Glaucium ALKALOIDS

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Plants of the genus *Glaucium* belong to the family Papaveraceae. The very name of the genus *Glaucium* (from the Greek word glaukos – bluish gray) was acquired because typical species of this genus have an unbroken bluish gray bloom [1]. At the present time, 13 species of *Glaucium* have been studied chemically.

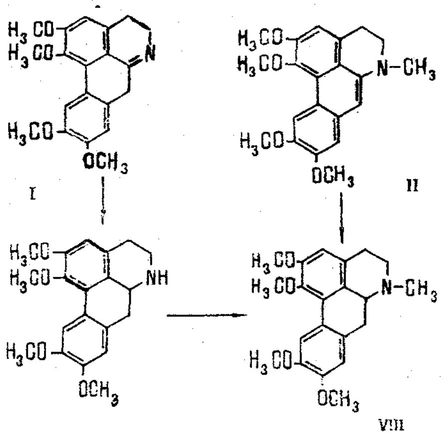
The alkaloid composition of the plants of this genus have been investigated in the Soviet Union, Czechoslovakia, Bulgaria, and Spain, and recently the results of work carried out in Iran have appeared in the literature [2-4]. The beginning of the chemical study of these plants was made in 1901 by Fischer who isolated glaucine from *G. luteum* [5]; the structure of this alkaloid was then established by Gadamer [6].

In our country, the alkaloid composition of the genus *Glaucium* was first studied by A. P. Orekhov, R. A. Konovalova, and S. Yu. Yunusov. From *G. fimbrilligerum* they isolated five alkaloids: sanguinarine, chelerythrine, corydine, protopine, and allocryptopine [7]. At the present time, from various species of *Glaucium* a number of isoquinoline alkaloids have been isolated among which there are representatives of the aporphine, benzisoquinoline, benzophenanthridine, protoberberine, and morphinan groups.

The greatest number of the alkaloids isolated belongs to the aporphine group. For example, while a total of 29 identified alkaloids have been isolated from *G. flavum*, 18 of them are aporphine compounds (Table 1). In addition, all the new alkaloids isolated recently from *Glaucium* also belong to this group.

Corydine and isocorydine are the most characteristic alkaloids of plants of this genus and are found in almost all species of *Glaucium*. However, they have not been detected in three species – *G. luteum* [5, 7], *G. leiocarpum* [8], *and G. grandiflorum* [9] – which is possibly due to an inadequate study of the alkaloid composition of these plants.

Another alkaloid frequently encountered is glaucine [3, 5-8, 10-18], the amount of which in some species reaches 50% of the combined alkaloids [9]. Glaucine (VIII) possesses an antitussive and sedative action [9] and is used in practical medicine.



Two new aporphine alkaloids have been isolated from *G. flavum*: dehydronorglaucine (I) and dehydroglaucine (II) [15, 19]. Their structures have been shown on the basis of spectral characteristics and conversion into glaucine (VIII).

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TABLE 1

Plant species	Alkaloids				Literature
	aporphine	protoberberine	benzophenanthridine	protopine	
1. *G. contortuplicatum***	Dicentrine				46
2. *G. corniculatum*	Corydine, isocorydine	Berberine, coptisine β-canadinemethohydroxide, β-stilopine methohydroxide	Sanguinarine, chelerythrine, chelidonine, dl-chelidonine, chelirubine	α-Allocryptopine, protopine	30, 31, 47, 49, 52
3. *G. elegans*	Corydine, isocorydine, glaucine, isoboldine, O-methylatheroline	Coptisine	Sanguinarine, chelerythrine, chelidonine, chelirubine, dihydrochelerythrine	Allocryptopine, protopine	10, 11, 32
4. *G. fimbrilligerum**	Corydine, isocorydine, corytuberine, N-methyllindcarpine, glaufine, isoboldine, glaufidine	Coptisine, berberine, isocorypalmine	Sanguinarine, chelerythrine, chelidonine, chelirubine	Allocryptopine, protopine	7, 33, 34, 35
5. *G. flavum*†	Corydine, isocorydine, corytuberine, magnoflorine, bulbocapnine, glaucine, 6,6a-dehydronorglaucine, isoboldine, O-methylatheroline, glauvine, thalicmidine, dicentrine	Coptisine, aurotensine	Sanguinarine, chelerythrine, chelidonine, chelirubine, bocconoline	Allocryptopine, protopine	2, 12, 13, 16, 17, 19, 43, 48, 52
5a. *G. flavum var. fulvum*			Norchelidonine		42
5b. *G. flavum var. leocarpum*	Dehydroglaucine, glaucine, isocorydine, O-methylatheroline		Chelidonine		15
5c. *G. flavum var. vestitum*	Dehydroglaucine, thalicmidine, cataline, corunnine, pontevedrine				14, 25
6. *G. grandiflorum*	Glaucine, isoboldine, thalicmidine, O-methylatheroline glauvine		Sanguinarine	Protopine	11

TABLE 1 (continued)

Plant species	Alkaloids				Literature
	aporphine	protoberberine	benzophenanthridine	protopine	
7. *G. leiocarpum*	Glaucine	Aurotensine		Protopine	8
8. *G. luteum*	Glaucine		Sanguinarine		
9. *G. oxylobum*	Corydine, glaucine, isobaldine, dl-isobaldine, domesticine, predicentrine, O-methylatheroline	Coptisine, berberine, scoulerine, aurotensine	Sanguinarine, chelerythrine, chelirubine, chelilutine	Allocryptopine, protopine	5, 7 3, 36 50
10. *G. pulchrum*	Corydine, isocorydine, bulbocapnine, N-methyllindcarpine			Protopine	18
11. *G. serpieri*	Isocorydine, glauvine	Aurotensine		Protopine	50
12. *G. squamigerum*	Corydine	Coptisine, berberine	Sanguinarine, chelerythrine	Allocryptopine, protopine	37, 52
13. *G. vitellinum*	Corydine, isocorydine, corytuberine, bulbocapnine, glaucine, dicentrine	Tetrahydropalmatine	Sanguinarine, chelerythrine, chelirubine	Allocryptopine, protopine, muramine	18, 38

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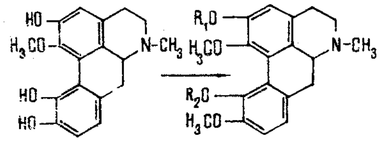
*In additon to the bases mentioned, the benzylisoquinoline alkaloids N-methylcoclaurine and reticuline have been isolated from this species.

† The morphinan alkaloid sinoacutine has also been isolated from *G. contortuplicatum*, and salutaridine and O-methylflavinantine from *G. flavum*.

The tetrasubstituted aporphine alkaloid glaufine (IX), containing three hydroxy groups and one methoxy group, has been isolated from *G. fimbrilligerum* [20]. According to its UV spectrum, glaufine is a 1,2,10,11-substituted aporphine alkaloid [21]. On the basis of the appearance of the NMR signal of the methoxy group in a strong field and the production by its methylation of N-methyllindcarpine (XII) and isocorydine (XIV), structure (IX) has been established for glaufine



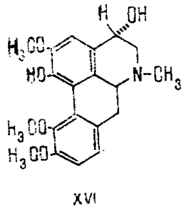
IX

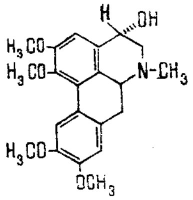
XII. $R_1=R_2=H$

XIV. $R_1=CH_3; R_2=H$

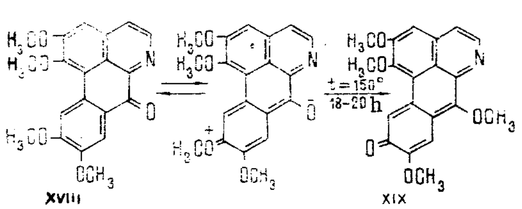
Two new pentasubstituted aporphine alkaloids have been isolated from plants of the genus *Glaucium* — cataline (XVII) and glaufine (XVI), each of which contains a hydroxy group in α orientation at C_4. This was established on the basis of a study of the NMR spectra of the bases themselves and of their acetyl derivative. An analysis of literature information shows that when the hydroxyl has the α orientation the geminal proton at C_4 gives a signal in the 4.43-4.47 region in the form of a poorly resolved triplet with a half-width of 5-6 Hz [22], while in the case of the β orientation of the hydroxy group it appears in the form of a quadruplet in the 4.93-5.00 ppm region (J_1 = 10 Hz, J_2 = 5.5-7 Hz) [23].

In these alkaloids, the aromatic proton at C_3 gives a signal in a weaker field than in the aporphines without a substituent in position 4 [24]. The catalytic hydrogenation of (XVII) with palladium on carbon gave glaucine, which confirms the 1,2,9,10-substituted structure of cataline [25].


XVI

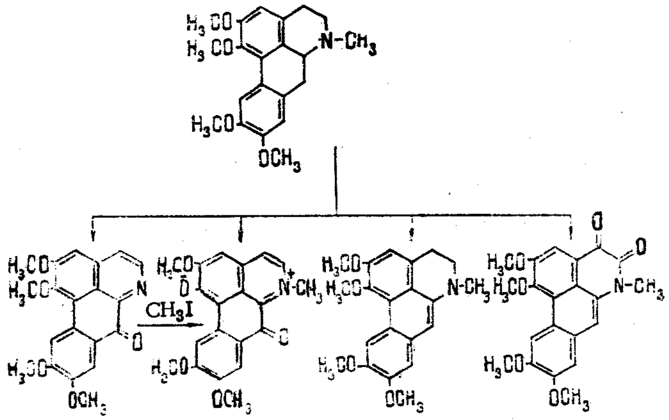

XVII

In contrast to cataline, glaufidine is a 1,2,10,11-substituted aporphine [26]. Four oxoaporphine alkaloids have recently been isolated: O-methylatheroline, glauvine, corunnine, and pontevedrine. O-Methylatheroline has been detected in five species of *Glaucium* [3, 9, 10, 13, 15]. The new alkaloid glauvine (XIX) has been isolated from the same plants, its prime source being *G. flavum* [13]. There is a report on the formation of glauvine from O-methylatheroline [13]. These two alkaloids have the same elementary composition and the same substituents, but differ in the locations of the carbonyl group and one methoxy group. In glauvine, the C=O group is present in position 10 and one of the methoxy groups in position 7.



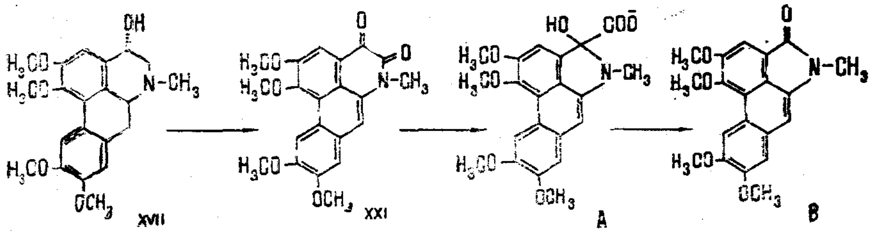
XVIII XIX

On thermolysis, O-methylatheroline (XVIII) is converted into glauvine. Corunnine (XX) isolated from *G. flavum* var. *vestitum* is 1-hydroxy-2,9,10-trimethoxy-7-oxoaporphine. The oxidation of glaucine chromium trioxide forms a mixture consisting of O-methylatheroline, corunnine, dehydroglaucine, and pontevedrine (XXI). Treatment of (XVIII) with methyl iodide also forms (XX) [14].



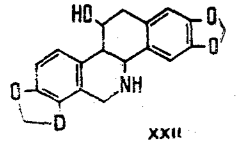
XVIII XX II XXI

In addition to corunnine, the dioxoaporphine alkaloid pontevedrine (XXI) has been isolated from *G. flavum*. The oxidation of cataline (XVII) has given pontevedrine, and the treatment of the latter with a methanolic solution of alkali leads through the benzilic acid (A) to compound B [27-29]. These facts have enabled structure (XX) with a double bond at 6a-7 and two carbonyl groups at C_4 and C_5 to be put forward for pontevedrine.



Pontevedrine is the first example of a 4,5-dioxoaporphine isolated from plants.

Another widespread group of bases in plants of the genus *Glaucium* is formed by the benzophenanthridine alkaloids. Sanguinarine and chelerythrine have been isolated from the majority of species [7, 9, 11, 16, 30-38]. A study of the pharmacological properties of these alkaloids has shown that they possess an antimicrobial action [39, 40] and also increase the intraocular pressure and exert a depressive action on the smooth musculature [41]. In recent times, only one new alkaloid belonging to this group of compounds has been found in these plants – norchelidonine (XXII) [12, 42].



XXII

The protoberberine alkaloids coptisine and berberine are found in almost all *Glaucium* plants [11, 30, 34, 36, 37, 43]. The methohydroxide of (–)-β-canadine (XXXVIII) and the methohydroxide (–)-β-stilopine (XXXVII) are new alkaloids of this group [31]. The representatives of the other groups of isoquinoline alkaloids are few. They include the protopine alkaloids protopine, allocryptopine, and muramine [18]. The first two alkaloids are found in almost all plants of the family Papaveraceae [41, 44, 45]. Three morphinan alkaloids are known: sinoacutine (XLII) [13, 46], salutaridine (XLIII [2, 4, 47]), which are enantiomers, and the methylflavinantine recently isolated from *G. flavum* [4]. This the first example of the finding of this alkaloid in the family Papaveraceae.

Two benzylisoquinoline alkaloids have been isolated from the plant *G. fimbrilligerum*: N-methylcoclaurine (XLV) [35] and reticuline (XLVI).

TABLE 2

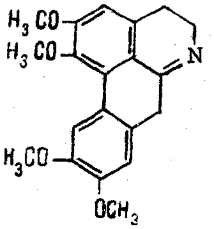
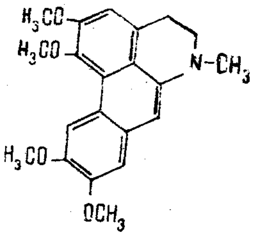
Alkaloid	Structural formula	Literature
I. 6,6a-Dehydronorglaucine, $C_{20}H_{21}NO_4$		15
II. Dehydroglaucine, $C_{21}H_{23}NO_4$, mp 133-134°C		15, 25

TABLE 2 (continued)

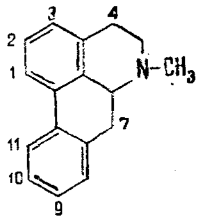
Alkaloid	Structural formula	Literature
Aporphine bases		
III. Domesticine, $C_{19}H_{19}NO_4$, mp 155-156°C (ether), $[\alpha]_D$ +56° (chloroform)	$R_1=OH$ $R_2=OCH_3$ $R_{9-10}=CH_2O_2$	36
IV. Isobaldine, $C_{19}H_{21}NO_4$, mp 126-127° (methanol), $[\alpha]_D$ +45° (chloroform)	$R_{1,9}=OH$ $R_{2,10}=OCH_3$	10, 11, 13, 17, 35, 36, 50
V. Dicentrine, $C_{20}H_{21}NO_4$, mp 168-169° (methanol), $[\alpha]_D$ +62° (chloroform)	$R_{1-2}=CH_2O_2$ $R_{9,10}=OCH_3$	2, 4, 46
VI. Thalicmidine, $C_{20}H_{23}NO_4$, mp 191-192° (methanol), $[\alpha]_D$ +40° (chloroform)	$R_1=OH$ $R_{2,9,10}=OCH_3$	9, 19, 25
VII. Predicentrine, $C_{20}H_{23}NO_4$, $[\alpha]_D$ +97° (ethanol)	$R_{1,9,10}=OCH_3$ $R_2=OH$	3
VIII. Glaucine, $C_{21}H_{25}NO_4$, mp 120-121° (ether), $[\alpha]_D$ +114° (ethanol)	$R_{1,2,9,10}=OCH_3$	3, 5, 8–18, 52
IX. Glaufine, $C_{18}H_{19}NO_4$, $[\alpha]_D$ +183° (methanol)	$R_1=OCH_3$ $R_{2,10,11}=OH$	20
X. Bulbocapnine, $C_{19}H_{19}NO_4$, mp 199-200° (ethanol), $[\alpha]_D$ +237° (chloroform)	$R_{1-2}=CH_2O_2$ $R_{10}=OCH_3$ $R_{11}=OH$	3, 4, 18
XI. Corytuberine, $C_{19}H_{21}NO_4$, mp 240-242°C (decomp., methanol), $[\alpha]_D$ +314° (methanol)	$R_{1,11}=OH$ $R_{2,10}=OCH_3$	33, 38, 43
XII. N-Methyllindcarpine, $C_{19}H_{21}NO_4$, mp 198-200°C (decomp., methanol), $[\alpha]_D$ +340° (methanol)	$R_{1,10}=OCH_3$ $R_{2,11}=OH$	18, 20
XIII. Corydine, $C_{20}H_{23}NO_4$, mp 149-150°C (methanol), $[\alpha]_D$ +204° (c 0.5; chloroform)	$R_1=OH$ $R_{2,10,11}=OCH_3$	7, 11, 18, 30, 38, 47, 49
XIV. Isocorydine, $C_{20}H_{23}NO_4$, mp 184-185°C (acetone), $[\alpha]_D$ +195° (chloroform)	$R_{1,2,10}=OCH_3$ $R_{11}=OH$	11, 13, 15, 18, 30 31, 33, 35, 38, 42
XV. Magnoflorine, $C_{20}H_{23}\overset{+}{N}O_4$, mp 249-250°C, $[\alpha]_D$ +197° (methanol)	$R_{1,11}=OH$ $R_{2,10}=OCH_3$ $>\overset{+}{N}<{CH_3 \atop CH_3}$	43
XVI. Glaufidine, $C_{20}H_{23}NO_5$, $[\alpha]_D$ +182° (methanol)	$R_1=OH$ $R_{2,10,11}=OCH_3$ $R_4=\alpha-OH$	26
XVII. Cataline, $C_{21}H_{25}NO_5$, mp 183-184°C, $[\alpha]_D$ +166° (chloroform)	$R_{1,2,9,10}=OCH_3$ $P_4=\alpha-OH$	25

TABLE 2 (continued)

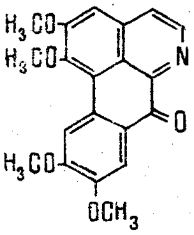
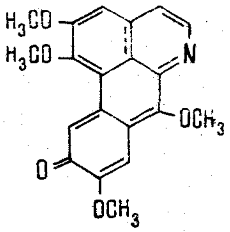
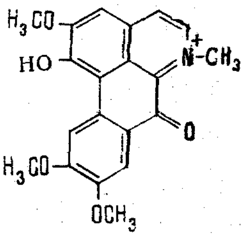
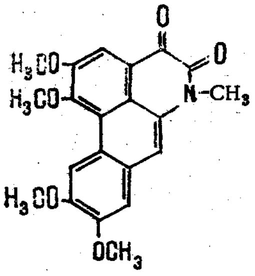
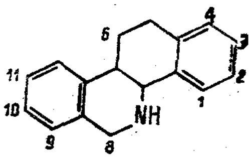
Alkaloid	Structural formula	Literature
Oxoaporphine bases		
XVIII. O-Methylatheroline, $C_{20}H_{17}NO_5$, mp 223-224°C		3, 9, 10, 13, 15
XIX. Glauvine, $C_{20}H_{17}NO_5$, mp 210-212°C (methanol)		9, 10, 13, 15
XX. Corunnine, $C_{20}H_{17}\overset{+}{N}O_5$, mp 255-257°C (ethanol)		14
XXI. Pontevedrine, $C_{21}H_{19}NO_6$, mp 269-271°C [chloroform — ethanol (1 : 1)]		14, 27
Benzophenanthridine bases		
		

TABLE 2 (continued)

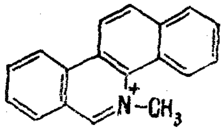
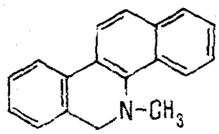
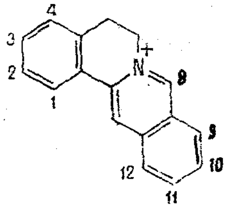
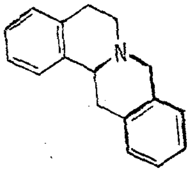
Alkaloid	Structural formula	Literature
XXII. Norchelidonine, $C_{19}H_{17}NO_5$, mp 198-199°C (methanol), $[\alpha]_D$ +100° (chloroform)	$R_{2-3}=R_{9-10}=CH_2O_2$ $R_6=OH$	12, 42
XXIII. Chelidonine, $C_{20}H_{19}NO_5$, mp 135-136°C (ethanol), $[\alpha]_D$ − 115° (ethanol)	$R_{2-3}=R_{9-10}=CH_2O_2$ $R_6=OH$ $>N-CH_3$	15, 12, 30, 31, 43
XXIV. dl-Chelidonine, mp 217-218°C (ethanol)	$R_{2-3}=R_{9-10}=CH_2O_2$ $R_6=OH$ $>N-CH_3$	11, 30, 35
XXV. Sanguinarine, $C_{20}H_{14}\overset{+}{N}O_4$, mp 242-243°C (methanol)	 $R_{2-3}R_{9-10}=CH_2O_2$	7, 9, 11, 16, 30—38, 43
XXVI. Chelerythrine, $C_{21}H_{18}\overset{+}{N}O_4$, mp 207-208°C (ether)	$R_{2-3}=CH_2O_2$ $R_{9,10}=OCH_3$	7, 9, 11, 16, 30—38, 43
XXVII. Chelirubine, $C_{21}H_{16}\overset{+}{N}O_5$, mp 257-258°C	$R_{2-3}=R_{9-10}=CH_2O_2$ $R_6=OCH_3$	11, 12, 30, 32, 34, 36, 38, 43
XXVIII. Chelilutine, $C_{22}N_{20}\overset{+}{N}O_5$, mp 229-230°C	$R_{2-3}=CH_2O_2$ $R_{6,9,10}=OCH_3$	36
XXIX. Dihydrochelythrine, $C_{21}H_{19}NO_4$, mp 164-165°C (methanol − chloroform)	 $R_{2-3}=CH_2O_2$ $R_{9,10}=OCH_3$	10
XXX. Bocconoline, $C_{22}H_{21}NO_5$, mp 221-222°C	$R_{2-3}=CH_2O_2$ $R_{9,10}=OCH_3$ R_8-CH_2OH	38
Protoberberine bases		
XXXI. Coptisine, $C_{19}H_{14}\overset{+}{N}O_4$, mp 216-218°C (ethanol)	$R_{2-3}=R_{9-10}=CH_2O_2$	11, 30, 31, 34, 36, 37, 43
XXXII. Berberine, $C_{20}H_{18}\overset{+}{N}O_4$, mp 145°C (aq. ethanol)	$R_{2-3}=CH_2O_2$ $R_{9,10}=OCH_3$	30, 31, 34, 36, 37
Tetrahydroprotoberberine bases		

TABLE 2 (continued)

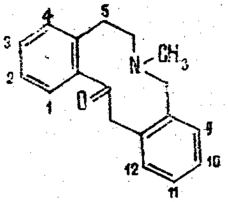
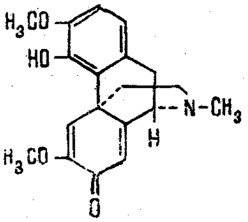
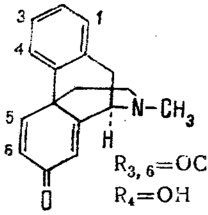
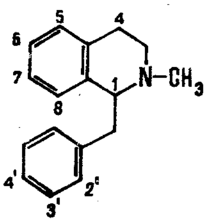
Alkaloid	Structural formula	Literature
XXXIII. Scoulerine, $C_{19}H_{21}NO_4$, mp 192-194°C (ethanol), $[\alpha]_D$ +260° (ethanol)	$R_{2,9}$=OH $R_{3,10}$=OCH_3	50
XXXIV. Aurotensine, $C_{19}H_{21}O_4$, mp 125-126°C, $[\alpha]_D$ +60° (ethanol)	$R_{2,9}$=OH $R_{3,10}$=OCH_3	12, 16, 48, 50, 51
XXXV. Isocorypalmine, $C_{20}H_{23}NO_4$ mp 231-232°C (methanol), $[\alpha]_D$ −282° (methanol)	R_2=OH $R_{3,9,10}$=OCH_3	34
XXXVI. Tetrahydropalmatine, $C_{21}H_{25}NO_4$, mp 141-142°C (ethanol), $[\alpha]_D$ −292° (chloroform)	$R_{2,3,9,10}$=OCH_3	18
XXXVII. Methohydroxide of (−)-β-stilopine, $C_{20}H_{20}\overset{+}{N}O_4$, mp 297-299°C (methanol), $[\alpha]_D$ −123° (methanol)	R_{2-3}=R_{9-10}=CH_2O_2 $>\overset{+}{N}-CH_3$	31
XXXVIII. Methohydroxide of (−)-β-canadine, $C_{21}H_{24}\overset{+}{N}O_4$, mp 260-261°C, (water), $[\alpha]_D$ −130° (methanol)	R_{2-3}=CH_2O_2 $R_{9,10}$=OCH_3 $>\overset{+}{N}-CH_3$	31
Protopine Bases		
XXXIX. Protopine, $C_{20}H_{19}NO_5$, mp 206-207°C (methanol)	R_{2-3}=R_{9-10}=CH_2O_2	2–4, 7–9, 11, 12, 15, 16, 18, 30–38, 49, 50, 52
XL. Allocryptopine, $C_{21}H_{23}NO_5$, mp 159-160°C (methanol)	R_{2-3}=CH_2O_2 $R_{9,10}$=OCH_3	7, 11, 1 , 30–38, 49, 52
XLI. Muramine, $C_{22}H_{27}NO_5$, mp 175-176°C (acetone)	$R_{2,3,9,10}$=OCH_3	18
Morphinan bases		
XLII. Sinoacutine, $C_{19}H_{21}NO_4$, mp 194-197°C (decomp.) $[\alpha]_D$ −117° (ethanol)		13, 46
XLIII. Salutaridine, $C_{19}H_{21}NO_4$, mp 198-199°C (ethanol)	 $R_{3,6}$=OCH_3 R_4=OH	3, 4

TABLE 2 (continued)

Alkaloid	Structure formula	Literature
XLIV. O-Methylflavinantine, $C_{20}N_{23}NO_4$	$R_{2,3,6}=OCH_3$	4
Benzylisoquinoline Bases		
XLV. N-Methylcoclaurine, $C_{18}H_{21}NO_3$, mp 132-133°C (methanol) $[\alpha]_D$ −62.3° (chloroform)	$R_{7,4'}=OH$ $R_6=OCH_3$	35
XLVI. Reticuline, $[\alpha]_D$ +47° (methanol)	$R_{7,3'}=OH$ $R_{6,4'}=OCH_3$	

At the present time, more than forty alkaloids have been isolated from *Glaucium* plants. Nine species of *Glaucium* grow on the territory of the USSR, of which *G. insigne* and *G. bracteatum* have not yet been studied for their alkaloid content, while some other species have been studied inadequately.

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