The Crystal and Molecular Structure of Delnudine Hydrochloride, C20H25NO3.HCl*

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The structure of delnudine hydrochloride, $C_{20}H_{25}NO_3$.HCl, has been determined from three-dimensional X-ray data collected at room temperature on a four-circle diffractometer using the $\theta/2\theta$ scan technique. It was refined by the least-squares method to $R = 3.8\%$ for 1834 observed reflexions. The crystals are monoclinic, space group $P2_1$ with two molecules in a unit cell of dimensions $a = 9.528$ (2), $b = 10.332$ (1), $c = 9.505$ (2) Å; $\beta = 108.50$ (2)°. The ring system of this alkaloid is a novel one but it is related to that of hetisine. The carbinolamine group has the following bond lengths: $N^+ - C$ 1.528 (4), $C-O$ 1.385 (4) Å.

Introduction

The alkaloid delnudine was isolated from the seeds of *Delphinium denudatum* by Götz & Wiesner (1969). From chemical and spectroscopic data they suspected that the structure was (I), *i.e.* a simple derivative of hetisine (II). The crystal structure of the latter alkaloid was determined in this laboratory by Przybylska (1963). Since the structure of delnudine was not fully known, it was decided to carry out this X-ray analysis. Its result, which showed the structure to be (III), was reported in a preliminary communication (Birnbaum, 1969). On the basis of this structure, Götz & Wiesner (1969) proposed a scheme for a biogenetic conversion of hetisine into delnudine.

$c = 9.505$ (2) Å; $\beta = 108.50$ (2)°. $V = 887.3$ Å³ [Mo $K\alpha_1$ ($\lambda = 0.70926$ Å) and Mo $K\alpha_2$ ($\lambda = 0.71354$ Å) radiations were used]. $D_m = 1.361$ g.cm⁻³ (by flotation in a mixture of dichloromethane and bromobenzene), $D_x = 1.362$ g.cm⁻³, $Z = 2$. $F(000) = 388$. Systematic absences: $0k0$ absent when k is odd. Space group $P2_1$. Linear absorption coefficient $\mu(\text{Mo } K\alpha) = 2.4$ cm⁻¹.

Experimental

A sample of delnudine (free base) was obtained from Professor Wiesner and many attempts were made to crystallize it. Finally, very small, irregularly shaped plates were grown by slow evaporation from an acetone–ethyl acetate solution. The space group was

(I)   (II)   (III)
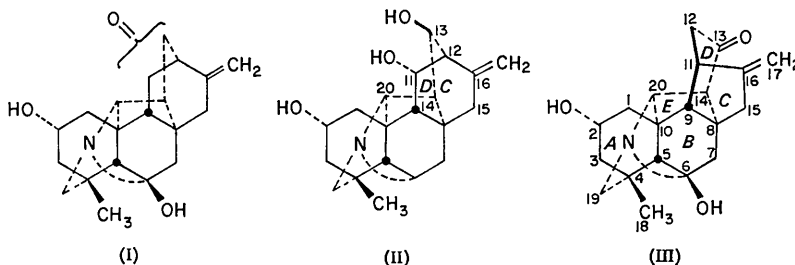
Crystal data

Delnudine hydrochloride, $C_{20}H_{25}NO_3$.HCl, F.W. 363.89. Monoclinic, $a = 9.528(2)$, $b = 10.332(1)$,

$P2_12_12_1$ with the approximate cell dimensions $a = 15.31$, $b = 22.65$, $c = 10.62$ Å (from precession photographs with Cu $K\alpha$ radiation) and two molecules in the asymmetric unit. However, these crystals were judged to be unsuitable for data collection and the hydrochloride derivative was therefore synthesized. Well developed,

Table 1. *Summary of the structure determination*

Cycle	Basis	Initial number of phases	$|E|_{min}$	x^*	Number of phases developed by tangent formula	$|E|_{min}$	$R(\%)$
1	Cl⁻ from Patterson						46.7
2	Cl⁻ + 6C from Fourier	190	1.5	0.3	376	1.3	43.2
3	Cl⁻ + 20C from E-map	207	1.5	0.6	383	1.3	28.3
4	Cl⁻ + 22C from E-map						26.6
5	Cl⁻ + 24C						25.0

* x is from the expression $|F_c| \geq x|F_o|$.

colourless, prismatic needles were obtained by crystallization from a mixture of 50 % ethanol and n-pentane. A crystal of dimensions $0.29 \times 0.39 \times 0.54$ mm was mounted along the b (needle) axis. The cell dimensions and the intensities were measured with an automated, four-circle diffractometer (Picker) equipped with a scintillation counter and a pulse-height analyser. Nb-filtered Mo $K\alpha$ radiation was employed. The intensity data were collected by the moving-crystal moving-counter ($\theta/2\theta$) scan technique. A $2.0°$ scan was used for reflexions with $2\theta \leq 35°$ and a $2.4°$ scan was used for those with $35° < 2\theta \leq 55°$. The background was measured for 20 sec on each side of the scan. When the count rate exceeded 20,000 c.p.s. the current was low-

Table 2. *Final fractional coordinates and thermal parameters* ($Å^2$)

(a) Non-hydrogen atoms

All values were multiplied by 10^4. The thermal-vibration expression used was

$$\exp[-2\pi^2(U^{11}h^2a^{*2} + U^{22}k^2b^{*2} + U^{33}l^2c^{*2} + 2U^{23}klb^*c^* + 2U^{13}hla^*c^* + 2U^{12}hka^*b^*)]$$

	x	y	z	U^{11}	U^{22}	U^{33}	$2U^{23}$	$2U^{13}$	$2U^{12}$
Cl⁻	2313 (1)	2500 (0)	4852 (1)	338 (3)	306 (3)	760 (6)	76 (9)	557 (7)	−6 (8)
C(1)	6358 (3)	886 (3)	6104 (4)	343 (15)	339 (16)	377 (16)	20 (26)	380 (26)	−196 (26)
C(2)	7487 (4)	518 (4)	7580 (4)	566 (21)	402 (20)	344 (17)	145 (29)	409 (31)	−200 (33)
C(3)	8071 (4)	1715 (4)	8480 (4)	585 (21)	486 (21)	287 (15)	−36 (31)	350 (29)	−66 (36)
C(4)	8559 (4)	2835 (3)	7694 (3)	381 (15)	340 (17)	304 (15)	−160 (25)	227 (25)	−52 (24)
C(5)	7394 (3)	3181 (3)	6186 (3)	264 (12)	241 (14)	377 (16)	−152 (25)	295 (23)	8 (23)
C(6)	8302 (3)	3844 (3)	5303 (3)	259 (12)	197 (12)	380 (16)	−43 (24)	174 (23)	−45 (22)
C(7)	7486 (3)	4107 (3)	3666 (4)	300 (14)	227 (14)	429 (17)	166 (25)	203 (24)	−4 (22)
C(8)	6671 (3)	2920 (3)	2896 (3)	266 (13)	243 (14)	331 (14)	96 (23)	129 (23)	75 (21)
C(9)	5729 (3)	2334 (3)	3778 (3)	185 (11)	230 (14)	400 (15)	29 (25)	146 (21)	37 (21)
C(10)	6874 (3)	1934 (3)	5242 (3)	215 (12)	209 (12)	321 (14)	19 (23)	257 (21)	23 (20)
C(11)	4814 (3)	1329 (3)	2667 (4)	270 (14)	301 (15)	426 (17)	8 (28)	53 (25)	−17 (25)
C(12)	5764 (4)	162 (3)	2506 (4)	460 (18)	232 (14)	337 (16)	−31 (26)	72 (26)	−22 (26)
C(13)	7144 (4)	631 (3)	2221 (3)	389 (16)	286 (15)	278 (14)	17 (26)	98 (26)	175 (25)
C(14)	7835 (3)	1827 (3)	3094 (3)	248 (12)	311 (15)	290 (14)	58 (25)	240 (22)	119 (24)
C(15)	5603 (4)	3084 (3)	1325 (4)	390 (16)	356 (17)	331 (16)	165 (29)	54 (26)	122 (29)
C(16)	4413 (4)	2096 (4)	1228 (5)	458 (19)	381 (18)	538 (21)	202 (33)	−241 (33)	15 (30)
C(17)	3216 (8)	1930 (7)	91 (9)	1140 (47)	1086 (51)	1284 (54)	1516 (93)	−1727 (88)	−1235 (86)
C(18)	8909 (5)	3974 (4)	8798 (4)	655 (24)	471 (21)	383 (19)	−356 (34)	261 (34)	−45 (38)
C(19)	9909 (3)	2505 (4)	7177 (3)	281 (12)	335 (14)	308 (14)	−27 (30)	50 (21)	−20 (30)
C(20)	8287 (3)	1614 (3)	4777 (3)	213 (11)	197 (12)	279 (13)	7 (21)	203 (20)	108 (19)
N(1)	9339 (2)	2680 (3)	5517 (3)	193 (9)	276 (12)	313 (12)	55 (20)	170 (17)	3 (19)
O(1)	8718 (3)	−185 (3)	7402 (3)	570 (15)	365 (12)	422 (13)	121 (22)	111 (22)	65 (23)
O(2)	9192 (3)	4877 (2)	5968 (3)	382 (12)	265 (10)	582 (15)	1 (21)	117 (21)	−200 (19)
O(3)	7649 (3)	135 (3)	1348 (3)	711 (17)	570 (17)	383 (13)	−244 (26)	437 (24)	307 (29)

(b) Hydrogen atoms

All values were multiplied by 10^3.

	x	y	z	U_{iso}
H(11)	550 (4)	121 (4)	634 (4)	42 (10)
H(12)	611 (4)	13 (4)	554 (4)	33 (9)
H(21)	695 (4)	−1 (4)	810 (4)	48 (11)
H(31)	883 (5)	149 (5)	936 (5)	55 (12)
H(32)	731 (5)	216 (5)	879 (5)	61 (13)
H(51)	667 (3)	364 (3)	622 (4)	25 (8)
H(71)	816 (4)	445 (3)	323 (4)	25 (8)
H(72)	685 (4)	487 (3)	361 (3)	26 (8)
H(91)	505 (4)	305 (4)	392 (4)	39 (10)
H(111)	391 (4)	100 (4)	293 (4)	32 (9)
H(121)	516 (4)	−40 (4)	172 (4)	35 (9)
H(122)	601 (4)	−32 (3)	339 (3)	23 (8)
H(141)	863 (4)	217 (4)	280 (4)	31 (9)
H(151)	595 (5)	292 (5)	56 (5)	64 (13)
H(152)	521 (4)	392 (4)	122 (4)	29 (8)
H(171)	311 (6)	232 (7)	−82 (6)	107 (20)
H(172)	285 (7)	125 (7)	−4 (6)	95 (20)
H(181)	900 (4)	473 (4)	836 (4)	43 (10)
H(182)	798 (5)	418 (5)	912 (5)	64 (14)
H(183)	967 (5)	369 (4)	960 (5)	50 (12)
H(191)	1066 (4)	319 (4)	749 (4)	32 (9)
H(192)	1023 (4)	159 (4)	737 (4)	37 (9)
H(201)	874 (3)	78 (3)	513 (3)	14 (7)
H(N11)	997 (5)	289 (4)	513 (5)	47 (11)
H(O11)	843 (6)	−84 (5)	701 (5)	73 (15)
H(O21)	881 (5)	547 (4)	585 (5)	52 (12)

Table 3. *Observed and calculated structure factors*

All *F* values were multiplied by 10. Unobserved reflexions are marked with an asterisk. The 001 reflexion was excluded from least-squares calculations.

K	|FO|	|FC|
H= 0, L= 0		
2	575	560
4	618	597
6	37	40
8	170	172
10	92	93
12	102	105
H= 0, L= 1		
0	735	815
1	248	213
2	385	370
3	62	58
4	246	247
5	133	135
6	212	212
7	86	88
8	157	160
9	62	70
10	98	100
11	61	59
12	103	102
13	38	24
H= 0, L= 2		
0	109	95
1	160	138
2	507	525
3	252	252
4	210	206
5	219	223
6	180	183
7	124	125
8	113	112
9	47	48
10	68	69
11	56	60
12	90	76
13	30*	6
H= 0, L= 3		
0	358	356
1	163	167
2	158	166
3	359	354
4	166	167
5	195	197
6	110	110
7	87	85
8	114	117
9	64	67
10	83	88
11	87	87
12	29	35
H= 0, L= 4		
0	193	186
1	237	226
2	278	274
3	102	100
4	153	159
5	49	48
6	81	80
7	65	65
8	126	125
9	81	91
10	154	162
11	96	98
12	44	42
H= 0, L= 5		
0	219	222
1	79	79
2	265	265
3	197	198
4	137	138
5	91	96
6	41	41
7	37	39
8	104	107
9	52	57
10	33	36
11	31	33
12	32	28
H= 0, L= 6		
0	35	45
1	103	104
2	199	200
3	40	39
4	159	160
5	136	147
6	52	54
7	124	127
8	26*	30
9	37	32
10	20*	24
11	30*	23
H= 0, L= 7		
0	66	60
1	22*	22
2	25	16
3	23*	12
4	37	44
5	66	65
6	118	116
7	26*	18
8	27*	11
9	29*	32
10	40	43
H= 0, L= 8		
0	262	268
1	24*	23
2	24*	28
3	72	74
4	55	56
5	26*	25
6	27*	26
7	31	37
8	29*	21
9	30*	25
H= 0, L= 9		
0	54	56
1	26*	1
2	38	37
3	26*	14
4	37	35
5	39	44
6	51	50
7	29*	27
8	47	48
H= 0, L= 10		
0	28*	25
1	38	40
2	52	44
3	29	33
4	29*	15
5	29*	22
6	30*	24
H= 0, L= 11		
0	29*	29
1	42	43
2	30*	30
3	40	46
4	30*	26
H= 1, L= 0		
0	91	72
1	771	700
2	344	331
3	527	538
4	193	193
5	183	180
6	75	70
7	243	243
8	131	132
9	230	235
10	144	148
11	89	86
12	58	55
13	30*	28
H= 1, L= 1		
0	451	416
1	430	415
2	176	169
3	489	515
4	168	161
5	279	281
6	114	118
7	234	238
8	76	76
9	183	185
10	79	85
11	63	66
12	31	26
13	30*	17
H= 1, L= -1		
0	160	143
1	776	746
2	124	118
3	119	110
4	191	185
5	186	182
6	42	42
7	91	92
8	92	92
9	125	130
10	25*	15
11	116	115
12	29*	20
13	43	36
H= 1, L= 2		
0	211	203
1	337	331
2	252	250
3	413	418
4	416	410
5	376	379
6	87	84
7	112	117
8	111	115
9	139	139
10	28	29
11	85	93
12	29*	10
13	31*	26
H= 1, L= -2		
0	71	67
1	684	661
2	128	108
3	313	311
4	338	321
5	203	208
6	64	65
7	59	60
8	141	145
9	59	60
10	25*	16
11	66	67
12	29*	17
13	30*	3
H= 1, L= 3		
0	358	345
1	575	588
2	247	236
3	283	289
4	105	96
5	251	252
6	96	97
7	179	179
8	46	44
9	38	36
10	51	49
11	77	70
12	29*	25
H= 1, L= -3		
0	80	62
1	214	238
2	144	137
3	262	260
4	17*	18
5	177	173
6	84	85
7	201	202
8	108	113
9	104	108
10	50	49
11	93	88
12	29*	7
13	41	38
H= 1, L= 4		
0	440	428
1	326	331
2	419	408
3	85	85
4	161	153
5	133	134
6	103	104
7	62	66
8	33	26
9	61	68
10	61	58
11	48	46
12	30*	16
H= 1, L= -4		
0	122	115
1	215	215
2	91	94
3	52	55
4	84	83
5	57	57
6	85	82
7	122	120
8	94	94
9	119	119
10	51	53
11	79	74
12	50	51
H= 1, L= 5		
0	253	255
1	92	87
2	75	66
3	142	141
4	117	118
5	253	258
6	164	167
7	152	150
8	94	95
9	98	105
10	42	36
11	59	61
H= 1, L= -5		
0	190	187
1	160	154
2	129	129
3	87	85
4	206	204
5	196	195
6	36	39
7	148	145
8	53	52
9	60	53
10	60	58
11	117	117
12	50	48
H= 1, L= 6		
0	21*	2
1	159	141
2	46	47
3	169	178
4	124	131
5	156	159
6	180	178
7	121	121
8	49	58
9	29	27
10	57	54
11	30*	16
H= 1, L= -6		
0	46	46
1	71	71
2	125	124
3	71	78
4	153	148
5	176	174
6	138	141
7	66	67
8	70	77
9	81	76
10	48	43
11	53	54
H= 1, L= 7		
0	51	47
1	41	40
2	76	79
3	93	91
4	24*	13
5	45	46
6	44	45
7	57	56
-8	34	31
9	38	32
10	33	38
H= 1, L= -7		
0	21*	9
1	116	114
2	35	30
3	138	139
4	124	128
5	65	67
6	144	144
7	74	73
8	90	87
9	60	64
10	39	38
H= 1, L= 8		
0	44	43
1	57	64
2	94	94
3	69	63
4	30	26
5	26*	18
6	27*	26
7	49	48
8	48	42
9	30*	26
H= 1, L= -8		
0	99	99
1	56	55
2	148	149
3	197	198
4	92	97
5	28	32
6	53	54
7	27*	16
8	37	44
9	55	47
10	31*	27
H= 1, L= 9		
0	51	50
1	48	46
2	51	51
3	77	70
4	49	53
5	69	59
6	38	36
7	46	39
8	50	46
H= 1, L= -9		
0	92	99
1	36	33
2	107	111
3	26*	14
4	26*	19
5	66	73
6	45	53
7	36	34
8	30*	17
H= 1, L= 10		
0	36	37
1	28	38
2	30	35
3	38	31
4	29*	18
5	48	45
6	49	47
H= 1, L= -10		
0	61	58
1	31	33
2	27*	24
3	38	36
4	28	31
5	38	33
6	30*	14
7	42	32
H= 1, L= 11		
0	30*	15
1	72	70
2	31	19
3	34	34
H= 1, L= -11		
0	29*	3
1	29*	12
2	52	53
3	30*	15
4	30*	23
5	31*	23
H= 2, L= 0		
0	647	625
1	230	230
2	124	117
3	247	250
4	62	59
5	225	215
6	186	183
7	54	56
8	118	123
9	117	121
10	65	66
11	27*	16
12	72	72
13	30*	23
H= 2, L= 1		
0	227	218
1	181	169
2	398	411
3	103	97
4	337	319
5	260	255
6	210	212
7	93	89
8	72	72
9	44	44
10	77	77
11	63	59
12	50	53
13	38	41
H= 2, L= -1		
0	861	884
1	437	415
2	558	520
3	58	53
4	225	229
5	121	121
6	92	97
7	54	55
8	100	106
9	138	141
10	40	38
11	44	46
12	65	65
13	38	30
H= 2, L= 2		
0	252	251
1	158	151
2	337	338
3	318	303
4	236	224
5	52	50
6	71	79
7	88	84
8	83	88
9	62	63
10	68	69
11	41	44
12	79	75
H= 2, L= -2		
0	247	221
1	456	449
2	358	350
3	285	280
4	444	439
5	56	59
6	84	88
7	37	38
8	288	295
9	159	151
10	111	115
11	27*	26
12	48	47
13	40	34
H= 2, L= 3		
0	244	246
1	234	223
2	260	252
3	74	65
4	226	232
5	64	61
6	46	45
7	41	40
8	90	89
9	88	90
10	85	86
11	28*	16
12	39	47
H= 2, L= -3		
0	224	226
1	65	57
2	201	199
3	232	220
4	179	170
5	75	77
6	161	161
7	158	159
8	235	239
9	130	132
10	156	152
11	28*	16
12	80	77
H= 2, L= 4		
0	64	41
1	148	142
2	168	164
3	77	77
4	75	70
5	147	146
6	86	87
7	35	31
8	58	65
9	92	95
10	53	60
11	30*	23
12	34	31
H= 2, L= -4		
0	277	273
1	158	160
2	266	273
3	124	120
4	203	200
5	142	139
6	203	196
7	149	146
8	63	57
9	30	37
10	119	118
11	78	75
12	30*	23
H= 2, L= 5		
0	60	56
1	198	199
2	132	128
3	298	305
4	154	152
5	189	196
6	40	44
7	26	25
8	26*	6
9	27*	27
10	36	33
11	30*	26
H= 2, L= -5		
0	273	266
1	220	214
2	40	39
3	121	112
4	285	293
5	21*	3
6	39	38
7	123	123
8	159	160
9	26*	15
10	85	80
11	104	97
12	53	50
H= 2, L= 6		
0	143	142
1	68	70
2	176	179
3	221	226
4	186	192
5	96	97
6	74	78
7	79	80
8	54	57
9	49	52
10	30*	27
H= 2, L= -6		
0	286	290
1	184	176
2	154	154
3	86	89
4	50	54
5	77	77
6	83	79
7	52	51
8	143	133
9	79	73
10	50	48
11	63	62
H= 2, L= 7		
0	193	201
1	159	162
2	29	36
3	69	80
4	93	94
5	52	61
6	28	26
7	27*	25
8	29	30
9	37	35
H= 2, L= -7		
0	80	79
1	105	109
2	155	155
3	124	121
4	110	109
5	97	96
6	210	209
7	74	73
8	28	26
9	28*	20
10	63	63
11	31*	10
H= 2, L= 8		
0	52	57
1	47	54
2	26*	15
3	26*	9
4	49	48
5	51	55
6	43	45
7	29*	7
8	44	36
H= 2, L= -8		
0	71	69
1	26	20
2	54	54
3	35	32
4	109	121
5	55	56
6	137	140
7	35	40
8	60	55
9	36	25
10	45	39
H= 2, L= 9		
0	34	36
1	37	35
2	64	61
3	42	37
4	28*	18
5	47	48
6	30*	30
7	71	73
H= 2, L= -9		
0	69	65
1	119	119
2	101	101
3	31	29
4	28	25
5	29	32
6	28*	26
7	29	31
8	30*	27
9	39	43
H= 2, L= 10		
0	39	34
1	35	35
2	45	39
3	52	52
4	38	36
5	35	40
H= 2, L= -10		
0	27*	12
1	60	59
2	53	50
3	29	32
4	45	39
5	28*	22
6	29*	15
7	30*	11
H= 2, L= -11		
0	36	27
1	41	41
2	29*	19
3	32	37
4	37	40
5	39*	33
H= 2, L= -12		
0	30*	12
1	33	32
2	45	37
H= 3, L= 0		
0	290	277
1	313	314
2	60	62
3	403	419
4	206	198
5	287	283
6	91	87
7	265	268
8	121	126
9	164	165
10	26*	28
11	44	48
12	38	36
H= 3, L= 1		
0	200	200
1	134	135
2	231	218
3	294	286
4	82	80
5	266	268
6	71	64
7	130	135
8	40	45
9	118	119
10	133	129
11	72	71
12	37	35
H= 3, L= -1		
0	249	258
1	408	420
2	143	164
3	355	384
4	221	225
5	324	326
6	33	36
7	184	187
8	126	125
9	254	245
10	89	90
11	82	81
12	42	47
13	94	83
H= 3, L= 2		
0	76	77
1	157	140
2	167	164
3	93	98
4	136	134
5	190	195
6	206	211
7	91	94
8	71	70
9	94	95
10	108	107
11	51	48
12	66	60
H= 3, L= -2		
0	16	18
1	589	604
2	106	97
3	262	259
4	138	135
5	275	276
6	131	136
7	136	128
8	90	92
9	138	136
10	90	95
11	127	125
12	59	60
H= 3, L= 3		
0	183	180
1	279	282
2	340	340
3	60	64
4	37	35
5	22*	15
6	77	81
7	126	128
8	25*	3
9	83	93
10	59	60
11	51	52
12	62	67
H= 3, L= -3		
0	473	463
1	394	389
2	198	188
3	330	331
4	50	48
5	223	222
6	52	55
7	128	124
8	166	168
9	71	73
10	53	51
11	67	62
12	37	35
H= 3, L= 4		
0	19*	7
1	108	111
2	20*	8
3	135	136
4	104	108
5	163	165
6	38	37
7	102	107
8	58	58
9	62	64
10	36	32
11	39	42
H= 3, L= -4		
0	23	24
1	202	199
2	231	222
3	390	382
4	123	123
5	151	150
6	151	153
7	178	175
8	113	119
9	88	94
10	47	48
11	58	56
12	30*	21
H= 3, L= 5		
0	143	144
1	21	28
2	161	161
3	274	280
4	117	120
5	66	65
6	146	150
7	86	87
8	27*	26
9	36	41
10	29*	23
11	31*	29
H= 3, L= -5		
0	114	114
1	137	143
2	59	61
3	83	82
4	123	120
5	34	37
6	107	98
7	105	98
8	37	47
9	72	71
10	76	76
11	120	118
12	47	52
H= 3, L= 6		
0	33	30
1	82	81
2	23*	8
3	94	96
4	51	55
5	57	63
6	58	64
7	54	57
8	28*	24
9	20*	8
10	50	44
H= 3, L= -6		
0	120	121
1	161	155
2	76	76
3	153	155
4	30	28
5	144	141
6	60	59
7	35	30
8	26*	22
9	84	76
10	34	37
11	99	100
H= 3, L= 7		
0	30	37
1	33	31
2	56	54
3	32	31
4	53	54
5	77	19
6	65	65
7	57	50
8	58	57
9	87	81
H= 3, L= -7		
0	45	46
1	127	127
2	157	159
3	187	190
4	122	124
5	103	103
6	41	43
7	37	38
8	62	63
9	64	61
10	44	36
11	76	70
H= 3, L= 8		
0	71	68
1	26*	15
2	27*	15
3	36	43
4	69	65
5	32	31
6	40	42
7	65	64
8	52	58
H= 3, L= -8		
0	41	40
1	39	47
2	75	77
3	149	148
4	52	50
5	57	60
6	26*	28
7	27*	19
8	75	70
9	29*	15
10	31*	7
H= 3, L= 9		
0	33	30
1	37	32
2	62	55
3	29*	18
4	29*	12
5	53	49
6	52	50
H= 3, L= -9		
0	99	89
1	87	83
2	100	102
3	53	55
4	81	78
5	72	69
6	28*	21
7	54	52
8	35	24
9	39	35
H= 3, L= 10		
0	30*	3
1	51	55
2	31	28
3	31*	28
H= 3, L= -10		
0	127	129
1	63	64
2	37	41
3	62	63
4	50	51
5	49	39
6	29*	17
7	36	35
H= 3, L= -11		
0	44	46
1	48	49
2	29*	24
3	63	55
4	29*	6
5	30*	25
6	31*	23
H= 3, L= -12		
0	30*	8
1	30*	28
2	36	36
H= 4, L= 0		
0	204	204
1	161	158
2	250	257
3	113	106
4	100	106
5	271	271
6	148	146
7	122	120
8	52	49
9	28	31
10	150	148
11	28*	13
12	59	58
H= 4, L= 1		
0	154	160
1	217	216
2	255	252
3	56	52
4	170	175
5	85	81
6	74	70
7	83	83
8	74	77
9	50	52
10	67	74
11	39	40
12	87	85
H= 4, L= -1		
0	15*	3
1	198	198
2	363	401
3	305	303
4	47	43
5	296	289
6	188	195
7	104	105
8	62	60
9	60	58
10	91	86
11	28*	12
12	30*	21
H= 4, L= 2		
0	189	190
1	52	53
2	195	188
3	157	152
4	308	319
5	91	94
6	50	54
7	44	54
8	92	101
9	26*	12
10	29	33
11	52	51
12	58	59
H= 4, L= -2		
0	182	175
1	147	136
2	382	375
3	324	329
4	239	228
5	122	120
6	166	166
7	75	75
8	110	106
9	73	74
10	42	40
11	47	39
12	30*	16
H= 4, L= 3		
0	257	258
1	123	130
2	72	75
3	120	120
4	239	247
5	255	268
6	113	110
7	142	150
8	56	55
9	32	45
10	57	59
11	30*	17
H= 4, L= -3		
0	154	150
1	139	137
2	130	122
3	299	291
4	170	174
5	54	57
6	119	122
7	27	34
8	128	129
9	93	89
10	71	74
11	49	46
12	65	60
H= 4, L= 4		
0	66	61
1	121	122
2	127	125
3	155	157
4	74	76
5	175	183
6	120	122
7	81	85
8	72	71
9	28*	28
10	79	67
11	47	45
H= 4, L= -4		
0	30*	15
1	127	124
2	224	226
3	83	84
4	47	47
5	44	46
6	258	287
7	32	36
8	98	101
9	34	34
10	128	136
11	43	43
12	34	36
H= 4, L= 5		
0	76	74
1	100	102
2	182	181
3	180	187
4	154	156
5	25*	10
6	26*	16
7	82	82
8	76	72
9	50	53
10	35	37
H= 4, L= -5		
0	162	162
1	54	49
2	102	105
3	37	39
4	159	167
5	149	156
6	133	140
7	34	31
8	122	131
9	27*	32
10	57	54
11	29*	24
H= 4, L= 6		
0	70	69
1	39	46
2	24*	16
3	110	109
4	73	74
5	60	63
6	46	42
7	28*	28
8	31	42
9	75	73
H= 4, L= -6		
0	127	130
1	77	73
2	106	105
3	38	39
4	162	158
5	108	111
6	67	64
7	51	50
8	154	155
9	27*	30
10	32	29
11	20*	25
H= 4, L= 7		
0	26*	17
1	29	26
2	45	46
3	26*	18
4	42	35
5	40	33
6	28	22
7	82	78
8	64	67
H= 4, L= -7		
0	67	66
1	93	86
2	152	155
3	112	106
4	85	88
5	71	69
6	102	105
7	26*	27
8	52	53
9	71	68
10	76	76
H= 4, L= 8		
0	28*	4
1	55	52
2	66	59
3	28*	21
4	29*	15
5	39	36
6	30*	29
H= 4, L= -8		
0	60	68
1	121	121
2	87	83
3	141	142
4	134	133
5	90	89
6	89	81
7	64	60
8	45	49
9	49	53
10	73	66
H= 4, L= 9		
0	44	39
1	30	31
2	71	67
3	30	35
4	31	26
H= 4, L= -9		
0	25*	14
1	47	43
2	112	114
3	33	38
4	44	44
5	65	73
6	46	47
7	87	88
8	71	68
9	31*	17
H= 4, L= -10		
0	27*	6
1	48	48
2	82	80
3	39	46
4	35	28
5	28*	5
6	36	34
7	71	70
H= 4, L= -11		
0	28*	16
1	59	51
2	53	53
3	29*	8
4	70	73
5	30*	26
6	31*	25
H= 4, L= -12		
0	30*	16
1	41	43
2	30*	19
3	34	29
H= 5, L= 0		
0	29	29
1	118	119
2	194	197
3	158	163
4	65	61
5	42	40
6	96	98
7	151	153
8	74	72
9	39	35
10	77	82
11	63	63
12	69	68
H= 5, L= 1		
0	35	36
1	162	168
2	208	207
3	26	22
4	29	26
5	51	55
6	134	132
7	77	77
8	66	61
9	75	76
10	104	102
11	68	64
H= 5, L= -1		
0	205	206
1	160	158
2	58	60
3	113	115
4	65	63
5	133	132
6	146	141
7	30	29
8	93	88
9	32	32
10	27*	23
11	53	55
12	70	66
H= 5, L= 2		
0	121	119
1	68	60
2	99	98
3	137	132
4	188	189
5	115	121
6	173	170
7	70	67
8	83	81
9	57	52
10	64	62
11	30*	19
H= 5, L= -2		
0	30	34
1	194	192
2	61	54
3	199	190
4	164	154
5	140	154
6	104	108
7	136	124
8	54	55
9	100	95
10	44	53
11	97	90
12	60	56
H= 5, L= 3		
0	100	98
1	37	38
2	64	66
3	46	46
4	123	119
5	169	170
6	117	119
7	39	33
8	59	56
9	28*	12
10	35	34
11	41	34
H= 5, L= -3		
0	18	17
1	467	468
2	33	28
3	305	285
4	139	132
5	133	134
6	51	48
7	110	108
8	70	73
9	126	123
10	51	48
11	97	93
12	41	38
H= 5, L= 4		
0	22*	11
1	94	91
2	85	87
3	86	83
4	73	72
5	57	58
6	78	25
7	74	68
8	94	96
9	35	30
10	47	48
H= 5, L= -4		
0	77	21
1	300	286
2	157	164
3	310	298
4	54	53
5	106	106
6	33	36
7	169	170
8	80	82
9	87	79
10	28*	14
11	58	58
H= 5, L= 5		
0	164	167
1	135	137
2	78	77
3	25*	22
4	72	69
5	35	37
6	40	40
7	52	47
8	57	55
9	30*	16
H= 5, L= -5		
0	29*	6
1	215	209
2	55	50
3	185	198
4	61	63
5	54	57
6	68	74
7	37	35
8	29	33
9	46	47
10	39	39
11	58	55
H= 5, L= 6		
0	139	137
1	60	56
2	104	101
3	32	24
4	54	50
5	69	64
6	95	89
7	34	22
8	53	47
H= 5, L= -6		
0	79	81
1	140	143
2	124	127
3	38	41
4	38	40
5	139	141
6	169	171
7	55	52
8	27*	7
9	67	68
10	33	30
11	50	50
H= 5, L= 7		
0	48	43
1	34	29
2	27*	28
3	65	59
4	47	45
5	29*	16
6	77	75
7	31*	9
H= 5, L= -7		
0	96	99
1	149	148
2	66	63
3	144	150
4	74	74
5	67	58
6	25*	28
7	31	33
8	28*	21
9	58	61
10	30*	17
H= 5, L= 8		
0	29*	8
1	51	46
2	39	29
3	29*	21
4	51	50
5	31*	9
H= 5, L= -8		
0	24*	6
1	153	162
2	53	56
3	106	108
4	59	62
5	26*	5
6	27*	25
7	92	92
8	46	38
9	50	46
H= 5, L= 9		
0	31*	0
1	31*	14
H= 5, L= -9		
0	87	91
1	130	129
2	56	54
3	61	63
4	26*	26
5	76	77
6	45	43
7	52	43
8	40	34
H= 5, L= -10		
0	50	49
1	70	69
2	27*	25
3	37	39
4	54	55
5	61	62
6	73	72
7	30*	6
H= 5, L= -11		
0	29*	13
1	82	79
2	37	34
3	86	86
4	32	28
5	30*	20
H= 5, L= -12		
0	39	33
1	58*	53
2	31*	14
H= 6, L= 0		
0	188	187
1	56	56
2	94	91
3	163	166
4	246	249
5	65	67
6	81	81
7	60	59
8	86	89
9	27*	9
10	79	78
11	54	50
H= 6, L= 1		
0	128	133
1	153	153
2	91	86
3	162	162
4	177	179
5	207	210
6	71	73
7	72	70
8	80	83
9	81	71
10	39	31
11	45	40
H= 6, L= -1		
0	36	36
1	74	73
2	127	127
3	110	109
4	202	204
5	76	71
6	185	188
7	52	52
8	81	78
9	33	34
10	99	92
11	34	40
H= 6, L= 2		
0	22*	19
1	55	53
2	156	164
3	122	121
4	109	110
5	130	135
6	146	150
7	123	126
8	41	34
9	51	50
10	47	45
H= 6, L= -2		
0	148	152
1	132	125
2	226	219
3	190	178
4	250	261
5	100	98
6	280	276
7	111	109
8	187	187
9	50	53
10	61	64
11	30*	20
H= 6, L= 3		
0	23*	23
1	121	119
2	43	40
3	24*	9
4	115	116
5	58	57
6	82	79
7	51	55
8	29*	12
9	30*	29
10	49	48
H= 6, L= -3		
0	170	181
1	121	118
2	124	131
3	110	103
4	244	249
5	109	102
6	192	198
7	24*	12
8	126	125
9	38	43
10	60	60
11	48	51
H= 6, L= 4		
0	226	237
1	111	110
2	72	75
3	43	46
4	78	80
5	59	60
6	41	46
7	31	28
8	29	27
9	33	31
H= 6, L= -4		
0	59	59
1	154	152
2	123	121
3	65	65
4	110	107
5	102	102
6	138	129
7	34	31
8	44	40
9	29	32
10	135	121
11	78	75
H= 6, L= 5		
0	45	46
1	49	51
2	35	34
3	50	51
4	31	23
5	27*	17
6	32	27
7	50	49
8	20*	21
H= 6, L= -5		
0	76	74
1	77	77
2	46	43
3	22*	19
4	114	120
5	44	37
6	156	159
7	58	53
8	71	68
9	28*	7
10	69	66
11	31	28
H= 6, L= 6		
0	27*	5
1	30	30
2	33	32
3	29	31
4	33	33
5	29*	18
6	55	47
7	56	49
H= 6, L= -6		
0	55	58
1	64	61
2	234	237
3	51	50
4	122	118
5	97	103

Table 3 (*cont.*)

K	|Fo|	|Fc|
H= 6, L= -6		
6	163	170
7	86	99
8	85	94
9	29*	26
10	58	57
H= 6, L= 7		
0	30	20
1	57	55
2	48	45
3	41	38
4	30*	6
5	30*	8
H= 6, L= -7		
0	94	91
1	133	141
2	135	134
3	71	75
4	76	76
5	84	88
6	118	119
7	49	53
8	85	81
9	29*	25
10	68	66
H= 6, L= 8		
0	43	36
1	58	59
2	33	35
H= 6, L= -8		
0	104	111
1	74	71
2	73	74
3	61	64
4	126	126
5	36	35
6	66	66
7	36	38
8	53	48
9	30*	15
H= 6, L= -9		
0	26*	20
1	57	54
2	70	70
3	49	46
4	38	37
5	29	29
6	28*	12
7	36	27
8	30*	30
H= 6, L= -10		
0	27*	5
1	27*	16
2	53	51
3	30	26
4	44	47
5	64	63
6	36	30
7	82	77
H= 6, L= -11		
0	29*	33
1	29*	26
2	38	35
3	30	29
4	52	51
5	61	61
H= 6, L= -12		
0	31*	5
1	40	36
H= 7, L= 0		
0	23	18
1	120	124
2	54	48
3	152	155
4	23*	22
5	35	40
6	68	69
7	32	28
8	69	69
9	78	77
10	52	54
H= 7, L= 1		
0	135	138
1	111	113
2	72	72
3	53	55
4	162	176
5	115	115
6	88	93
7	75	77
8	28*	19
9	53	52
10	34	40
H= 7, L= -1		
0	118	114
1	190	190
2	110	106
3	155	156
4	92	87
5	101	99
6	99	100
7	113	105
8	89	88
9	74	69
10	42	36
H= 7, L= 2		
0	154	154
1	63	64
2	35	38
3	42	39
4	226	230
5	63	65
6	50	52
7	83	81
8	63	67
9	87	82
H= 7, L= -2		
0	137	132
1	201	200
2	106	109
3	70	66
4	135	140
5	122	120
6	194	197
7	30	31
8	103	101
9	33	28
10	62	64
11	46	43
H= 7, L= 3		
0	25*	2
1	215	218
2	65	68
3	141	140
4	72	70
5	27*	12
6	70	70
7	53	50
8	102	103
9	50	47
H= 7, L= -3		
0	51	50
1	59	61
2	66	71
3	22*	14
4	159	162
5	111	114
6	120	119
7	26*	13
8	83	78
9	32	36
10	59	60
11	40	42
H= 7, L= 4		
0	26*	9
1	144	145
2	119	122
3	44	39
4	27*	20
5	43	42
6	43	42
7	29*	19
8	30*	27
H= 7, L= -4		
0	25	32
1	157	155
2	223	234
3	90	89
4	27	24
5	98	93
6	62	63
7	90	92
8	27*	16
9	28*	29
10	33	39
H= 7, L= 5		
0	31	27
1	42	39
2	29	33
3	28*	6
4	44	40
5	36	31
6	30*	26
7	39	37
H= 7, L= -5		
0	22*	7
1	242	249
2	154	158
3	145	148
4	41	39
5	57	59
6	53	52
7	81	83
8	28*	15
9	29*	10
10	30*	26
H= 7, L= 6		
0	29*	31
1	29*	16
2	30	24
3	29	19
4	36	40
5	44	41
H= 7, L= -6		
0	93	94
1	215	214
2	59	56
3	68	71
4	82	87
5	91	91
6	62	63
7	84	84
8	28*	19
9	54	61
10	31*	10
H= 7, L= 7		
0	49	53
1	30	25
2	30*	16
3	31*	16
H= 7, L= -7		
0	29	13
1	84	81
2	70	72
3	77	78
4	25*	17
5	99	97
6	30	36
7	79	79
8	51	49
9	57	60
H= 7, L= -8		
0	70	75
1	59	56
2	71	74
3	118	118
4	54	55
5	27*	22
6	40	37
7	49	48
8	34	28
H= 7, L= -9		
0	41	43
1	32	26
2	37	40
3	27	24
4	46	41
5	50	53
6	29*	8
7	47	47
H= 7, L= -10		
0	28	26
1	35	37
2	47	41
3	56	55
4	37	37
5	41	46
6	41	40
H= 7, L= -11		
0	29*	20
1	51	48
2	45	47
3	45	44
4	53	52
H= 8, L= 0		
0	24*	5
1	123	124
2	73	70
3	67	74
4	64	63
5	52	53
6	103	101
7	76	78
8	49	51
9	73	68
H= 8, L= 1		
0	24*	4
1	50	51
2	107	109
3	42	43
4	54	57
5	50	56
6	46	45
7	78	75
8	42	45
9	69	70
H= 8, L= -1		
0	23	17
1	179	184
2	53	53
3	168	166
4	44	41
5	91	92
6	70	72
7	63	59
8	53	53
9	48	44
10	71	74
H= 8, L= 2		
0	92	94
1	124	124
2	42	44
3	51	55
4	52	53
5	55	55
6	28*	16
7	29*	22
8	41	38
H= 8, L= -2		
0	46	49
1	98	99
2	23*	18
3	101	104
4	101	105
5	38	36
6	49	54
7	27*	18
8	69	67
9	30	33
10	60	58
H= 8, L= 3		
0	95	97
1	77	79
2	62	65
3	52	54
4	34	35
5	83	82
6	29*	24
7	31	32
8	37	35
H= 8, L= -3		
0	26	24
1	93	92
2	75	74
3	90	89
4	165	174
5	60	58
6	92	96
7	51	51
8	85	85
9	29*	6
10	42	43
H= 8, L= 4		
0	35	34
1	110	112
2	28*	21
3	37	35
4	44	36
5	47	49
6	30*	31
H= 8, L= -4		
0	149	142
1	73	74
2	165	160
3	81	81
4	180	185
5	31	28
6	64	66
7	27	32
8	60	60
9	55	53
10	88	91
H= 8, L= 5		
0	29*	23
1	64	62
2	35	33
3	57	56
4	41	45
5	30*	34
H= 8, L= -5		
0	61	57
1	78	76
2	62	58
3	44	39
4	145	140
5	43	43
6	36	38
7	52	49
8	120	114
9	72	68
H= 8, L= 6		
0	30*	9
1	30*	21
2	32	37
H= 8, L= -6		
0	48	56
1	108	105
2	97	98
3	27	22
4	80	77
5	26*	6
6	67	75
7	42	38
8	77	81
9	41	34
H= 8, L= -7		
0	41	42
1	41	36
2	142	141
3	32	36
4	96	92
5	30	24
6	91	90
7	47	47
8	30	38
H= 8, L= -8		
0	137	135
1	38	43
2	44	45
3	51	50
4	80	80
5	29	26
6	63	70
7	61	59
8	32	34
H= 8, L= -9		
0	99	103
1	28*	7
2	97	97
3	28*	27
4	39	38
5	29*	12
6	51	53
7	31*	22
H= 8, L= -10		
0	66	68
1	29*	31
2	95	99
3	36	36
4	30*	29
5	30*	24
H= 8, L= -11		
0	30*	23
1	31	27
2	30*	25
3	31*	21
H= 9, L= 0		
0	97	96
1	67	70
2	102	103
3	119	115
4	137	136
5	27*	25
6	39	44
7	29	20
8	35	27
H= 9, L= 1		
0	61	64
1	26*	17
2	77	81
3	69	64
4	111	110
5	48	50
6	62	62
7	66	64
8	51	43
H= 9, L= -1		
0	88	89
1	132	137
2	41	38
3	140	150
4	50	89
5	52	61
6	68	63
7	29*	15
8	47	40
H= 9, L= 2		
0	27*	6
1	29	27
2	78	75
3	60	62
4	28*	27
5	29*	24
6	51	48
7	31*	13
H= 9, L= -2		
0	48	50
1	139	139
2	25*	23
3	105	106
4	38	42
5	84	85
6	69	75
7	74	70
8	30*	22
9	47	48
H= 9, L= 3		
0	88	86
1	28*	15
2	43	44
3	29*	16
4	29*	32
5	30*	32
6	45	41
H= 9, L= -3		
0	115	116
1	157	162
2	25*	32
3	66	65
4	111	114
5	76	78
6	28*	26
7	67	70
8	57	55
9	31*	28
H= 9, L= 4		
0	34	35
1	44	40
2	30*	18
3	57	53
4	39	42
H= 9, L= -4		
0	48	51
1	98	100
2	73	72
3	56	55
4	121	121
5	58	57
6	43	42
7	105	105
8	42	45
9	64	60
H= 9, L= 5		
0	86	90
1	69	61
H= 9, L= -5		
0	81	81
1	106	106
2	152	152
3	54	51
4	92	92
5	59	51
6	52	55
7	83	81
8	47	48
H= 9, L= -6		
0	127	124
1	124	131
2	106	106
3	92	95
4	90	91
5	71	71
6	49	44
7	29*	21
8	31	30
H= 9, L= -7		
0	50	54
1	32	38
2	61	58
3	71	72
4	42	43
5	83	83
6	43	46
7	89	92
H= 9, L= -8		
0	37	43
1	33	37
2	39	40
3	86	86
4	40	39
5	59	54
6	63	64
7	94	92
H= 9, L= -9		
0	33	31
1	41	39
2	45	51
3	57	57
4	36	42
5	61	60
H= 9, L= -10		
0	30*	29
1	38	39
2	50	48
3	52	42
4	31*	21
H= 10, L= 0		
0	70	66
1	59	59
2	59	66
3	28*	16
4	57	55
5	59	58
6	52	49
7	52	52
H= 10, L= 1		
0	99	96
1	97	95
2	41	38
3	32	35
4	39	25
5	42	37
6	39	27
H= 10, L= -1		
0	27*	18
1	79	79
2	48	45
3	43	44
4	28*	5
5	74	77
6	83	82
7	32	35
H= 10, L= 2		
0	88	86
1	82	81
2	32	27
3	45	44
4	30*	19
5	32	36
H= 10, L= -2		
0	34	31
1	180	178
2	27*	26
3	94	93
4	28*	16
5	28*	21
6	29*	22
7	69	70
H= 10, L= 3		
0	42	39
1	89	85
2	42	33
3	41	40
H= 10, L= -3		
0	26*	8
1	115	113
2	78	80
3	105	105
4	34	35
5	53	51
6	29*	5
7	86	86
H= 10, L= -4		
0	27*	16
1	82	77
2	78	75
3	72	77
4	31	39
5	101	97
6	29*	2
7	85	89
H= 10, L= -5		
0	40	41
1	43	44
2	56	54
3	47	48
4	77	79
5	71	71
6	29*	7
7	87	88
H= 10, L= -6		
0	28	23
1	27*	16
2	39	41
3	31	19
4	62	61
5	24	44
6	39	45
7	91	91
H= 10, L= -7		
0	119	114
1	45	39
2	72	72
3	29*	20
4	41	34
5	30*	14
6	33	34
H= 10, L= -8		
0	162	160
1	37	37
2	72	69
3	29*	21
4	40	38
5	42	43
H= 10, L= -9		
0	46	47
1	33	23
2	30	38
3	30	31
4	31*	20
H= 10, L= -10		
0	31*	10
H= 11, L= 0		
0	39	34
1	65	61
2	62	63
3	39	34
4	73	76
H= 11, L= 1		
0	30*	2
1	74	72
2	61	57
3	31*	15
H= 11, L= -1		
0	69	70
1	29*	27
2	96	95
3	29*	22
4	72	73
5	31*	12
H= 11, L= -2		
0	29*	19
1	50	51
2	79	80
3	32	23
4	69	69
5	61	62
H= 11, L= -3		
0	36	29
1	33	36
2	49	40
3	29*	23
4	63	61
5	56	60
6	82	81
H= 11, L= -4		
0	58	63
1	46	45
2	63	63
3	40	43
4	46	48
5	56	59
6	31*	14
H= 11, L= -5		
0	54	54
1	62	63
2	66	62
3	54	51
4	37	28
5	74	73
H= 11, L= -6		
0	73	67
1	69	63
2	38	38
3	53	51
4	53	55
5	65	66
H= 11, L= -7		
0	29*	7
1	29*	13
2	45	42
3	62	69
4	39	44
H= 11, L= -8		
0	34	29
1	56	57
2	30*	6
3	39	46
H= 12, L= -1		
0	85	84
H= 12, L= -2		
0	54	52
1	31	33
2	36	31
H= 12, L= -3		
0	30*	17
1	39	38
2	52	54
3	46	40
H= 12, L= -4		
0	76	74
1	30*	16
2	83	84
3	31*	24
H= 12, L= -5		
0	100	98
1	50	52
2	63	66
H= 12, L= -6		
0	48	47
1	67	60
2	41	43

ered and when necessary brass attenuation filters were used. A standard reflexion, 600, was measured after every 20–40 reflexions and used for scaling the data. There was no appreciable change in the intensity of this reflexion during the data collection. Of the 2151 independent reflexions with $2\theta \leq 55°$, 1835 (85 %) were observed above threshold value. The threshold was taken as a net count of 150 or 10 % of the background, whichever was higher. The intensities were corrected for Lorentz and polarization factors but an absorption correction was considered unnecessary since the minimum and maximum transmission coefficients (μt) were 0·07 and 0·13 respectively.

Structure determination

The structure was solved by a combination of the heavy-atom method ($\sum_H f^2_H / \sum_L f^2_L = 0{\cdot}33$ at $\sin\theta = 0$) and tangent refinement (Karle, 1968). A summary of the structure determination can be seen in Table 1. From a sharpened Patterson synthesis the position of the chloride ion was determined to be (0·23, 0·25, 0·48) (the y coordinate was arbitrarily chosen as 0·25). A subsequent Fourier synthesis showed six peaks situated at bonding distances from each other which were considered to represent carbon atoms. It turned out later that one of these peaks was spurious and that another was a 'ghost peak' due to the pseudo-mirror plane. The phases based on Cl^- and the assumed six carbon atoms were refined by the tangent formula and all but four atoms were located from the subsequent E map. Two more atoms were identified in the next E map and the last two atoms, C(16) and C(17), were located from a difference Fourier synthesis. The scale factor and the isotropic thermal parameter ($B = 2{\cdot}96$ Å^2) used in the structure factor calculations had been derived from a Wilson plot.

Structure refinement

Coordinates, thermal parameters, and the scale factor were refined by the block-diagonal approximation of the least-squares method, minimizing the expression $\sum w(|F_o| - |F_c|)^2$. The matrices used were 9×9 for coordinates and thermal parameters (4×4 for isotropic thermal motion) of each atom. The scale factor and the overall isotropic thermal parameter were refined by a 2×2 matrix. A shift factor of 0·8 was used throughout the refinement.

The nitrogen and oxygen atoms were identified as such from a difference Fourier synthesis calculated after the third cycle. A low-angle ($\sin\theta/\lambda \leq 0{\cdot}40$ Å^{-1}) difference Fourier synthesis was calculated after the 8th cycle ($R = 8{\cdot}0$ %) and the positions of all hydrogen atoms except H(N11) were indicated by peaks varying

in heights from 0·22 to 0·46 e.Å^{-3}. H(N11) was placed in a calculated position.

The following weighting scheme, chosen so as to reflect the trends in $|\Delta F|$'s, was introduced at this stage: $\sqrt{w} = \sqrt{w_1} \cdot \sqrt{w_2}$ where $\sqrt{w_1} = (|F_o|/p_1)p_2$ when $|F_o| \leq p_1$, $\sqrt{w_1} = (p_1/|F_o|)^{1/2}$ when $|F_o| > p_1$, $\sqrt{w_2} = (\sin^2 \theta/p_3)^{1/2}$ when $\sin^2 \theta \leq p_3$, $\sqrt{w_2} = 1$ when $\sin^2 \theta > p_3$. The parameters were adjusted during the refinement in order to make $w(\Delta F)^2$ as linear as possible with respect to $|F_o|$ and to $\sin^2 \theta$. The final values were $p_1 = 10·0$, $p_2 = 0·12$ and $p_3 = 0·022$.

The refinement converged to an R value of 3·76% ($R' = 4·40\%$) for 1834 observed reflexions. The strongest reflexion, 001, was affected by extinction and therefore excluded from the refinement. After the last cycle of refinement the average shift in the parameters was $0·06\sigma$ and no shift was greater than $0·3\sigma$.

A difference Fourier synthesis was calculated based on the final parameters but excluding the contribution of the hydrogen atoms to F_c. The refined positions of the hydrogen atoms are all on well-defined peaks except H(171) and H(172). This can be attributed to the pronounced anisotropic thermal vibration of these two atoms. A final difference Fourier synthesis was also computed. It showed no significant features although there was an indication of bonding electrons at several bonds.

The scattering factors of Hanson, Herman, Lea & Skillman (1964) were used throughout the analysis for carbon, nitrogen and oxygen atoms. The values for Cl⁻ were obtained from Cromer & Waber (1965) and Cromer (1965), and those for bonded hydrogen atoms were taken from Stewart, Davidson & Simpson (1965).

The final coordinates and thermal parameters are listed in Table 2. The listed estimated standard devia-tion values were obtained from the inverse of the least-squares matrix. Table 3 shows the observed and calculated structure factors. The unobserved reflexions are marked with an asterisk. They were given the threshold value but were excluded from the least-squares refinement. A summary of the agreement be-tween $|F_o|$ and $|F_c|$ can be seen in Table 4.

Thermal vibration

The thermal vibration of the cation was analysed in terms of a rigid body (Cruickshank, 1956). The strongly vibrating C(17) and all the hydrogen atoms were ex-cluded. The calculation showed that the molecule vi-brates only approximately as a rigid body since $\sigma(U_{\text{calc}})$ ($\simeq 0·005$ Å^2) was larger than the e.s.d.'s of the observed U^{ij} (0·001–0·002 Å^2). The maximum libration occurred about an axis which was perpendicular to the C(10)–C(20) bond and in the (010) plane. It was small (3·7°) and consequently the corrections to the positional par-ameters (Cruickshank, 1961) were also small. The cor-rections in bond lengths (0·001–0·005 Å) were con-sidered insignificant and they were therefore ignored.

Description and discussion of the structure

The molecular structure and conformation can be seen in Fig. 1. The bond lengths and angles for non-hydro-gen atoms are shown in Fig. 2. The range of e.s.d.'s for these bond lengths is 0·004–0·005 Å (except for C(16)–C(17) which has an e.s.d. of 0·009 Å). For the valency angles the e.s.d.'s vary between 0·2 and 0·3°. The two angles associated with C(17) have e.s.d.'s of 0·5°. These e.s.d.'s were derived from the block-diagonal approx-imation of the least squares and correlations between

Table 4. *Agreement summary*

1834 observed reflexions in the range $1·58 \leq |F_o| \leq 86·07$

Category	Limits	Number
1	$|\Delta F|/|F_o| \leq 2R(0·075)$, or $|\Delta F| \leq |F_{th}|$	1822
2	$2R < |\Delta F|/|F_o| \leq 3R(0·113)$, or $|F_{th}| < |\Delta F| \leq 2|F_{th}|$	10
3	$3R < |\Delta F|/|F_o| \leq 4R(0·150)$, or $2|F_{th}| < |\Delta F| \leq 3|F_{th}|$	2

316 unobserved reflexions

Category	Limits	Number
1	$|F_c| \leq |F_{th}|$	291
2	$|F_{th}| < |F_c| \leq 1·5|F_{th}|$	25

$|F_{th}|$ is the threshold amplitude as defined in the text. Its range is $1·54 \leq |F_{th}| \leq 3·08$
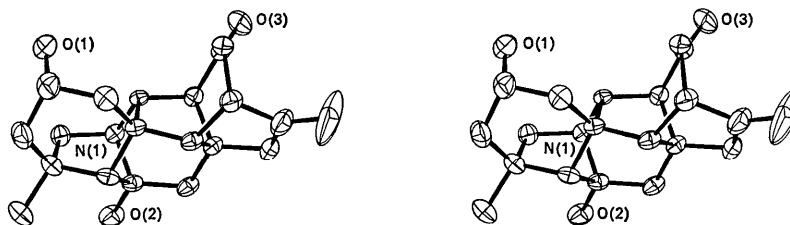

Fig. 1. Stereoscopic view of delnudine. The thermal ellipsoids enclose 50% probability.

the x and z coordinates and between atoms were not taken into account. They may therefore be somewhat underestimated. Table 5 shows bond lengths involving hydrogen atoms. The bond angles involving hydrogen atoms do not differ from usually observed values and they are not listed.

Table 5. Bond lengths involving hydrogen atoms

Their e.s.d.'s are: Csp^3–H 0·03–0·05 Å; Csp^2–H 0·06–0·07 Å; N–H and O–H 0·05 Å.

C(1)—H(11)	0·97 Å	C(18)—H(181)	0·91 Å
C(1)—H(12)	0·94	C(18)–H(182)	1·05
C(2)—H(21)	0·98	C(18)—H(183)	0·92
C(3)—H(31)	0·95	C(19)–H(191)	0·99
C(3)—H(32)	0·98	C(19)–H(192)	1·00
C(5)—H(51)	0·85	C(20)—H(201)	0·98
C(7)—H(71)	0·94		
C(7)—H(72)	0·99	C(17)–H(171)	0·94
C(9)—H(91)	1·02	C(17)–H(172)	0·77
C(11)–H(111)	1·03		
C(12)–H(121)	0·98	N(1)—H(N11)	0·83
C(12)–H(122)	0·94		
C(14)–H(141)	0·96	O(1)—H(O11)	0·78
C(15)–H(151)	0·91	O(2)—H(O21)	0·70
C(15)–H(152)	0·93		

Conformation

The ring system of delnudine has not been observed previously. It is related to that of hetisine (II) (Przybylska, 1963), only two of the rings (C and D) being different in the two alkaloids. The following ring junctions are present: A/B and B/C trans, B/D, C/E and D/F cis.

All six-membered rings occur in the chair form, except the one in the azabicyclo[2,2,1]heptane system which is necessarily boat-shaped. All five-membered rings are envelope-shaped. The ring system is very rigid and only the conformation of ring A is not determined by fusion with other rings. There are several bicyclic

systems fused to each other and this imposes considerable strain on the molecule. Many of the rings are therefore distorted from ideal conformations. This can be seen from the torsional angles (Table 6) and the mean planes (Table 7). For the purpose of comparisons it may be noted that the torsional angles in the energetically most favorable cyclohexane chair are $\pm 55·8°$ (Bucourt & Hainaut, 1965).
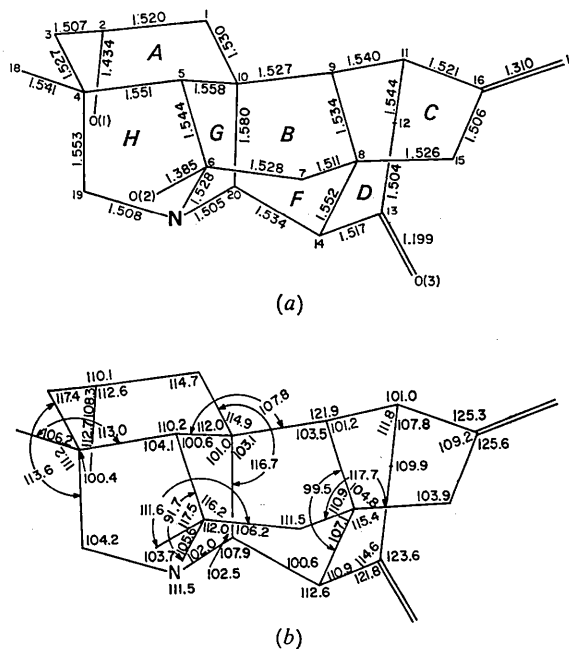

(a)

(b)

Fig. 2. (a) Bond lengths. The range of their e.s.d.'s is 0·004–0·005 Å, except for C(16)–C(17) which has an e.s.d. of 0·009 Å. (b) Valency angles. Their e.s.d.'s vary between 0·2 and 0·3°, except for the two angles associated with C(17) which have e.s.d.'s of 0·5°.

Table 6. Torsional angles

For definition see Klyne & Prelog (1960). The e.s.d.'s are 0·3° (Huber, 1961).

Ring A		Ring D		Ring G	
C(10)-C(1)—C(2)—C(3)	$+50·3°$	C(14)-C(8)—C(19)-C(11)	$-77·4°$	C(5)—C(6)—N(1)—C(20)	$+60·5°$
C(1)—C(2)—C(3)—C(4)	$-47·6$	C(8)—C(9)—C(11)-C(12)	$+72·1$	C(6)—N(1)—C(20)-C(10)	$-38·8$
C(2)—C(3)—C(4)—C(5)	$+48·7$	C(9)—C(11)-C(12)-C(13)	$-50·3$	N(1)—C(20)-C(10)-C(5)	$+0·6$
C(3)—C(4)—C(5)—C(10)	$-48·9$	C(11)-C(12)-C(13)-C(14)	$+39·0$	C(20)-C(10)-C(5)—C(6)	$+37·1$
C(4)—C(5)—C(10)-C(1)	$+52·5$	C(12)-C(13)-C(14)-C(8)	$-51·9$	C(10)-C(5)—C(6)—N(1)	$-59·1$
C(5)—C(10)-C(1)—C(2)	$-55·0$	C(13)-C(14)-C(8)—C(9)	$+70·4$		

Ring B		Ring E		Ring H	
C(10)-C(5)—C(6)—C(7)	$+56·5$	C(14)-C(8)—C(9)—C(10)	$+49·6$	C(5)—C(6)—N(1)—C(19)	$-55·4$
C(5)—C(6)—C(7)—C(8)	$-48·9$	C(8)—C(9)—C(10)-C(20)	$-29·8$	C(6)—N(1)—C(19)-C(4)	$+36·8$
C(6)—C(7)—C(8)—C(9)	$+49·8$	C(9)—C(10)-C(20)-C(14)	$-1·1$	N(1)—C(19)-C(4)—C(5)	$-0·9$
C(7)—C(8)—C(9)—C(10)	$-63·0$	C(10)-C(20)-C(14)-C(8)	$+30·9$	C(19)-C(4)—C(5)—C(6)	$-35·0$
C(8)—C(9)—C(10)-C(5)	$+76·4$	C(20)-C(14)-C(8)—C(9)	$-48·9$	C(4)—C(5)—C(6)—N(1)	$+55·1$
C(9)—C(10)-C(5)—C(6)	$-70·6$				

Ring C		Ring F			
C(8)—C(9)—C(11)-C(16)	$-42·4$	N(1)—C(6)—C(7)—C(8)	$+54·6$	C(19)-C(4)—C(5)—C(10)	$+72·2$
C(9)—C(11)-C(16)-C(15)	$+28·6$	C(6)—C(7)—C(8)—C(14)	$-57·8$	C(20)-N(1)—C(19)-C(4)	$-72·1$
C(11)-C(16)-C(15)-C(8)	$-2·9$	C(7)—C(8)—C(14)-C(20)	$+66·5$	C(4)—C(5)—C(10)-C(20)	$-72·4$
C(16)-C(15)-C(8)—C(9)	$-24·5$	C(8)—C(14)-C(20)-N(1)	$-78·4$	C(10)-C(20)-N(1)—C(19)	$+71·3$
C(15)-C(8)—C(9)—C(11)	$+42·2$	C(14)-C(20)-N(1)—C(6)	$+73·1$		
		C(20)-N(1)—C(6)—C(7)	$-58·7$		

There is non-bonded interaction between the axial hydroxyl group in ring A and two axially oriented carbon atoms [C(19) and C(20)]. As a result, the C(2)–O(1) bond is bent away from the other two axial bonds and ring A is flattened. This can be seen from torsional and valency angles [C(2)–C(1)–C(10) and C(2)–C(3)–C(4) are considerably larger than tetrahedral]. Another demonstration of this bending can be obtained by comparing the non-bonded distances C(2)$\cdots$C(4) and C(2)$\cdots$C(10) [2·592 (5) and 2·568 (5) Å] with the considerably longer ones O(1)$\cdots$C(19) and O(1)$\cdots$C(20) [3·035 (5) and 3·033 (4) Å]. The two equatorial bonds C(4)–C(18) and C(6)–O(2) are similarly disposed, the non-bonded distances being 2·441 (4) Å for C(4)$\cdots$C(6) and 2·939 (5) Å for C(18)$\cdots$O(2). The mean planes of rings A and B are not parallel but form an angle of 20·4° with each other, a consequence of the fusion of these two rings with the azabicyclo[2,2,1]heptane system.

Ring D is considerably flattened at C(12). Although this is not indicated by the valency angle, it does result in an abnormally small torsional angle at C(12)–C(13), viz. 39·0° as compared with 51·4° calculated for a cyclohexanone chair (Bucourt & Hainaut, 1967). This flattening may be caused by a repulsion between two methylene groups, C(1) and C(12). The distance between H(12) and H(122) is only 2·06 (5) Å. The distortion at C(12) is the most likely reason for the lack of planarity in the carbonyl group (cf. plane 1).

Neither ring C nor ring E have ideal envelope conformations. In the former C(8) is significantly displaced from plane 2, and in the latter the four atoms C(9), C(10), C(20), C(14) are not coplanar ($\chi^2 = 17 \cdot 1$). The displacements of the atoms C(9) (0·729 Å from plane 2) and C(8) (0·763 Å from plane 3) are somewhat larger than the calculated values 0·61 and 0·70 Å (Sim, 1965) based on the mean angles of the two rings (104·0° in ring C and 102·6° in ring E).

There is a very small *contra* twist (Altona & Sundaralingam, 1970) of the azabicyclo[2,2,1]heptane system. The dihedral angle of 114·7° between planes 4 and 5 agrees with the value 113·2° found in 5-norbornene-2,3-*endo*-dicarboxylic anhydride (Destro, Filippini, Gramaccioli & Simonetta, 1969). Since C(6) is also part of rings B and F it is displaced further away from plane 4 than from plane 5. This can also be seen from the larger absolute values of the torsional angles in ring G than in ring H. (See Altona & Sundaralingam, 1970, for a discussion of torsional angles in bicyclo[2,2,1]-heptane systems.) The valency angles in this part of the molecule are fairly close to the ones in other bicyclo-[2,2,1]heptane systems (Sim, 1965) and to calculated values (Gleicher & Schleyer, 1967; Allinger, Hirsch, Miller, Tyminski & Van-Catledge, 1968).

The agreement between the valency angles which are common in delnudine and hetisine is very good. More than half of them differ by 1° or less (the e.s.d. in the angles of hetisine hydrobromide was given as close to 1°). Only two angles may differ significantly, viz. C(4)–C(5)–C(10) and C(1)–C(10)–C(5).

Bond lengths

Most bond lengths agree with usually observed values. However, several of them require some comments. The single bonds C(5)–C(10) (1·558 Å) and C(10)–C(20) (1·580 Å) are significantly longer than $1 \cdot 537 \pm 0 \cdot 005$ Å

Table 7. *Least-squares planes*

Plane 1 (Carbonyl group)		Plane 2 (ring C)		Plane 3 (ring E)		Plane 4 (ring G)		Plane 5 (ring H)	
	$\varDelta$		$\varDelta$		$\varDelta$		$\varDelta$		$\varDelta$
C(12)	0·004 Å	C(11)	−0·001 Å	C(9)	−0·005 Å	C(5)	0·003 Å	C(4)	0·006 Å
C(13)	−0·011	C(15)	−0·001	C(10)	0·007	C(10)	−0·004	C(5)	−0·004
C(14)	0·003	C(16)	0·003	C(20)	−0·007	C(20)	0·003	N(1)	0·002
O(3)	0·004	C(17)	−0·004	C(14)	0·005	N(1)	−0·002	C(19)	−0·006
		C(8)*	−0·081	C(8)*	−0·763	C(6)*	0·927	C(6)*	−0·875
		C(9)*	−0·729						
		H(171)*	0·15						
		H(172)*	0·31						
R.m.s.d. = 0·006		r.m.s.d. = 0·002		r.m.s.d. = 0·006		r.m.s.d. = 0·003		r.m.s.d. = 0·005	
$\chi^2 = 15 \cdot 8$		$\chi^2 = 0 \cdot 7$		$\chi^2 = 17 \cdot 1$		$\chi^2 = 4 \cdot 8$		$\chi^2 = 8 \cdot 5$	

* Atoms excluded from the calculation of the plane.

Equations of the planes are of the type $hx + ky + lz = m$ where x, y, z are fractional monoclinic coordinates.
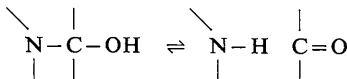
	h(Å)	k(Å)	l(Å)	m(Å)
1:	−3·316	6·075	−5·540	−3·202
2:	6·117	−6·871	−5·382	0·596
3:	−2·525	−9·826	−0·634	−3·973
4:	−0·962	5·817	−7·087	−3·249
5:	−2·582	−9·877	−0·210	−5·174

Some dihedral angles between planes

$\angle 2,3$	$\angle 3,4$	$\angle 4,5$
58·5°	112·3°	114·7°

(Sutton, 1965). (Note that the uncertainty indicated by Sutton is 2σ.) However, both bonds are between two highly substituted carbon atoms and such long bonds have been observed in the past (Birnbaum & Ferguson, 1969). The two parallel bonds C(2)–C(3) (1·507 Å) and C(7)–C(8) (1·511 Å) are shorter than usual. There is no obvious explanation for this deviation.

Two of the N^+–C bonds, N(1)–C(19) (1·508 Å) and N(1)–C(20) (1·505 Å) are equal while the third, N(1)–C(6) (1·528 Å) is 4·4σ longer than the mean of the other two. It is also significantly longer than the weighted mean value 1·499 (2) Å calculated from 30 bonds in various structures where the nitrogen atom is trisubstituted (Birnbaum, 1967), while only 2·8σ longer than the average of three such bonds (1·511 (5) Å) in lappaconine hydrobromide (Birnbaum, 1970). (Note that in calculating the average value of 1·479 Å for the N^+–C bond Sutton (1965) ignored the substitution of the nitrogen atom.) The increased length of the N(1)–C(6) bond, together with the observation that the C(6)–O(2) bond (1·385 Å) is 0·041 Å (8·9σ) shorter than the value (1·426 ± 0·005 Å) given by Sutton (1965) can be explained by the fact that the free delnudine base is a carbinolamine. As is frequently the case with such substances, there is an equilibrium between the two tautomeric forms of delnudine (Götz & Wiesner, 1969):

$$\overset{\displaystyle |}{\underset{\displaystyle |}{N}}-\overset{\displaystyle |}{\underset{\displaystyle |}{C}}-OH \rightleftharpoons \overset{\displaystyle |}{N}-H \quad \overset{\displaystyle |}{C}=O$$

A small amount of the salt of the keto tautomer in the crystal could explain the abnormal bond lengths. In this case a hydrogen atom would have to be inserted between N(1) and C(6). This could only be accomplished by pushing these atoms away from each other. Their thermal parameters indicate that this does not occur. A better explanation of the observed bond lengths may be offered by postulating that only the hydroxy tautomer is present in the crystal and that it exists as a resonance hybrid with a small contribution from the canonical form (b) to the structure.

$$H—\overset{\displaystyle |}{\underset{\displaystyle |}{\overset{\oplus}{N}}}—\overset{\displaystyle |}{\underset{\displaystyle .O—H}{C}}— \quad \leftrightarrow \quad H—\overset{\displaystyle |}{\underset{\displaystyle |}{N}}: \quad \overset{\displaystyle |}{\underset{\displaystyle \overset{\oplus}{O}—H}{C}}—$$

$$(a) \qquad\qquad (b)$$
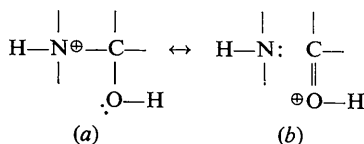
Hydrogen bonding and packing

The relevant bond lengths and angles are shown in Fig. 3. All hydrogen atoms which are capable of forming hydrogen bonds [H(O11), H(O21) and H(N11)] are donated to the chloride ion. The three $H\cdots Cl^-$ distances are about 0·6 Å shorter than the sum of the van der Waals radii (H 1·2, Cl^- 1·8 Å). In general hydrogen bonds are not linear and the bend may be as high as 30° (Donohue, 1968). The $H–O\cdots Cl^-$ and $H–N\cdots Cl^-$ angles lie within this range. The three $Cl^-\cdots H$ bonds are approximately perpendicular to each other and the symmetry around the Cl^- ion is thus close to C_{3v}.

The hydrogen bonds can also be seen in Fig. 4, which shows the molecular arrangement parallel to (010). O(1) and O(2) are not hydrogen bonded to the same Cl^- ion, as it may appear from the Figure, but to two different ones separated by b. The molecules are thus linked together via hydrogen bonds to the Cl^- ion and form spirals around one of the twofold screw axes (at $x=0$, $z=\frac{1}{2}$) in each unit cell. This packing arrangement explains the fact that delnudine hydrochloride crystallizes in the form of needles elongated in the [010] direction.

In addition to hydrogen bonds there are three intermolecular contacts shorter than 3·5 Å, viz. $O(2)\cdots O(3)$ 3·281 (4) Å, $C(20)\cdots O(2)$ 3·251 (4) Å and $C(13)\cdots O(2)$ 3·456 (4) Å.

Absolute configuration

The refinement had converged to $R=3·85\%$ and $R'=4·57\%$. When the $\Delta f''$ correction (0·19 e) to the Cl^- scattering curve was introduced into the least-squares refinement the value of R decreased to 3·76% and that of R' to 4·40%. During this refinement the y coordinates of all the non-heavy atoms moved in one direction by an average distance of 0·005 Å. After final convergence the R and R' for the opposite absolute configuration were calculated and their values were found to be 3·93% and 4·59%. The ratio of the weighted R factors for the two absolute configurations is 1·043 and by interpolation of the table for testing this ratio at 0·5% significance level (Hamilton, 1965) it was found that $\mathscr{R}_{1,1834,0.005}=1·002$. This shows clearly that the absolute configuration of delnudine chosen originally is the correct one. It is the same as that of related alkaloids, e.g. hetisine (Przybylska, 1963), *Garrya* and *Aconitum* alkaloids (ApSimon & Edwards, 1962; Vorbrueggen & Djerassi, 1962). All drawings and diagrams are presented with respect to a right-handed
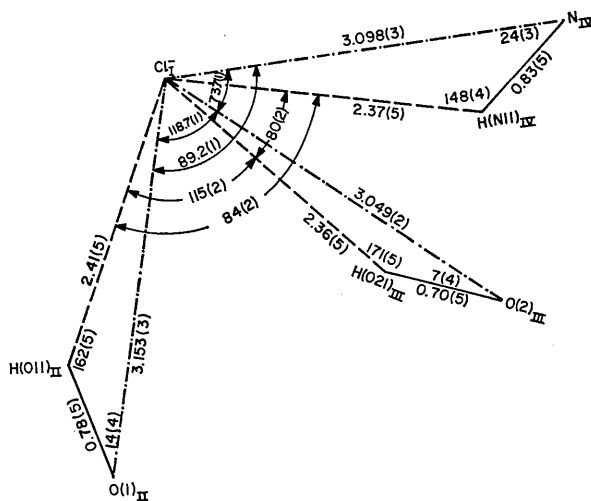

Fig. 3. Bond lengths and angles for the hydrogen bonds. The Roman numbers refer to the following positions: I (x,y,z); II $(1-x, y-\frac{1}{2}, 1-z)$; III $(1-x, y+\frac{1}{2}, 1-z)$; IV $(1+x, y, z)$.
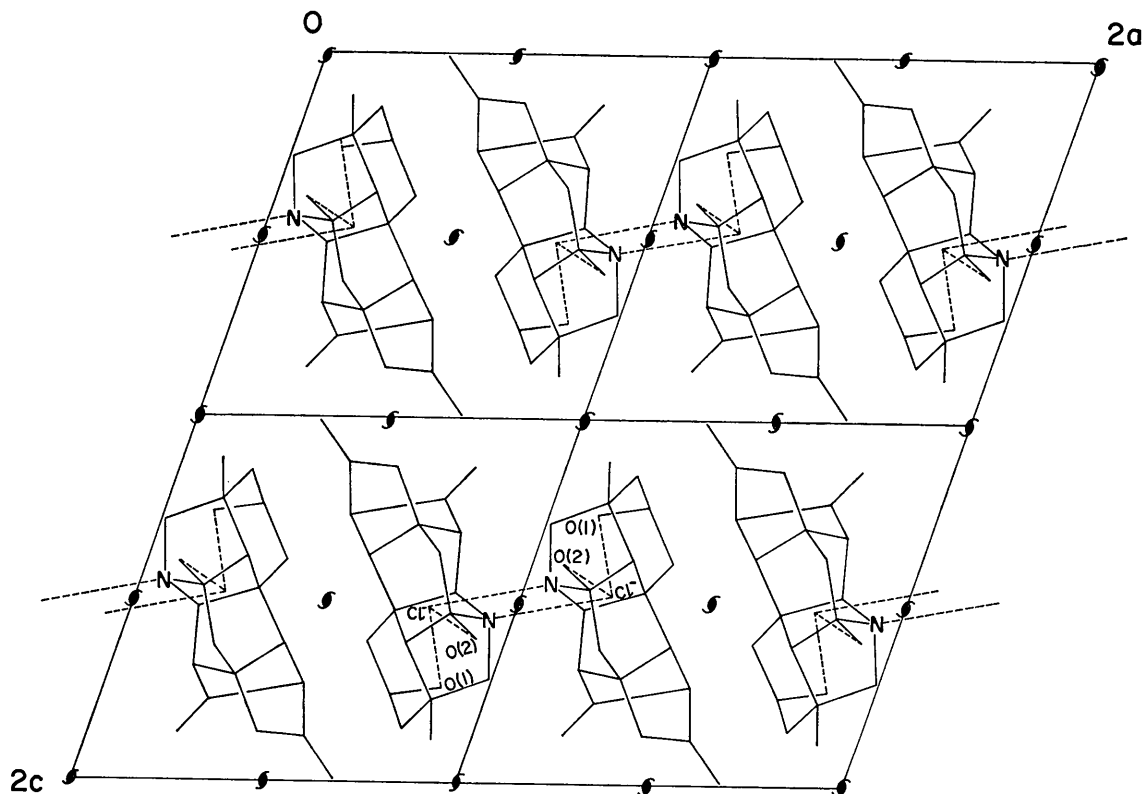
Fig. 4. Packing diagram viewed along the *b* axis. Hydrogen bonds are marked with broken lines.

set of crystal axes and show the correct absolute configuration.

I wish to thank Professor K. Wiesner for suggesting this problem and for supplying me with delnudine. I am also indebted to Dr Maria Przybylska for her constant encouragement and interest during this work. The use of the computer programs of Ahmed, Hall, Pippy & Huber (1966) and of *ORTEP* (Johnson, 1965) is gratefully acknowledged.

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