GOULD–JACOBS REACTION OF 5- AND 6-AMINO-2-SUBSTITUTED BENZOXAZOLES. III. REACTION WITH 3-ETHOXY-2-CYANOPROPENONITRILE AND ETHYL 3-ETHOXY-2-CYANOPROPENOATE

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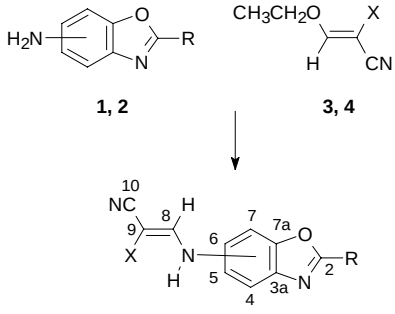
Nucleophilic reaction of 5-amino- (**1**) and 6-amino-2-substituted benzoxazoles (**2**) with 3-ethoxy-2-cyanopropenonitrile (**3**) afforded the respective benzoxazolylaminomethylenemalononitriles **5** and **6**. The amino derivatives **1** and **2** reacted with ethyl 3-ethoxy-2-cyanopropenoate (**4**) to give the corresponding esters of benzoxazolylamino-2-cyanopropenoic acid **7** and **8**, respectively. The products **7** on thermal cyclization at 250–260 °C in a mixture of diphenyl ether and biphenyl afforded a mixture of angularly and linearly annelated 5-nitrile-4-oxooxazolo[4,5-*f*]quinoline **9** and 7-nitrile-8-oxooxazolo[5,4-*g*]quinoline **10**; under the same conditions compounds **8** were converted into 8-nitrile-9-oxooxazolo[5,4-*f*]quinolines **11** and 6-nitrile-5-oxooxazolo[4,5-*g*]quinolines **12**.
Key words: Gould–Jacobs reaction; Oxazoloquinolones.

In our previous communications we studied reactions of 5- and 6-amino-2-substituted benzoxazoles with diethyl ethoxymethylenepropanedioate[1], 3-ethoxymethylene-2,4-pentanedione and ethyl 2-ethoxymethylene-3-oxobutanoate[2], and the subsequent thermal cyclization of the obtained substitution products, containing an ethyl ester group in the molecule. Similar transformation has been mentioned in a Japanese patent[3], describing reaction of 6-amino-2-methylbenzoxazole with ethyl ethoxymethylenepropanedioate (EMME) and cyclization of the arising substitution product under exclusive formation of the angularly annelated quinolone derivative.

In the present study we describe the nucleophilic substitution reaction of 5-amino- (**1**) and 6-amino- (**2**) 2-substituted benzoxazoles with 3-ethoxy-2-cyanopropenonitrile (**3**) leading to substitution products **5** and **6** (Scheme 1). The reaction of amino derivatives **1** and **2** with ethyl 3-ethoxy-2-cyanopropenoate (**4**), leading to the corresponding derivatives **7** and **8**, required an about 15 min reflux in ethanol. We did not observe any difference in reactivity of the 5-amino (**1**) and 6-amino (**2**) derivatives. The physicochemical properties of the obtained derivatives **5–8** are summarized in Table I, their IR and UV spectra in Table II.

^{1}H NMR spectra of substitution products **5–8** exhibited doublets of protons H-4 and H-7 and doublets of doublets due to protons H-5 or H-6. In the spectra of derivatives **5** and **6** the H-8 signals appeared as singlets[4], in the spectra of **7** and **8** as doublets.

Whereas the antiperiplanar arrangement (*Z*) of the H-8 protons (^3J = 12.0–14.0 Hz) can be stabilized by intramolecular hydrogen bond between the NH proton and the ester carbonyl, the (*E*)-arrangement is preferred sterically. The observed ratio of geometric isomers was about 2 : 1 but we could not decide whether in favour of the (*Z*)- or (*E*)-isomer because of insolubility of the compounds which had to be measured in DMSO at 80 °C. In the spectra of compounds **5** and **6** the signals of protons NH and H-8 were shifted downfield relative to those of derivatives **7** and **8**. This difference is due to a strong bond polarization resulting from the presence of two proton accepting nitrile groups. The ^{1}H NMR spectra of compounds **5–8** are given in Table III.



In formulae : **1, 5, 7** : 5-substitution
2, 6, 8 : 6-substitution
3, 5, 6 : X = CN
4, 7, 8 : X = $COOC_2H_5$

5-8: **a**, R = H
b, R = CH_3
c, R = C_6H_5

SCHEME 1

In the ^{13}C NMR spectra of derivatives **5** and **6** (Table IV) the nitrile signals appear in the region 113.0–116.0 ppm. The strong acceptor influence appeared mainly as a shift of the C-9 signal (51.0–52.0 ppm). The spectra of compounds **7** and **8** (Table IV) exhibited the C-9 signal at about 73.0–74.0 ppm. The acceptor groups had a much weaker influence on the C-8 signal[5] (differences of only 2.0–3.0 ppm).

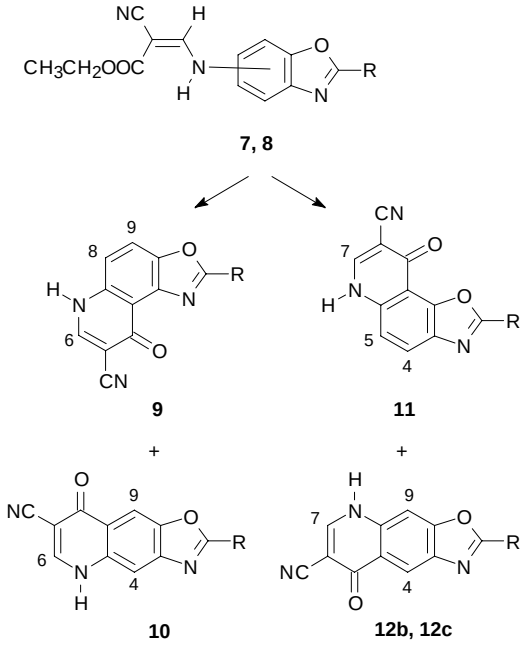
Mass spectra of the products **5–8** are given in Table V. In all cases the spectra contain the molecular ion. Derivatives **5a**, **5b** and **6a**, **6b** lose fragment *m/z* 65, derivatives **5c** and **6c** fragment *m/z* 76, to give 5-amino and 6-amino-2-phenylbenzoxazole. The different fragmentation paths are probably influenced by the substituent bonded to the oxazole nucleus and its ability to stabilize the arising fragment. Characteristic fragmentation of derivatives **7** and **8** consists in the preferential loss of ethanol (*m/z* 46) followed by loss of the nitrile group together with the substituent at the position 2 of the oxazole nucleus.

The thermal cyclization of the products **7** and **8** took place in a mixture of diphenyl ether and biphenyl at 250–260 °C (Scheme 2). The best results were achieved with

TABLE I
Melting points, yields and analyses of compounds **5–8**

Compound	M.p., °C Yield, %	Formula M.w.	Calculated/Found		
			% C	% H	% N
5a	238–240	$C_{11}H_6N_4O$	62.86	2.88	26.65
	63	210.2	62.79	2.82	26.60
5b	229–231	$C_{12}H_8N_4O$	64.28	3.60	24.99
	98	224.2	64.23	3.57	24.91
5c	265–266	$C_{17}H_{10}N_4O$	71.32	3.52	19.57
	94	286.3	71.24	3.46	19.49
6a	130–135	$C_{11}H_6N_4O$	62.86	2.88	26.65
	68	210.2	62.80	2.82	26.70
6b	260–263	$C_{12}H_8N_4O$	64.28	3.60	24.99
	64	224.2	64.23	3.54	25.03
6c	226–230	$C_{17}H_{10}N_4O$	71.32	3.52	19.57
	92	286.3	71.28	3.46	19.50
7a	209–210	$C_{13}H_{11}N_3O_3$	60.70	4.31	16.33
	52	257.2	60.61	4.27	16.28
7b	150–151	$C_{14}H_{13}N_3O_3$	61.99	4.83	15.49
	65	271.3	61.87	4.78	15.41
7c	178	$C_{19}H_{15}N_3O_3$	68.46	4.54	12.61
	71	333.3	68.17	4.48	12.55
8a	168–169	$C_{13}H_{11}N_3O_3$	60.70	4.31	16.33
	64	257.2	60.61	4.25	16.38
8b	164–165	$C_{14}H_{13}N_3O_3$	61.99	4.83	15.49
	65	271.3	61.91	4.78	15.52
8c	190–191	$C_{19}H_{15}N_3O_3$	68.46	4.54	12.61
	87	333.3	68.39	4.47	12.73

concentrations 1 g of the compound in 80–150 g of the solvent. About a half of the solvent was distilled from the reaction mixture during the reaction and the mixture was then allowed to stand overnight in the cold. The cyclic products were obtained as very fine brown powders. Because the reaction could not be followed analytically, the given reaction times do not reflect the time actually needed for its completion. Due to the high dilution employed, the cyclization products remained dissolved. Their precipitation with nonpolar solvents such as hexane was not advantageous because the solvent precipitated also the unreacted material.



In formulae **7-12** : **a**, R = H
b, R = CH_3
c, R = C_6H_5

SCHEME 2

In most cases, the reaction gave mixtures of the angularly and linearly annelated oxazoloquinolones. The only exception was the cyclization of derivative **8a** leading solely to the angularly annelated product **11a**. The low yields of the cyclizations (10–20%) are presumably due to decomposition of the oxazole nucleus during the cyclization, incomplete reaction, and also a still dissolved part of the product. The cyclic products melted invaribly above 310 °C and were very insoluble which made separation by crystallization or chromatography impossible. The physicochemical constants and IR spectra of compounds **9–12** are summarized in Table VI, the ^{1}H NMR spectra of the

cyclocondensation products are given in Table VII. In the mixtures of compounds **9** and **10**, the angularly annelated derivatives are characterized by doublets of the H-8 and H-9 protons whereas for linearly annelated products the singlets of protons H-4 and H-9 are typical. We observed a doubling of signals of the proton and the methyl group in position 2 of the oxazole nucleus. The derivative **11a** was isolated in the pure state; its spectrum exhibited two doublets of H-4 and H-5 (^3J = 9.8 Hz). The H-2 signal was observed as singlet at 8.69 ppm. Spectra of mixtures **11b** + **12b**, and **11c** + **12c** exhibited H-4, H-5 doublets, and also singlets due to H-4 and H-9 protons. We also observed a doubling of the signal of methyl bonded to the oxazole nucleus, as well as signals of protons H-7. From the proton spectra it was not possible to determine correctly whether the cyclic products exist in the oxo or enol form. In both cases the system is conjugated. In the angularly annelated derivatives the enol form may be additionally stabi-

TABLE II
Infrared and UV spectra of substitution products **5–8** (i = inflex)

Compound or mixture	IR spectra					UV spectra		
	ν(NH)	ν(CO)	ν(CN)	other				
5a	3 240	1 670	2 300	1 640	1 618	209 (3.23)	235 (3.05) 297i (3.28)	312 (3.39)
5b	3 240	1 670	2 300	1 640	1 615	208 (3.41)	235 (3.13) 295i (3.30)	314 (3.39)
5c	3 220	1 655	2 300	1 640	1 620	–	–	–
6a	3 220	1 685	2 220	1 650	1 620	–	248i (2.82)	335 (3.16)
6b	3 440	1 670	2 230	1 645	1 615	–	238i (2.80)	322 (3.34)
6c	3 220	1 660	2 220	1 630	1 560	–	–	–
7a	–	1 675	2 220	1 670	1 650	208 (3.31)	234 (3.11) 295i (3.33)	318 (3.38)
7b	–	1 675	2 220	1 670	1 650	208 (3.41)	235 (3.13) 295i (3.30)	319 (3.47)
7c	–	1 675	2 220	1 655	1 620	–	274 (3.35)	327 (3.58)
8a	–	1 680	2 220	1 620	1 580	–	244 (3.26) 290i (3.03)	325 (3.54)
8b	–	1 680	2 215	1 620	1 560	–	242 (2.87) 292 (3.03)	326 (3.47)
8c	–	1 670	2 210	1 620	1 565	–	222 (3.26) 246 (3.06)	342 (3.67)

lized by hydrogen bonding between the hydroxy group and the nitrogen or oxygen atom of the oxazole nucleus. However, the situation is complicated by the fact that in the studied isomer mixtures different oxo–enol population may exist for each isomer[6]. The angular-to-linear ratio of isomers was invariably 2 : 1 as determined from comparison of integrated intensities of doublets of protons H-4, H-5 or H-8, H-9 with those of the H-4 and H-9 singlets, and also from comparison of H-2 integrals with those of the methyl signals.

Mass spectra of all cyclocondensation products exhibit molecular ion peaks and almost in all cases also an ion *m/z* 170, which indicates that the oxazole nucleus is split first and only then gradual fragmentation takes place. The mass spectra are given in Table VIII.

TABLE III
^{1}H NMR spectral data for substitution products **5–8**

Compound[a]	H-4	H-5	H-6	H-7	NH	H-8	CH_2	CH_3	R
5a	7.71	–	6.60	7.51	10.57	8.54		–	7.89
5b	7.72	–	7.39	7.61	10.95	8.42	–	–	2.60
5c	7.86	–	7.48	7.77	11.14	8.50	–	–	8.20–8.17 7.63–7.61
6a	8.13	7.75	–	7.91	11.18	8.56	–	–	8.68
6b	7.74	7.40	–	7.63	11.14	8.61	–	–	2.59
6c	7.90	7.50	–	7.78	11.29	8.57	–	–	8.18–8.15 7.62–7.60
7a	7.84	–	7.53	7.80	10.55	8.40	4.23	1.30	8.51
7b	7.52	–	7.32	7.69	10.79 10.90	8.25 8.36	4.17 4.21	1.24 1.29	2.59
7c	7.94	–	7.45	7.80	10.83 11.00	8.34 8.50	4.21 4.26	1.26 1.31	8.19–8.16 7.62–7.59
8a	8.05	7.71	–	7.86	11.09	8.31	4.25	1.35	8.52
8b	7.81	7.39	–	7.63	10.79 10.97	8.22 8.37	4.17 4.23	1.23 1.26	2.59
8c	7.97	7.47	–	7.75	10.83 11.02	8.36 8.48	4.18 4.24	1.22 1.19	8.16–8.13 7.61–7.59

[a] **5a–5c**: ^4J(4,6) = 2.0, ^3J((6,7) = 9.0; **6a–6c**: ^4J(5,7) = 2.0, ^3J(4,5) = 9.0; **7a–7c:** ^4J(4,6) = 2.0, ^3J(6,7), ^3J(H8-NH) = 12.0; **8a–8c**: ^4J(5,7) = 3.0, ^3J(4,5) = 9.0, ^3J(H8-NH) = 14.0.

TABLE IV
^{13}C NMR spectral data for substitution products **5–8**

Compound	C-2	C-3a	C-4	C-5	C-6	C-7	C-7a	C-8	C-9	C-10	C-11	C-12	C-13	R[a]
5a	156.25	142.85	111.38	138.32	114.22	109.52	146.82	155.25	51.73	113.96	116.56	–	–	–
5b	164.95	141.68	113.65	136.51	115.92	110.28	148.12	155.88	51.47	115.21	116.28	–	–	13.66
5c	163.59	142.22	111.30	136.38	116.69	109.13	147.84	156.68	51.85	114.06	116.33	–	–	–
6a	159.82	144.33	108.32	120.32	136.39	104.92	147.42	155.97	51.02	114.09	116.37	–	–	–
6b	165.43	147.94	110.77	115.66	136.19	108.57	141.88	156.32	51.45	113.87	116.53	–	–	14.15
6c	162.65	150.65	116.01	120.11	137.24	100.63	138.77	155.90	52.35	114.07	116.33	–	–	–
7a	162.16	137.66	106.20	126.20	114.44	103.99	141.91	152.14	73.22	113.96	163.50	60.05	13.36	–
7b	163.23	141.81	108.60	135.99	111.32	102.58	147.62	153.11	73.99	115.26	165.50	60.03	13.86	23.18
7c	163.51	142.22	111.32	136.96	116.63	109.02	147.76	153.53	74.18	115.74	166.26	60.04	13.81	–
8a	163.52	139.21	109.18	116.41	135.81	103.41	147.49	152.99	73.24	115.63	164.39	60.15	13.98	–
8b	164.41	141.94	110.75	115.60	136.12	108.45	147.91	153.72	73.75	115.91	166.33	60.32	14.25	24.11
8c	163.30	138.49	111.33	115.80	136.63	100.29	147.60	153.28	74.68	114.21	166.95	60.33	13.85	–

[a] **5c**: 1* = 126.09, (2*,6*) = 127.30, (3*,5*) = 129.25, 4* = 132.01; **6c**: 1* = 126.18, (2*,6*) = 127.24, (3*,5*) = 129.30, 4* = 131.94; **7c**: 1* = 126.18, (2*,6*) = 127.27, (3*,5*) = 129.21, 4* = 132.01; **8c**: 1* = 128.85, (2*,6*) = 129.03, (3*,5*) = 129.92, 4* = 131.61.

EXPERIMENTAL

^{1}H and ^{13}C NMR spectra (δ, ppm; *J*, Hz) were measured on a Varian VXR-300 (300 MHz for ^{1}H and 75 MHz for ^{13}C) spectrometer, IR spectra (ν, cm^{-1}) were taken on an M-80 (Zeiss, Jena) spectrometer using the KBr technique (1 mg in 300 mg of KBr). UV spectra were obtained with a UV-VIS M-40 (Zeiss, Jena) spectrophotometer in methanol, concentration 10^{-4} mol dm^{-3}. Mass spectra were taken on an MS 902S (AEI Kratos) instrument. The starting aminooxazoles **1** and **2** were prepared according to ref.[2].

5- and 6-(2-Substituted Benzoxazolyl)aminomethylenemalononitriles **5** and **6** and Ethyl 3-[5- and 6-(2-Substituted Benzoxazolyl)amino]-2-cyanopropenoates **7** and **8**

To an alcoholic solution of aminobenzoxazole **1** or **2** (0.01 mol) was added 3-ethoxy-2-cyanopropenonitrile (EMMN) or ethyl 3-ethoxy-2-cyanopropenoate (EMCA) (0.01 mol). The mixture was allowed to stand at ambient temperature (compounds **5** and **6**) or was heated at reflux for 15 min

TABLE V
Mass spectral data for substitution products **5–8**

Compound	*m/z* (relative intensity, %)
5a	($M^{+\bullet}$ 210, 100), 182 (42), 155 (10), 145 (51), 128 (12), 118 (20), 101 (10), 77 (10), 63 (20), 52 (16), 51 (16), 28 (10)
5b	($M^{+\bullet}$ 224, 100), 195 (17), 183 (5), 159 (17), 155 (17), 128 (19), 109 (5), 77 (10), 63 (19), 51 (19), 28 (10)
5c	($M^{+\bullet}$ 286, 100), 258 (18), 238 (56), 210 (54), 155 (27), 128 (18), 104 (20), 91 (18), 79 (51), 63 (10), 52 (29), 51 (29), 28 (14)
6a	($M^{+\bullet}$ 210, 100), 182 (42), 155 (10), 145 (51), 128 (12), 118 (20), 101 (10), 77 (10), 63 (18), 52 (16), 51 (16), 28 (10)
6b	($M^{+\bullet}$ 224, 100), 195 (18), 159 (17), 155 (24), 148 (46), 128 (17), 91 (10), 79 (58), 63 (24), 52 (41), 41 (17), 28 (17)
6c	($M^{+\bullet}$ 286, 65), 238 (65), 223 (100), 210 (45), 77 (20), 63 (16), 43 (18), 28 (10)
7a	($M^{+\bullet}$ 257, 84), 212 (25), 211 (100), 184 (40), 157 (10), 145 (45), 129 (11), 91 (7), 63 (10), 52 (18), 29 (20)
7b	($M^{+\bullet}$ 271, 85), 225 (100), 198 (30), 184 (12), 159 (30), 148 (25), 91 (12), 79 (20), 63 (12), 52 (22), 29 (22)
7c	($M^{+\bullet}$ 333, 94), 287 (100), 260 (32), 259 (32), 221 (11), 184 (17), 157 (14), 156 (17), 91 (17), 77 (20), 63 (11), 52 (17), 28 (38)
8a	($M^{+\bullet}$ 257, 60), 212 (22), 211 (100), 184 (22), 183 (15), 157 (10), 145 (40), 90 (20), 68 (10), 63 (17), 52 (17), 51 (15), 29 (33)
8b	($M^{+\bullet}$ 271, 79), 225 (100), 198 (29), 184 (13), 159 (29), 129 (13), 91 (10), 63 (10), 52 (15), 29 (13)
8c	($M^{+\bullet}$ 333, 78), 287 (100), 260 (26), 259 (28), 221 (18), 184 (10), 157 (10), 156 (12), 91 (16), 77 (10), 63 (12), 28 (26)

TABLE VI
Melting points, yields, analyses and IR spectra of compounds **9–12**

Compound	M.p., °C Yield, %	Formula M.w.	Calculated/Found			IR spectra		
			% C	% H	% N	ν(CN)	ν(C=O)	ν(C=C)
9a, **10a**	–	$C_{11}H_5N_3O_2$	62.56	2.39	19.91	2 210	1 631	1 549
	10	211.2	62.50	2.35	20.00			1 504
9b, **10b**	–	$C_{12}H_7N_3O_2$	64.00	3.13	18.66	2 212	1 631	1 577
	12	225.2	63.74	3.15	18.60			1 541
9c, **10c**	–	$C_{17}H_9N_3O_2$	71.08	3.16	14.63	2 216	1 633	1 610
	20	287.3	71.13	3.20	14.69			1 506
11a	>300	$C_{11}H_5N_3O_2$	62.56	2.39	19.91	2 222	1 622	1 543
	15	211.2	62.50	2.42	19.81			1 514
11b, **12b**	–	$C_{12}H_7N_3O_2$	64.00	3.13	18.66	2 206	1 633	1 608
	10	225.2	63.59	3.01	19.06			1 539
11c, **12c**	–	$C_{17}H_9N_3O_2$	71.08	3.16	14.63	2 222	1 630	1 583
	15	287.3	71.00	3.05	14.57			1 545

TABLE VII
^{1}H NMR spectral data for cyclocondensation products **9–12**

Compound[a]	H-4	H-5	H-6	H-7	H-8	H-9	NH	R
9a	–	–	8.48 s	–	7.47 d	7.85 d	10.83 s	8.71 s
10a	7.95 s		8.53 s		–	7.95 s	10.78 s	8.71 s
9b	–	–	8.62 s	–	7.58 d	8.05 d	8.61 s	2.68 s
10b	7.83 s		8.62 s		–	8.25 s	8.67 s	2.65 s
9c	–	–	8.60 s	–	[b]	8.24 d	8.85 s	8.27–8.25
10c	7.99 s		8.60 s		–	8.38 s	8.85 s	7.66–7.61
11a	8.15 d	9.38 d	–	8.70 s	–	–	8.85 s	8.69 s
11b	7.93 d	7.61 d	–	8.69 s	–	–	8.88 s	2.63 s
12b	8.12 s			8.69 s		8.46 s	8.88 s	2.66 s
11c	8.17 d	[b]	–	8.72 s	–	–	9.12 s	8.24–8.22
12c	8.41 s			8.72 s		7.92 s	9.12 s	7.66–7.64

[a] **9a**: $^3J(8,9)$ = 9.0; **9b**: $^3J(8.9)$ = 9.3; **9c**: $^3J(8,9)$ = 9.2; **11a**: $^3J(4,5)$ = 9.8; **11b**: $^3J(4,5)$ = 9.1. [b] Signals in a multiplet.

(compounds **7** and **8**). The deposited product was collected by filtration and in the case of compounds **7** and **8** purified by crystallization (chloroform, hexane). Physicochemical constants of the products are given in Table I, IR and UV spectra in Table II, ^{1}H NMR spectra in Table III, ^{13}C NMR spectra in Table IV and mass spectra in Table V.

Preparation of Cyclocondensation Products **9–12**

A mixture of the substitution product **7a–7c**, **8a–8c** and Dowtherm (80–150 g) was heated to the boil (250–260 °C) while about half of the solvent was distilled off. After cooling, the crude product was collected, washed with ether to remove the remaining solvent, and dried in vacuo at 100 °C. Melting points, yields and analyses are summarized in Table VI, ^{1}H NMR spectral data in Table VII and mass spectral data in Table VIII.

TABLE VIII
Mass spectra of cyclocondensation products **9–12**

Compound	*m/z* (relative intensity, %)
9a, **10a**	($M^{+\bullet}$ 211, 100), 184 (43), 170 (12), 154 (14), 145 (50), 129 (24), 118 (12), 51 (24), 29 (26), 27 (20)
9b, **10b**	($M^{+\bullet}$ 225, 26), 170 (97), 154 (100), 142 (29), 141 (36), 105 (14), 77 (45), 63 (12), 51 (40), 44 (25), 39 (12)
9c, **10c**	($M^{+\bullet}$ 287, 100), 259 (27), 221 (10), 170 (12), 77 (33), 57 (16), 51 (14), 41 (16), 28 (70)
11a	($M^{+\bullet}$ 211, 100), 183 (24), 170 (71), 154 (74), 145 (29), 134 (50), 128 (24), 77 (50), 63 (18), 51 (42), 44 (18), 39 (26), 28 (42)
11b, **12b**	($M^{+\bullet}$ 225, 20), 170 (100), 154 (30), 142 (38), 141 (45), 115 (25), 77 (30), 51 (28), 39 (915), 28 (23)
11c, **12c**	($M^{+\bullet}$ 287, 100), 260 (10), 259 (12), 221 (13), 166 (12), 77 (18), 63 (20), 52 (12), 51 (10), 29 (916), 28 (10), 27 (12)

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