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Berberis ALKALOIDS.

XVII. INVESTIGATION OF THE ALKALOIDS OF Berberis heteropoda

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The alkaloid composition of young shoots and leaves of Berberis heteropoda Schrenk has been studied. In addition to known alkaloids, two new ones have been isolated – N-methyldihydroberberine and 8-oxoberberrubine, the structures of which were established by chemical transformations and a study of spectral properties. Of known alkaloids, berbamunine, aromoline, glaucine, thalicmidine, isocorydine, and reticuline have been found in this plant for the first time. Pseudopalmitine and laudanosine have been found for the first time in plants of the genus Berberis.

Continuing an investigation of the alkaloid composition of plants of the genus Berberis, we have studied young shoots and leaves of B. heteropoda collected in the fruit-bearing phase in the Dzhungarian Ala-Tau, in the environs of the settlement of Sarybel' (Alma-Ata province) [1]. The ethanolic extraction of the young shoots yielded 2.5% of total alkaloids.

The separation of the ether fraction on a column of silica gel led to the isolation of laudanosine [2], oxyacanthine, berbamunine, and aromoline [3], which were identified by their physicochemical properties, spectral characteristics, and comparison with authentic samples. By separation according to solubility and chromatography on a column of alumina, from the total quaternary alkaloids we isolated berberine, magnoflorine, columbamine, and jatrorrhizine in the form of their chlorides [4]. In addition to those mentioned above, we isolated another two alkaloids: base (I) with mp 212-213°C (chloroform) and base (II) with mp 210-212°C. The UV spectrum of base (I) had absorption bands at $\lambda_{max}^{C_2H_5OH}$ 263, 288, 307 (shoulder), 330, 379 nm ($\log \varepsilon$ 4.14; 4.45; 4.32; 4.10; 3.66), which are characteristic for protoberberine salts [5]. The PMR spectrum of (I), taken in $CDCl_3$, showed signals at (δ scale, ppm) 3.97 (3H, s, OCH_3), 3.99 (3H, s, OCH_3), 4.03 (3H, s, OCH_3), 4.14 (3H, s, OCH_3), 3.25 (2H, t, CH_2), 4.97 (2H, t, CH_2), 6.85 (1H, s), 7.48 (1H, s), 7.90 (2H, s), 8.65 (1H, s), 9.75 (1H, s). According to TLC and its IR spectrum, (I) was different from palmatine chloride [4]. The facts enabled (I) to be identified as pseudopalmatine chloride [5].

The UV spectrum of base (II) had absorption bands at $\lambda_{max}^{C_2H_5OH}$ 241, 350 nm ($\log \varepsilon$ 4.37; 3.92), which are characteristic for dihydroprotoberberines [6]. Its mass spectrum showed the peaks of ions with m/z 351, 337, 337, 336, 321, 320, 308, 307, 292, 278. The PMR spectrum, taken in DMSO-d_6 revealed signals at (δ scale, ppm) 6.05 (2H, s, OCH_2O), 4.05 (3H, s, OCH_3), 4.17 (3H, s, OCH_3), 3.45 (3H, br.s, $\overset{+}{N}CH_3$), 3.22-4.98 (6H, m), 6.49 (1H, s), 6.78(1H, s), 7.44 (2H, s), 7.85 (1H, s). The reduction of (II) with $NaBH_4$ gave (±)-N-methyltetrahydroberberine (III), which, on thermal demethylation at 300°C in vacuum, gave (±)-tetrahydroberberine (IV) [7], identical with an authentic sample (see Scheme 1).

Thus, base (II) was N-methyldihydroberberine, which has been synthesized previously [8].

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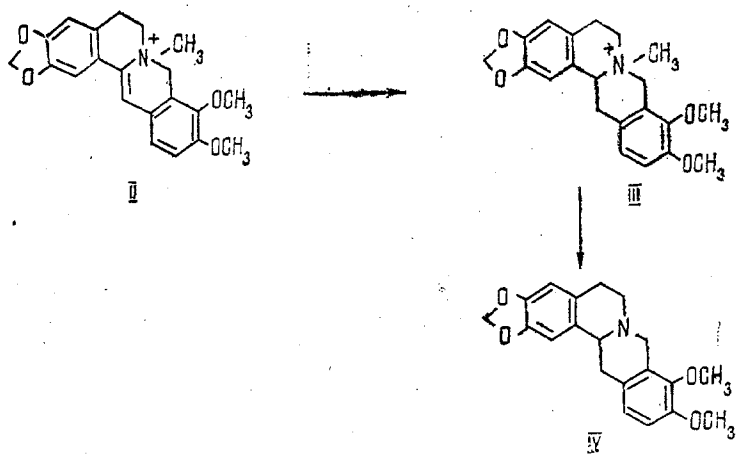
Scheme 1



Chloroform extraction of the leaves yielded 0.24% of total alkaloids. By chromatography on a column of alumina, glaucine, oxyacanthine, thalicmidine, isocorydine, and reticuline [9], and base (V) with mp 251-252°C, composition $C_{19}H_{15}NO_5$ were isolated. The UV spectrum of (V) had absorption maxima at $\lambda_{max}^{C_2H_5OH}$ 280 and 363 nm (log ε 3.97 and 3.94), which are characteristic for 8-oxoprotoberberines [10]. The IR spectrum had absorption bands at ν_{max}^{KBr} (cm^{-1}) 3450 (OH), 1650 (C=O), 1490 ($ArOCH_2O$).

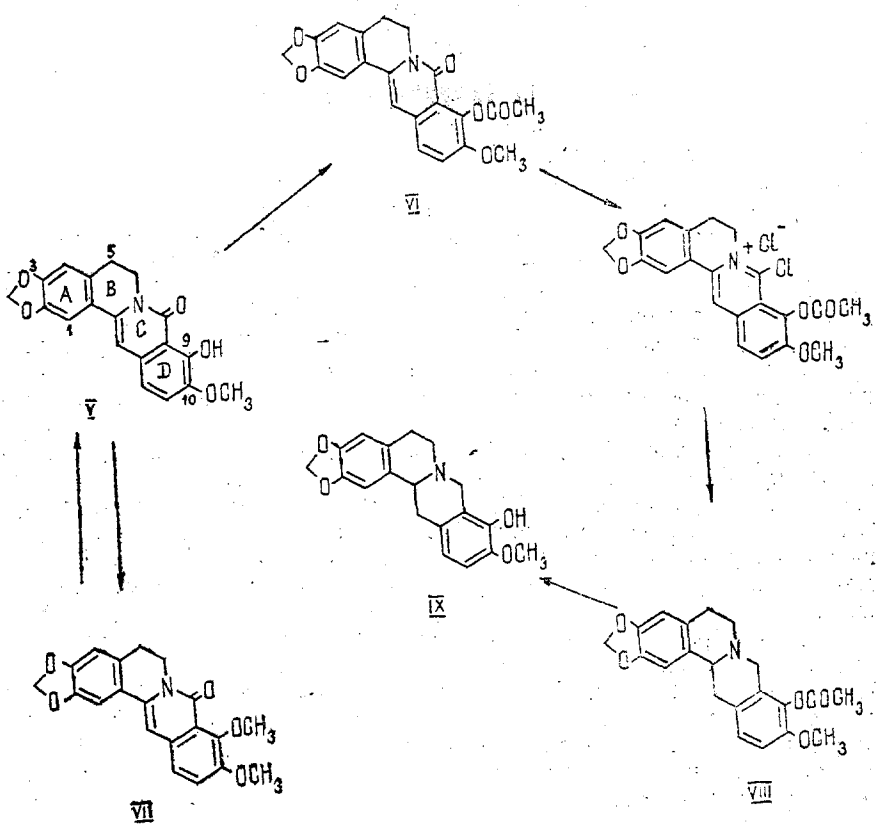
The mass spectrum of (V) contained the peaks of ions with m/z (%) 337 (M^+ 100), 323(22), 322(50), 308(13), 294(63), 279(27), which are characteristic for 8-oxoprotoberberines [11]. In the PMR spectrum taken in $CDCl_3$, signals were observed in the form of a two-proton singlet from a methylenedioxy group at 5.92 ppm, in the form of a three-proton singlet from a methoxy group at 3.88 ppm, and in the forms of two two-proton triplets at 2.83 and 4.17 ppm from two pairs of methylene protons. In the region of aromatic protons three one-proton singlets were observed at 6.60, 6.71, and 7.08 ppm, two one-proton doublets at 6.86 and 7.19 ppm with ^3J = 8.5 Hz, and a one-proton singlet at 12.75 ppm from a hydroxy group present in the peri-position to a carbonyl group [15]. According to the facts given, (V) was assigned to the protoberberine alkaloids having an amide carbonyl and phenolic hydroxyl.

The acetylation of (V) with acetic anhydride in acetone in the presence of sodium acetate gave the O-acetyl derivative (VI). When (V) was methylated with dimethyl sulfate, 8-oxoberberine (VII), identical with an authentic sample was obtained. Thus, (V) had a methylenedioxy group in the 2,3 position and hydroxy and methoxy substituents in positions 9 and 10. To confirm the mutual positions of the substituents in ring D, we obtained (V) from (VII) under conditions described in the literature [12, 13] (see Scheme 2).

According to the results presented, base (V) was 8-oxoberberrubine. However, it must be mentioned that Perkin et al. [14] and Govindachari et al. [12] obtained the latter from 8-oxoberberine (VII) and called it isooxyberberine. According to Govindachari et al. [12], isooxyberberine was insoluble in $CDCl_3$ and also in DMSO. For this reason, these authors recorded the PMR spectrum of isooxyberberine in trifluoroacetic acid, while the alkaloid (V) that we had isolated was readily soluble in chloroform and we therefore established the position of the hydroxy group by the following chemical transformations and by NOE measurements in the PMR spectrum of (V).

When (VI) was treated with phosphorus oxychloride (Scheme 2), followed by reduction of the product obtained with sodium tetrahydroborate, O-acetyltetrahydroberberrubine (VIII) was obtained. Hydrolysis of the latter with hydrochloric aid gave (±)-tetrahydroberberrubine [2], identical with an authentic sample according to its IR spectrum and the absence of a depression of the melting point of a mixed sample.

By measuring the intramolecular nuclear Overhauser effect (NOE) in the PMR spectrum of (V) we established the mutual positions of the substituents in ring D and also made an assignment of a number of protons signals. When the signal of the methylene protons at 2.83 ppm was irradiated, a strong increase (about 75%) in the singlet signal of the aromatic proton at 6.60 ppm was observed. At the same time, its integral intensity increased by 23%, which showed both the close spatial arrangement of the corresponding protons and the presence of a weak scalar interaction between them. On this basis, the singlet signal at 6.60 ppm had to be assigned to the proton at C-4, and the triplet signal at 2.83 ppm to the methylene protons at C-5. The protons of the methylene group at C-6 formed the triplet signal at 4.17 ppm.



Scheme 2

The appearance of the signals of the methylene protons in the form of triplets showed the equivalence of the geminal protons and their fairly rapid conversion around the C_5-C_6 bond.

When the signal of the methoxy group at 3.88 ppm was irradiated, a NOE of 15% was observed on the doublet signal at 7.19 ppm. This made it possible to assign the OCH_3 group to the C-10 position, and the doublet at 7.19 ppm to C-11. Consequently, the second doublet, at 6.86 ppm, had to be assigned to the proton at C-12. The remaining two singlet signals of aromatic protons at 7.08 and 6.71 ppm exhibited MOEs with one another of 25-30%. This confirmed their very close spatial position and corresponded to substitution in the C-1 and C-13 positions.

The observed NOE of 11% between the protons responsible for the doublet at 6.86 ppm and the singlet at 6.71 ppm made it possible to assign the latter to the proton at C-13, and the singlet at 7.08 ppm to the proton at C-1. On the whole, the results of the NOE measurements in the case of (V) unambiguously showed the positions of the OH and OCH groups in ring D at C-9 and C-10, respectively, which was in complete harmony with the results of the chemical transformations.

Thus, compound (V) was 8-oxoberberrubine. As a result of the investigation of young shoots and leaves of B. heteropoda, a total of 18 alkaloids were isolated. Of them, N-methyldihydroberberine and 8-oxoberberrubine were new natural bases; it was the first time that berbamumine, aromoline, glaucine, thalicmidine, isocoridine, and reticuline had been isolated from this plant, and the first time that pseudopalmitine and laudanosine had been isolated from plants of the genus Berberis.

EXPERIMENTAL

The composition of mixtures of alkaloids and the individuality of the compounds were monitored in a fixed thin layer of type LS 5/40 silica gel with 13% of gypsum in the solvent systems: 1) chloroform–methanol–conc. HCl (50:50:0.1); 2) chloroform–methanol (9:1; 95:5; 99:5 [sic]); and 3) benzene–ethanol (9:1). For column chromatography we used type KCK silica gel and alumina (Brockmann). UV spectra w4ere taken on a Hitachi spectrophotometer (in ethanol), IR spectra on a UR-20 spectrometer (KBr), mass spectra on a Mkh-1310 spectrometer with a system for direct introduction into the ion source, and PMR spectra on a Tesla BS-567A instrument. Hexamethyldisiloxane (HMDS) was used as internal standard.

Isolation and Separation of the Total Alkaloids from Young Shoots of B. heteropoda. The comminuted raw material (2.5 kg) was wetted with a 5% solution of acetic acid in ethanol in a ratio of 1:1. Then ethanol was added in a ratio to raw material of extractant of 1:4 and the mixture was boiled under reflux for 4 h. After cooling, the extract was decanted off, and the raw material was treated with ethanol (in a ratio of raw material to extractant of 1:3) and the mixture was boiled under reflux for 2 h. Then the solution was cooled, the extract was decanted off and combined with the preceding fraction, and the whole was evaporated under reduced pressure to a viscous mass. The latter was cooled to room temperature and was diluted with 0.5 liter of distilled water, filtered, made alkaline with conc. NH_4OH to pH 9, and extracted with ether and with chloroform. This gave 1.24 g of ether function (A) and 18.1 g of chloroform fraction (B).

The aqueous alkaline mother solution was acidified with conc. HCl to pH 1, and 4.5 g of crystalline KI was added. This led to the immediate formation of a bright yellow precipitate, which was filtered off and was washed with water, ethanol, and acetone. It was then dried, giving 7.75 g of berberine iodide. On standing, the acid fitrate deposited a brown resin of other quaternary iodides, which was separated off by decantation after a day. The resin was washed with water and was dissolved in methanol. This led to the crystallization of 10.25 of magnoflorine iodide. The mother solution was evaporated and the dried, giving 14 g of a mixture of quaternary iodides (C).

Separation of Mixture A. The ether fraction (6.2 g) was chromatographed on a column of silica gel. The alkaloids were eluted with chloroform and with mixtures of chloroform and methanol in various ratios. The fractions eluted by chloroform yielded 0.2 g of laudanosine, mp 87-88°C (benzene), and the fractions eluted by chloroform–methanol (98:2 and 97.3) gave 3.2 g of oxyacanthine, mp 217-218°C (hexane) and 1.4 g of berbamunine, mp 190-191°C (acetone). The 96:4 fractions yielded 0.6 g of aromoline, mp 196-197°C (methanol). Mass spectrum, m/z (%): 594 (M^+, 100), 593(55), 382(47), 381(85), 368(9), 367(42), 192(17), 191.5(19), 191(75), 174(17). IR spectrum (ν_{max}^{KBr}, cm^{-1}): 3400 OH. UV spectrum ($\lambda_{max}^{C_2H_5OH}$, nm): 228 (shoulder), 284 ($\log \varepsilon$ 4.65; 3.93).

Separation of the Mixture C. a) Conversion of the Mixture C into Chlorides. The alkaloid mixture C (14 g) was dissolved in 100 ml of methanol, and the solution was passed slowly through a column (2 × 110 cm) containing the anion-exchange resin IRA-400 (Cl form). The eluates obtained were combined and evaporated, to give 10.1 g of a mixture of chlorides (D) in the form of a yellow powder.

b) Separation of the Mixture D. The mixture of quaternary chlorides (D) (10.1 g) was chromatographed on a column of alumina (Brockmann activity grade II) at a ratio of the total alkaloids to the adsorbent of 1:40. The alkaloids were eluted with chloroform and with mixtures of chloroform and methanol in various ratios. The sections eluted by chloroform–methanol (99:1 and 98:1) yielded 2.4 g of berberine chloride, mp 204-205°C (methanol) and 0.8 g of pseudopalmitine chloride, mp 212-213°C ($CHCl_3$); the 97:3 fraction yielded 0.9 g of columbamine chloride, mp 238-239°C (methanol); the 95:5 fractions 0.7 g of jatrorrhizine chloride, mp 205-206°C (methanol); and the 94:6 fraction 0.14 g of N-methyldihydroberberine chloride, mp 211-212°C (methanol).

Isolation and Separation of the Total Alkaloids from the Leaves of B. heteropoda. The air-dry leaves (1.8 kg) were first wetted with an 8% solution of NH_4OH and were then extracted with chloroform (three extractions). The chloroform extract was evaporated to a volume of 700 ml, and the alkaloids were extracted with 5% hydrochloric acid. The acid solution was washed with ether and was then made alkaline with conc. NH_4OH to pH 9 and the alkaloids were extracted successively with ether and with chloroform. After evaporation of the solvents, 1.76 g of ether fraction (E) and 2.56 g of chloroform fraction (F) were obtained.

Separation of Mixture E. The mixture (1.76 g) was chromatographed on a column of silica gel. The alkaloids were eluted with chloroform and with mixtures of chloroform and methanol in various ratios. The chloroform fraction yielded 0.67 g of glaucine, and the fractions eluted with chloroform–methanol (98:2 and 97:3) gave 0.35 g of oxyacanthine and 0.47 g of thalicmidine.

Separation of Mixture F. Mixture F from the leaves (2.2 g) was chromatographed on a column of silica gel under similar conditions to the separation of mixture E, giving 0.15 g of 8-oxoberberrubine, 0.37 g of isocorydine, 0.49 g of reticuline, and 0.54 g of oxyacanthine.

Reduction of N-Methyldihydroberberine Chloride (II). A suspension of 80 mg of (II) in 15 ml of methanol was treated at 5°C with 80 mg of $NaBH_4$ in portions. The reaction mixture

was left at room temperature for 3 h, and then the solvent was evaporated off. The residue was dissolved in methanol with heating, and the solution was filtered and was left in the refrigerator overnight. The crystals that had deposited were separated off and were washed with methanol and dried. This gave 30 mg of (±)-N-methyltetrahydroberberine (III) with mp 258-260°C.

Demethylation of (±)-N-Methyltetrahydroberberine (III). Compound (III) (30 mg) was heated in vacuum at 300°C for 5 h. Then the tube was cooled and opened, and the residue was dissolved in chloroform and chromatographed on a column of silica gel. On elution with benzene–ethanol (95:5), an individual substance (12 mg) was isolated, and after recrystallization from methanol this had mp 169-170°C. From the absence of a depression of the melting point of a mixed sample, the demethylation product was identified as (±)-tetrahydroberberine (IV).

Acetylation of 8-Oxoberberrubine (V). A mixture of 50 mg of (V), 50 mg of anhydrous CH_3COONa, and 1 ml of acetic anhydride was boiled for 4 h. Then the solution was cooled and was treated with 10 ml of water. The precipitate that deposited was separated off and was crystallized from acetic acid. This gave 20 mg of acetyl-8-oxoberberrubine (VI) with mp 259-260°C. IR spectrum: ν_{max}^{KBr} 1735 cm^{-1}.

Methylation of 8-Oxoberberrubine (V). A suspension of 60 mg of (V) in 10 ml of dry acetone containing 40 mg of anhydrous K_2CO_3 and 1 ml of freshly distilled dimethyl sulfate was boiled in the water bath under reflux for 48 h. Then it was cooled, the acetone was evaporated off, and the residue was dissolved in 10 ml of water and extracted with chloroform. The chloroform solution was washed with water, dried over Na_2SO_4, and evaporated. The residue was crystallized from methanol, which gave 10 mg of a substance with mp 200°C identical with an authentic sample of 8-oxoberberine (VII) according to TLC, IR spectroscopy, and the absence of a depression of the melting point in a mixed sample.

Preparation of 8-Oxoberberrubine (V). a) Compound (V) was obtained by the method of Perkin et al. [12], mp 245-246°C, M^+ 337, IR spectrum: ν_{max}^{KBr}, cm^{-1}: 3400 (OH), 1650 (C=O).

b) Berberine chloride (1 g) was suspended in 10 ml of 20% NaOH solution, and the mixture was boiled on a hotplate for 2 h. After cooling, the precipitate that had deposited was separated off, washed with water, and dried in vacuum. It was then dissolved in 20 ml of chloroform, and dry gaseous HCl was passed through it. The precipitate that deposited (dihydroberberine chloride) was separated off, the chloroform solution was evaporated, and the residue was chromatographed on a column of silica gel. Elution with chloroform gave an individual substance (70 mg) which, after crystallization from acetic anhydride had mp 251-252°C, and was identical with 8-oxoberberrubine according to the absence of a depression of the melting point of a mixture and also according to spectral characteristics. Elution of the column with chloroform gave 0.25 g of (VII), in addition [13].

Preparation of O-Acetyltetrahydroberberrubine (VIII). A suspension of 45 mg of O-acetyl-8-oxoberberrubine (VI) in 1 ml of phosphorus oxychloride was heated under reflux for 2 h [16]. Then the solution was cooled, the brown residue that had deposited was separated off and was washed with chloroform and, without further purification, was dissolved in 115 ml of methanol and was treated in portions with 50 mg of $NaBH_4$. Reduction was carried out at room temperature for 0.5 h, and then the reaction mixture was boiled in the water bath under reflux for 0.5 h. After cooling, the solution was diluted with 20 ml of water and was extracted with chloroform. The chloroform solution was dried over Na_2SO_4, filtered, and evaporated. The residue was treated with acetone, which led to the isolation of 26 mg of O-acetyltetrahydroberberrubine (VIII) with mp 157-159°C.

Saponification of O-Acetyltetrahydroberberrubine (VIII). A solution of 26 mg of (VII) in 5 ml of methanol were treated with 5 ml of conc. HCl. The mixture was boiled in the water bath under reflux for 3 h. After cooling the solution was made alkaline with 5% NaOH solution and was washed with ether (2 × 50 ml). Then the alkaline solution was saturated with NH_4Cl and was extracted with ether (2 × 50 ml). The ethereal extracts were dried over Na_2SO_4, filtered, and evaporated. Treatment of the residue with methanol yielded 13 mg of (±)-tetrahydroberberrubine (IX) with mp 171-172°C.

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ALKALOIDS OF THE MONGOLIAN FLORA.

IV. TURCOSINE – A NEW ALKALOID FROM Aconitum turczaninowi

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UDC 547.944/945

As the result of a further study of the total alkaloids from the epigeal part of Aconitum turczaninowi we have isolated beiwutine and delcaroline and also a new alkaloid, which has been called turcosine. The structure of turcosine as 6β,16β-dimethoxy-4β-methoxymethyl-1α,7β,8β,10β,14α-pentahydroxy-N-ethylaconitane has been shown from a study of the IR, mass, PMR, and ^{13}C NMR spectra of the alkaloid and of the triacetate obtained from it.

In the preceding communication [1] we described the isolation from the epigeal part of Aconitum turczaninowi Worosch of aconitine, delsoline, delcosine, and lepenine and the new alkaloid tursoline. Continuing the study of the alkaloid composition of this plant, we have obtained beiwutine and delcaroline and a new alkaloid, which we have called turcosine (I).

Turcosine (I) has the composition $C_{24}H_{39}NO_8$ (M^+ 469.26664, HRMS). In the IR spectrum of alkaloid there were absorption bands of hydroxy groups at 3300-3530 cm^{-1} and of ether bonds at 1090 and 1100 cm^{-1}. The PMR spectrum of turcosine contained the signals of a N-ethyl group and of three methoxy groups and also the signals of a C-6-α proton at 3.99 ppm in the form of a doublet (J = 2 Hz) [2] and of a C-14-β proton in the form of a doublet of doublets at 4.62 ppm ($J_1 = J_2 = 5$ HZ). The mass spectrum of the alkaloid was close to that of delcosine, but differed from it by 16 m.u., i.e., by the presence of an additional hydroxy group. In the mass spectrum of turcosine, just as in the case of delcosine, the maximum peak was that of the $M^+ - 15$ ion, and there were also the ions $M^+ - 17$ (41%) and $M^+ - 33$ (38%), showing the presence of a hydroxy group at C-1 and a methoxy group at C-6 and of a 7,8-diol system [3].

In a comparison of the PMR spectra of turcosine (I) and delcosine (III) it was observed that the signal of the C-14-β proton was shifted downfield by 0.52 ppm (in the PMR spectrum of delosine the signal appears at 4.10 ppm), which is connected with the presence of a hydroxy group at C-10 [4].

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