Annelation of drotaverine by *p*-quinones to form hydroxyindolo- and hydroxybenzindoloisoquinoline derivatives

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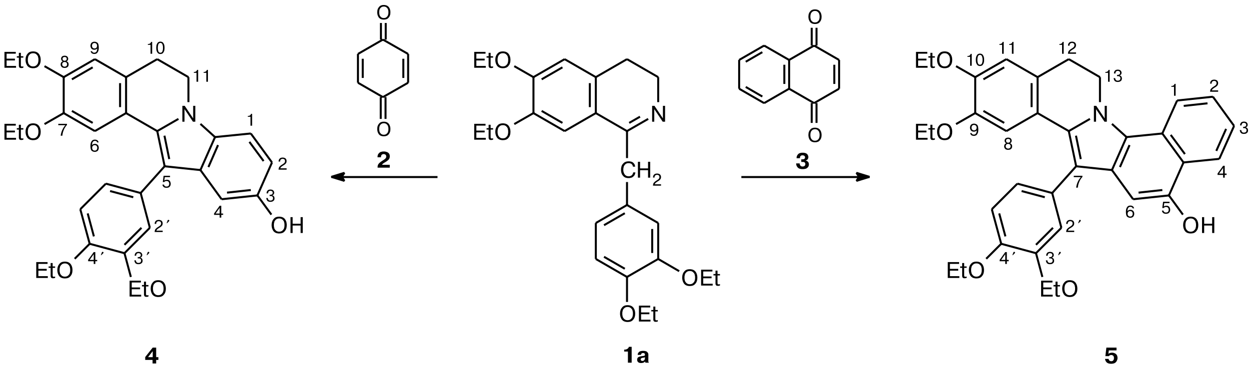
1-(3,4-Diethoxybenzyl)-6,7-diethoxy-3,4-dihydroisoquinoline (drotaverine, **1a**) reacts with *p*-benzoquinone (**2**) and *p*-naphthoquinone (**3**) in nitromethane or during fusion to give 5-(3,4-diethoxyphenyl)-7,8-diethoxy-3-hydroxy-5*a*,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (**4**) and 7-(3,4-diethoxyphenyl)-9,10-diethoxy-5-hydroxy-7*a*,12,13,14-tetrahydrobenz[*g*]indolo[2,1-*a*]isoquinoline (**5**), respectively. Compounds **4** and **5** are smoothly alkylated at the oxygen atom in the presence of bases. The structure of one alkylation product, *viz.*, 3-allyloxy-5-(3,4-diethoxyphenyl)-7,8-diethoxy-5*a*,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline, was established by X-ray diffraction analysis.

Key words: drotaverine, *p*-quinones, Nenitzescu reaction, indolotetrahydroisoquinoline derivatives, alkylation, X-ray diffraction analysis.

The reaction of *p*-quinones with enamines (Nenitzescu reaction) provides a general method for the synthesis of 5-hydroxyindole and 5-hydroxybenzofuran derivatives.[1—4] We studied the reactions of *p*-quinones with drotaverine, which is a well-known spasmolytic agent widely used in medicine as hydrochloride. Annelation of drotaverine in reactions with quinones gave promise that the resulting polycyclic systems could serve as a basis for the preparation of new physiologically active compounds. According to the ^{1}H NMR spectroscopic data, the drotaverine base has the structure of cyclic azomethine **1a**. Quantum-chemical calculations performed by the *ab initio* method (RHF/3-21G) and the density functional theory (DFT, B3LYP/3-21G)[5,6] also demonstrated that the azomethine form of drotaverine **1a** is energetically more favorable than enamine form **1b**. However, drotaverine base readily reacts with *p*-benzoquinone (**2**) and *p*-naphthoquinone (**3**) in nitromethane to give Nenitzescu reaction products, *viz.*, 5-(3´,4´-diethoxyphenyl)-7,8-diethoxy-3-hydroxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (**4**) and 7-(3´,4´-diethoxyphenyl)benz[*g*]-9,10-diethoxy-5-hydroxy-7*a*,12,13,14-tetrahydroindolo[2,1-*a*]isoquinoline (**5**), respectively, in moderate yields (Scheme 1).

The reaction is accompanied by heat evolution and bright coloring characteristic of the formation of radical ion salts. The possibility of the formation of such salts

Scheme 1



4 **1a** **5**

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upon electron transfer from a donor molecule to a quinoid compound is a well-known fact. The bright-brown reaction mixture rapidly turns dirty-brown. Even if the reaction is performed under argon, it affords a large amount of resinification products as by-products, which is typical of reactions involving radical steps. Nitromethane was chosen as a conventional solvent used in the Nenitzescu reaction.[3]

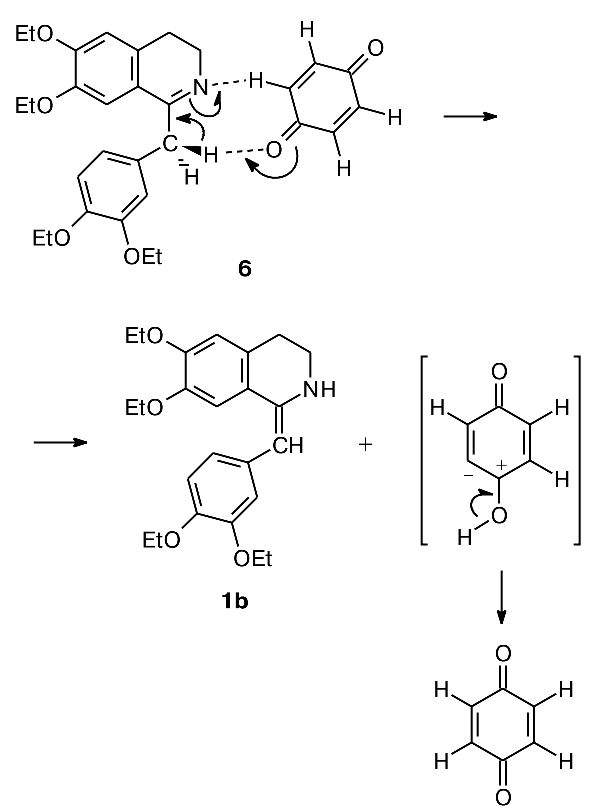
Condensation can be carried out also without a solvent by fusing an equimolar mixture of drotaverine and quinone. The appearance of a bright-crimson color was observed already during grinding of the reagents in a mortar. After heating of the mixture to 100—120 °C, a rapid exothermic reaction started. This procedure substantially simplifies the synthesis, while the yields of the final products are not decreased.

The form in which drotaverine reacts with quinones remains an open question, because it cannot be ruled out that the reaction is accompanied by the transformation of drotaverine into enamine form **1b**. Due to unfavorable steric conditions of 1,3-proton transfer, intramolecular isomerization of azomethine structure **1a** to enamine structure **1b** requires that a high energy barrier be overcome[7] and, consequently, this process seems to be unlikely. However, the low-barrier proton transfer can proceed if tautomerization occurs by an intermolecular mechanism.[7]

Calculations by the *ab initio* (RHF/3-21G) and DFT (B3LYP/3-21G) methods suggested that the reaction of the azomethine form of drotaverine with *p*-benzoquinone or *p*-naphthoquinone can afford H-bonded bimolecular complexes **6** and **6´**, respectively (Fig. 1), corresponding to energy minima ($\lambda = 0$) on potential energy surfaces.

The N...HC and CH...O interatomic distances in complexes **6** and **6´** calculated by the RHF method are 2.29 and 2.38 Å, respectively, and correspond to weak hydrogen bonds. The hydrogen bonds calculated by the DFT method are slightly shorter (N...HC, 2.14 Å; CH...O, 2.24 Å). The calculated stabilization energies for the H-bonded complexes are relatively high compared to the total energy of the isolated monomers. The complexation energies calculated by the RHF method are 8.2 (complex **6**) and 8.1 kcal mol^{-1} (complex **6´**). The energies calculated by DFT are slightly higher (11.3 and 11.2 kcal mol^{-1}, respectively). The formation of the sterically unhindered eight-membered ring with two virtually linear hydrogen-bond bridges in H-bonded complexes **6** and **6´** provides favorable steric conditions[8,9] for a low-barrier proton transfer and the transformation of drotaverine **1a** into enamine form **1b** (Scheme 2).

Scheme 2



Therefore, quinone can serve as the isomerizing agent, which is then involved in the Nenitzescu reaction to give enamine **1b**. In other words, molten quinone serves not only as the reagent but also as the solvent, in which drotaverine is transformed into the enamine form. In our opinion, the further mechanism involves the one-electron transfer to form a radical-ion pair **7** (**7´**),[5] which undergoes condensation to yield the hydroquinone adduct **8** (**8´**)[6] and then gives the final dihydroindoloisoquinoline **4** (or **5** in the reaction with naphthoquinone) (Scheme 3).

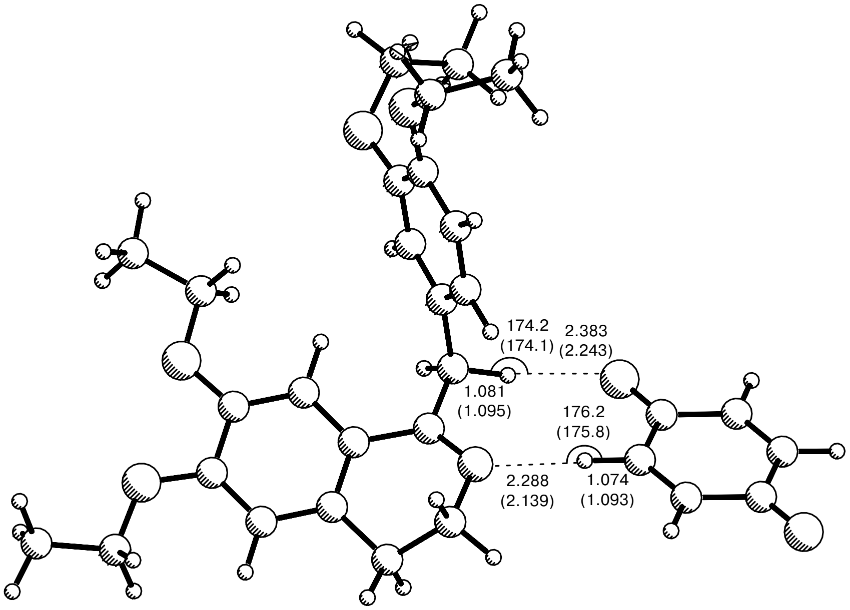
Compounds **4** and **5** are smoothly alkylated at the oxygen atom of the OH group by a wide range of halogen derivatives **9a—i** in the presence of bases to give compounds **10a—i** and **11a—g**, respectively. Heating of compounds **4** and **5** with acetic anhydride affords acetoxy derivatives **10j** and **11h**, respectively (Scheme 4).

Compound **5** is aminomethylated at position 6 (Scheme 5). Compound **4** is not involved in the analogous reaction.

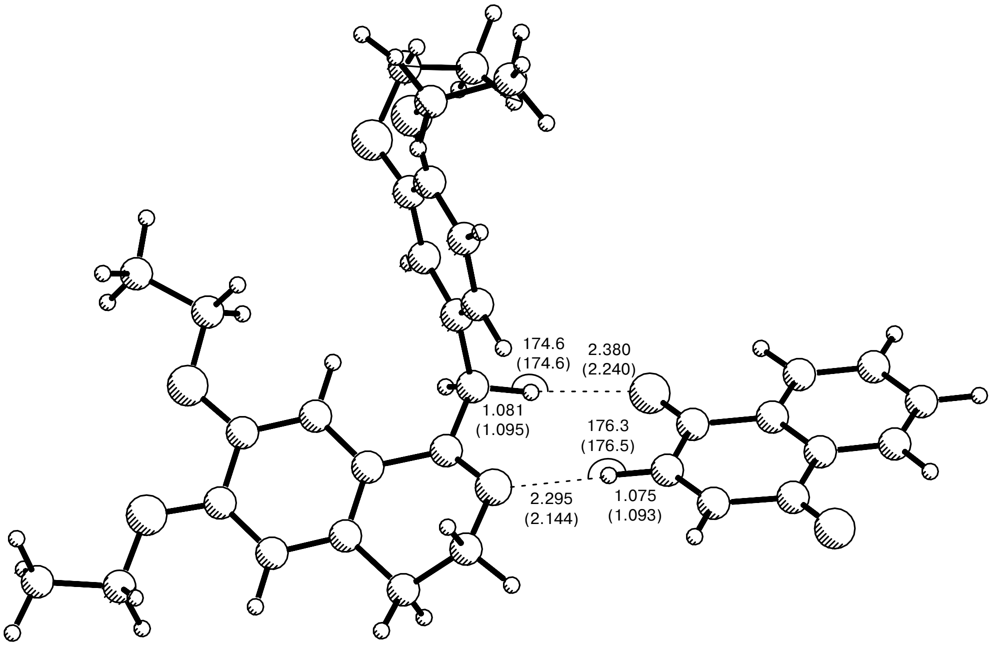
The ^1H NMR spectroscopic data for compounds **1a**, **4**, **10** and **5**, **11**, **12** are given in Tables 1 and 2, respectively.

The structure of 3-allyloxy-5-(3,4-diethoxyphenyl)-7,8-diethoxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (**10h**) was established by X-ray diffraction study (crystals were grown from CH_3CN). The molecular structure is shown in Fig. 2. Selected bond lengths and bond angles are given in Table 3.

Molecule **10h** consists of four fused rings and the phenyl moiety linked to the five-membered hetero-



6 (λ = 0)



6´ (λ = 0)

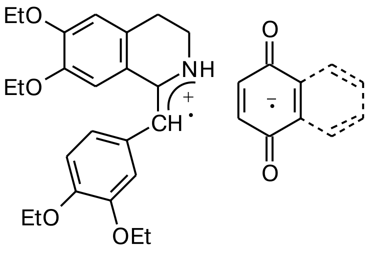
Fig. 1. Structures of complexes **6** and **6**´ calculated by the RHF/3-21G and B3LYP/3-21G methods. The bond lengths and bond angles are given in Å and deg.

cycle through the C(12)—C(17) single bond. The conformation of the polycyclic system is shown in the lateral projection (Fig. 3). In particular, it can be seen that the molecule is slightly twisted. The dihedral angle between two peripheral aromatic rings, *viz.*, C(10)C(11)C(13)C(14)C(15)C(16) (ring D) and C(2)C(3)C(4)C(5)C(6)C(7) (ring C), is 13.1°. The five-membered heterocycle (ring A) is planar. The dihedral angle between the planes of the rings A and D is 1.3°.

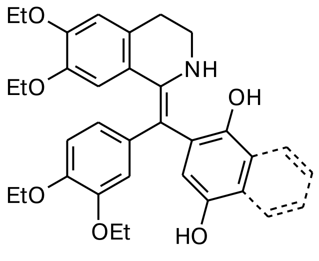
The six-membered heterocycle (B) is nonplanar and adopts a sofa conformation with the C(8) and C(9) atoms deviating from the plane through the other four atoms, N(1)C(1)C(2)C(7), in opposite directions by +0.264 and −0.408 Å, respectively. The aryl substituent at the C(12)

Scheme 3

$$\mathbf{1b} + \mathbf{2} \longrightarrow [\mathbf{7} \longrightarrow \mathbf{8}] \xrightarrow{-H_2O} \mathbf{4} \qquad \mathbf{1b} + \mathbf{3} \longrightarrow [\mathbf{7'} \longrightarrow \mathbf{8'}] \xrightarrow{-H_2O} \mathbf{5}$$

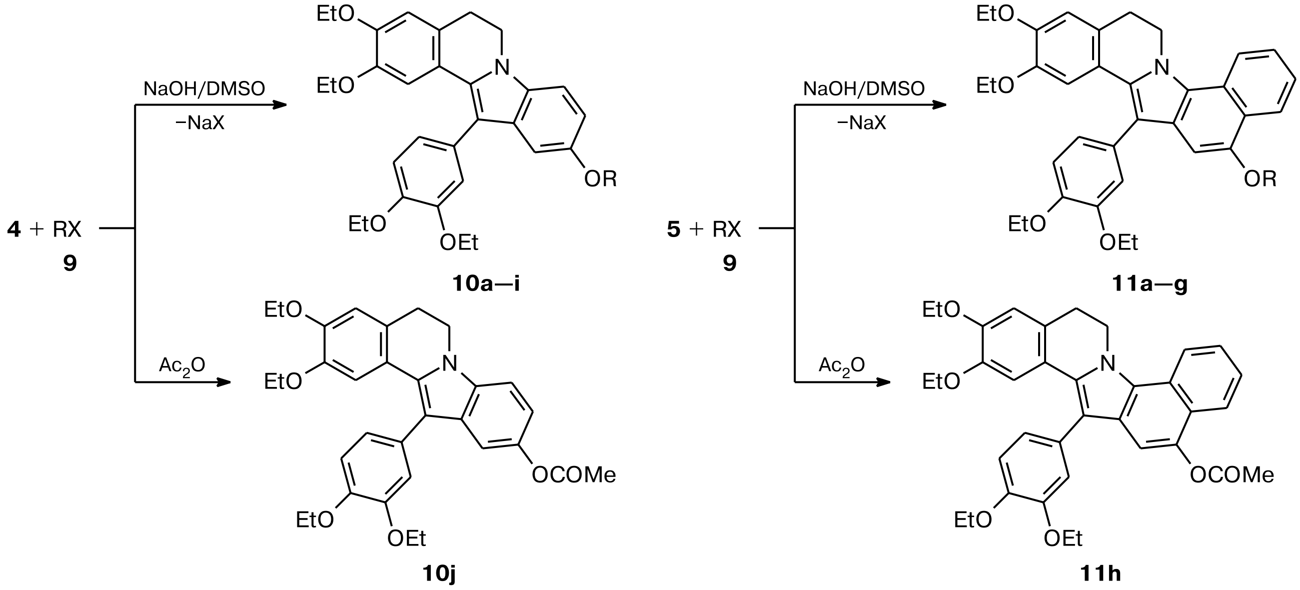


7, 7′



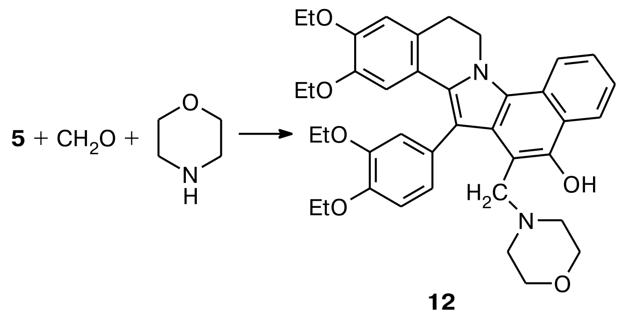
8, 8′

Scheme 4



9, **10**, **11:** R = Me (**a**), Et (**b**), Pr (**c**), Pr^i (**d**), Bu (**e**), Bu^i (**f**), PhCH$_2$ (**g**).
9, **10:** R = CH$_2$=CHCH$_2$ (**h**), HC≡CCH$_2$ (**i**). **11g:** R = HC≡CCH$_2$.

Scheme 5



12

atom (ring E) is twisted out of the plane of the ring A by 129.7°, which virtually excludes conjugation of the latter with the cyclic system.

The bond length distribution in the cyclic system is usual within the experimental error.[10,11] The disorder of two ethoxy groups over two positions is indicative of a low energy of their conjugation with the corresponding aromatic systems. The presence of disordered groups in the crystals of this compound even at low temperature is responsible for low accuracy of the results obtained in the present study, and hence the detailed discussion of the geometry makes no sense.

Table 1. ^{1}H NMR spectra of compounds **1a**, **4**, and **10a—j**

Compound	$CDCl_3$, δ (J/Hz)
1a	1.25—1.55 (m, 12 H, 4 Me); 2.62 (t, 2 H, C(4)H$_2$, J = 7.5); 3.71 (t, 2 H, C(3)H$_2$, J = 7.5); 3.85—4.20 (m, 8 H, 4 CH$_2$); 3.92 (s, 2 H, CH$_2$), 6.63 (s, 1 H, C(2′)H; 6.72—6.92 (m, 2 H, C(5′)H+C(6′)H); 6.78 (s, 1 H, C(5)H); 6.99 (s, 1 H, C(8)H)
4	1.19 (t, 3 H, Me, J = 7.0); 1.35—1.50 (m, 9 H, 3 Me), 3.10 (t, 2 H, C(10)H$_2$, J = 6.45); 3.62 (q, 2 H, CH$_2$, J = 7.0); 3.95—4.20 (m, 8 H, C(11)H$_2$+3 CH$_2$); 4.46 (s, 1 H, OH); 6.72 (s, 1 H, C(9)H); 6.77 (dd, 1 H, C(2)H, ^3J = 8.6, ^4J = 2.4); 6.88 (d, 1 H, C(4)H, J = 2.4); 6.90—7.00 (m, 3 H, CH$_{arom}$); 6.96 (s, 1 H, C(6)H); 7.17 (d, 1 H, C(1)H, J = 8.6)
10a	1.19 (t, 3 H, Me, J = 7.0), 1.38—1.45 (m, 9 H, 3 Me); 3.10 (t, 2 H, C(10)H$_2$, J = 6.45); 3.62 (q, 2 H, CH$_2$, J = 7.0); 3.78 (s, 3 H, OMe); 4.00—4.20 (m, 8 H, C(11)H$_2$+3CH$_2$); 6.72 (s, 1 H, C(9)H); 6.85 (dd, 1 H, C(2)H, ^3J = 8.7, ^4J = 2.4); 6.93 (d, 1 H, C(4)H, J = 2.4); 6.96 (s, 1 H, C(6)H); 6.97—7.10 (m, 3 H, CH$_{arom}$); 7.21 (d, 1 H, C(1)H, J = 8.7)
10b	1.19 (t, 3 H, Me, J = 7.0); 1.35—1.55 (m, 12 H, 4 Me); 3.10 (t, 2 H, C(10)H$_2$, J = 6.4); 3.63 (q, 2 H, CH$_2$, J = 7.0); 3.95—4.20 (m, 10 H, C(11)H$_2$+4 CH$_2$); 6.72 (s, 1 H, C(9)H); 6.86 (dd, 1 H, C(2)H, ^3J = 8.8, ^4J = 2.4); 6.93 (d, 1 H, C(4)H, J = 2.4); 6.95—7.05 (m, 4 H, CH$_{arom}$); 7.20 (d, 1 H, C(1)H, J = 8.8)
10c	1.00 (t, 3 H, Me, J = 7.4); 1.19 (t, 3 H, Me, J = 7.0); 1.35—1.50 (m, 9 H, 3 Me); 1.77 (m, 2 H, CH$_2$); 3.10 (t, 2 H, C(10)H$_2$, J = 6.4); 3.65 (q, 2 H, CH$_2$, J = 7.0); 3.88 (t, 2 H, CH$_2$, J = 6.6); 4.00—4.20 (m, 8 H, C(11)H$_2$+3 CH$_2$); 6.72 (s, 1 H, C(9)H); 6.86 (dd, 1 H, C(2)H, ^3J = 8.7, ^4J = 2.4); 6.93 (d, 1 H, C(4)H, J = 2.4); 6.95—7.05 (m, 4 H, CH$_{arom}$); 7.20 (d, 1 H, C(1)H, J = 8.7)
10d	1.19 (t, 3 H, Me, J = 7.0); 1.28 (d, 6 H, 2 Me, J = 6.0); 1.35—1.55 (m, 9 H, 3 Me); 3.10 (t, 2 H, C(10)H$_2$, J = 6.4); 3.62 (q, 2 H, CH$_2$, J = 7.0); 4.00—4.20 (m, 8 H, C(11)H$_2$+3 CH$_2$); 4.44 (quint, 1 H, CH, J = 6.0); 6.71 (s, 1 H, C(9)H); 6.85 (dd, 1 H, C(2)H, ^3J = 8.8, ^4J = 2.35); 6.90—7.10 (m, 5 H, CH$_{arom}$); 7.20 (d, 1 H, C(1)H)
10e	0.94 (t, 3 H, Me, J = 7.4); 1.19 (m, 4 H, CH$_3$+H); 1.35—1.55 (m, 10 H, 3 Me+H); 1.73 (m, 2 H, CH$_2$); 3.10 (t, 2 H, C(10)H$_2$, J = 6.45); 3.62 (q, 2 H, CH$_2$, J = 7.0); 3.92 (t, 2 H, CH$_2$, J = 7.0); 4.00—4.20 (m, 8 H, 4 CH$_2$); 6.72 (s, 1 H, C(9)H); 6.86 (dd, 1 H, C(2)H, ^3J = 8.8, ^4J = 2.4); 6.93 (d, 1 H, C(4)H, J = 2.4); 6.95—7.05 (m, 4 H, CH$_{arom}$); 7.20 (d, 1 H, C(1)H, J = 8.8)
10f	1.00 (d, 6 H, 2 Me, J = 6.65); 1.22 (t, 3 H, Me, J = 7.0); 1.35—1.55 (m, 9 H, 3 Me); 2.05 (quint, 1 H, CH, J = 6.5); 3.09 (t, 2 H, C(10)H$_2$, J = 6.4); 3.63 (q, 2 H, CH$_2$, J = 7.0); 3.68 (d, 2 H, CH$_2$, J = 6.65); 4.00—4.20 (m, 8 H, C(11)H$_2$+3 CH$_2$); 6.72 (s, 1 H, C(9)H); 6.86 (dd, 1 H, C(2)H, ^3J = 8.7, ^4J = 2.3); 6.92 (d, 1 H, C(4)H, J = 2.3); 6.95—7.05 (m, 4 H, CH$_{arom}$); 7.20 (d, 1 H, C(1)H, J = 8.7)
10g	1.19 (t, 3 H, Me, J = 7.0); 1.35—1.50 (m, 9 H, 3 Me); 3.10 (t, 2 H, C(10)H$_2$, J = 6.4); 3.63 (q, 2 H, CH$_2$, J = 7.0); 3.95—4.20 (m, 8 H, C(11)H$_2$+3 CH$_2$); 5.03 (s, 2 H, OCH$_2$Ph); 6.72 (s, 1 H, C(9)H); 6.90—7.05 (m, 6 H, CH$_{arom}$); 7.20— 7.45 (m, 6 H, CH$_{arom}$)
10h	1.19 (t, 3 H, Me, J = 7.0); 1.30—1.50 (m, 9 H, 3 Me); 3.10 (t, 2 H, C(10)H$_2$, J = 6.4); 3.63 (q, 2 H, CH$_2$, J = 7.0); 4.00—4.20 (m, 8 H, C(11)H$_2$+3 CH$_2$); 4.49 (d, 2 H, OCH$_2$C=, J = 5.35); 5.18—5.44 (m, 2 H, =CH$_2$); 6.05 (m, 1 H, —CH=); 6.72 (s, 1 H, C(9)H); 6.90 (dd, 1 H, C(2)H, ^3J = 8.7, ^4J = 2.4); 6.86—7.05 (m, 5 H, CH$_{arom}$); 7.21 (d, 1 H, C(1)H, J = 8.7)
10i	1.20 (t, 3 H, Me, J = 7.0); 1.30—1.50 (m, 9 H, 3 Me); 2.45 (t, 1 H, ≡CH, J = 2.3); 3.09 (t, 2 H, C(10)H$_2$, J = 6.3); 3.62 (q, 2 H, CH$_2$, J = 7.0); 4.00—4.20 (m, 8 H, C(11)H$_2$+3 CH$_2$); 4.65 (d, 2 H, OCH$_2$C≡, J = 2.3); 6.72 (s, 1 H, C(9)H); 6.92 (dd, 1 H, C(2)H, ^3J = 8.7, ^4J = 2.4); 6.94—7.05 (m, 5 H, CH$_{arom}$); 7.23 (d, 1 H, C(1)H, J = 8.7)
10j	1.19 (t, 3 H, Me, J = 7.0); 1.30—1.50 (m, 9 H, 3 Me); 2.27 (s, 3 H, MeCO); 3.10 (t, 2 H, C(10)H$_2$, J = 6.3); 3.62 (q, 2 H, CH$_2$, J = 7.0); 3.65—4.25 (m, 8 H, C(11)H$_2$+3 CH$_2$); 6.73 (s, 1 H, C(9)H); 6.90 (dd, 1 H, C(2)H, ^3J = 8.7, ^4J = 2.1); 6.90—7.05 (m, 4 H, CH$_{arom}$); 7.14 (d, 1 H, C(4)H, J = 2.1); 7.28 (d, 1 H, C(1)H, J = 8.7)

Experimental

The IR spectra were measured on a Specord IR-75 instrument in Nujol mulls. The ^{1}H NMR spectra were recorded on a Varian UNITY-300 spectrometer. Drotaverine hydrochloride was provided by ZAO EMPILS FOKh (Rostov-on-Don). The base was isolated by treatment of a methanolic solution of drotaverine hydrochloride with an aqueous ammonia solution. Allyl bromide was used as the alkylating agent. Benzo- and naphthoquinones were purchased from Fluka.

5-(3,4-Diethoxyphenyl)-7,8-diethoxy-3-hydroxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (4). Drotaverine (19.7 g, 0.05 mol) and *p*-benzoquinone (5.4 g, 0.05 mol) were ground in a mortar, placed in a 250-mL conical flask, and heated on a boiling water bath. After completion of a vigorous exothermic reaction, MeOH (100 mL) was added. The reaction mixture was refluxed and stirred with a rod until the solid residue was completely dissolved. The solution was cooled and kept on ice for several hours. The precipitate that formed was filtered off, washed with cold MeOH, and dried. The yield was 8 g (33%), m.p. 95—100 °C (from MeOH), solidification, repeated

Table 2. ^{1}H NMR spectra of compounds **5**, **11a—h**, and **12**

Compound	$CDCl_3$, δ (*J*/Hz)
5	1.18 (t, 3 H, Me, *J* = 7.0); 1.30—1.50 (m, 9 H, 3 Me); 3.15 (t, 2 H, C(12)H_2, *J* = 6.5); 3.62 (q, 2 H, CH_2, *J* = 7.0); 4.00—4.20 (m, 6 H, 3 CH_2); 4.82 (t, 2 H, C(13)H_2, *J* = 6.5); 5.05 (s, 1 H, OH); 6.76 (s, 1 H, C(11)H); 6.86 (s, 1 H, C(2´)H); 6.93 (s, 1 H, C(6)H); 6.95—7.05 (m, 3 H, CH_{arom}); 7.40—7.60 (m, 2 H, C(2)H+C(3)H); 8.25—8.45 (m, 2 H, C(1)H+C(4)H)
11a	1.20 (t, 3 H, Me, *J* = 7.0); 1.35—1.55 (m, 9 H, 3 Me); 3.14 (t, 2 H, C(12)H_2, *J* = 6.5); 3.63 (q, 2 H, CH_2, *J* = 7.0); 3.93 (s, 3 H, OMe); 4.00—4.25 (m, 6 H, 3 CH_2); 4.81 (t, 2 H, C(13)H_2, *J* = 6.5); 6.76 (s, 1 H, C(11)H); 6.86 (s, 1 H, C(2´)H); 6.90 (s, 1 H, C(6)H); 6.95—7.05 (m, 2 H, CH_{arom}); 7.07 (s, 1 H, C(8)H); 7.35—7.60 (m, 2 H, C(2)H+C(3)H); 8.36 (m, 2 H, C(1)H+C(4)H)
11b	1.19 (t, 3 H, Me, *J* = 7.0); 1.35—1.55 (m, 9 H, 3 Me); 3.14 (t, 2 H, C(12)H_2, *J* = 6.5); 3.62 (q, 2 H, CH_2, *J* = 7.0); 4.00—4.20 (m, 8 H, 4 CH_2); 4.81 (t, 2 H, C(13)H_2, *J* = 6.5); 6.75 (s, 1 H, C(11)H); 6.85 (s, 1 H, C(2´)H); 6.90 (s, 1 H, C(6)H); 6.95—7.04 (m, 2 H, CH_{arom}); 7.05 (s, 1 H, C(8)H); 7.35—7.60 (m, 2 H, C(2)H+C(3)H); 8.30—8.50 (m, 2 H, C(1)H+C(4)H)
11c	1.09 (t, 3 H, Me, *J* = 7.4); 1.19 (t, 3 H, Me, *J* = 7.0); 1.35—1.55 (m, 9 H, 3 Me); 1.91 (m, 2 H, CH_2); 3.14 (unres. t, C(12)H_2); 3.63 (q, 2 H, CH_2, *J* = 7.0); 3.90—4.30 (m, 8 H, 4 CH_2); 4.81 (t, 2 H, C(13)H_2, *J* = 6.5); 6.75 (s, 1 H, C(11)H); 6.86 (s, 1 H, C(2´)H); 6.89 (s, 1 H, C(6)H); 6.95—7.05 (m, 2 H, CH_{arom}); 7.06 (s, 1 H, C(8)H); 7.35—7.60 (m, 2 H, C(2)H+C(3)H); 8.30—8.50 (m, 2 H, C(1)H+C(4)H)
11d	1.20 (t, 3 H, Me, *J* = 7.0); 1.38 (d, 6 H, 2 Me, *J* = 6.0); 1.40—1.60 (m, 9 H, 3 Me); 3.14 (t, 2 H, C(12)H_2, *J* = 6.5); 3.63 (q, 2 H, CH_2, *J* = 7.0); 4.00—4.25 (m, 6 H, 3 CH_2); 4.60 (quint, 1 H, CH, *J* = 6.0); 4.81 (t, 2 H, C(13)H_2, *J* = 6.5); 6.75 (s, 1 H, C(11)H); 6.86 (s, 1 H, C(2´)H); 6.97 (s, 1 H, C(6)H); 7.06 (s, 1 H, C(8)H); 7.35—7.60 (m, 2 H, C(2)H+C(3)H); 8.37 (m, 2 H, C(1)H+C(4)H)
11e	0.98 (t, 3 H, Me, *J* = 7.4); 1.20 (t, 3 H, Me, *J* = 7.0); 1.30—1.70 (m, 11 H, CH_2+3 Me); 1.87 (m, 2 H, CH_2); 3.14 (unres. t, C(12)H_2); 3.63 (q, 2 H, CH_2, *J* = 7.0); 4.00—4.25 (m, 8 H, 4 CH_2); 4.81 (t, 2 H, C(13)H_2, *J* = 6.5); 6.75 (s, 1 H, C(11)H); 6.86 (s, 1 H, C(2´)H); 6.89 (s, 1 H, C(6)H); 6.95—7.05 (m, 2 H, CH_{arom}); 7.06 (s, 1 H, C(8)H); 7.35—7.60 (m, 2 H, C(2)H+C(3)H); 8.30—8.50 (m, 2 H, C(1)H+C(4)H)
11f	1.09 (br.d, 6 H, 2 Me); 1.20 (t, 3 H, Me, *J* = 7.0); 1.30—1.60 (m, 9 H, 3 Me); 2.21 (m, 1 H, CH), 3.14 (unres. t, 2 H, C(12)H_2); 3.63 (q, 2 H, CH_2, *J* = 7.0); 3.81 (d, 2 H, CH_2, *J* = 6.3); 4.00—4.20 (m, 6 H, 3 CH_2); 4.81 (t, 2 H, C(13)H, *J* = 6.5); 6.75 (s, 1 H, C(11)H); 6.86 (s, 1 H, C(2´)H); 6.87 (s, 1 H, C(6)H); 6.95—7.05 (m, 2 H, CH_{arom}); 7.06 (s, 1 H, C(8)H); 7.35—7.60 (m, 2 H, C(2)H+C(3)H); 8.30—8.50 (m, 2 H, C(1)H+C(4)H)
11g	1.20 (t, 3 H, Me, *J* = 7.0); 1.30—1.60 (m, 9 H, 3 Me); 2.46 (t, 1 H, ≡CH, *J* = 2.35); 3.14 (t, 2 H, C(12)H_2, *J* = 6.5); 3.63 (q, 2 H, CH_2, *J* = 7.0); 4.00—4.20 (m, 6 H, 3 CH_2); 4.80 (m, 4 H, C(13)H_2+CH_2C≡); 6.76 (s, 1 H, C(11)H); 6.87 (s, 1 H, C(2´)H); 6.99 (s, 1 H, C(6)H); 7.00—7.10 (m, 2 H, CH_{arom}); 7.05 (s, 1 H, C(8)H); 7.40—7.60 (m, 2 H, C(2)H+C(3)H); 8.38 (m, 2 H, C(1)H+C(4)H)
11h	1.19 (t, 3 H, Me, *J* = 7.0); 1.30—1.60 (m, 9 H, 3 Me); 2.42 (s, 3 H, $COCH_3$); 3.14 (t, 2 H, C(12)H_2, *J* = 6.5); 3.63 (q, 2 H, CH_2, *J* = 7.0); 3.90—4.30 (m, 6 H, 3 CH_2); 4.84 (t, 2 H, C(13)H_2, *J* = 6.5); 6.76 (s, 1 H, C(11)H); 6.88 (s, 1 H, C(2´)H); 6.90—7.10 (m, 3 H, CH_{arom}); 7.32 (s, 1 H, C(6)H); 7.40—7.60 (m, 2 H, C(2)H+C(3)H); 7.89 (d, 1 H, C(1)H, *J* = 8.5); 8.43 (d, 1 H, C(4)H, *J* = 8.5)
12	1.15 (t, 3 H, Me, *J* = 7.0); 1.30—1.60 (m, 9 H, 3 Me); 1.87 (m, 2 H, CH_2); 2.40 (unres. t, 4 H, $(CH_2)_2N$); 3.10 (t, 2 H, C(12)H_2, *J* = 6.5); 3.56 (m, 4 H, 2 CH_2); 3.68 (unres. t, 4 H, $(CH_2)_2O$); 3.90—4.30 (m, 6 H, 3 CH_2); 4.83 (m, 2 H, CH_2); 6.58 (s, 1 H, C(11)H); 6.72 (s, 1 H, C(2´)H); 6.90—7.10 (m, 3 H, CH_{arom}); 7.30—7.60 (m, 2 H, C(2)H+C(3)H); 8.35 (m, 2 H, C(1)H+C(4)H); 11.80 (s, 1 H, OH)

melting at 175—180 °C. IR, ν/cm^{-1}: 3390 br (OH), 1615, 1600, 1575, 1548 (arom.). Found (%): C, 73.51; H, 6.39; N, 3.11. $C_{30}H_{33}NO_5$. Calculated (%): C, 73.90; H, 6.82; N, 2.87.

7-(3´,4´-Diethoxyphenyl)-9,10-diethoxy-5-hydroxy-7a,12,13,14-tetrahydrobenz[*g*]indolo[2,1-*a*]isoquinoline (5). Drotaverine (16.0 g, 0.04 mol) and *p*-naphthoquinone (7.0 g, 0.044 mol) were ground in a mortar and fused in a 250-mL conical flask on a boiling water bath. After completion of the exothermic reaction, during which the temperature raised to 145—150 °C, the mixture was slightly cooled, MeOH (150 mL) was added, and the mixture was refluxed with grinding using a glass rod until the reaction product turned into a powdered precipitate. Then the mixture was cooled with cold water and the precipitate was filtered off and dried. The yield was 14.5 g (68%); m.p. 130—135 °C (from CH_3CN), solidification, repeated melting at 190 °C. IR, ν/cm^{-1}: 3465 (OH), 1620, 1600, 1580, 1540 (arom.). Found (%): C, 75.60; H, 6.52; N, 3.00. $C_{34}H_{35}NO_5$. Calculated (%): C, 75.95; H, 6.56; N, 2.61.

5-(3,4-Diethoxyphenyl)-7,8-diethoxy-3-methoxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (10a). Sodium hydroxide (0.24 g, 0.006 mol) was ground in a 25-mL conical flask under DMSO (5 mL). Then compound **4** (1.46 g, 0.003 mol) was added, the mixture was stirred with a glass rod for 10 min, and CH_3I (0.4 mL, 100% excess) was added. After 12 h, CH_3OH (10 mL) was added and the mixture was cooled with ice. The precipitate that formed was filtered off and recrystallized from Pr^{i}OH (50 mL). The colorless compound, m.p. 145 °C. The yield was 1.05 g (70%). Found (%): C, 74.47;

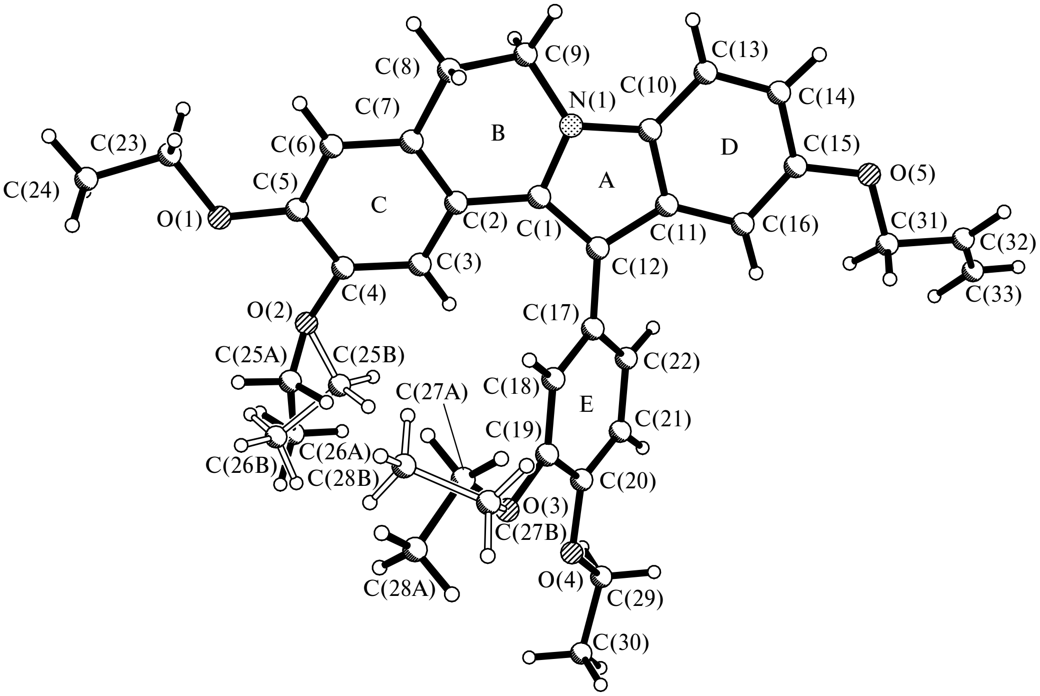


Fig. 2. Structure of molecule **10h**. The bonds for the second position of the disordered ethyl groups are indicated by empty lines.

H, 7.23; N, 2.60. $C_{31}H_{35}NO_5$. Calculated (%): C, 74.23; H, 7.03; N, 2.79.

5-(3,4-Diethoxyphenyl)-3,7,8-triethoxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (10b) was prepared analogously to **10a** by the reaction with EtI. After 12 h, the precipitate that formed was chromatographed on a column with Al_2O_3 in $CHCl_3$, the solvent was evaporated, and the residue was triturated with MeOH. The resulting colorless compound was filtered off and recrystallized from Pr^iOH (125 mL); m.p. 155 °C. The yield was 1.0 g (65%). Found (%): C, 74.30; H, 6.95; N, 2.83. $C_{32}H_{37}NO_5$. Calculated (%): C, 74.54; H, 7.23; N, 2.74.

5-(3,4-Diethoxyphenyl)-7,8-diethoxy-3-propoxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (10c) was prepared analogously to **10b** by the reaction with PrBr and chromatographed with hot $CHCl_3$. After column chromatography, the compound was recrystallized from Pr^iOH. The colorless

Table 3. Selected geometric parameters of molecule **10h**

Bond	*d*/Å	Bond	*d*/Å	Bond angle	ω/deg	Bond angle	ω/deg
O(1)—C(5)	1.374(5)	C(7)—C(8)	1.493(6)	N(1)—C(1)—C(2)	117.8(4)	C(10)—C(11)—C(16)	119.1(4)
O(2)—C(4)	1.366(5)	C(8)—C(9)	1.509(6)	C(12)—C(1)—C(2)	133.6(4)	C(10)—C(11)—C(12)	108.2(4)
O(3)—C(19)	1.370(5)	C(10)—C(13)	1.383(6)	C(7)—C(2)—C(3)	118.9(4)	C(16)—C(11)—C(12)	132.7(4)
O(4)—C(20)	1.368(5)	C(10)—C(11)	1.388(5)	C(7)—C(2)—C(1)	118.6(4)	C(1)—C(12)—C(11)	106.0(4)
O(5)—C(15)	1.376(5)	C(11)—C(16)	1.422(6)	C(3)—C(2)—C(1)	122.5(4)	C(1)—C(12)—C(17)	128.8(4)
N(1)—C(1)	1.382(5)	C(11)—C(12)	1.435(6)	C(4)—C(3)—C(2)	121.4(4)	C(11)—C(12)—C(17)	125.1(4)
N(1)—C(10)	1.384(5)	C(12)—C(17)	1.460(6)	C(6)—C(5)—C(4)	118.6(4)	C(14)—C(13)—C(10)	117.6(5)
N(1)—C(9)	1.454(5)	C(13)—C(14)	1.355(6)	C(7)—C(6)—C(5)	121.7(4)	C(13)—C(14)—C(15)	120.8(5)
C(1)—C(12)	1.405(5)	C(14)—C(15)	1.378(6)	C(6)—C(7)—C(2)	119.3(4)	O(5)—C(15)—C(14)	113.6(5)
C(1)—C(2)	1.457(6)	C(15)—C(16)	1.378(6)	C(6)—C(7)—C(8)	120.2(4)	O(5)—C(15)—C(16)	122.9(5)
C(2)—C(7)	1.391(5)	C(17)—C(22)	1.382(6)	C(2)—C(7)—C(8)	120.5(4)	C(14)—C(15)—C(16)	123.4(5)
C(2)—C(3)	1.394(5)	C(17)—C(18)	1.406(6)	C(7)—C(8)—C(9)	111.0(4)	C(15)—C(16)—C(11)	116.0(5)
C(3)—C(4)	1.370(5)	C(18)—C(19)	1.369(6)	N(1)—C(9)—C(8)	108.9(4)	C(22)—C(17)—C(18)	117.4(4)
C(4)—C(5)	1.387(6)	C(19)—C(20)	1.419(6)	C(13)—C(10)—N(1)	129.2(4)	C(22)—C(17)—C(12)	122.6(5)
C(5)—C(6)	1.387(6)	C(20)—C(21)	1.359(6)	C(13)—C(10)—C(11)	122.9(4)	C(18)—C(17)—C(12)	119.9(4)
C(6)—C(7)	1.381(6)	C(21)—C(22)	1.383(6)	N(1)—C(10)—C(11)	107.9(4)	C(19)—C(18)—C(17)	121.6(5)
				C(1)—N(1)—C(10)	109.3(4)	C(18)—C(19)—C(20)	119.6(5)
				C(1)—N(1)—C(9)	122.6(4)	C(21)—C(20)—C(19)	118.5(4)
				C(10)—N(1)—C(9)	126.3(4)	C(20)—C(21)—C(22)	121.6(5)
				N(1)—C(1)—C(12)	108.5(4)	C(17)—C(22)—C(21)	121.1(5)

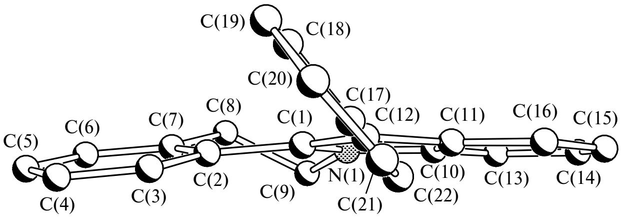


Fig. 3. Side projection of molecule **10h** showing the conformation of the cyclic system. All substituents in the cyclic system, except for the C(17)—C(22) benzene ring, are omitted for clarity.

compound, m.p. 145 °C. The yield was 0.85 g (46%). Found (%): C, 74.65; H, 7.30; N, 2.50. $C_{33}H_{39}NO_5$. Calculated (%): C, 74.83; H, 7.42; N, 2.66.

5-(3,4-Diethoxyphenyl)-7,8-diethoxy-3-isopropoxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (10d) was prepared analogously by the reaction with Pr^{i}Br. After column chromatography, the compound was recrystallized from Pr^{i}OH (60 mL). Colorless crystals, m.p. 160—162 °C. The yield was 0.75 g (41%). Found (%): C, 74.90; H, 7.05; N, 2.74. $C_{33}H_{39}NO_5$. Calculated (%): C, 74.83; H, 7.42; N, 2.66.

3-Butoxy-5-(3,4-diethoxyphenyl)-7,8-diethoxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (10e) was prepared analogously by the reaction with BuI. The column was eluted with hot $CHCl_3$, and the product was recrystallized from Pr^{i}OH (15 mL). The colorless compound, m.p. 125—128 °C. The yield was 0.9 g (55%). Found (%): C, 74.83; H, 7.26; N, 2.64. $C_{34}H_{41}NO_5$. Calculated (%): C, 75.11; H, 7.60; N, 2.58.

5-(3,4-Diethoxyphenyl)-7,8-diethoxy-3-isobutoxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (10f) was prepared analogously by the reaction with Bu^{i}Br and recrystallized from a mixture of CH_3CN (80 mL) and Pr^{i}OH (15 mL). The colorless compound, m.p. 155 °C. The yield was 0.72 g (44%). Found (%): C, 75.11; H, 7.43; N, 2.70. $C_{34}H_{41}NO_5$. Calculated (%): C, 75.11; H, 7.60; N, 2.58.

3-Benzyloxy-5-(3,4-diethoxyphenyl)-7,8-diethoxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (10g) was prepared analogously by the reaction with $ClCH_2C_6H_5$ and recrystallized from CH_3CN (20 mL). The colorless compound, m.p. 174—175 °C. The yield was 1.0 g (58%). Found (%): C, 77.13; H, 6.52; N, 2.78. $C_{37}H_{39}NO_5$. Calculated (%): C, 76.92; H, 6.80; N, 2.43.

3-Allyloxy-5-(3,4-diethoxyphenyl)-7,8-diethoxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (10h) was prepared analogously by the reaction with allyl bromide in DMSO (3 mL) for 3 h and recrystallized from Pr^{i}OH (40 mL). The colorless compound, m.p. 133—135 °C. The yield was 0.85 g (54%). Found (%): C, 74.70; H, 6.72; N, 2.76. $C_{33}H_{37}NO_5$. Calculated (%): C, 75.11; H, 7.07; N, 2.65.

5-(3,4-Diethoxyphenyl)-7,8-diethoxy-3-propargyloxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (10i) was prepared analogously by the reaction with propargyl bromide and recrystallized from Pr^{i}OH (40 mL). The colorless compound, m.p. 135 °C. The yield was 1.05 g (67%). IR, ν/cm^{-1}: 2115 (C≡C), 3300, 3294 (≡CH). Found (%): C, 75.07; H, 6.40; N, 2.84. $C_{33}H_{35}NO_5$. Calculated (%): C, 75.41; H, 6.71; N, 2.66.

3-Acetoxy-5-(3,4-diethoxyphenyl)-7,8-diethoxy-5a,10,11,12-tetrahydroindolo[2,1-*a*]isoquinoline (10j). Compound **4** (2 g, 0.004 mol) was refluxed in a mixture of Ac_2O (5 mL) and pyridine (0.1 mL) for 1 h and then poured into cold water (100 mL). The flocculent precipitate that formed upon trituration with a rod was filtered off, washed with water, dried, and passed through a column with Al_2O_3 in $CHCl_3$. The solvent was evaporated. The residue was triturated with MeOH, filtered off, and recrystallized from Pr^{i}OH (40 mL). The colorless compound, m.p. 150—152 °C. The yield was 1.3 g (60%). IR, ν/cm^{-1}: 1734, 1754 (CO). Found (%): C, 72.60; H, 6.45; N, 2.73. $C_{32}H_{35}NO_6$. Calculated (%): C, 72.57; H, 6.66; N, 2.64.

7-(3,4-Diethoxyphenyl)-9,10-diethoxy-5-methoxy-7a,12,13,14-tetrahydrobenz[*g*]indolo[2,1-*a*]isoquinoline (11a). Sodium hydroxide (0.16 g, 0.004 mol) was ground under DMSO (10 mL). Then compound **5** (1.1 g, 0.002 mol) was added, the mixture was triturated for 10 min, and CH_3I (0.4 mL, 0.0065 mol) was added. The mixture was triturated with a glass rod, warmed to 40 °C, and kept for 2 h. The precipitate that formed was filtered off, washed with hot H_2O and MeOH, and recrystallized from CH_3CN. The colorless compound, m.p. 155—160 °C. The yield was 0.68 g (60%). Found (%): C, 75.87; H, 7.08; N, 2.36. $C_{35}H_{37}NO_5$. Calculated (%): C, 76.20; H, 6.76; N, 2.54.

7-(3,4-Diethoxyphenyl)-5,9,10-triethoxy-7a,12,13,14-tetrahydrobenz[*g*]indolo[2,1-*a*]isoquinoline (11b). Sodium hydroxide (0.32 g, 0.008 mol) was triturated with DMSO (15 mL). Then compound **5** (2.15 g, 0.004 mol) was added, the mixture was triturated for 10 min, C_2H_5I (0.7 mL, 0.0086 mol) was added, and the mixture was heated to 40 °C and kept for 2 h. The precipitate that formed was filtered off, washed with MeOH, and dried. The pale precipitate that formed upon treatment of the filtrate with water was filtered off, washed with cold MeOH, and dried. The precipitates were combined, dissolved in $CHCl_3$, and passed through a column with Al_2O_3. The first colorless fraction was discarded. The solvent was evaporated, and the residue was recrystallized from a 3 : 1 Pr^{i}OH—CH_3CN mixture. The yellowish compound, m.p. 185 °C. The yield was 0.8 g (35%). Found (%): C, 76.00; H, 7.31; N, 2.57. $C_{36}H_{39}NO_5$. Calculated (%): C, 76.43; H, 6.95; N, 2.48.

7-(3,4-Diethoxyphenyl)-9,10-diethoxy-5-propoxy-7a,12,13,14-tetrahydrobenz[*g*]indolo[2,1-*a*]isoquinoline (11c). Sodium hydroxide (0.24 g, 0.006 mol) was ground in DMSO (10 mL). Then compound **5** (1.65 g, 0.003 mol) was added. The mixture was triturated for 10 min, PrBr (0.55 mL, 0.006 mol) was added, and the mixture was triturated and kept for 12 h. Then MeOH (10 mL) was added. The precipitate was filtered off, washed with MeOH, dried, dissolved in $CHCl_3$, and passed through a column with Al_2O_3. The solvent was evaporated. The residue was triturated with MeOH, filtered off, washed with MeOH, and dried. The colorless compound, m.p. 193—195 °C (from Pr^{i}OH). The yield was 1.16 g (65%). Found (%): C, 76.81; H, 7.29; N, 2.33. $C_{37}H_{41}NO_5$. Calculated (%): C, 76.66; H, 7.13; N, 2.42.

7-(3,4-Diethoxyphenyl)-9,10-diethoxy-5-isopropoxy-7a,12,13,14-tetrahydrobenz[*g*]indolo[2,1-*a*]isoquinoline (11d) was prepared analogously by the reaction with Pr^{i}Br (100% excess). After chromatography on a column with Al_2O_3, the compound was recrystallized from a 3 : 1 CH_3CN—Pr^{i}OH mixture. The colorless crystals, m.p. 200—203 °C. The yield was 0.85 g (48%). Found (%): C, 76.55; H, 7.37; N, 2.31. $C_{37}H_{41}NO_5$. Calculated (%): C, 76.66; H, 7.13; N, 2.42.

5-Butoxy-7-(3,4-diethoxyphenyl)-9,10-diethoxy-7a,12,13,14-tetrahydrobenz[*g*]indolo[2,1-*a*]isoquinoline (11e)

was prepared analogously by the reaction with BuI (100% excess). After chromatography on a column with Al_2O_3, the compound was recrystallized from ethyl acetate. The colorless compound, m.p. 182 °C. The yield was 0.86 g (47%). Found (%): C, 76.53; H, 7.62; N, 2.40. $C_{38}H_{43}NO_5$. Calculated (%): C, 76.87; H, 7.30; N, 2.36.

7-(3,4-Diethoxyphenyl)-9,10-diethoxy-5-isobutoxy-7a,12,13,14-tetrahydrobenz[*g*]indolo[2,1-*a*]isoquinoline (11f) was prepared analogously by the reaction with Bu^{i}Br (100% excess). After chromatography on a column with Al_2O_3, the compound was recrystallized from CH_3CN. The colorless compound, m.p. 190—195 °C. The yield was 0.72 g (39%). Found (%): C, 77.13; H, 7.48; N, 2.42. $C_{38}H_{43}NO_5$. Calculated (%): C, 76.87; H, 7.30; N, 2.36.

7-(3,4-Diethoxyphenyl)-9,10-diethoxy-5-propargyloxy-7a,12,13,14-tetrahydrobenz[*g*]indolo[2,1-*a*]isoquinoline (11g). Sodium hydroxide (0.16 g, 0.004 mol) was ground in DMSO (5 mL). Then compound **5** (1.0 g, 0.0019 mol) was added, the mixture was triturated for 10 min, and a 80% propargyl bromide solution (0.3 mL, 0.318 g, 0.0027 mol) in toluene was added. The mixture was heated at 90 °C and stirred with a rod until the solid residue was completely dissolved. Then MeOH (10 mL) was added, the mixture was cooled on ice, and the precipitate was filtered off and recrystallized from a 1 : 1 CH_3CN—Pr^{i}OH mixture. The colorless compound, m.p. 175—178 °C. The yield was 0.5 g (47%). IR, ν/cm^{-1}: 2127 (C≡C), 3253 (≡CH). Found (%): C, 77.00; H, 6.40; N, 2.31. $C_{37}H_{37}NO_5$. Calculated (%): C, 77.19; H, 6.48; N, 2.43.

5-Acetoxy-7-(3,4-diethoxyphenyl)-9,10-diethoxy-7a,12,13,14-tetrahydrobenz[*g*]indolo[2,1-*a*]isoquinoline (11h). Compound **5** (1.3 g, 0.0024 mol) was refluxed in a mixture of Ac_2O (5 mL) and pyridine (0.1 mL) for 15 min, poured into cold water (100 mL), and stirred with a rod until a flocculent precipitate was obtained. The precipitate was filtered off, recrystallized from EtOH (75 mL), and chromatographed on a column with Al_2O_3 in $CHCl_3$. The solvent was evaporated. The residue was triturated with MeOH, filtered off, washed with MeOH, and dried *in vacuo*. The colorless compound, m.p. 140—143 °C. The yield was 0.7 g (50%). IR, ν/cm^{-1}: 1767 (CO). Found (%): C, 74.13; H, 6.72; N, 2.69. $C_{36}H_{37}NO_6$. Calculated (%): C, 74.59; H, 6.43; N, 2.42.

7-(3,4-Diethoxyphenyl)-9,10-diethoxy-5-hydroxy-6-morpholinomethyl-7a,12,13,14-tetrahydrobenz[*g*]indolo[2,1-*a*]isoquinoline (12). A mixture of paraformaldehyde (0.07 g, 0.0023 mol) and morpholine (0.5 mL, 0.0055 mol) in Pr^{i}OH (5 mL) was refluxed until the solid residue was dissolved. Then compound **5** (1 g, 0.0019 mol) was added. The mixture was refluxed for 1.5 h, filtered before cooling, and cooled on ice. After 2 h, the precipitate that formed was filtered off, dissolved in $CHCl_3$, and passed through a column with Al_2O_3. The solvent was removed. The residue was triturated with CH_3CN, filtered off, and recrystallized from CH_3CN. The colorless compound, m.p. 150 °C. The yield was 0.52 g (44%). Found (%): C, 71.23; H, 7.58; N, 4.50. $C_{35}H_{42}N_2O_6$. Calculated (%): C, 71.65; H, 7.22; N, 4.77.

X-ray diffraction analysis of compound 10h. Principal crystallographic parameters and results of X-ray diffraction analysis are given in Table 4.

Table 4. Crystallographic parameters of compound **10h**

Parameter	**10h**
Molecular formula	$C_{33}H_{37}NO_5$
Molecular weight	527.64
Crystal system	Monoclinic
Space group	$P2_1/n$
a/Å	14.840(4)
b/Å	7.219(2)
c/Å	25.334(7)
β/deg	97.079(7)
V/Å^3	2693.3(14)
Z	4
ρ_{calc}/g cm^{-1}	1.301
F(000)	1128
λ(MoK_α)/mm^{-1}	0.087
Crystal dimensions/mm	0.34×0.24×0.18
Scanning mode	ω
Scan range, θ/deg	1.51—25.00
Ranges of indices of measured reflections	$-16 \leq h \leq 17$, $-8 \leq k \leq 7$, $-30 \leq l \leq 29$
Number of measured reflections	16816
Number of independent reflections	4762 (R_{int} = 0.1116)
Reflections with $I > 2\sigma(I)$	3854
Absorption correction	Semiempirical (SADABS)
Transmission, min/max	0.478/1.000
Variables in refinement	350
R factors based on reflections with $I > 2\sigma(I)$ and all reflections	$R_1 = 0.0657$, $wR_2 = 0.1491$ $R_1 = 0.1923$, $wR_2 = 0.2060$
Goodness-of-fit on F^2	0.823
Extinction coefficient	0.0000
Residual electron density, min/max, e/Å^3	−0.337/0.263

A prismatic colorless single crystal was coated with a perfluorinated oil and mounted on a Bruker SMART diffractometer equipped with a CCD area detector at 120 K. The reflections were scanned with a counting time of 10 s per step.

The structure was solved by direct methods and refined by the least-squares method against F^2. Analysis of a difference electron density synthesis revealed disorder of the ethyl groups of two ethoxy substituents over two positions with occupancies of ~30 and 70% for both groups. All hydrogen atoms were located from a difference electron density synthesis and then refined using a riding model.

The final refinement of the structure was carried out by the full-matrix least-squares method with anisotropic displacement parameters for all nonhydrogen atoms. All calculations were carried out using the SHELXS-86[12] and SHELXS-97[13] program packages.

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