    92
24   101L    21
26   219   -196

 H =  9, K =  0
 2    93L   -13
 4   276    232
 6   303    206
 8   100L    -6
10   198    204
12   279    219
14   282    237
16   113L   -68
18   211   -204
20   111L    59
22   104L    43
24   170    132

 H = 10, K =  0

 L    FO     FC
 0   412    393
 2   110     94
 4   263   -171
 6   103L    69
 8   208    246
10   911   -851
12   570   -511
14   435   -391
16   437    395
18   228   -205
20   107L   -93
22   189    139

 H = 11, K =  0
 2   177    155
 4   315    279
 6   109L    35
 8   110L    67
10   113L    66
12   505   -496
14   592   -590
16   256    226

 H = 12, K =  0
 0   226    211
 2   111L   -42
 4   112L   -55
 6   175    144
 8   170   -147
10   399    357
12   113L    -9
14   207   -190

 H = 13, K =  0
 2   292   -261
 4   252    235
 6   164   -145
 8   113L   -43
10   168    159
12   110L   106
14   106L   -48
16    99L    27
18    91L   -42
20    99    -87

 H = 14, K =  0
 0   568    503
 2   113L   -15
 4   112L     8
 6   146    139
 8   223   -188
10    89    -79
12   104     97
14   114   -113
16   110     77
18   144    111
20   103    -97

 H = 15, K =  0
 2   107L    26
 4   106L    19
 6   104L    69
 8   102L   -29
10    97L   -37
12    92L   -11
14    84L   -74
16   152   -101

 H = 16, K =  0
 0    97L    34
 2    97L     3
 4    97    -80
 6    93L    25
 8   193   -172
10    84L    43
12    77L    49
14    97     93

 H = 17, K =  0
 2    83L   -25
 4   101    109
 6   224    199

 H = 18, K =  0
 0    64L     0
 2    63L   -44
 4    96    -78

 H =  1, K =  1
 1   126    130
 2   531    582
 3    55    -28
 5   827E   991
 6   135    154
 7   405   -508
 8   261    262
 9   300   -304
10    52    -79
11   324    322
12   270    278
13    50     55
14    66L    35
15    55     35
16   135   -126
17    76L    32
18    80L   -27
19   114   -119
20    85L   -12
21    87L     5
22   111   -114
23    89L   -15
24   249    280
25   151   -181
26    85L    11
27   117    142
28   114    143
29    69     79

 H =  2, K =  1
 0  1217E -1490
 1   297   -340
 2   302    325
 3   714E  -995
 4   742   -780
 5   340   -361
 6   115    -98
 7   182   -170
 8   368   -330
 9   432    396
10   787   -767
11   460    461
12    69     61
13   326   -302
14    63L   -31
15   234    217
16   321    292
17    75     67
18   113    115
19    81    -71
20    80L     7

 L    FO     FC
20    68L     7
21    91L   -54
22   100   -113
23   196    244
24   112   -126
25    69L   -23
26    67L   -53
27    64L   -34
28    60L     0
29   119    150

 H =  3, K =  1
 1   312   -267
 2   806    866
 3   272   -259
 4   165    197
 5   472    492
 6   599    678
 7    87     99
 8   546    558
 9   691   -672
10   667E  -696
11   949    998
12   466   -437
14   782   -806
15   576   -681
16   276   -281
17   140    162
18    65L    34
19    67L   -33
20    68L    23
21    70L   -44
22    96     96
23    71L    56
24    70L   -55
25    69L     6
26    66L     6
27    90    -92
28   115   -122

 H =  4, K =  1
 0   775E  -902
 1   261    277
 2   198   -194
 3   235   -226
 4   426   -427
 5   192   -214
 6   315    308
 7    51    -38
 8    44L    13
 9   733   -571
10   971    994
11   608   -604
12    91     92
13   340    323
14   425    429
15   216   -225
16   243   -239
17    64L   -46
18    66L    63
19    96     46
20    49L     0
21   111     83

 H =  5, K =  1
 1   323   -308
 2   311    303
 3   375   -379
 4   123    101
 5   180   -164
 6   590    562
 7   165   -147
 8   186    196
 9    85     49
10   144    124
11    53L    15
12    55L    56
13   128    164
14   315    264
15   305    261
16   135   -141
17   237    254
18    69L   105
19    69L    26
21    71L    36
22    71L    41
23    70L    50
24   119   -135
25    66L    42
26    64L   -62
27    60L   -74
28   107   -104

 H =  6, K =  1
 0   694    732
 1   264    306
 2   305   -277
 3   591    592
 4   316    336
 5    81    -60
 6    59    -46
 7   116    131
 8   356    345
 9    93     88
10    54L    41
11   167    155
12    71     49
13    60L   -65
14   139   -117
15    64L    40
16    66L   -20
17    68L    70
18   129   -155
19    70L    55
20    71L   -19
21    71L   -32
22    70L     7
23    69   -117
24    67     96
25    64L    -1
26    61L   -35
27    57L   -12
28    52L     4
29    98   -124

 H =  7, K =  1
 1    97   -105
 2   129    178
 3   226    229
 4   193    188
 5   270    250
 6    52L   -23
 7   190   -200
 8    77    100
 9    56L   -22
10    58L     6
11   140    136
12    61L    -3
13    63L    -2
14    65L    45
15    66L    19
16    68L   -93
17    69L    -7
18    70L    20
19    71L     2

 L    FO     FC
20    71L   -50
21    71L   -52
22    69L   -29
23    83    -86
24   122    160

 H =  8, K =  1
 0   171   -181
 1   186   -185
 2    76    -69
 3    54L    -1
 4   102   -101
 5    78     91
 6    80    105
 7   201   -209
 8    59L    30
 9   170    169
10    62L   -79
11    63L    30
12    64L    41
13    66L   -38
14   126   -120
15   108   -107
16    70L   -39
17    71L   -51
18    71L    63
19    71L     5
20   111   -112

 H =  9, K =  1
 1    58    -53
 2   117    138
 3    59L   -55
 4   181   -183
 5   127    109
 6   190    208
 7   107   -101
 8    63L   -14
 9    64L    39
10   138    116
11    66    -78
12   202    220
13    68     69
14    69L    22
15   131    134
16    71     66
17    71    -54

 H = 10, K =  1
 0   205    230
 1   140   -130
 2    63L     4
 3    63L    -4
 4   100    -95
 5    78     79
 6    55L    26
 7   245   -270
 8   220   -235
 9   378    382
10   337   -369
11   341    392
12    70L     3
13   395   -424
14   123   -150
15   236    279
16   212    251
17   111    126
18    69L   -11
19   144   -136

 H = 11, K =  1
 1    66L   -19
 2   200    216
 3   116   -120
 4   143    155
 5   203    210
 6   108     82
 7    68L    16
 8   317    299
 9   345   -378
10   308   -346
11   462    525
12   188   -239
13   100    -96
14   410   -447
15   389   -424
16   170   -204

 H = 12, K =  1
 0    98    125
 1    69L    34
 2    70L   -45
 3   121    154
 4    70L   -20
 5    99    -93
 6    86     74
 7    86    -73
 8    71L    28
 9   166   -152
10   296    356
11   235   -258
12   111    134
13   110     92
14   155    169

 H = 13, K =  1
 1    71L   -53
 2   233    274
 3   100     92
 4   142    160
 5   201    224
 6   159    191
 7   173   -198
 8    71L    19
 9    70L   -11
10    70L   -12
11    69L    89
12    69L   -49
13    67L    18
14    65L    25
15    64L    59
16    61L   -72
17    59L    74
18    56L    43
19    53L     0
20    49L    26
21    86    -71
22    88     91
23    76    -55

 H = 14, K =  1
 0   393   -432
 1   121   -143
 2   171   -213
 3   121   -136
 4    98   -178
 5    85     80
 6    97    146
 7    68L   -78
 8    93     91
 9    67L   -75
10   114    136
11    45L     1

 L    FO     FC
12    89    102
13    67L   -37
14    60L    -1
15    57L    40
16    55L   -74
17    51L    -7
18    49L   -36
19    44L    14
20    88   -125
21    76     91

 H = 15, K =  1
 1    66L    -7
 2    66L   -21
 3   129   -145
 4   127   -131

 H =  0, K =  2
 0    45L    29
 1   375    417
 2   514E  -704
 3  2062E -2630
 4  2338E -3013
 5   162   -159
 6  1273E  1494
 7   294    316
 8   435    414
 9   445   -447
10    58L    42
11   475E  -534
12   332    347
13   358    362
14    71L   -57
15   124   -107
16    80    -57
17   107   -102
18    71L  -216
19   632    690
20   802E  -939
21   152    164
22   134    132
23    77     86
24    91L    25
25   172    144
26    86L   -18
27    82L    16
28   232    265
29   292    392
30   165    194

 H =  1, K =  2
 1   619   -630
 2   336    355
 3   508   -570
 4  2333E -2950
 5  1889E -2144
 6   801   -884
 7   194    201
 8    42    -40
 9    60L   -28
10   339    371
11    66L    -4
12   339    302
13    73L   -36
14    60     49
15   108    -86
16    84L   -55
17    87L   -11
18   122   -129
19   167   -166
20   353   -342
21   491    527
22   316   -301
23    99L    34
24    98L   -68
25   126     89
26   168    151
27   181   -179
28   184   -190
29    76L   -21
30   144    135

 H =  2, K =  2
 0   305    308
 1   133    129
 2   529   -600
 3   572    599
 5   586   -546
 6   362    355
 7   197    179
 8   442    401
 9   742   -718
10   784    711
11   182   -136
12   178   -189
13   352    333
14    72L   -17
15   158   -142
16   115    147
17    82L    62
18   171   -171
19   127   -115
20   190    205
21   153   -162
22    77L    14
23    77L    86
24    76L    30
25    75    -92
26    72L   -15
27   103   -103
28    78    -90
29   136   -170
30   101    -95

 H =  3, K =  2
 1    69     76
 2   176    142
 3    67     41
 4    60    -68
 5   201    179
 6   591   -565
 7   179    200
 8    73     54
 9   265    221
10   171    146
11   274   -241
12    57L    11
13    60L     5
14   266    265
15   229    216
16    67     83
17    70L     0
18   278    282
19    76L    50
20   120   -116
21   109   -125

 H =  4, K =  2
 0   183    193
 1   338    340
 2    40L   -13
 3   198   -191
 4   229    230
 5   291   -234

 L    FO     FC
 6   231   -201
 7   214    150
 8   150   -172
 9   116   -125
10    54L    11
11    57L    16
12    59L    60
13   115     95
14    64L   -12
15   115   -115
16   155    197
17   124    153
18   116    110

 H =  5, K =  2
 1    44     47
 2    45L   -15
 3   130   -128
 4    47L    -9
 5    76    -92
 6   165    191
 7   146   -140
 8   223   -204
 9    55L    13
10    57L   -15
11    60    -62
12    62    -64
13    64     70
14    66    -71
15    69     34
16    71    -22
17    73    112
18   118    108
19    93     72

 H =  6, K =  2
 0    60    -85
 1   116    130
 2   234   -249
 3   348    323
 4   232    243
 5   149   -152
 6   115    -77
 7    68     48
 8   162    118
 9   110     97
10    61L    52
11    63L    25
12   130    126
13    67L    85
14    69L   -75
15   113   -103
16    73L   -47
17    75L    27
18    76L   -48
19    77L    12
20    77L   115
21    95   -126

 H =  7, K =  2
 1   134    108
 2   310   -270
 3   181    164
 4   726    683
 5   687    580
 6   396    376
 7   127   -100
 8    62L   -62
 9   109   -116
10   215   -245
11    66L    43
12    68L     7
13    70L    15
14    72L   -11
15    74L   -64
16    75L    19
17    76L    52
18    77L   -30
19    77L    81
20   144    156
21   170   -223
22   129    142

 H =  8, K =  2
 0    60L     1
 1   147    143
 2   357   -347
 3   445   -379
 4   552   -509
 5    62L   -32
 6   179    183
 7   327    328
 8   114    121
 9    67L    18
10   194   -174
11   141    141
12   161    169
13    73L   -43
14    75L   -47
15    76L    76
16    77L    39
17    77L    -1
18    77L   -76
19   361    343
20   142   -157

 H =  9, K =  2
 1    65L   -15
 2   221   -209
 3    66L    55
 4   401   -389
 5   287   -249
 6   117    116
 7    69L   -17
 8    70L    62
 9   231   -244
10    72L   -83
11   359    339
12    91     92
13   200   -207
14    76L    22
15   362   -389
16   122   -115
17   199   -202
18   290   -340
19   146   -152
20    74L    44
21   124    139

 H = 10, K =  2
 0    70L    58
 1    70L     5
 2   157   -153
 3   111   -100
 4   303   -288
 5    71L   -23
 6   414    385
 7   267   -254
 8   446    474
 9   496   -522
10   469    499
11   243   -265
12   265   -286
13   256    276

 L    FO     FC
14   190    180
15   122   -124
16    77L   -16
17    76L   -25
18   306   -268
19   163   -167
20    70L   -62
21    67L    62
22   101     98

 H = 11, K =  2
 1    74L    85
 2    74    -77
 3   210   -205
 4    75L    -6
 5   270    269
 6   409   -441
 7   131    139
 8    76L   -75
 9    77L   -22
10   189    196
11    77L   -63
12   122   -100
13    77L    48
14   172    206
15   132    151
16   159    189
17   179    175
18   133    121
19    69L   -44
20   127   -127

 H = 12, K =  2
 0    77L    46
 1    77L    -1
 2    77L    47
 3    77L   -45
 4    77L   -12
 5   189   -199
 6   134   -144
 7   173    177
 8    77L   -84
 9    77L   -31
10    77L   -44
11    77L    -3
12    76L    39
13    75L    61
14    74L   -49
15    73L   -29
16   133    181
17    97    108
18    93    109
19    77     80
20    93    -84
21    54L   -64
22    49L   -14
23    64     58

 H = 13, K =  2
 1    95     97
 2    77L   -63
 3    77    -84
 4    95    110
 5    77     69
 6    94     94
 7    77L   -41
 8   143   -146
 9    76L   -34
10    75     86
11    74L    14
12    73L   -42
13    71L    -9
14   110   -103
15    67L   -70
16    65L    63
17    62     61
18    58L    48
19    55L    20
20    50L    67
21    66    -74
22   101     73

 H = 14, K =  2
 0    93     84
 1   131    160
 2   141   -190
 3   119   -135
 4   106    -95
 5    75L   -69
 6   128    153
 7    73L    46
 8    73L   -32
 9    71L   -58
10    70L    79
11    69L   -42
12    67L    77
13    65L    69
14    63L   -93
15    60L   -71
16    57L   -33
17    53L   -14
18    49L   -15
19    76     57
20    84    -90

 H = 15, K =  2
 1    71L    -4
 2    71L   -32
 3    70L   -29
 4    96    -82

 H = 16, K =  2
 0    63L    -8
 1    77     68
 2   109    -84
 3    77     55
 4    62L    23
 5    61L   -22
 6    60L    12

 H = 17, K =  2
 1    53L     3
 2   111   -124

 H =  1, K =  3
 1    88L   -10
 2   440    481
 3   369    425
 4   728E  -829
 5   587   -582
 6    70     61
 7   410   -476
 8   371   -344
 9   370   -336
10   168   -168
11   112    -93
12   154    119
13    83L    12
14    86L   -31
15    90L    37
16   133   -113
17    96L   -61

 L    FO     FC
18   244    236
19   102L   -37
20   112    -85
21   228    219

 H =  2, K =  3
 0   225   -259
 1    75    -68
 2   397   -443
 3   233    240
 4   230    253
 5    77    105
 6   508    496
 7   207   -175
 8   487    509
 9    64L    23
10    68L    15
12   186    171
13    63    -88
14   128    -89
15   139   -129
16    87L   -53
17    90L    83
18   192   -169
19   476   -473
20   144    132

 H =  3, K =  3
 1   122   -128
 2    66     71
 3    69     59
 4   769E  -797
 5   990E  1051
 6   698   -633
 7    49L    17
 8   405    418
 9   152    128
10   313   -283
11    73     52
12   242    227
13    65L   -25
14   232    207
15   228   -190
16    73L    27
17   119     87
18   317    284
19   244    214
20   138   -129
21    80L   -78
22    80L  -114
23    79L    74
24    77L    30
25    74L    82
26    71L   -35
27    66L    -5
28    60L    28
29    54L   -24
30   125    142

 H =  4, K =  3
 0   114   -111
 1    53    -20
 2    44L   -36
 3   243    227
 4   226    216
 5   102     73
 6   762   -831
 7   783    720
 8   245   -187
 9    57L   -50
10   256   -261
11    62L     2
12   144     95
13    67L    39
14   156    130
15   114     94
16    74L    43
17   133   -107
18   270    248
19   292    292
20   277    253
21    80L   -56
22    80L    13
23    78L    33
24    76L   -22
25    73L   -32
26    69L   -12
27    64L   -31
28    58L   -11
29    51L    45
30    81    -88

 H =  5, K =  3
 1    49L   -47
 2    49L    35
 3   281    269
 4    51L   -13
 5    53L     3
 6   452    425
 7    93    100
 8   326    303
 9    60L    82
10    63L   -21
11    65L   -57
12   329    329
13   184   -181
14   125    121
15    74L   -62
16    76L    26
17   206   -214
18   210   -189

 H =  6, K =  3
 0   326    333
 1   245    229
 2   363    339
 3   250   -263
 4   194   -190
 5    57     -7
 6    59L    25
 7   209   -197
 8    62L   -23
 9   197    210
10    66    -69
11    68    -73
12   202   -180
13   126    102
14    75L   -16
15    76L   -64
16    78L     7
17    79L     0
18    80L  -101
19    80L    58
20   179   -204

 H =  7, K =  3
 1    50L    -9
 2   141    114
 3   296    312
 4   213   -186
 5   283   -271
 6   157   -127
 7   243   -214
 8   164   -176

 L    FO     FC
 9   109   -113

 H =  8, K =  3
 0   228    226
 1    65L    20
 2   113    -85
 3    66L    38
 4    67L   -76
 5    68    -84
 6   154    159
 7    70L    31
 8   251   -230
 9   162   -147
10   117     93
11   141    124
12    77    -80

 H =  9, K =  3
 1    70L     3
 2    71L    65
 3   167    172
 4    71L   -21
 5   120   -102
 6   127    115
 7    74L   -64
 8   125   -117
 9    76L   -29
10    77L    20
11   124   -127
12    79L     2
13    80L   -50
14    80L    19
15   127    121
16    80L   -69
17   126   -117
18    78L    71
19    77L    10
20    74L    44
21   124    144

 H = 10, K =  3
 0    75L    80
 1    75    -49
 2   141    -97
 3   130    137
 4   170   -157
 5    76L   -39
 6   451    509
 7   364   -339
 8   192    177
 9   131    123
10    80L    -6
11    80L   -53
12   139    143
13    80L   -88
14   197   -185
15    80L   -65
16    79L   -14
17    77L   102
18   173   -132
19   284   -249
20   210   -184

 H = 11, K =  3
 1    78L   -34
 2    78L    84
 3    79L     4
 4   440   -423
 5   463    465
 6   327   -316
 7    80L   -10
 8   139    146
 9    80L   -14
10   133   -131
11    80L    19
12   126    101
13    80L    81
14    78L    47
15   134   -134
16    75L    18
17   116    125
18   276    276
19   166    139
20    91    -80

 H = 12, K =  3
 0   179    176
 1   133    141
 2   127   -112
 3    80L    -1
 4    80L    22
 5    80L     9
 6   230   -210
 7   276    286
 8   150   -137
 9    98    122
10    79L   -33
11    79L   -31
12    77L    30
13    76L   -17
14    75L    25
15    73L    47
16    71L   -31
17    68L    -2
18    64L    63
19   271    216

 H = 13, K =  3
 1    80L   -89
 2    80     81
 3   149    134
 4   246   -269
 5   194   -193
 6    79L   -73
 7   184   -182
 8    77L   -75
 9    77L   -60
10    76L    22
11    75L    50
12   155    133
13    71L   -45
14    69L    39
15    66L   -69
16    63L    56
17    60L   -71
18    56L   -51
19    51L    30
20   118   -101
21    73     66

 H = 14, K =  3
 0    94    106
 1    77L   -42
 2    76L    48
 3    76L    37
 4    93     93
 5    75L    40
 6    75L   -39
 7    74L    22
 8   218   -189
 9    71L   -36
10   148   -126
11    68L    77

 L    FO     FC
12    66L    44
13    64L    18
14    61L   -46
15    57L    -3
16   140    130
17   149   -120
18   121    105
19    89    -76

 H = 15, K =  3
 1    71L    25
 2    70L    54
 3    70L    41
 4    69L    51
 5    68L    43
 6   155    150
 7    66L    17
 8   126     98
 9    63L   -41
10   130   -113
11    59L    11
12    49     40

 H =  0, K =  4
 0   622    670
 1    69L   -66
 3   285   -407
 4   342   -370
 5   309   -325
 6   144   -142
 7   665    733
 8   568   -592
 9  1002E -1119
10   820   -882
11    93    -35
12   146    -99
13   289    290
14   265    254
15   265   -271
16   717E   826
17   544E  -651
18   210    219
19   119   -128
20   179    180
21   116   -116
22   147   -160
23    68L   -62
24    66L     0
25    63L    44
26    59L   -17
27    55L    32
28   129   -122
29    43L     5
30    93    121

 H =  1, K =  4
 1    50     48
 2    40    -13
 3    35    -10
 4   178    202
 5   120    106
 6   108    -69
 7   518    561
 8  1164E  1233
 9   565    578
10    49    -73
11   172   -173
12    75L   -30
13    78L   -54
14   184   -169
15   486    541
16   322   -357
17   134   -107
18   216    199
19    91L    51
20    91L   -42
21    70     83
22    69L    -2
23    83     58

 H =  2, K =  4
 0    45     22
 1   111    121
 2   101     88
 3    84     71
 4   134    147
 5   293    323
 6   400   -400
 7   300    290
 8   546    574
 9   325    336
10   181    138
11    67L   -19
12   111   -127
13   217    203
14    75L    25
15   117   -125
16   236   -237
17   241    242
18    68L   -58
19   110     73
20    86     87
21    70L    25
22   119    117
23   107     96
24    65L    -5
25    62L   -17
26    58L   -31
27    54L   -13
28    93    -33
29    41L    83
30    66    -75

 H =  3, K =  4
 0    35L     0
 1    56     55
 2   369    401
 3   134   -129
 4   212    226
 5   266   -278
 6   219    223
 7    46L   -47
 8    48L   -12
 9   123    138
10    53L    15
11   135   -138
12    57L   -24
14   201   -184
15    79     83
16   162    171
17    68L   -48
18   150   -161
19    70L   -79
20    99    111
13    60L    48

 H =  4, K =  4
 0   251   -284
 1   206    215
 2    41L     5
 3   134    153
 4    44L   -34
 5    45L    68
 6   110    -95
```

Table 3 (*cont.*)

L	FO	FC
7	49L	1
8	51L	-33
9	92	-68
10	146	142
11	57L	7
12	180	-168
13	124	-135
14	78	83
H = 5, K = 4		
1	138	-135
2	123	-112
3	47L	48
4	48L	36
5	177	-177
6	51L	-9
7	174	181
8	54L	35
9	56L	7
10	58L	41
11	60L	31
12	103	82
H = 6, K = 4		
0	51	-20
1	125	133
2	230	239
3	52L	31
4	53L	-5
5	54L	-9
6	55L	39
7	56L	-16
8	82	79
9	197	206
10	97	99
11	109	146
12	137	-119
13	66L	-38
14	67L	35
15	68L	-61
16	110	-115
17	121	142
H = 7, K = 4		
1	79	-85
2	56L	-4
3	56	-46
4	172	-101
5	96	87
6	111	112
7	282	-285
8	389	-407
9	209	-179
10	64L	-15
11	147	139
12	67L	12
13	68L	-8
14	119	126
15	177	-204
H = 8, K = 4		
0	207	203
1	60L	41
2	121	124
3	149	-137
4	106	-94
5	98	-48
6	104	80
7	64L	-17
8	113	-105
9	132	-95
10	313	-324
11	68L	15
12	68L	-81
13	125	141
14	70L	57
15	110	-108
16	231	246
17	170	-174
18	69L	9
19	133	-158
20	64L	-14
21	62L	-86
22	152	-146
23	117	-119
H = 9, K = 4		
1	91	46
2	227	-233
3	205	214
4	234	-238
5	355	387
6	217	-221
7	67L	-44
8	303	318
9	68L	55
10	208	-211
11	69L	91
12	70L	-32
13	70L	15
14	70L	0
15	69L	3
16	154	-197
17	67L	73
15	139	133
H = 10, K = 4		
0	179	166
1	241	-284
2	83	69
3	68L	16
4	96	-97
5	211	218
6	294	-324
7	229	268
8	197	213
9	156	-168
10	70L	-17
11	70L	-20
12	70L	-91
13	69L	58
14	84	106
15	68L	2
16	66L	-2
17	64L	-53
18	108	82
19	163	153
20	129	136
21	102	121
22	167	183
23	98	114
H = 11, K = 4		
1	70L	54
2	326	345
3	249	-288
4	171	184
5	198	-197
6	185	189
7	70L	46
8	70L	-6
9	121	-117
10	69L	-33
11	69L	19
12	68L	-45
13	67L	8
14	66L	-73
15	64L	50
16	62L	57
17	60L	34
18	57L	-52
19	76	-101
H = 12, K = 4		
0	110	-101
1	185	203
2	99	-109
3	70L	9
4	70	59
5	69L	57
6	69L	-79
7	69L	84
8	68L	69
9	68L	-12
10	67L	66
11	66L	-7
12	92	-98
H = 13, K = 4		
1	68L	-13
2	68L	-94
3	69L	-80
4	67L	-6
5	67L	43
6	66L	-27
7	66L	27
8	65L	-38
9	64L	-29
10	63L	66
11	75	92
H = 14, K = 4		
0	64L	-8
1	64L	56
2	110	105
3	63L	3
4	62	-70
5	88	-85
6	61L	43
7	135	128
8	59L	-45
9	82	-71
10	59	-63
11	54L	-16
12	52L	17
13	50L	24
14	47L	-1
15	92	-82
16	75	56
17	57	-39
H = 15, K = 4		
1	57L	-55
2	57L	34
3	56L	-12
4	55L	5
5	55L	-26
6	85	104
7	53L	30
8	85	74
9	70	84
H = 16, K = 4		
0	48L	28
1	71	61
2	91	65
H = 1, K = 5		
0	24L	0
2	110	-110
3	201	199
4	63	-75
5	150	-173
6	141	-117
7	119	85
8	215	188
9	301	305
10	83L	33
11	92	82
12	206	213
13	148	140
14	204	-181
15	222	211
16	96L	-9
17	206	-176
18	71L	51
19	72L	37
20	71L	-63
21	70L	41
22	68L	-53
23	81	-82
24	63L	-10
25	59L	25
26	54L	-14
27	48L	-24
28	70	45
H = 2, K = 5		
0	258	292
1	260	-274
1	299	-274
2	169	179
3	64	54
4	89	-98
5	74	-55
6	135	133
7	204	-193
8	96	-91
9	275	264
10	76L	39
11	177	144
12	81L	-26
13	163	150
14	165	182
15	128	105
16	275	-251
17	113	90
18	134	116
19	113	-96
20	71L	25
21	70L	-43
22	118	105
23	123	114
24	62L	-21
25	58L	3
26	138	-121
27	91	69
H = 3, K = 5		
1	316	-369
2	663	679
3	121	-95
4	99	47
5	114	-108
6	225	230
7	89	-78
8	54L	-30
9	102	-90
10	131	-120
11	130	-115
12	252	-217
13	104	105
14	95	-83
15	130	-107
16	71L	4
17	71L	34
18	102	72
19	88	92
20	71L	70
21	69	-84
22	67	59
23	64L	47
24	175	155
25	57L	-12
26	51L	-3
27	45L	34
28	69	-61
H = 4, K = 5		
0	457	-492
1	278	278
2	47L	33
3	168	-190
4	207	163
5	157	111
6	99	-65
7	55L	45
8	57L	-6
9	132	118
10	106	54
11	63L	21
12	65L	28
13	67L	53
14	69L	18
15	70L	30
16	87	-54
17	72L	47
18	135	-122
19	71L	-39
20	70L	34
21	68L	-55
22	181	-150
23	189	-165
24	59L	-44
25	54L	-29
26	159	117
27	115	-97
H = 5, K = 5		
0	51L	0
1	174	150
2	196	-178
3	337	345
4	54L	-33
5	96	74
6	90	54
7	166	-132
8	128	-112
9	62L	-47
10	78	-68
11	114	-84
12	112	-96
13	69L	19
14	86	82
15	123	-107
16	72L	43
17	72L	-34
18	71L	-66
19	71L	-31
20	69L	21
21	67L	-12
22	64L	73
23	182	-183
24	98	-54
H = 6, K = 5		
0	57	-54
1	106	-102
2	107	87
3	92	76
4	97	-97
5	104	-81
6	81L	35
7	99	60
8	101	86
9	300	-265
10	67L	-11
11	118	-125
12	155	-135
13	150	-145
14	71L	5
15	72L	-37
16	124	116
17	71L	-71
18	71L	-50
H = 7, K = 5		
1	62L	-49
2	108	-90
3	62L	-9
4	63L	14
5	64L	25
6	113	-123
7	66L	-42
8	67L	41
9	152	122
10	120	102
11	70L	47
12	87	-80
13	71L	49
14	124	-128
15	113	94
16	71L	11
17	99	-67
18	69L	42
19	67L	70
20	65L	-69
21	62L	37
22	101	84
23	54L	48
24	49L	0
25	42L	31
26	47	39
H = 8, K = 5		
0	93	79
1	81	54
2	162	-134
3	66L	-14
4	67L	64
5	68L	-22
6	108	-86
7	69L	30
8	110	82
9	71L	16
10	71L	16
11	71L	-29
12	124	108
13	113	92
14	113	-98
15	71L	-31
16	70L	20
17	68L	-25
18	66L	20
19	64L	53
20	61L	-12
21	57L	-21
22	53L	10
23	49L	14
24	42L	36
25	49	49
H = 9, K = 5		
1	98	-77
2	197	-155
3	197	178
4	70L	17
5	157	-121
6	112	-61
7	159	127
8	71L	-46
9	102	73
10	72L	15
11	72L	6
12	124	132
13	71L	57
14	70L	-45
15	69L	17
16	67L	54
17	65L	-57
18	63L	-27
19	60L	-56
20	56L	-11
21	52L	32
22	47L	-23
23	80	-80
24	67	-73
H = 10, K = 5		
0	434	422
1	71	-72
2	71	-65
3	113	81
4	160	-142
5	113	-106
6	124	118
7	72L	-28
8	102	-73
9	71L	-24
10	71L	-26
11	71L	55
12	70L	-19
13	69L	28
14	67L	30
15	66L	-17
16	64L	-7
17	61L	75
18	58L	56
19	55L	-37
20	50L	-15
21	45L	12
22	116	110
H = 11, K = 5		
1	241	-229
2	390	415
3	208	-182
4	71L	34
5	71L	-20
6	113	89
7	71L	-58
8	71L	24
9	70L	-33
10	69L	-26
11	68L	-22
12	94	-71
13	103	66
14	63L	-11
15	61L	7
16	58L	-13
17	55L	-12
18	51L	36
19	91	74
20	42L	40
21	93	-106
H = 12, K = 5		
0	240	-224
1	225	205
2	70L	36
3	110	-93
4	69L	0
6	68L	32
7	107	73
8	95	-57
9	66L	-20
10	65L	-55
11	63L	-47
12	62L	-47
13	60L	-42
14	57L	29
15	54L	-24
16	108	83
17	47L	-29
18	90	-78
19	69	69
H = 13, K = 5		
1	66L	-2
2	66L	3
3	197	183
4	65L	-12
5	64L	-48
6	64L	2
7	63L	-25
8	161	99
9	105	72
10	59L	37
11	99	49
12	116	-91
13	90	66
14	159	-127
15	88	47
16	41L	6
17	60	-44
H = 14, K = 5		
0	128	-76
1	60L	-16
2	60L	-13
3	59L	23
4	59L	34
5	59L	19
6	57L	51
7	55L	-28
8	54L	42
9	52L	59
10	151	135
11	49L	1
12	103	85
13	94	88
14	77	-71
H = 0, K = 6		
0	109	-121
1	139	-155
2	93L	50
3	93L	16
4	56	-52
5	105	97
6	61	-41
7	104	101
8	251	260
9	83L	-1
10	192	210
11	99	-96
12	428	404
13	670F	860
14	109	112
15	72L	-42
16	113	-98
17	103	-63
18	73L	16
19	72L	-18
20	70L	-23
21	69L	37
22	65L	-10
23	61L	17
24	76	80
H = 1, K = 6		
0	28L	0
1	128	-147
2	73L	9
3	75L	-13
4	180	-171
5	79L	40
6	81L	36
7	155	137
8	86L	48
9	226	204
10	144	99
11	486	-548
12	351	354
13	102	-95
14	311	315
15	72L	46
16	122	106
17	103	-77
18	73L	-48
19	111	-108
20	70L	-13
21	68L	3
22	65L	-26
23	77	71
H = 2, K = 6		
0	232	-248
1	65L	21
2	66L	36
3	188	-144
3	160	-144
4	69L	-33
5	119	94
6	74L	2
7	76L	-29
8	78L	8
9	94	-97
10	82L	-10
11	59	43
12	372	-398
13	308	-331
14	110	103
15	91	73
16	124	-104
17	73L	52
18	73L	-35
19	71L	41
20	70L	15
21	67L	23
22	64L	-64
23	60L	-49
24	55L	-8
25	86	-75
H = 3, K = 6		
1	140	124
2	103	-96
3	49L	19
4	51L	2
5	75	-60
6	70	-35
7	82	44
8	101	-105
9	62L	45
10	92	66
11	95	-84
12	212	-234
H = 4, K = 6		
0	52L	53
1	52L	-46
2	163	154
3	54L	-14
4	55L	20
5	85	-65
6	140	-126
7	95	-92
8	106	95
9	65L	14
10	113	-123
11	107	87
12	109	107
13	71L	-34
14	102	-93
15	73L	5
16	103	103
H = 5, K = 6		
1	128	145
2	59L	-30
3	84	75
4	86	74
5	62L	16
6	63L	43
7	58	-57
8	66L	28
9	68L	-5
10	69L	0
11	71L	-4
12	102	90
H = 6, K = 6		
0	133	130
1	89	-74
2	133	-115
3	91	76
4	65L	34
5	66L	-34
6	67L	-12
7	106	-99
8	88	54
9	70L	3
10	71L	-39
11	72L	8
12	146	-145
13	196	-199
H = 7, K = 6		
1	67L	-16
2	68L	-18
4	69L	43
5	98	-66
5	76	-66
6	70L	-15
7	77	-51
8	71L	24
9	121	-108
10	73L	-62
11	215	215
12	73L	-78
13	103	54
14	112	-109
H = 8, K = 6		
0	155	158
1	110	-88
2	110	118
3	142	127
4	101	-75
5	72L	-13
6	102	63
7	73L	-24
8	108	75
9	73L	-56
10	92	62
11	73L	-44
12	112	103
13	201	199
H = 9, K = 6		
1	287	-349
2	194	174
3	73L	-17
4	146	-116
5	113	98
6	73L	8
7	73L	-30
8	73L	10
9	92	65
10	72L	-21
11	90	-65
12	109	92
H = 10, K = 6		
0	288	-318
1	73L	-61
2	183	-173
3	223	-251
4	73L	-23
5	194	230
6	72L	-23
7	72L	26
8	71L	50
9	71L	-54
10	70L	17
11	68L	30
12	95	-78
H = 11, K = 6		
1	206	179
2	110	-59
3	123	-96
4	134	-83
5	70L	-23
6	69L	2
H = 12, K = 6		
0	67L	37
1	67L	-5
2	243	219
H = 13, K = 6		
1	95	70
H = 14, K = 6		
0	52L	0
1	52L	16
2	90	-74
3	51L	11
4	51L	33
5	76	-40
6	48L	-28
7	46L	33
8	75	60
H = 15, K = 6		
1	38L	14
2	68	-32
H = 1, K = 7		
1	152L	1
2	57	-56
3	84	113
4	84	95
5	84L	-30
6	85L	32
7	47	-54
8	88L	17
9	89L	28
10	138	136
11	199	-229
H = 2, K = 7		
0	69L	70
1	69L	-21
2	192	-250
3	67	-56
4	65	-39
5	69	44
6	66	38
7	77L	17
8	84	89
9	80L	-15
10	91	-87
11	80	70
12	82L	43
13	219	-275
14	102	-105
15	62L	-13
16	109	-117
H = 3, K = 7		
0	43L	0
1	102	64
3	46L	-11
4	102	-98
5	49L	-43
6	110	-65
7	53L	50
8	55L	2
9	57L	-15
10	59L	3
11	99	111
12	61L	-34
13	62L	-11
14	62L	30
15	62L	-2
16	61L	65
17	60L	-70
18	58L	-45
19	55L	34
20	53L	-20
21	91	65
22	117	-120
H = 4, K = 7		
0	49L	21
1	49L	-13
2	212	255
3	168	207
4	140	150
5	53L	-16
6	55L	-13
7	56L	-31
8	57L	-56
9	59L	52
10	60L	-17
11	61L	-10
12	62L	-41
13	62L	-40
14	62L	31
15	61L	11
16	60L	-15
17	58L	13
18	57L	-19
19	54L	7
20	51L	20
21	47L	-16
22	42L	-42
23	91	34
H = 5, K = 7		
1	115	-131
2	97	-88
3	91	77
4	55L	20
5	66	53
6	67	-64
7	59L	16
8	60L	-37
9	60L	-39
10	61L	-64
11	124	126
12	93	-82
13	62L	19
14	61L	-23
15	81	-76
H = 6, K = 7		
0	57L	-13
1	95	-97
2	95	-98
3	68	-57
4	59L	-1
5	81	-91
7	60L	-45
8	61L	60
9	62L	-36
10	62L	22
11	62L	-32
12	61L	7
13	108	140
14	60L	57
15	59L	6
16	86	98
H = 7, K = 7		
1	60L	-15
2	60L	-30
3	57	-67
4	61L	6
5	61L	1
6	61L	-21
8	62L	15
9	62L	-57
10	93	102
11	82	-63
12	60L	23
13	59L	35
14	58L	-24
15	56L	30
16	63	72
17	49	64
H = 8, K = 7		
0	102	-108
1	93	108
2	106	109
3	93	-84
H = 9, K = 7		
1	62L	-2
2	93	-82
3	61L	8
4	101	109
H = 10, K = 7		
0	70	69
1	60L	10
2	197	-203
3	110	-80
4	119	-113
H = 11, K = 7		
1	108	100
2	153	142
3	55L	20
4	102	-66
H = 12, K = 7		
0	51L	15
1	133L	48
3	130	136
4	91	83
H = 0, K = 8		
0	93	103
1	101L	-30
2	101L	-5
3	119	-137
4	101L	28
5	100L	17
6	158	-211
7	173	242
8	179	-216
H = 1, K = 8		
1	152L	29
2	152L	12
3	152L	-9
4	151L	9
5	151L	-41
6	150L	79
7	149L	3
8	165	-176
9	163	158
10	144L	-80
11	141L	-47
12	138L	-69
13	134L	-90
14	130L	17
15	124L	12
16	146	-133
17	182	-199
18	143	-149
H = 2, K = 8		
0	142L	-56
1	142L	-71
2	142L	45
3	141L	97
4	141L	59
5	141L	52
6	140L	49
7	160	-118
8	210	190
H = 2, K = 9		
0	125L	13
1	125L	0
2	124L	15
3	124L	-61
4	122L	-28
5	121L	29
6	207	166

down to the nine reflections shown in Table 1. Subsequent calculations utilized the 208 $|E|$'s $\geq 1{\cdot}805$ (this number of reflections gave 2400 $\sum_2$ triples, the storage limit of *PHASER* on a 131*K* by the IBM 360/40 computer). Although there were some strong relationships which could have been used to reduce the number of 'arbitrary' signs (Table 1) to three, we chose to treat all six as unknowns and 64 (2^6) sign pyramids were computed. It had been our previous experience (*e.g.* Ammon & Jensen, 1967; Ammon & Sundaralingam, 1966) that the correct solution often could be pinpointed by the use of several simple criteria: *e.g.* relative number of + and − phases, number of undetermined phases, and number of correct and incorrect $\sum_2$ triples. Of the 64 solutions, nos. 6 and 16 were superior to all of the others, and of these two, the latter appeared to be the single 'best' solution (Table 2). Using the *PHASER* subprogram *ADDPHAS*, the initial set of 197 signs from no. 16 was expanded to 850 signs ($|E| \geq 0{\cdot}5$); an E map computed with these phases did not reveal the structure.

Frequently, the starting sets of phases for the few 'best' solutions differ from each other only in the phases of one or two reflections, and the generated sets of phases are virtually identical. A comparison of the 194 phases common to solutions 6 and 16 revealed that the sets of signs were different for 106 reflections. The 194 signs from no. 6 were then expanded to include the $|E|$'s from 1·8–1·3. From these 204 additional data, 171 new signs were accepted according to the probability criteria, and an E map was calculated with the 365 terms. All 26 of the carbon, nitrogen and oxygen atoms were clearly discernible from this synthesis and a subsequent structure factor calculation gave an R ($R=\sum|F_o|-|F_c||/\sum|F_o|$) of 0·31. A comparison of the original determined phases with those obtained from the fully refined structure showed that 3 of the original 194, and 9 of the second set of 171, were incorrect.

All least-squares computations were done using the *X-ray* 67 (1967) subprogram *ORFLS*, which collects the full matrix of the normal equations. Although all of the carbon, nitrogen and oxygen parameters could be varied simultaneously with the program in the isotropic temperature factor mode, computer storage limitations permitted simultaneous refinement of only 12 atoms with anisotropic temperature factors. Thus one overall 'cycle' of anisotropic refinement was achieved by three individual cycles of refinement in which the positional and thermal parameters for 12 atoms were varied during each cycle. The quantity minimized was $\sum w(F_o-F_c)^2$. During the final cycles of refinement, a Hughes (1941) weighting scheme was applied ($4F_{\rm min}=36{\cdot}0$); the unobserved data were given unit weights if $|F_c|>|F_o|$ and zero weights if $|F_c|\le|F_o|$. Hydrogen atoms were located from a difference synthesis and were refined with individual isotropic temperature factors. The average and maximum parameter shifts from the last cycle of heavy atom refinement were $0{\cdot}119\sigma$ and $0{\cdot}409\sigma$ and the R index was 0·10. For 73 of the 933 unobserved reflections, the calculated structure amplitudes were larger that the corresponding minimum observed amplitudes. The atomic scattering factors used were carbon, nitrogen and oxygen (Berghuis, Haanappel, Potters, Loopstra, MacGillavry & Veenendaal, 1955); hydrogen (Stewart, Davidson & Simpson, 1965). The measured and calculated structure factors are given in Table 3.

The atomic coordinates and thermal parameters are given in Table 4; bond lengths and angles between the

Table 4. *Fractional coordinates and temperature factors* (Å^2) *for the carbon, oxygen, nitrogen and hydrogen atoms*

Estimated standard deviations are given in parentheses.

The anisotropic thermal parameters are in the form $\exp\left[-0{\cdot}25(h^2B_{11}a^{*2}\ldots 2klB_{23}b^*c^*)\right]$.

	x	y	z	B or B_{11}	B_{22}	B_{33}	B_{12}	B_{13}	B_{23}
O(7)	0·0081 (3)	0·2192 (9)	0·4510 (2)	2·6 (2)	16·9 (6)	9·2 (4)	−0·2 (3)	0·8 (2)	−6·7 (4)
O(12)	0·3744 (2)	0·2656 (5)	0·4414 (2)	2·9 (2)	7·1 (3)	3·8 (2)	−0·1 (2)	0·2 (1)	1·6 (2)
O(14)	0·5080 (2)	−0·0936 (6)	0·3213 (2)	2·3 (2)	9·2 (3)	4·8 (2)	0·0 (2)	1·1 (2)	−2·1 (2)
N(5)	0·1384 (3)	−0·0272 (6)	0·3220 (2)	3·2 (2)	4·7 (3)	3·9 (3)	0·0 (2)	−0·9 (2)	−0·1 (2)
N(6)	0·0972 (3)	0·0541 (6)	0·3717 (2)	2·7 (2)	5·5 (3)	4·0 (3)	0·2 (2)	−0·2 (2)	−0·5 (2)
C(1)	0·4016 (4)	−0·2296 (8)	0·2400 (3)	4·8 (3)	4·5 (3)	4·7 (3)	0·3 (3)	1·1 (3)	−0·3 (3)
C(2)	0·3441 (5)	−0·2902 (9)	0·2017 (3)	6·2 (4)	5·8 (4)	3·5 (3)	−0·3 (3)	0·9 (3)	−1·0 (3)
C(3)	0·2492 (5)	−0·2621 (9)	0·2041 (3)	5·7 (3)	6·2 (4)	3·7 (3)	−0·1 (3)	−0·1 (3)	−0·7 (3)
C(4)	0·2104 (4)	−0·1796 (8)	0·2450 (3)	4·3 (3)	4·7 (3)	3·7 (3)	0·1 (2)	−0·8 (2)	−0·5 (3)
C(4*a*)	0·2672 (4)	−0·1136 (7)	0·2848 (2)	3·9 (3)	4·0 (3)	2·7 (3)	0·2 (2)	−0·3 (2)	0·0 (2)
C(4*b*)	0·2292 (3)	−0·0249 (7)	0·3285 (2)	3·0 (2)	4·1 (3)	3·2 (3)	−0·2 (2)	0·2 (2)	−0·0 (2)
C(6)	−0·0061 (4)	0·0227 (10)	0·3698 (3)	2·1 (2)	9·1 (5)	9·1 (5)	−0·4 (3)	0·6 (3)	−3·4 (4)
C(6*a*)	0·1439 (4)	0·1369 (7)	0·4083 (2)	3·2 (2)	4·0 (3)	4·0 (3)	0·2 (2)	−0·7 (2)	0·4 (2)
C(7)	0·0926 (4)	0·2223 (9)	0·4494 (3)	3·9 (3)	6·6 (4)	4·7 (4)	0·4 (3)	0·3 (3)	0·2 (3)
C(7*a*)	0·1430 (4)	0·3098 (8)	0·4904 (2)	4·1 (3)	5·0 (3)	3·2 (3)	0·8 (2)	0·4 (2)	0·3 (3)
C(8)	0·0939 (4)	0·3865 (8)	0·5307 (3)	4·2 (3)	5·2 (5)	4·3 (3)	1·0 (3)	0·8 (3)	0·7 (3)
C(9)	0·1437 (5)	0·4687 (9)	0·5690 (3)	6·3 (4)	5·7 (4)	4·0 (4)	0·9 (3)	1·1 (3)	0·1 (3)
C(10)	0·2405 (5)	0·4789 (8)	0·5678 (2)	6·3 (4)	5·5 (4)	3·4 (3)	1·2 (3)	0·6 (3)	0·6 (3)
C(11)	0·2874 (4)	0·4011 (8)	0·5270 (2)	4·8 (3)	4·6 (3)	2·8 (2)	0·0 (3)	0·4 (2)	−0·2 (2)
C(11*a*)	0·2389 (4)	0·3222 (7)	0·4883 (2)	4·1 (3)	4·1 (3)	3·1 (3)	1·1 (2)	0·4 (2)	0·2 (2)
C(12)	0·2915 (3)	0·2407 (8)	0·4444 (2)	2·8 (2)	5·1 (3)	3·7 (3)	0·3 (2)	−0·2 (2)	0·8 (3)
C(12*a*)	0·2417 (4)	0·1426 (7)	0·4060 (2)	3·0 (2)	4·1 (3)	2·5 (2)	0·7 (2)	0·5 (2)	0·8 (2)
C(12*b*)	0·2885 (3)	0·0538 (7)	0·3662 (2)	2·9 (2)	3·6 (3)	3·3 (3)	−0·1 (2)	0·0 (2)	0·2 (2)
C(14)	0·4248 (4)	−0·0662 (8)	0·3219 (2)	3·4 (2)	5·0 (3)	4·0 (3)	0·5 (2)	0·3 (2)	0·1 (3)
C(14*a*)	0·3632 (4)	−0·1371 (7)	0·2814 (2)	3·7 (3)	4·3 (3)	2·5 (3)	−0·2 (2)	0·6 (2)	0·1 (2)
H(1)	0·463 (3)	−0·256 (5)	0·237 (2)	3·0 (2)					
H(2)	0·389 (4)	−0·381 (8)	0·181 (2)	8·0 (1)					
H(3)	0·209 (4)	−0·360 (9)	0·188 (3)	8·0 (2)					
H(4)	0·133 (5)	−0·158 (9)	0·248 (3)	10·0 (3)					
H(6*a*)	−0·003 (3)	0·028 (6)	0·316 (2)	5·0 (2)					
H(6*b*)	−0·048 (6)	−0·067 (11)	0·379 (3)	12·0 (2)					
H(6*c*)	−0·045 (5)	0·104 (10)	0·377 (3)	10·0 (1)					
H(8)	0·017 (5)	0·371 (9)	0·529 (3)	9·0 (2)					
H(9)	0·100 (4)	0·499 (8)	0·599 (2)	8·0 (3)					
H(10)	0·269 (7)	0·544 (12)	0·591 (4)	15·0 (2)					
H(11)	0·348 (5)	0·420 (10)	0·506 (3)	10·0 (1)					
H(13)	0·422 (3)	0·095 (6)	0·381 (2)	4·0 (2)					

carbon, nitrogen and oxygen atoms are shown in Fig. 1; bond lengths and angles involving the hydrogen atoms are reported in Tables 5 and 6.

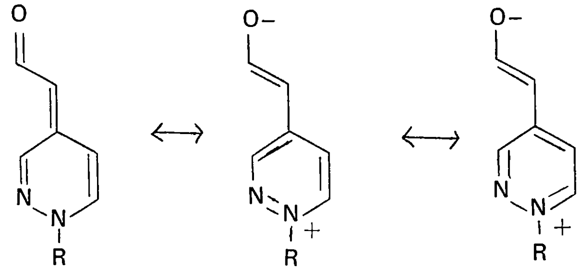
Table 5. *Bond lengths and estimated standard deviations to hydrogen*

	Length	e.s.d.
C(1)—H(1)	0·92 Å	0·04 Å
C(2)—H(2)	1·12	0·06
C(3)—H(3)	1·08	0·07
C(4)—H(4)	1·13	0·07
C(6)—H(6*a*)	1·39	0·05
C(6)—H(6*b*)	0·98	0·09
C(6)—H(6*c*)	0·89	0·08
C(8)—H(8)	1·12	0·07
C(9)—H(9)	1·02	0·06
C(10)-H(10)	0·89	0·10
C(11)-H(11)	1·04	0·07
C(13)-H(13)	0·90	0·05

Discussion

With the exception of O(12), O(14) and the methyl carbon, all of the atoms in a molecule are reasonably coplanar and deviate on the average 0·073 Å from the mean molecular plane. Least-squares planes were calculated for each of the six-membered rings and for several combinations of rings, and in all cases examined the groups of atoms showed significant deviations from planarity.* Of the six-membered rings, the atoms in the two benzene rings showed the highest degrees of coplanarity.

Bond lengths in the central portion of the molecule, depicted by structure (IV), differ substantially from the idealized values for single and double bonds. With the expection of the C(14)–O(14) length of 1·223 Å, which is normal for a ketonic carbonyl group, all of the bonds written in (IV) as formal double bonds are longer than one would expect for these bond types. In a similar way, all of the formal single bonds appear to be shorter than structure (IV) would predict. It is tempting to speculate that canonical forms (V) and (VI) may make a large contribution to the ground state resonance hybrid and thus are responsible for these bond length discrepancies. The distances found for the C=N–N–C moiety are close to those observed in other molecules which show substantial π-electron delocalization, *e.g.* 1,2,4-triazole (Goldstein, Ladell & Abowitz, 1969) and 2-methyl-3-aza-5-trifluoracetyl-



(IV) (V) (VI)

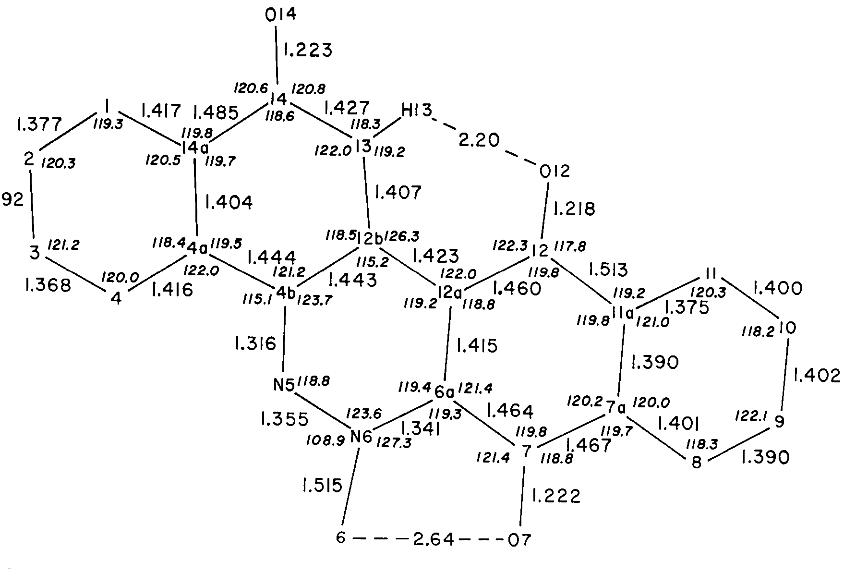


Fig. 1. Bond lengths (Å, larger numbers) and angles (°) for the carbon, oxygen and nitrogen atoms. Two close intramolecular approaches are also shown. E.s.d.'s in lengths and angles range from 0·006–0·010 Å and 0·5–0·6°, respectively.

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* The least-squares planes were computed with the programs of Ahmed, Hall, Pippy & Saunderson (1967). The χ^2 quantity ($\chi^2 = \sum_{i=i}^{n} d_i^2/\sigma_i^2$, where d_i and σ_i refer to the ith atom's distance from the plane and the standard deviation of the distance) was computed for each plane and used as a statistical test of coplanarity.

2*H*-2-pyrindine.[†] Correction of the C(14)–O(14) bond length for the riding motion of one atom upon another (Busing & Levy, 1964), increased the distance from 1·223 to 1.248 Å.

Most of the other bond lengths in (III*b*) have normal values. The average distances in the two benzene rings are 1·396 Å and 1·393 Å.

Perhaps the most interesting structural feature in this molecule arises from the juxtaposition of the methyl and oxygen substituents on the cinnoline moiety This arrangement is responsible for the C(6)···O(7) approach of 2·64 Å, which is *ca.* 0·8 Å shorter than the sum of the van der Waals radii of methyl and oxygen (3·4 Å). The strain created by this interaction is evident in the two exocyclic angles at N(6) (127·3° and 108·9°); an increase in the C(6)–N(6)–C(6*a*) angle by 9·2°, and concomitant decrease in C(6)–N(6)–N(5), from the conformation with two equal exocyclic anles of 118·1°, has permitted an increase in the methyl–oxygen distance of 0·24 Å (from 2·40 Å). This close approach is probably also responsible for the 0·13 Å displacement of the methyl group from the mean plane of O(7)–C(7)–C(6*a*)–N(6)–N(5) and for the CH$_3$–N bond length, 1·515 Å, which is about 0·04 Å longer than the values usually found for the C(sp^3)–N linkage.

The bond length and angles of the carbonyl group appear to show little effect of the methyl–oxygen interaction. However, the magnitude of B_{22} for O(7), 16·9 Å^2, is considerably larger than the *B*'s for any of the other atoms. This observation, in conjunction with the relatively small angle between the *b* axis and the normal to the molecular plane, suggests that the size of the term is not due to any large thermal motion of the oxygen atom, but to a positional disorder of the atom.[*] The disorder could be caused by the methyl–oxygen interaction which forces the oxygen atom to lie above the methyl group in some molecules and

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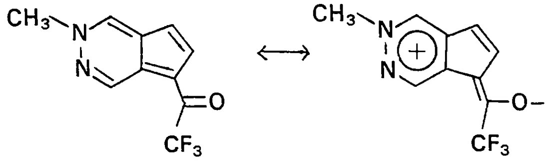
[†] Bond distances in this pyrindine derivative suggest that the dipolar structure shown below is important to the ground state structure of the molecule (Ammon, Watts, Anderson, Forkey, Grina & Johnson, 1970).



The ultraviolet and visible spectral properties of compounds like (IV) have been attributed to transitions involving dipolar structures (*e.g.* (V) and (VI); Hand & Cohen, 1967).

[*] We are grateful to a referee for pointing out this possibility.

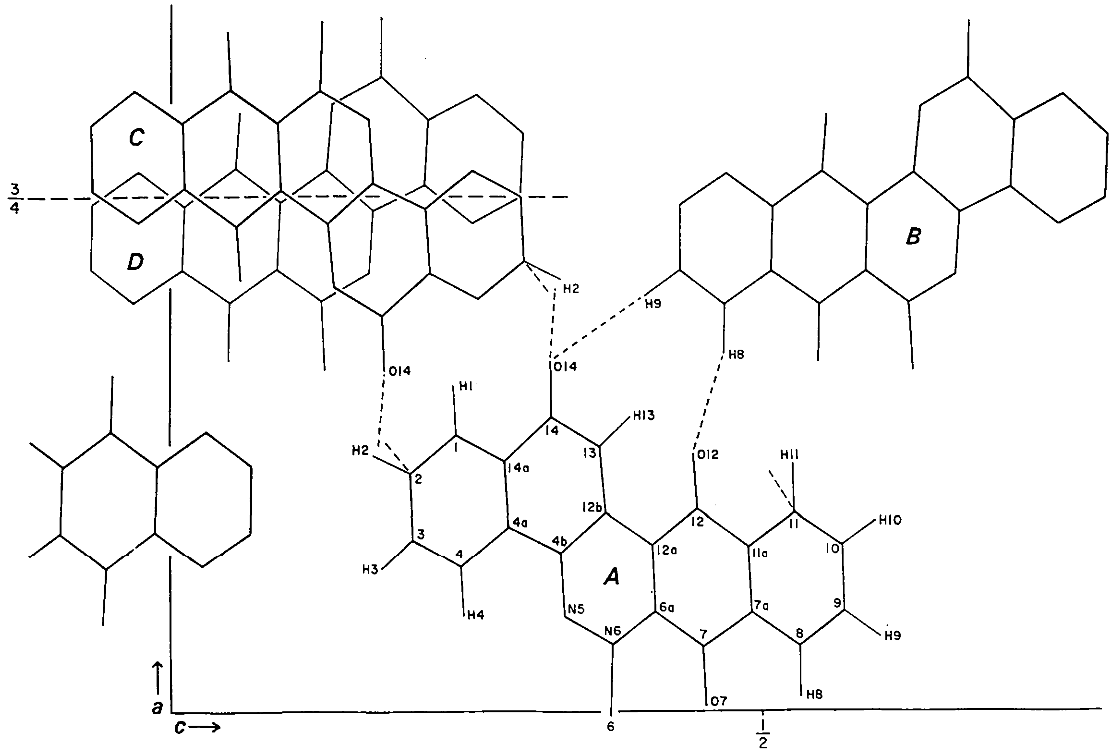


Fig. 2. A drawing of a portion of the unit cell viewed along the *b* axis. The nine hydrogen atoms which are linked to trigonal carbon are shown at their idealized positions. The observed positions of H(11) and H(2) are also shown with dashed lines from C(11) and C(2). O···H intermolecular contacts discussed in the text are designated with dashed lines. The relative positions of the molecules are: *A* at x, y, z; *B* at $\frac{1}{2}+x, \frac{1}{2}-y, 1-z$; *C* at $1-x, \frac{1}{2}+y, \frac{1}{2}-z$; *D* at $\frac{1}{2}+x, y, \frac{1}{2}-z$. The dashed line separating *C* and *D* shows one position of the *b*-glide plane.

Table 6. *Bond angles and estimated standard deviations to hydrogen*

	Angle	E.s.d.		Angle	E.s.d.
H(1)—C(1)-C(2)	116·0°	3·0°	H(6*a*)-C(6)—H(6*c*)	101·0°	5·0°
H(1)—C(1)-C(14*a*)	124·0	3·0	H(6*b*)-C(6)—H(6*c*)	97·0	7·0
H(2)—C(2)-C(1)	103·0	3·0	H(8)—C(8)—C(7*a*)	115·0	4·0
H(2)—C(2)-C(3)	134·0	3·0	H(8)—C(8)—C(9)	126·0	4·0
H(3)—C(3)-C(2)	113·0	4·0	H(9)—C(9)—C(8)	109·0	4·0
H(3)—C(3)-C(4)	116·0	4·0	H(9)—C(9)—C(10)	128·0	4·0
H(4)—C(4)-C(3)	123·0	4·0	H(10)-C(10)-C(9)	119·0	6·0
H(4)—C(4)-C(4*a*)	117·0	4·0	H(10)-C(10)-C(11)	122·0	6·0
H(6*a*)-C(6)-N(6)	90·0	2·0	H(11)-C(11)-C(10)	137·0	4·0
H(6*b*)-C(6)-N(6)	137·0	5·0	H(11)-C(11)-C(11*a*)	97·0	4·0
H(6*c*)-C(6)-N(6)	119·0	5·0	H(13)-C(13)-C(12*b*)	119·0	3·0
H(6*a*)-C(6)-H(6*b*)	106·0	5·0	H(13)-C(13)-C(14)	118·0	3·0

below the methyl group in other molecules. The r.m.s. amplitude of vibration of O(7) normal to the O(7)–C(7)–C(6*a*)–N(6)–C(6) plane is 0·5 Å.

Another short intramolecular distance is observed between H(13) and O(12). The H···O separation of 2·20 Å, which is appreciably less than the 2·6 Å sum of the hydrogen and oxygen van der Waals radii, is decreased to the very low value of 2·12 Å when the observed C(13)–H(13) distance of 0·90 Å is corrected to the more normal C–H length of 1·05 Å. In view of this short H···O approach and the fact that the hydrogen atom is linked to carbon, the question can be raised whether the H···O interaction is attractive (*i.e.* a hydrogen bond) or repulsive. Unfortunately, the standard deviations in the C–C–H angles are so large as to make any conclusions based on these angles extremely tenuous. The difference in the distances of H(13) and O(12) from the mean plane of the three central rings is 0·5 Å and the C(13)–H(13)···O(12) angle is 124°.

Fig. 2 shows part of the unit cell viewed along *b*. The average molecular plane intersects the *ac* plane at an angle of about 33°, and the long axis of the molecule is almost parallel to the *bc* plane. The molecules are located at *ca.* $\frac{1}{4}$ and $\frac{3}{4}$ in *x*, and thus are cut by the *b*-glide plane which is approximately normal to the molecular plane. The *b* glide transforms one molecule (at x,y,z) into a second molecule (at $\frac{1}{2}-x$, $\frac{1}{2}+y,z$), which is almost directly above the first one (*e.g.* C and D in Fig. 2), and leads to an arrangement in which the closest intermolecular contacts along *b* are between parallel rings. Assuming that the length of the *b* axis is solely dictated by these contacts, and taking into account the 33° angle between *b* and the plane of the molecule, the van der Waals 'thickness' of the molecule was computed as 3·45 Å.

The intermolecular contacts between hydrogen and oxygen were evaluated using (*a*) the hydrogen positions obtained from least-squares refinement, and (*b*) idealized hydrogen positions for the nine atoms linked to trigonal carbon (the C–H distances were set to 1·05 Å and the C–H vectors were assumed to bisect the appropriate C–C–C angles). Using the idealized hydrogens, only two H···O contacts were found to be shorter than the 2·6 Å sum of the hydrogen and oxygen van der Waals radii: O(14)···H(9) and O(12)···H(8) were both 2·54 Å (between molecules *A* and *B* in Fig. 2). The observed O(14)···H(2) distance of 2·30 Å, between molecules *A* and *C*, was calculated at 2·63 Å for the idealized case.

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