Synthesis and selected properties of *N*-substituted pyrrolo[2,1-*c*]-1,3-diazacycloalkano[1,2-*a*]pyrazinones

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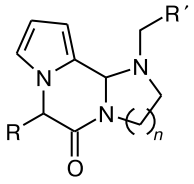
The reactions of methyl α-(2-formyl-1*H*-pyrrol-1-yl)carboxylates with *N*-substituted aliphatic 1,2-, 1,3-, and 1,4-diamines afford new pyrrole-containing heterocyclic systems: 1,2,3,10b-tetrahydroimidazo[1,2-*a*]pyrrolo[2,1-*c*]pyrazin-5(6*H*)-ones, 1,3,4,11b-tetrahydro-2*H*-pyrrolo[2′,1′:3,4]pyrazino[1,2-*a*]pyrimidin-6(7*H*)-ones, and 1,2,3,4,5,12b-hexahydropyrrolo[2′,1′:3,4]pyrazino[1,2-*a*][1,3]diazepin-7(8*H*)-ones. The reduction of these compounds with different reagents was studied.

Key words: aliphatic diamines, aldehyde esters, aminals, pyrrolo[2,1-*c*]-1,3-diazacycloalkano[1,2-*a*]pyrazinones, heterocyclization, hydrogenolysis, X-ray diffraction study, NMR spectroscopy.

The reactions of aldehyde acids, keto acids, and their esters with amino alcohols, amino thiols, and diamines provide a convenient route to bicyclic saturated and partially unsaturated heterocyclic systems.[1—5] These reactions are used for the synthesis of biologically active compounds[2,3] and alkaloids[4,6] and are employed in the highly stereoselective synthesis of drugs of natural origin.[7,8] These reactions lied at the basis of the present study. The following derivatives of the previously unknown heterocyclic systems were synthesized: 1,2,3,10b-tetrahydroimidazo-[1,2-*a*]pyrrolo[2,1-*c*]pyrazines (**1**), 1,3,4,11b-tetrahydro-2*H*-pyrrolo[2′,1′:3,4]pyrazino[1,2-*a*]pyrimidines (**2**), and 1,2,3,4,5,12b-hexahydropyrrolo[2′,1′:3,4]pyrazino-[1,2-*a*][1,3]diazepines (**3**). These compounds are structurally similar to a series of compounds with a broad spectrum of biological activities (antiviral, nootropic, spasmolytic antitussive, anxiolytic, *etc.*).[2,9,10] Hence, these compounds are of interest as potential new drugs. In addition, due to the presence of the aminal moiety, heterocycles **1—3** are of interest in terms of chemical transformations, primarily, under reduction conditions.



1—3

n = 1 (**1**), 2 (**2**), 3 (**3**)

Heterocycles **1—3** were synthesized starting from methyl α-(2-formyl-1*H*-pyrrol-1-yl)carboxylates (**4**). We developed a new procedure for the synthesis of these compounds based on the condensation of 2,5-dimethoxy-2-dimethoxymethyltetrahydrofuran (acetal **5**)[11] with amino acid methyl ester hydrochlorides **6** in an aqueous phosphate buffer (pH 4.7) (Scheme 1). The five-membered heterocycle in this reaction is formed due to the fact that in an acidic medium the hydrolysis of the starting acetal **5** containing three reaction centers initially occurs at the least stable endocyclic ketal group. The resulting carbocation reacts with the amino group of ester **6** followed by the closure of the five-membered ring, which is transformed into pyrrole.[12,13]

The developed method for the synthesis of esters **4** has an advantage over the procedures described previously[14—16] because it has a general character and allows the one-pot synthesis of these compounds in good yield.

N-Substituted diamines **7—9** served as the second components for the synthesis of heterocycles **1**, **2**, and **3**, respectively.

N-(Arylmethyl)substituted diamines **7—9** were synthesized by the reduction of the condensation products of the corresponding aromatic aldehydes **10** with unsubstituted diamines **11** (Scheme 2).[17,18] The reactions involved the hydrogenation of a mixture of the starting compounds in methanol in the presence of a palladium catalyst under atmospheric pressure. To reduce the formation of by-prod-

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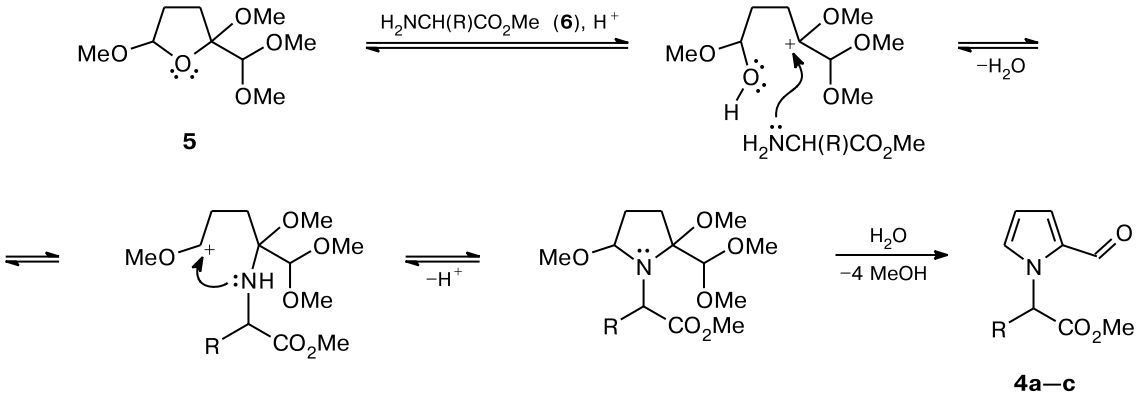
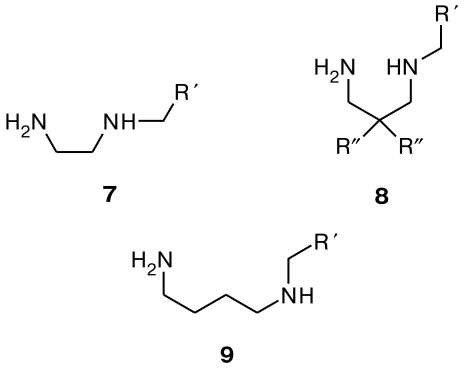
Scheme 1



4: R = H (**a**), Me (**b**), CH_2Ph (**c**)



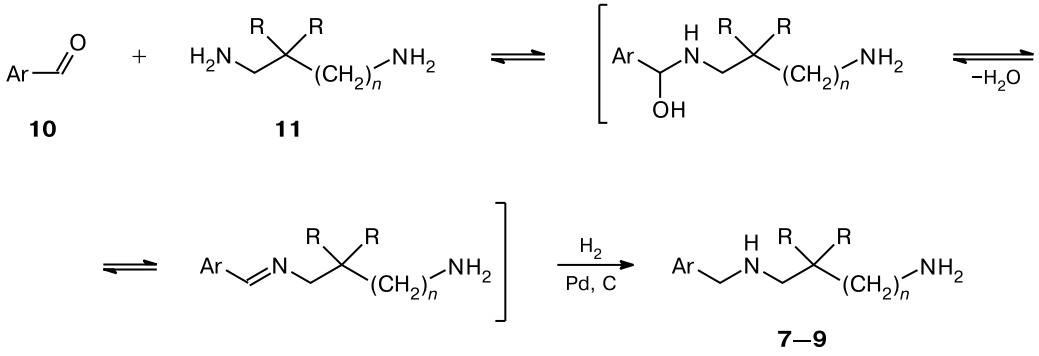
ucts, the threefold molar excess of diamines **11** was used. Target compounds **7—9** were prepared in 65—80% yields.

Tricyclic fused systems **1**, **2**, and **3** were accessed by the reactions of aldehyde esters **4** with diamines **7—9** (Scheme 3). Previously, such reactions have been performed by prolonged boiling of a mixture of the starting oxo acids or their esters and diamines[1,5,19] in aromatic solvents. We also showed that heterocyclic compounds **1** and **2** can be prepared in high yields simply by refluxing esters **4** with diamines **7** and **8** in a ratio of 1 : 1.1 in ethanolic solutions for 1 h.

The synthesis of compounds **3** required more drastic conditions. Thus, these compounds were synthesized by refluxing esters **4** with diamines **9** in xylene for 7 h.

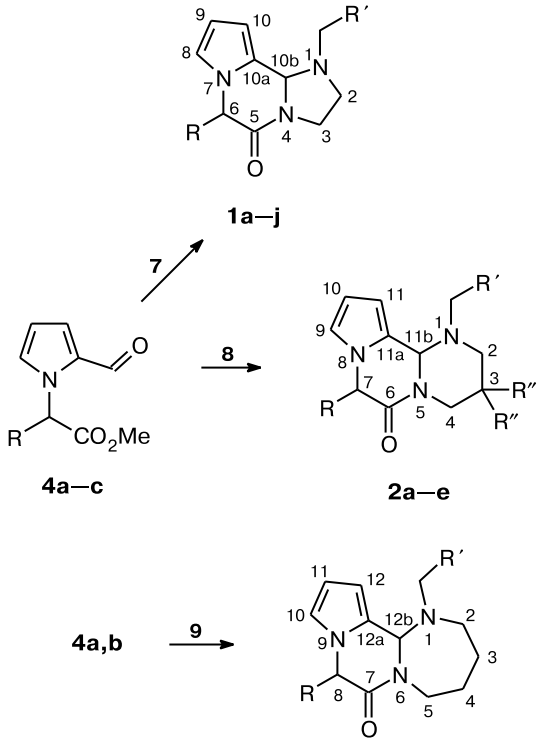
We suggest that the reaction giving rise to tricyclic systems **1**, **2**, and **3** occurs by the mechanism presented in Scheme 4. Initially, aldehyde ester **4** reacts with diamines **7—9** to give hemiaminal **12** at the primary amino group. Compound **12** exists in the equilibrium with imine form **13**. The second amino group adds at the imine bond to form cyclic aminal **14**, which is acylated by the ester group.

Scheme 2



Compound	Ar	*n*	R	Compound	Ar	*n*	R
7a	Ph	0	H	**8a**	3,4-$(MeO)_2C_6H_3$	1	H
7b	3,4-$(MeO)_2C_6H_3$	0	H	**8b**	3,4-$(MeO)_2C_6H_3$	1	Me
7c	3,4,5-$(MeO)_3C_6H_2$	0	H	**9a**	Ph	2	H
7d	2-Furyl	0	H	**9b**	3,4-$(MeO)_2C_6H_3$	2	H

Scheme 3



Compound	R	R′
1a	H	Ph
1b	H	3,4-$(MeO)_2C_6H_3$
1c	H	3,4,5-$(MeO)_3C_6H_2$
1d	H	2-Furyl
1e	H	Me
1f	H	CH_2OH
1g	H	CH_2NH_2
1h	Me	Ph
1i	Me	3,4,5-$(MeO)_3C_6H_2$
1j	CH_2Ph	3,4-$(MeO)_2C_6H_3$

Compound	R	R′	R″
2a	H	H	H
2b	H	3,4-$(MeO)_2C_6H_3$	H
2c	Me	3,4-$(MeO)_2C_6H_3$	H
2d	CH_2Ph	3,4-$(MeO)_2C_6H_3$	H
2e	H	3,4-$(MeO)_2C_6H_3$	Me

Compound	R	R′
3a	H	Ph
3b	H	3,4-$(MeO)_2C_6H_3$
3c	Me	Ph

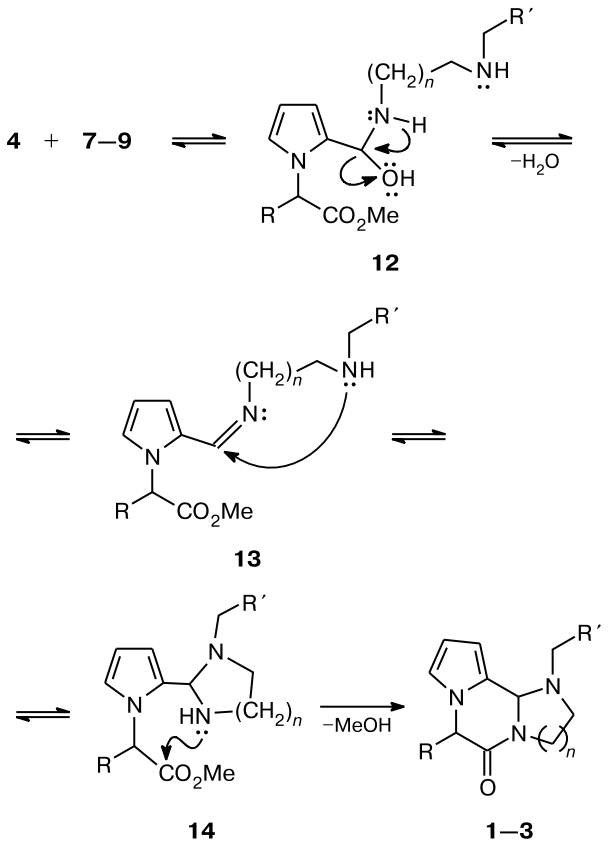
The structures of compounds **1—3** were confirmed by ^{1}H NMR spectroscopy, elemental analysis, and (for some compounds) mass spectrometry. The structures of compounds **1c** and **2d** were determined by X-ray diffraction (see the Experimental section).

Tricyclic structures **1c** and **2d** crystallized as racemates in the centrosymmetric space group $P\bar{1}$ (Fig. 1). Compound **1c** crystallized with one water molecule per formula unit. The asymmetric unit of compound **2d** contains two independent molecules (hereinafter, **2dA** and **2dB**).

In the crystal structure of **1c**, the water molecule forms strong hydrogen bonds with the N(1) (O...N, 3.0618(13) Å; O—H...N, 167.0(18)°) and O(13) (O...O, 2.8457(12) Å; O—H...O 173.5(19)°) atoms, resulting in the formation of hydrogen-bonded centrosymmetric dimers of **1c**, which are, in turn, linked together by weak van der Waals interactions.

Compound **2d** crystallized as the *trans* diastereomer. The conformations of independent molecules **2dA** and **2dB** in the crystal structure of **2d** are very similar and differ only in the C(6)—C(7)—C(14)—C(15) torsion angle (60.08(11)° and 63.84(12)° for molecules **2dA** and **2dB**, respectively). This leads to the difference in the atomic coordinates of the phenyl moieties after the superposition of the molecules (0.4 Å for the carbon atoms of the phenyl ring and no larger than 0.1 Å for the other nonhydrogen atoms of the molecules). The systems of contacts formed by **2dA** and **2dB** in the crystal structure are also similar. The crystals of **2d** are stabilized primarily by van der Waals

Scheme 4



n = 1 (**1**), 2 (**2**), 3 (**3**)

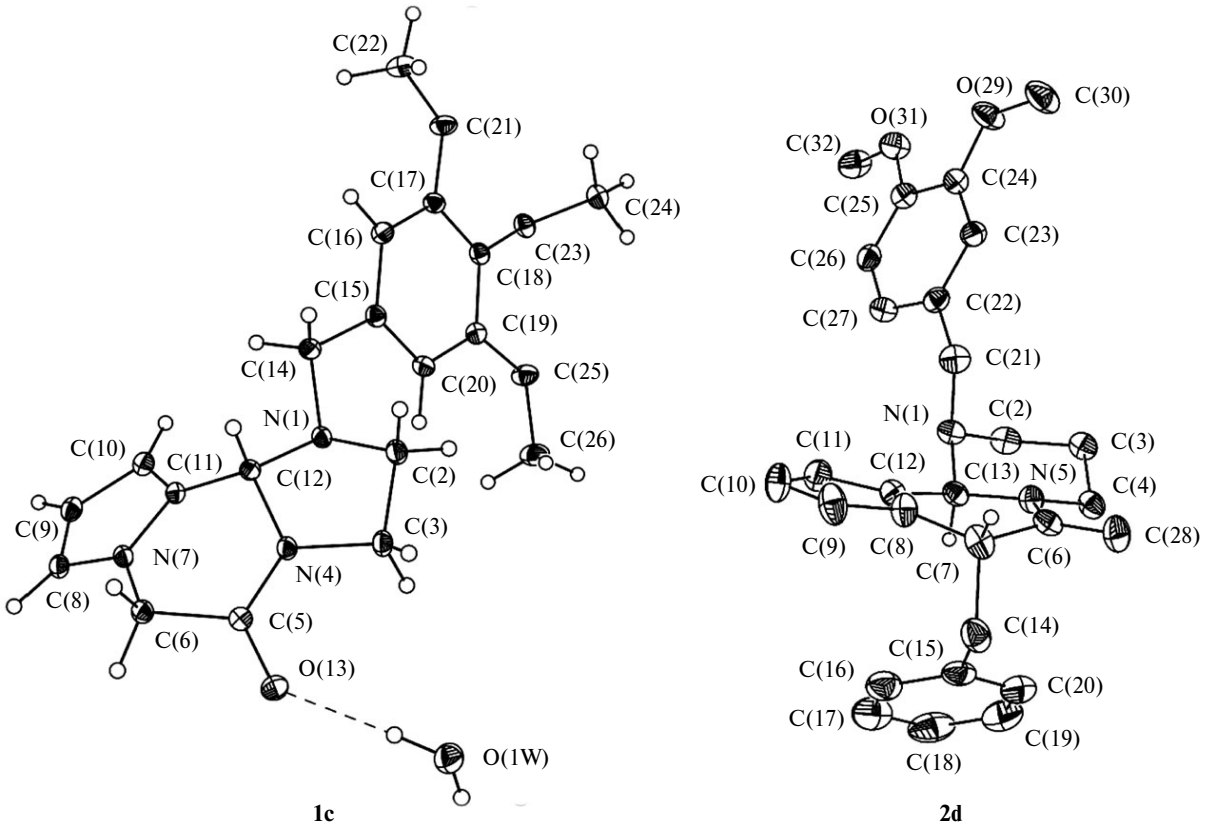


Fig. 1. Molecular structures of **1c** and **2d** in the crystals. The atoms are represented by thermal ellipsoids (p = 50%). Some hydrogen atoms of molecule **2d** are not shown.

C—H...O and H...H interactions, but there are also weak stacking interactions between the phenyl and 3,4-dimethoxyphenyl moieties of the crystallographically independent molecules. The shortest C...C distances for these interactions are 3.429(2) Å and 3.535(2) Å for two symmetrically nonequivalent interactions **2dA**...**2dB**; the distances between the centers of the rings are 3.723 and 3.764 Å, respectively; the angle between the planes involved in the stacking of the rings is at most 9°.

Like in compound **1c**, the N(1) atom in **2d** is highly pyramidalized (the deviations of the substituents from the plane are larger than 0.4 Å). It should be noted that the substituent at the N(1) atom in compound **1c** is in a pseudoequatorial position, whereas the corresponding atom in **2d** is in a pseudoaxial position. The pseudoaxial orientation of the substituent in compound **2d** results in the appearance of the generalized anomeric effect in the lp-N(1)—C(13)—N(5) system, which leads to an elongation of the C(13)—N(5) bond in **2d** compared to the equivalent C(12)—N(4) bond in **1c** by 0.024(5) Å and a shortening of the N(1)—C(13) bond compared to the equivalent N(1)—C(12) bond by 0.022(2) Å. As a result, the N(1)—C(12) and C(12)—N(4) bonds in **1c** are virtually equalized (1.473(2) Å), whereas the difference in the bond lengths in **2d** are 0.04 and 0.05 Å for molecules **2dA** and **2dB**, respectively. These values are typical of the generalized anomeric effect.[20]

In the ^{1}H NMR spectra of compounds **1**, the signals for the protons of the ethylene chain deserve consideration. Due to the rigidity of the heterocycle, these signals form an ABCD system, resulting in a considerable difference in their chemical shifts. As an example, Fig. 2 shows a fragment of the ^{1}H NMR spectrum of **1g** containing signals of this system, which do not overlap with other signals.

To assign these signals, we used the X-ray diffraction data for the structure of compound **1c** (Fig. 3), which gives the similar spectrum of the ethylene chain, and the Karplus—Conroy curve,[21] which shows the dependence of the vicinal spin-spin coupling constants $^3J_{H,H}$ on the corresponding dihedral angles.

The signals for all protons are observed as doublets of doublets of doublets. The signals at δ 2.72 and 3.76 belong to the protons H(2a) and H(3b), respectively (see Fig. 3). The dihedral angles corresponding to the vicinal constants of these protons are about 30° and 150°, with the result that the geminal coupling constants are similar to the vicinal constants (8—11 Hz). The dihedral angle between the bonds formed by the protons H(2b) (δ 3.45) and H(3a) (δ 3.58) with the corresponding carbon atoms is close to

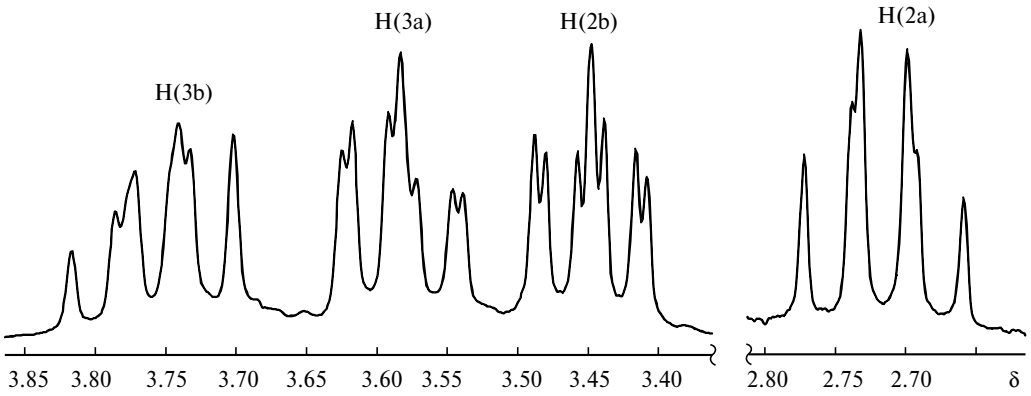


Fig. 2. Fragments of the ^{1}H NMR spectrum of compound **1g** showing signals for the protons of the ethylene chain.

90°, resulting in the spin-spin coupling between H(2b) and H(3a) as small as 2.0 Hz. It should be noted that there is a strong difference in the chemical shifts of the protons H(2a) and H(2b) (0.73 ppm for **1g** and 0.4—0.7 ppm for other compounds **1**).

This situation is apparently attributed to the pseudoequatorial position of the substituent at the amine nitrogen atom (Fig. 4), due to which the lone electron pair of the nitrogen atom shields the nearby protons in a different fashion. Based on the published data,[22] the pseudoaxial lone pair of the nitrogen atom shields the vicinal axial proton (in the structure under consideration, the proton H(2a)) by up to 1 ppm due to the n-σ* interaction, with no effect on the equatorial proton (H(2b)).

In compound **2**, the protons of the propylene chain form an ABCDEF system. To interpret the ^{1}H NMR spectrum of this moiety of **2** (Fig. 5), the dihedral angles between the C—H bonds of this system were determined from the X-ray diffraction data for **2d**. The angles corresponding to pairs of the axial protons H(2a)—H(3b) and H(3b)—H(4a) are about 165°; the angles corresponding to pairs of the equatorial protons H(2b)—H(3a) and H(3a)—H(4b) and to pairs of the axial and equatorial protons vary from 45 to 70° (Fig. 6). Based on the data from the Karplus—Conroy curve, the vicinal spin-spin coupling between the axial protons should be 9.5—14 Hz, and the coupling between the equatorial protons and between the axial and equatorial protons should be 1—5 Hz.

The terminal axial protons H(2a) (δ 2.68) and H(4a) (δ 2.43) have two large spin-spin coupling constants (geminal and vicinal). The central axial proton H(3b) (δ 2.04) has three constants (one geminal and two vicinal), due to which it appears as a broad multiplet. The equatorial protons H(2b) (δ 2.82), H(3a) (δ 1.18), and H(4b) (δ 4.71) have one (geminal) constant each. The geminal spin-spin coupling constants and the vicinal spin-spin coupling constants between the axial protons are approximately equal (13 Hz).

It should be noted that there is a very large difference in the chemical shifts of the protons of the $C(4)H_2$ group ($\Delta\delta = 2.3$ ppm). This can be attributed to the fact that the proton H(4b) is almost in one plane with the carbonyl bond of the amide group (the X-ray diffraction data). It is known[5] that in this case the proton is most efficiently deshielded by this group, resulting in a considerable downfield shift of this signal. At the same time, the spatial position of the proton H(4a) is such that the deshielding effect of the amide group is reduced. As a result, the signal for H(4a) is observed at even higher field compared to the signals for the protons of the $C(2)H_2$ group. The chemical shifts of the protons of the $C(3)H_2$ group are also substantially different. This fact can be attributed to the stronger deshielding effect of the nitrogen atoms on the proton

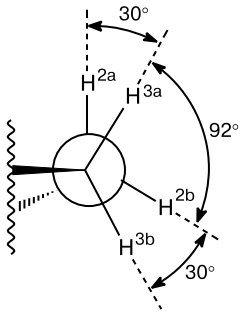


Fig. 3. Dihedral angles between the C—H bonds in the ethylene chain of compound **1c** determined by X-ray diffraction.

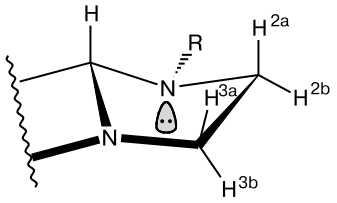


Fig. 4. Stereostructure of the tetrahydroimidazole moiety in compounds **1** according to the ^{1}H NMR spectroscopic data.

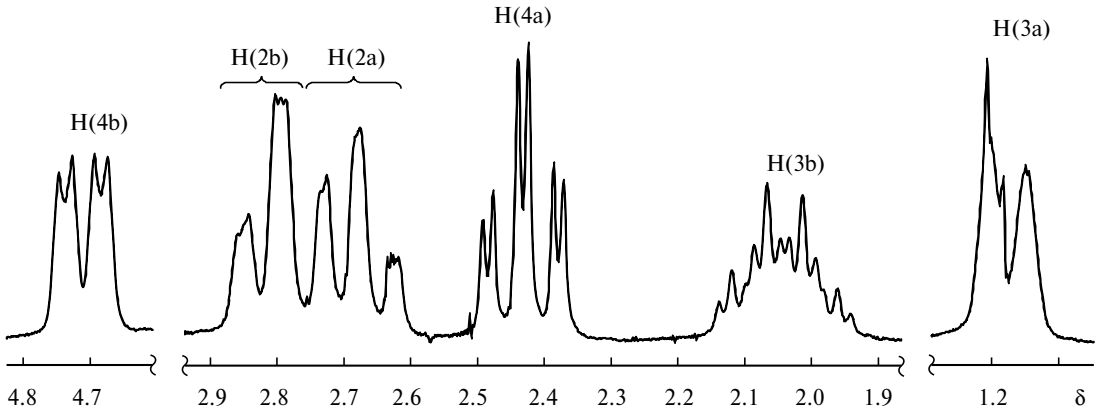


Fig. 5. Fragments of the ^{1}H NMR spectrum of compound **2d** showing signals for the protons of the propylene chain.

H(3b), which is spatially closer to the nitrogen atoms than H(3a).

In the spectra of **3**, the protons of the butylene chain form a complex system of multiplets, among which only signals for the protons of the methylene group bound to the amide nitrogen atom (H(5a) and H(5b)) are clearly distinguished; the other signals overlap with each other and are unresolved. The protons H(5a) and H(5b), like those in compounds **2**, essentially differ in the chemical shifts ($\Delta\delta$ = 0.6—0.8 ppm), which indicates that the lower-field proton (H(5b)) is approximately in the plane of the amide group.

There are two chiral centers in tricyclic compounds **1**—**3** containing substituents in the pyrazine ring. Hence, the synthesis of these compounds can afford two diastereomers (**A** and **B**, see Scheme 5). The X-ray diffraction study of one of these compounds (**2d**) showed that in the crystalline state it exists as the pure diastereomer **A** (see Fig. 1). However, according to the ^{1}H NMR spectroscopic data, some of these structures showed that most of these compounds exist in solution as mixtures of diastereomers **A** and **B**. It appeared that their ratio depends on the nature of the solvent and, for some compounds, on the time of the storage of their solutions. Table 1 gives the ratios of the pairs of diastereomers for heterocycles **1**—**3** containing substituents in the pyrazine ring, which were determined from the ^{1}H NMR spectra of solutions in $CDCl_3$ and DMSO-d_6 recorded immediately* after the dissolution of crystalline samples and after the storage of the solutions for one day and one week. The largest difference in the chemical shifts of the corresponding protons in pairs of diastereomers, which were used for the determination of their ratios, was observed for the protons of the methyl ($\Delta\delta$ up to 0.2 ppm) or methylene groups ($\Delta\delta$ up to 0.3 ppm) in the pyrazine ring and for the protons bound to the chiral C atoms ($\Delta\delta$ up to 0.15 ppm for the proton in the pyrazine ring; $\Delta\delta$ up to 1.5 ppm for the proton bound to the aminal C atom). The observed interconversion of the diastereomers can be attributed to the possibility of enolization of the amide carbonyl (see Scheme 5), which is facilitated due to the conjugation of the enol that formed with the aromatic pyrrole ring. Apparently, this transformation re-

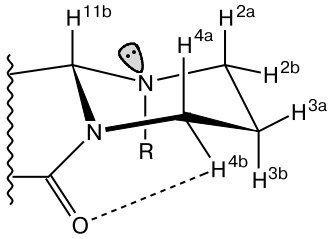
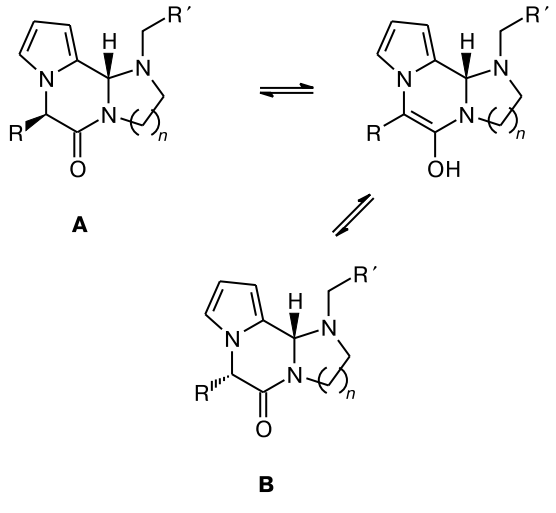


Fig. 6. Stereostructure of the hexahydropyrimidine moiety in compounds **2** according to the ^{1}H NMR spectroscopic data.

Scheme 5



n = 1—3

* The ^{1}H NMR spectra were recorded within 120—150 s after the dissolution of the samples.

Table 1. Ratio of diastereomers of heterocycles **1—3** containing substituents in the pyrazine ring depending on the time of the storage (t_1, t_2, t_3)* of their solutions in $CDCl_3$ and DMSO-d_6

Compound	$CDCl_3$			DMSO-d_6		
	t_1	t_2	t_3	t_1	t_2	t_3
1h	1 : 2.5	1 : 2.5	1 : 2.5	1 : 10	1 : 9	1 : 3
1i	1 : 2.4	1 : 2.4	1 : 2.4	3 : 7	3.5 : 7	1 : 1
1j	1 : 3	10 : 1	14 : 1	**	1 : 8	1 : 4
2c	1 : 5.5	1 : 1.7	2 : 1	1 : 6	1 : 6	1 : 5.5
2d	**	**	**	**	**	**
3c	1 : 1.5	1 : 1.5	1 : 1.5	1 : 1.7	1 : 1.7	1 : 1.6

* t_1, 120—150 s; t_2, 1 day; t_3, 1 week.
** One isomer.

sults in the thermodynamically most favorable ratio of the diastereomers for each structure in a particular solvent. It should be noted that in the present study we did not determine the relative configurations of the observed diastereomers.

In the present study, we investigated the reduction of selected representatives of heterocyclic compounds **1** and **2** (Scheme 6). It was shown that the catalytic hydrogenation of these compounds on 10% Pd/C under atmospheric pressure affords 1,2-dihydropyrrolo[1,2-*a*]pyrazin-3(4*H*)-one derivatives **15**, *i.e.*, the aminal moiety of the molecules is subjected to hydrogenolysis. The hydrogenation of compounds containing arylmethyl substituents in position 1 is accompanied by the elimination of these substituents as a result of the hydrogenolysis to give *N*-unsubstituted 2-aminoalkyl-1,2-dihydropyrrolo[1,2-*a*]pyrazin-3(4*H*)-ones **16**.

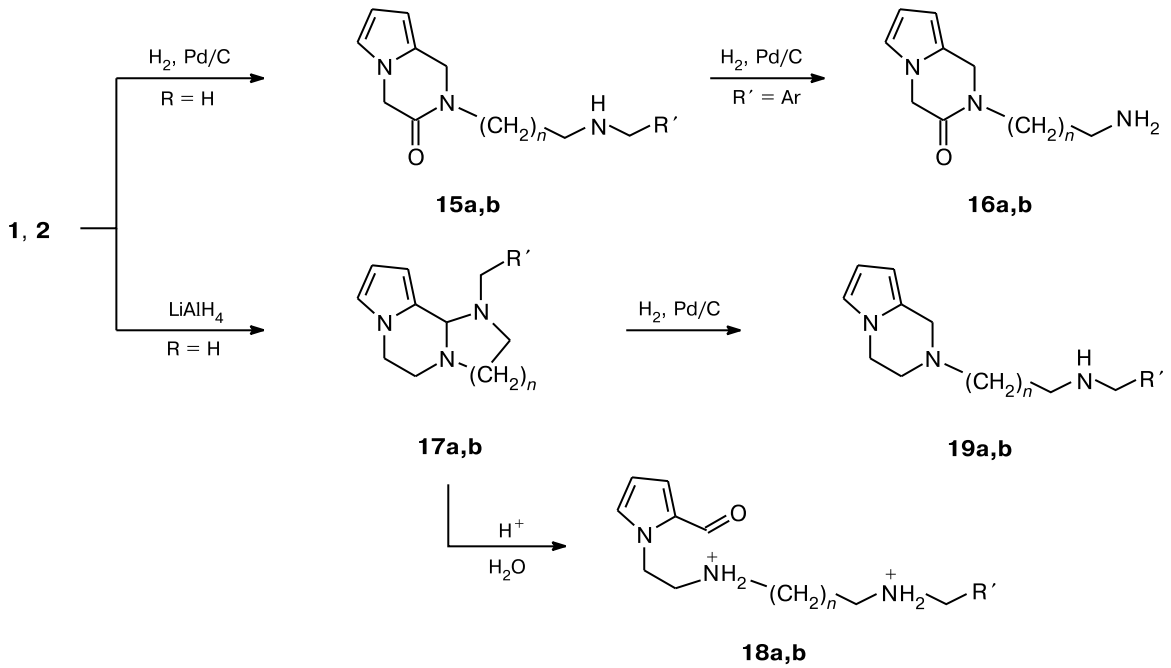
Then we investigated the reactions of compounds **1** and **2** with lithium aluminum hydride. These reactions were found to lead only to the reduction of the carbonyl group, whereas the bicyclic aminal system of compounds **17** remains intact. According to the ^{1}H NMR spectroscopic data, heterocycles **17** are hydrolyzed in an acidic medium to form, most likely, acyclic amino aldehydes **18** (Scheme 6).

Tricyclic aminals **17** readily undergo hydrogenation on the palladium catalyst to form 1,2,3,4-tetrahydropyrrolo[1,2-*a*]pyrazine derivatives **19**.

The structures of the reduction products of heterocycles **1** and **2** were confirmed by ^{1}H NMR spectroscopy and elemental analysis.

To sum up, we synthesized the previously unknown heterocyclic compounds **1—3**. The interconversion of the diastereomers of the derivatives containing two chiral centers in solution was observed. The hydrogenation of such compounds on the palladium catalyst leads to the hydrogenolysis of the aminal moiety and the opening of one diazacycloalkane moiety, whereas the treatment of these compounds with lithium aluminum hydride results in the reduction of the carbonyl group, the aminal function being intact. The results of the present study are promising for preparative organic and medical chemistry.

Scheme 6



R′ = Me, n = 1 (**a**); R′ = H, n = 2 (**b**)

Experimental

The ^{1}H NMR spectra were recorded on a Bruker AC-250 spectrometer in DMSO-d_6 and $CDCl_3$ with the use of signals of the residual protons of the solvents (δ 2.50 and 7.24, respectively) as the internal standard. The melting points were determined on a Kofler hot-stage apparatus and are uncorrected. The mass spectra were measured on a Kratos MS-300 instrument (EI, 70 eV). The course of the reactions and the purity of the reaction products were monitored by TLC using a toluene—acetone—heptane—triethylamine system (14 : 9 : 3 : 1) on Kieselgel 60 F_{254} plates with detection by UV light. Starting 2,5-dimethoxy-2-dimethoxymethyltetrahydrofuran (**5**) was synthesized according to a known procedure.[11]

Synthesis of methyl α-(2-formyl-1*H*-pyrrol-1-yl)carboxylates (4) (general procedure). 2,5-Dimethoxy-2-dimethoxymethyltetrahydrofuran (**5**) (51.56 g, 0.25 mol) was added to a solution of NaOH (11.0 g, 0.275 mol), H_3PO_4 (16 mL, 0.275 mol), and amino acid methyl ester hydrochloride **6** (0.275 mol) in water (150 mL). The reaction mixture was refluxed for 25 min. The product was extracted with toluene, the extract was washed with a solution of K_2CO_3 (12.5 g, 90 mmol) in water (60 mL) and then with water (50 mL), filtered through a paper filter, and concentrated to dryness. The residue was distilled off.

Methyl (2-formyl-1*H*-pyrrol-1-yl)acetate (4a) was synthesized by the reaction of acetal **5** with glycine methyl ester hydrochloride (**6a**). The yield was 70%, pale-pink oil, b.p. 98—101 °C (1.5 Torr), n_D^{20} = 1.5421. Found (%): C, 57.53; H, 5.28; N, 8.37. $C_8H_9NO_3$. Calculated (%): C, 57.48; H, 5.43; N, 8.38. ^{1}H NMR ($CDCl_3$), δ: 3.73 (s, 3 H, OMe); 5.02 (s, 2 H, CH_2); 6.24 (m, 1 H, H(4)); 6.90 (m, 1 H, H(3)); 6.95 (m, 1 H, H(5)); 9.48 (s, 1 H, CHO).

Methyl 2-(2-formyl-1*H*-pyrrol-1-yl)propionate (4b) was synthesized by the reaction of acetal **5** with alanine methyl ester hydrochloride (**6b**). The yield was 75%, pale-yellow oil, b.p. 105—107 °C (1.5 Torr), n_D^{20} = 1.5323. Found (%): C, 59.59; H, 6.22; N, 7.89. $C_9H_{11}NO_3$. Calculated (%): C, 59.66; H, 6.12; N, 7.73. ^{1}H NMR ($CDCl_3$), δ: 1.74 (d, 3 H, Me, ^3J = 7.2 Hz); 3.74 (s, 3 H, OMe); 5.88 (q, 1 H, CH, ^3J = 7.2 Hz); 6.30 (m, 1 H, H(4)); 6.98 (m, 1 H, H(3)); 7.17 (m, 1 H, H(5)); 9.50 (s, 1 H, CHO).

Methyl 2-(2-formyl-1*H*-pyrrol-1-yl)-3-phenylpropionate (4c) was synthesized by the reaction of acetal **5** with phenylalanine methyl ester hydrochloride (**6c**). The yield was 67%, pale-yellow crystals, m.p. 38—40 °C. Found (%): C, 69.70; H, 5.96; N, 5.30. $C_{15}H_{15}NO_3$. Calculated (%): C, 70.02; H, 5.88; N, 5.44. ^{1}H NMR ($CDCl_3$), δ: 3.29 (dd, 1 H, $\underline{H}_a(CH_b)Ph$, ^2J = 13.6 Hz, ^3J = 9.6 Hz); 3.53 (dd, 1 H, $\underline{H}_b(CH_a)Ph$, ^2J = 13.6 Hz, ^3J = 5.0 Hz); 3.74 (s, 3 H, OMe); 6.07 (m, 1 H, $C\underline{H}CO_2Me$); 6.21 (m, 1 H, H(4)); 6.98—7.27 (m, 7 H, H(3), H(5), Ph); 9.43 (s, 1 H, CHO).

Synthesis of *N*-(arylmethyl)-substituted 1,2-, 1,3-, and 1,4-diamines 7—9 (general procedure). The palladium on carbon catalyst (0.3 g, 10% Pd) was added to a solution of aromatic aldehyde (0.1 mol) and diamine (0.3 mol) in methanol (100 mL). The reaction mixture was hydrogenated until the theoretical amount of hydrogen was consumed. The catalyst was filtered off, and the filtrate was concentrated to dryness. The residue was distilled.

***N*-Benzyl-1,2-ethanediamine (7a)** was synthesized by the reaction of benzaldehyde with 1,2-ethanediamine. The yield was 78%, colorless oil, b.p. 100—102°C (1.5 Torr) (see lit. data[17]: b.p. 100 °C (0.1 Torr)), n_D^{20}= 1.5386 (see lit. data[17]: n_D^{20}= = 1.5417).

***N*-(3,4-Dimethoxybenzyl)-1,2-ethanediamine (7b)** was synthesized by the reaction of 3,4-dimethoxybenzaldehyde with 1,2-ethanediamine. The yield was 75%, colorless oil, b.p. 157—159 °C (1.5 Torr) (see lit. data[18]: 168—170 °C (4 Torr)), n_D^{20} = 1.5480.

***N*-(3,4,5-Trimethoxybenzyl)-1,2-ethanediamine (7c)** was synthesized by the reaction of 3,4,5-trimethoxybenzaldehyde with 1,2-ethanediamine. The yield was 65%, colorless oil, b.p. 171—173 °C (1.5 Torr), n_D^{20} = 1.5422. Found (%): C, 59.70; H, 8.42; N, 11.47. $C_{12}H_{20}N_2O_3$. Calculated (%): C, 59.98; H, 8.39; N, 11.66. ^{1}H NMR ($CDCl_3$), δ: 1.47 (s, 3 H, NH_2, NH); 2.66 (t, 2 H, $C\underline{H}_2NH_2$, ^3J = 5.8 Hz); 2.79 (s, 2 H, $C\underline{H}_2NH$, ^3J = 5.8 Hz); 3.70 (s, 2 H, $C\underline{H}_2Ar$); 3.78 (s, 3 H, 4-OMe); 3.82 (s, 6 H, 3-OMe, 5-OMe); 6.53 (s, 2 H, Ar).

***N*-(2-Furylmethyl)-1,2-ethanediamine (7d)** was synthesized by the reaction of 2-furylaldehyde with 1,2-ethanediamine. The yield was 81%, colorless oil, b.p. 70 °C (1 Torr) (see lit. data[23]: 68—70 °C (2 Torr)), n_D^{20} = 1.5021 (see lit. data[23]: n_D^{20} = 1.5029).

***N*-(3,4-Dimethoxybenzyl)-1,3-propanediamine (8a)** was synthesized by the reaction of 3,4-dimethoxybenzaldehyde with 1,3-propanediamine. The yield was 74%, colorless oil, b.p. 173—175 °C (2.5 Torr), n_D^{20} = 1.5418. Found (%): C, 64.53; H, 8.96; N, 12.37. $C_{12}H_{20}N_2O_2$. Calculated (%): C, 64.26; H, 8.99; N, 12.49. ^{1}H NMR ($CDCl_3$), δ: 1.52—1.69 (m, 5 H, NH_2, NH, $NH_2CH_2C\underline{H}_2$); 2.64 and 2.72 (both t, 2 H each, $C\underline{H}_2NH_2$, $C\underline{H}_2NH$, ^3J = 6.8 Hz, ^3J = 6.9 Hz); 3.68 (s, 2 H, $C\underline{H}_2Ar$); 3.81 and 3.83 (both s, 3 H each, 2 OMe); 6.73—6.85 (m, 3 H, Ar).

***N*-(3,4-Dimethoxybenzyl)-2,2-dimethyl-1,3-propanediamine (8b)** was synthesized by the reaction of 3,4-dimethoxybenzaldehyde with 2,2-dimethyl-1,3-propanediamine. The yield was 72%, colorless oil, b.p. 175—177 °C (2.5 Torr), n_D^{20} = 1.5300. Found (%): C, 66.71; H, 9.39; N, 11.07. $C_{14}H_{24}N_2O_2$. Calculated (%): C, 66.63; H, 9.59; N, 11.10. ^{1}H NMR ($CDCl_3$), δ: 0.85 (s, 6 H, 2 Me); 1.67 (s, 3 H, NH_2, NH); 2.38 (s, 2 H, $C\underline{H}_2NH$); 2.53 (s, 2 H, $C\underline{H}_2NH_2$); 3.71 (s, 2 H, $C\underline{H}_2Ar$); 3.85 and 3.87 (both s, 3 H each, 2 OMe); 6.76—6.91 (m, 3 H, Ar).

***N*-Benzyl-1,4-butanediamine (9a)** was synthesized by the reaction of benzaldehyde with 1,4-butanediamine. The yield was 73%, colorless oil, b.p. 131—133 °C (2 Torr), n_D^{20} = 1.5260. Found (%): C, 73.99; H, 10.32; N, 15.65. $C_{11}H_{18}N_2$. Calculated (%): C, 74.11; H, 10.18; N, 15.71. ^{1}H NMR ($CDCl_3$), δ: 1.21 (s, 3 H, NH_2, NH); 1.46 (m, 4 H, $C\underline{H}_2C\underline{H}_2CH_2NH_2$); 2.61 (m, 4 H, $C\underline{H}_2NH$, $C\underline{H}_2NH_2$); 3.71 (s, 2 H, $C\underline{H}_2Ph$); 7.12—7.33 (m, 5 H, Ph).

***N*-(3,4-Dimethoxybenzyl)-1,4-butanediamine (9b)** was synthesized by the reaction of 3,4-dimethoxybenzaldehyde with 1,4-butanediamine. The yield was 77%, colorless oil, b.p. 184—186 °C (2 Torr), n_D^{20} = 1.5378. Found (%): C, 65.36; H, 9.39; N, 11.80. $C_{13}H_{22}N_2O_2$. Calculated (%): C, 65.52; H, 9.30; N, 11.75. ^{1}H NMR ($CDCl_3$), δ: 1.35—1.54 (m, 7 H, NH_2, NH, $C\underline{H}_2C\underline{H}_2CH_2NH_2$); 2.59 and 2.65 (both t, 2 H each, $C\underline{H}_2NH_2$, $C\underline{H}_2NH$, ^3J = 6.9 Hz, ^3J = 6.6 Hz); 3.69 (s, 2 H, $C\underline{H}_2Ar$); 3.81 and 3.83 (both s, 3 H each, 2 OMe); 6.74—6.85 (m, 3 H, Ar).

Reaction of methyl α-(2-formyl-1*H*-pyrrol-1-yl)carboxylates (4) with 1,2- and 1,3-diamines 7 and 8 (general procedure). A solution of methyl α-(2-formyl-1*H*-pyrrol-1-yl)carboxylate (**4**) (20 mmol) and diamine **7** or **8** (22 mmol) in ethanol (30 mL) was

refluxed for 1 h. The reaction mixture was kept at −10 °C for 2 days. The precipitate that formed was filtered off, washed with cold ethanol, and recrystallized if necessary.

1-Benzyl-1,2,3,10b-tetrahydroimidazo[1,2-*a*]pyrrolo[2,1-*c*]-pyrazin-5(6*H*)-one (1a) was synthesized by the reaction of aldehyde ester **4a** with diamine **7a**. The yield was 69%, white crystals, m.p. 106—108 °C. Found (%): C, 71.60; H, 6.50; N, 15.57. $C_{16}H_{17}N_3O$. Calculated (%): C, 71.89; H, 6.41; N, 15.72. ^{1}H NMR ($CDCl_3$), δ: 2.73 (ddd, 1 H, $H_aC(2)$, $^2J_{2a,2b}$ = 10.5 Hz, $^3J_{2a,3b}$ = 11.2 Hz, $^3J_{2a,3a}$ = 8.5 Hz); 3.23 (ddd, 1 H, $H_bC(2)$, $^2J_{2b,2a}$ = 10.5 Hz, $^3J_{2b,3b}$ = 7.5 Hz, $^3J_{2b,3a}$ = 1.8 Hz); 3.56 (ddd, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 11.3 Hz, $^3J_{3a,2a}$ = 8.5 Hz, $^3J_{3a,2b}$ = 1.8 Hz); 3.60 (d, 1 H, $\underline{H}_a(CH_b)Ph$, ^2J = 13.2 Hz); 3.74 (ddd, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = 11.3 Hz, $^3J_{3b,2a}$ = 11.2 Hz, $^3J_{3b,2b}$ = 7.5 Hz); 3.99 (d, 1 H, $\underline{H}_b(CH_a)Ph$, ^2J = 13.2 Hz); 4.58 and 4.65 (both d, 1 H each, $H_aC(6)$ and $H_bC(6)$, ^2J = 17.7 Hz); 5.12 (s, 1 H, H(10b)); 6.24 (m, 1 H, H(9)); 6.28 (m, 1 H, H(10)); 6.67 (m, 1 H, H(8)); 7.22—7.38 (m, 5 H, Ph).

1-(3,4-Dimethoxybenzyl)-1,2,3,10b-tetrahydroimidazo[1,2-*a*]-pyrrolo[2,1-*c*]pyrazin-5(6*H*)-one (1b) was synthesized by the reaction of aldehyde ester **4a** with diamine **7b**. The yield was 80%, white crystals, m.p. 99—101 °C. Found (%): C, 66.33; H, 6.64; N, 13.03. $C_{18}H_{21}N_3O_3$. Calculated (%): C, 66.04; H, 6.47; N, 12.84. ^{1}H NMR ($CDCl_3$), δ: 2.72 (ddd, 1 H, $H_aC(2)$, $^2J_{2a,2b}$ = 10.9 Hz, $^3J_{2a,3b}$ = 11.3 Hz, $^3J_{2a,3a}$ = 8.4 Hz); 3.22 (ddd, 1 H, $H_bC(2)$, $^2J_{2b,2a}$ = 10.9 Hz, $^3J_{2b,3b}$ = 7.5 Hz, $^3J_{2b,3a}$ = 1.6 Hz); 3.52 (d, 1 H, $\underline{H}_a(CH_b)Ar$, ^2J = 12.3 Hz); 3.59 (ddd, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 11.6 Hz, $^3J_{3a,2a}$ = 8.4 Hz, $^3J_{3a,2b}$ = 1.6 Hz); 3.76 (ddd, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = 11.6 Hz, $^3J_{3b,2a}$ = 11.3 Hz, $^3J_{3b,2b}$ = 7.5 Hz); 3.88 and 3.89 (both s, 3 H each, 2 OMe); 3.97 (d, 1 H, $\underline{H}_b(CH_a)Ar$, ^2J = 12.3 Hz); 4.61 and 4.68 (both d, 1 H each, $H_aC(6)$ and $H_bC(6)$, ^2J = 17.8 Hz); 5.12 (s, 1 H, H(10b)); 6.30 (m, 1 H, H(9)); 6.34 (m, 1 H, H(10)); 6.70 (m, 1 H, H(8)); 6.82 and 6.88 (both d, 1 H each, $H_{Ar}(5)$ and $H_{Ar}(6)$, ^3J = 8.1 Hz); 6.92 (s, 1 H, $H_{Ar}(2)$).

1-(3,4,5-Trimethoxybenzyl)-1,2,3,10b-tetrahydroimidazo-[1,2-*a*]pyrrolo[2,1-*c*]pyrazin-5(6*H*)-one (1c) was synthesized by the reaction of aldehyde ester **4a** with diamine **7c**. The yield was 86%, white crystals, m.p. 129—131 °C. Found (%): C, 60.93; H, 6.46; N, 11.08. $C_{19}H_{23}N_3O_4 \cdot H_2O$. Calculated (%): C, 60.79; H, 6.71; N, 11.19. ^{1}H NMR ($CDCl_3$), δ: 2.72 (ddd, 1 H, $H_aC(2)$, $^2J_{2a,2b}$ = 10.9 Hz, $^3J_{2a,3b}$ = 11.3 Hz, $^3J_{2a,3a}$ = 8.7 Hz); 3.24 (ddd, 1 H, $H_bC(2)$, $^2J_{2b,2a}$ = 10.9 Hz, $^3J_{2b,3b}$ = 7.5 Hz, $^3J_{2b,3a}$ = 1.7 Hz); 3.49 (d, 1 H, $\underline{H}_a(CH_b)Ar$, ^2J = 13.3 Hz); 3.56 (ddd, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 11.6 Hz, $^3J_{3a,2a}$ = 8.7 Hz, $^3J_{3a,2b}$ = 1.7 Hz); 3.75 (ddd, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = 11.6 Hz, $^3J_{3b,2a}$ = 11.3 Hz, $^3J_{3b,2b}$ = 7.5 Hz); 3.82 (s, 3 H, 4-OMe); 3.84 (s, 6 H, 3-OMe, 5-OMe); 3.92 (d, 1 H, $\underline{H}_b(CH_a)Ar$, ^2J = 13.3 Hz); 4.58 and 4.66 (both d, 1 H each, $H_aC(6)$ and $H_bC(6)$, ^2J = 17.5 Hz); 5.11 (s, 1 H, H(10b)); 6.27 (m, 2 H, H(9), H(10)); 6.58 (s, 2 H, Ar); 6.67 (m, 1 H, H(8)).

1-(2-Furylmethyl)-1,2,3,10b-tetrahydroimidazo[1,2-*a*]pyrrolo[2,1-*c*]pyrazin-5(6*H*)-one (1d) was synthesized by the reaction of aldehyde ester **4a** with diamine **7d**. The yield was 76%, pale-yellow crystals, m.p. 100—102 °C. Found (%): C, 65.46; H, 5.96; N, 16.02. $C_{14}H_{15}N_3O_2$. Calculated (%): C, 65.36; H, 5.88; N, 16.33. ^{1}H NMR ($CDCl_3$), δ: 2.94 (ddd, 1 H, $H_aC(2)$, $^2J_{2a,2b}$ = 11.0 Hz, $^3J_{2a,3b}$ = 11.2 Hz, $^3J_{2a,3a}$ = 8.5 Hz); 3.35 (ddd, 1 H, $H_bC(2)$, $^2J_{2b,2a}$ = 11.0 Hz, $^3J_{2b,3b}$ = 7.8 Hz, $^3J_{2b,3a}$ = 2.0 Hz); 3.52 (ddd, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 11.4 Hz, $^3J_{3a,2a}$ = 8.5 Hz, $^3J_{3a,2b}$ = 2.0 Hz); 3.77 (ddd, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = 11.4 Hz, $^3J_{3b,2a}$ = 11.2 Hz, $^3J_{3b,2b}$ = 7.8 Hz); 3.81 (d, 1 H, $\underline{H}_a(CH_b)Fur$, ^2J = 14.4 Hz); 3.90 (d, 1 H, $\underline{H}_b(CH_a)Fur$, ^2J = 14.4 Hz); 4.56 and 4.63 (both d, 1 H each, $H_aC(6)$ and $H_bC(6)$, ^2J = 17.2 Hz); 5.13 (s, 1 H, H(10b)); 6.20 (m, 1 H, $H_{Fur}(3)$); 6.30 (m, 1 H, $H_{Fur}(4)$); 6.32 (m, 1 H, H(9)); 6.36 (m, 1 H, H(10)); 6.67 (m, 1 H, H(8)); 7.39 (m, 1 H, $H_{Fur}(5)$).

1-Ethyl-1,2,3,10b-tetrahydroimidazo[1,2-*a*]pyrrolo[2,1-*c*]-pyrazin-5(6*H*)-one (1e) was synthesized by the reaction of aldehyde ester **4a** with *N*-ethyl-1,2-ethanediamine. The yield was 86%, white crystals, m.p. 123—124 °C (recrystallization from toluene). Found (%): C, 63.77; H, 7.61; N, 20.80. $C_{11}H_{15}N_3O$. Calculated (%): C, 64.37; H, 7.37; N, 20.47. ^{1}H NMR ($CDCl_3$), δ: 1.20 (t, 3 H, Me, ^3J = 7.2 Hz); 2.45 (dq, 1 H, $\underline{H}_a(CH_b)Me$, ^2J = 11.9 Hz, ^3J = 7.2 Hz); 2.62 (ddd, 1 H, $H_aC(2)$, $^2J_{2a,2b}$ = = 10.0 Hz, $^3J_{2a,3b}$ = 9.8 Hz, $^3J_{2a,3a}$ = 8.4 Hz); 3.07 (dq, 1 H, $\underline{H}_b(CH_a)Me$, ^2J = 11.9 Hz, ^3J = 7.2 Hz); 3.49 (ddd, 1 H, $H_bC(2)$, $^2J_{2b,2a}$ = 10.0 Hz, $^3J_{2b,3b}$ = 7.7 Hz, $^3J_{2b,3a}$ = 1.9 Hz); 3.60 (ddd, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 11.0 Hz, $^3J_{3a,2a}$ = 8.4 Hz, $^3J_{3a,2b}$ = 1.9 Hz); 3.77 (ddd, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = 11.0 Hz, $^3J_{3b,2a}$ = 9.8 Hz, $^3J_{3b,2b}$ = 7.7 Hz); 4.53 and 4.61 (both d, 1 H each, $H_aC(6)$ and $H_bC(6)$, ^2J = 17.6 Hz); 4.86 (s, 1 H, H(10b)); 6.20—6.26 (m, 2 H, H(9), H(10)); 6.62 (m, 1 H, H(8)).

1-(2-Hydroxyethyl)-1,2,3,10b-tetrahydroimidazo[1,2-*a*]-pyrrolo[2,1-*c*]pyrazin-5(6*H*)-one (1f) was synthesized by the reaction of aldehyde ester **4a** with *N*-(2-hydroxyethyl)-1,2-ethanediamine. The yield was 73%, white crystals, m.p. 82—84 °C. MS, *m/z*: 221 $[M]^+$, 190 $[M - CH_2OH]^+$, 176 $[M - CH_2CH_2OH]^+$. Found (%): C, 59.76; H, 6.76; N, 19.09. $C_{11}H_{15}N_3O_2$. Calculated (%): C, 59.71; H, 6.83; N, 18.99. ^{1}H NMR ($CDCl_3$), δ: 2.47 (s, 1 H, OH); 2.64 (m, 1 H, $\underline{H}_a(CH_b)CH_2OH$); 2.81 (ddd, 1 H, $H_aC(2)$, $^2J_{2a,2b}$ = 10.5 Hz, $^3J_{2a,3b}$ = 11.2 Hz, $^3J_{2a,3a}$ = 8.5 Hz); 3.02 (m, 1 H, $\underline{H}_b(CH_a)CH_2OH$); 3.51 (ddd, 1 H, $H_bC(2)$, $^2J_{2b,2a}$ = 10.5 Hz, $^3J_{2b,3b}$ = 7.7 Hz, $^3J_{2b,3a}$ = 1.8 Hz); 3.61 (ddd, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 11.4 Hz, $^3J_{3a,2a}$ = 8.5 Hz, $^3J_{3a,2b}$ = 1.8 Hz); 3.69—3.82 (m, 3 H, $H_bC(3)$, $C\underline{H}_2OH$); 4.54 and 4.61 (both d, 1 H each, $H_aC(6)$ and $H_bC(6)$, ^2J = 17.5 Hz); 5.07 (s, 1 H, H(10b)); 6.25 (m, 2 H, H(9), H(10)); 6.64 (m, 1 H, H(8)).

1-(2-Aminoethyl)-1,2,3,10b-tetrahydroimidazo[1,2-*a*]pyrrolo[2,1-*c*]pyrazin-5(6*H*)-one (1g) was synthesized by the reaction of aldehyde ester **4a** with *N*-(2-aminoethyl)-1,2-ethanediamine. The yield was 76%, white crystals, m.p. 67—69 °C. MS, *m/z*: 220 $[M]^+$, 190 $[M - CH_2NH_2]^+$, 176 $[M - CH_2CH_2NH_2]^+$. Found (%): C, 59.89; H, 7.31; N, 25.59. $C_{11}H_{16}N_4O$. Calculated (%): C, 59.98; H, 7.32; N, 25.44. ^{1}H NMR ($CDCl_3$), δ: 1.70 (s, 2 H, NH_2); 2.50 (m, 1 H, $\underline{H}_a(CH_b)CH_2NH_2$); 2.72 (ddd, 1 H, $H_aC(2)$, $^2J_{2a,2b}$ = 10.4 Hz, $^3J_{2a,3b}$ = 10.9 Hz, $^3J_{2a,3a}$ = = 8.3 Hz); 2.84—2.98 (m, 3 H, $\underline{H}_b(CH_a)$ CH_2NH_2, $C\underline{H}_2NH_2$); 3.45 (ddd, 1 H, $H_bC(2)$, $^2J_{2b,2a}$ = 10.4 Hz, $^3J_{2b,3b}$ = 7.7 Hz, $^3J_{2b,3a}$ = 2.0 Hz); 3.58 (ddd, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 11.4 Hz, $^3J_{3a,2a}$ = 8.3 Hz, $^3J_{3a,2b}$ = 2.0 Hz); 3.76 (ddd, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = 11.4 Hz, $^3J_{3b,2a}$ = 10.9 Hz, $^3J_{3b,2b}$ = 7.7 Hz); 4.54 and 4.61 (both d, 1 H each, $H_aC(6)$ and $H_bC(6)$, ^2J = 17.5 Hz); 4.98 (s, 1 H, H(10b)); 6.25 (m, 2 H, H(9), H(10)); 6.63 (m, 1 H, H(8)).

1-Benzyl-6-methyl-1,2,3,10b-tetrahydroimidazo[1,2-*a*]pyrrolo[2,1-*c*]pyrazin-5(6*H*)-one (1h). The reaction of aldehyde ester **4b** with diamine **7a** afforded compound **1h** as a mixture of diastereomers (see Table 1). The yield was 75%, white powder, m.p. 93—95 °C. Found (%): C, 72.27; H, 6.73; N, 14.96. $C_{17}H_{19}N_3O$. Calculated (%): C, 72.57; H, 6.81; N, 14.93. ^{1}H NMR ($CDCl_3$), δ (the signals of the minor diastereomer are given in italic type): 1.63, *1.82* (d, 3 H, Me, ^3J = 7.1 Hz);

2.72 (ddd, 1 H, $H_aC(2)$, $^2J_{2a,2b}$ = 10.7 Hz, $^3J_{2a,3b}$ = 11.2 Hz, $^3J_{2a,3a}$ = 8.5 Hz); 3.23 (ddd, 1 H, $H_bC(2)$, $^2J_{2b,2a}$ = 10.7 Hz, $^3J_{2b,3b}$ =7.5 Hz, $^3J_{2b,3a}$ = 1.7 Hz); 3.56 (ddd, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 11.4 Hz, $^3J_{3a,2a}$ = 8.5 Hz, $^3J_{3a,2b}$ = 1.7 Hz); 3.59 (d, 1 H, $\underline{H}_a(CH_b)Ph$, ^2J = 13.1 Hz); 3.70 (ddd, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = = 11.4 Hz, $^3J_{3b,2a}$ = 11.2 Hz, $^3J_{3b,2b}$ = 7.5 Hz); 4.06 (d, 1 H, $\underline{H}_b(CH_a)Ph$, ^2J = 13.1 Hz); *4.57*, 4.72 (q, 1 H, C<u>H</u>Me, ^3J = 7.1 Hz); *5.11*, 5.13 (s, 1 H, H(10b)); 6.29 (m, 1 H, H(9)); 6.32 (m, 1 H, H(10)); 6.69, *6.78* (m, 1 H, H(8)); 7.24—7.42 (m, 5 H, Ph).

6-Methyl-1-(3,4,5-trimethoxybenzyl)-1,2,3,10b-tetrahydroimidazo[1,2-*a*]pyrrolo[2,1-*c*]pyrazin-5(6*H*)-one (1i). The reaction of aldehyde ester **4b** with diamine **7c** afforded compound **1i** as a mixture of diastereomers (see Table 1). The yield was 70%, white powder, m.p. 63—65 °C. Found (%): C, 64.37; H, 6.61; N, 11.11. $C_{20}H_{25}N_3O_4$. Calculated (%): C, 64.67; H, 6.78; N, 11.31. ^{1}H NMR ($CDCl_3$), δ (the signals of the minor diastereomer are given in italic type): 1.61, *1.80* (d, 3 H, Me, ^3J = 7.1 Hz); 2.70 (ddd, 1 H, $H_aC(2)$, $^2J_{2a,2b}$ = 10.5 Hz, $^3J_{2a,3b}$ = 11.1 Hz, $^3J_{2a,3a}$ = 8.5 Hz); 3.24 (ddd, 1 H, $H_bC(2)$, $^2J_{2b,2a}$ = 10.5 Hz, $^3J_{2b,3b}$ = 7.6 Hz, $^3J_{2b,3a}$ = 1.7 Hz); 3.48 (d, 1 H, $\underline{H}_a(CH_b)Ph$, ^2J = 13.1 Hz); 3.56 (ddd, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 11.4 Hz, $^3J_{3a,2a}$ = 8.5 Hz, $^3J_{3a,2b}$ = 1.7 Hz); 3.71 (ddd, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = 11.4 Hz, $^3J_{3b,2a}$ = 11.1 Hz, $^3J_{3b,2b}$ = 7.6 Hz); 3.82 (s, 3 H, 4-OMe); 3.84 (s, 6 H, 3-OMe, 5-OMe); 3.97 (d, 1 H, $\underline{H}_b(CH_a)Ph$, ^2J = 13.1 Hz); *4.57*, 4.71 (q, 1 H, C<u>H</u>Me, ^3J = 7.1 Hz); *5.07*, 5.10 (s, 1 H, H(10b)); 6.27 (m, 2 H, H(9), H(10)); 6.68 (s, 2 H, Ar); 6.68, *6.76* (m, 1 H, H(8)).

6-Benzyl-1-(3,4-dimethoxybenzyl)-1,2,3,10b-tetrahydroimidazo[1,2-*a*]pyrrolo[2,1-*c*]pyrazin-5(6*H*)-one (1j). The reaction of aldehyde ester **4c** with diamine **7b** afforded compound **1j** as a mixture of diastereomers (see Table 1). The yield was 72%, white crystals, m.p. 72—74 °C. Found (%): C, 71.95; H, 6.32; N, 10.15. $C_{25}H_{27}N_3O_3$. Calculated (%): C, 71.92; H, 6.52; N, 10.06. MS, *m/z*: 417 $[M]^+$, 326 $[M - CH_2Ph]^+$, 266 $[M - CH_2Ph(OCH_3)_2\text{-}3,4]^+$, 175 $[M - CH_2Ph - CH_2Ph(OCH_3)_2\text{-}3,4]^+$. The ^{1}H NMR spectrum of the first diastereomer of **1j**, which was obtained by subtraction of the spectrum recorded within one week after the dissolution of crystalline compound **1j** in $CDCl_3$ from the spectrum recorded immediately after the dissolution ($CDCl_3$), δ: 1.79 (d, H, $\underline{H}_a(CH_b)Ar$, ^2J = 13.3 Hz); 2.77 (ddd, 1 H, $H_aC(2)$, $^2J_{2a,2b}$ = 12.0 Hz, $^3J_{2a,3b}$ = 11.4 Hz, $^3J_{2a,3a}$ = 8.7 Hz); 2.89 (ddd, 1 H, $H_bC(2)$, $^2J_{2b,2a}$ = 12.0 Hz, $^3J_{2b,3b}$ =8.0 Hz, $^3J_{2b,3a}$ = 2.1 Hz); 2.99 (d, 1 H, $\underline{H}_b(CH_a)Ar$, ^2J = 13.3 Hz); 3.33 (ddd, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 11.5 Hz, $^3J_{3a,2a}$ = 8.7 Hz, $^3J_{3a,2b}$ = 2.1 Hz); 3.47 (dd, 1 H, $\underline{H}_a(CH_b)Ph$, ^2J = 14.0 Hz, ^3J = 4.2 Hz); 3.55 (ddd, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = = 11.5 Hz, $^3J_{3b,2a}$ = 11.4 Hz, $^3J_{3b,2b}$ = 8.0 Hz); 3.68 (dd, $\underline{H}_b(CH_a)Ph$, ^2J = 14.0 Hz, ^3J = 4.2 Hz); 3.84 and 3.86 (both s, 3 H each, 2 OMe); 5.04 (t, 1 H, C<u>H</u>CH_2Ph, ^3J = 4.2 Hz); 5.20 (s, 1 H, H(10b)); 6.21 (m, 1 H, H(10)); 6.39 (m, 1 H, H(9)); 6.54 (d, 1 H, $H_{Ar}(5)$, ^3J = 8.1 Hz); 6.67 (m, 1 H, H(8)); 6.74 (d, 1 H, $H_{Ar}(6)$, ^3J = 8.1 Hz); 6.90—7.11 (m, 6 H, Ph, $H_{Ar}(2)$).

The ^{1}H NMR spectrum of the second diastereomer of **1j**, which was obtained by subtraction of the spectrum recorded immediately after the dissolution of crystalline compound **1j** in $CDCl_3$ from the spectrum recorded within one week after the dissolution ($CDCl_3$), δ: 2.39 (ddd, 1 H, $H_aC(2)$, $^2J_{2a,2b}$ = 10.9 Hz, $^3J_{2a,3b}$ = 11.1 Hz, $^3J_{2a,3a}$ = 8.5 Hz); 3.05 (ddd, 1 H, $H_bC(2)$, $^2J_{2b,2a}$ = 10.9 Hz, $^3J_{2b,3b}$ =7.6 Hz, $^3J_{2b,3a}$ = 1.9 Hz); 3.22—3.39 (m, 4 H, C<u>H</u>$_2$Ph, $\underline{H}_a(CH_b)Ar$, $H^aC(3)$); 3.59 (ddd, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = 11.4 Hz, $^3J_{3b,2a}$ = 11.1 Hz, $^3J_{3b,2b}$ =7.6 Hz); 3.67 (d, 1 H, $\underline{H}_b(CH_a)Ar$, ^2J = 13.2 Hz); 3.70 (s, 1 H, H(10b)); 3.84 and 3.85 (both s, 3 H each, 2 OMe); 4.95 (t, 1 H, C<u>H</u>CH_2Ph, ^3J = 4.5 Hz); 6.14 (m, 1 H, H(10)); 6.28 (m, 1 H, H(9)); 6.64 (m, 1 H, H(8)); 6.71—6.86 (m, 5 H, Ar, $H_{Ph}(2)$, $H_{Ph}(6)$); 7.09—7.26 (m, 3 H, $H_{Ph}(3)$, $H_{Ph}(4)$, $H_{Ph}(5)$).

1-Methyl-1,3,4,11b-tetrahydro-2*H*-pyrrolo[2´,1´:3,4]pyrazino[1,2-*a*]pyrimidin-6(7*H*)-one (2a) was synthesized by the reaction of aldehyde ester **4a** with *N*-methyl-1,3-propanediamine. The yield was 93%, white powder, m.p. 58—60 °C. Found (%): C, 64.28; H, 7.57; N, 20.65. $C_{11}H_{15}N_3O$. Calculated (%): C, 64.37; H, 7.37; N, 20.47. ^{1}H NMR ($CDCl_3$), δ: 1.40 (dm, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 13.7 Hz); 2.04 (m, 1 H, $H_bC(3)$); 2.10 (s, 3 H, Me); 2.81 (ddd, 1 H, $H_aC(4)$, $^2J_{4a,4b}$ = $^3J_{4a,3b}$ = 13.1 Hz, $^3J_{4a,3a}$ = 3.3 Hz); 2.93—3.07 (m, 2 H, $H_2C(2)$); 4.57 and 4.65 (both d, 1 H each, $H_aC(7)$ and $H_bC(7)$, ^2J = 18.1 Hz); 4.78 (ddm, 1 H, $H_bC(4)$, $^2J_{4b,4a}$ = 13.1 Hz, $^3J_{4b,3b}$ = 4.9 Hz); 5.24 (s, 1 H, H(11b)); 6.10 (m, 1 H, H(11)); 6.22 (m, 1 H, H(10)); 6.55 (m, 1 H, H(9)).

1-(3,4-Dimethoxybenzyl)-1,3,4,11b-tetrahydro-2*H*-pyrrolo[2´,1´:3,4]-pyrazino[1,2-*a*]pyrimidin-6(7*H*)-one (2b) was synthesized by the reaction of aldehyde ester **4a** with diamine **8a**. The yield was 82%, white crystals, m.p. 108—109 °C. Found (%): C, 66.74; H, 6.80; N, 11.97. $C_{19}H_{23}N_3O_3$. Calculated (%): C, 66.84; H, 6.79; N, 12.31. ^{1}H NMR ($CDCl_3$), δ: 1.34 (d, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 13.5 Hz); 2.10 (ddm, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = = 13.5 Hz, $^3J_{3b,4a}$ = 13.2 Hz); 2.93 (ddd, 1 H, $H_aC(4)$, $^2J_{4a,4b}$ = = $^3J_{4a,3b}$ = 13.2 Hz, $^3J_{4a,3a}$ = 3.3 Hz); 2.95—3.02 (m, 2 H, $H_2C(2)$); 3.30 (d, 1 H, $\underline{H}_a(CH_b)Ar$, ^2J = 12.7 Hz); 3.47 (d, 1 H, $\underline{H}_b(CH_a)Ar$, ^2J = 12.7 Hz); 3.83 and 3.84 (both, 3 H each, 2 OMe); 4.65 and 4.74 (both d, 1 H each, $H_aC(7)$ and $H_bC(7)$, ^2J = 18.2 Hz); 4.90 (dd, 1 H, $H_bC(4)$, $^2J_{4b,4a}$ = 13.2 Hz, $^3J_{4b,3b}$ = = 4.5 Hz); 5.68 (s, 1 H, H(11b)); 6.27 (m, 2 H, H(10), H(11)); 6.60 (m, 1 H, H(9)); 6.67—6.79 (m, 3 H, Ar).

1-(3,4-Dimethoxybenzyl)-7-methyl-1,3,4,11b-tetrahydro-2*H*-pyrrolo[2´,1´:3,4]pyrazino[1,2-*a*]pyrimidin-6(7*H*)-one (2c). The reaction of aldehyde ester **4b** with diamine **8a** afforded compound **2c** as a mixture of diastereomers (see Table 1). The yield was 80%, white crystals, m.p. 71—73 °C. Found (%): C, 67.60; H, 7.38; N, 11.68. $C_{20}H_{25}N_3O_3$. Calculated (%): C, 67.58; H, 7.09; N, 11.82. MS, *m/z*: 355 $[M]^+$, 340 $[M - Me]^+$, 204 $[M - CH_2Ph(OMe)_2\text{-}3,4]^+$. The ^{1}H NMR spectrum of the first diastereomer of **2c**, which was obtained by subtraction of the spectrum recorded within one week after the dissolution of crystalline compound **2c** in $CDCl_3$ from the spectrum recorded immediately after the dissolution ($CDCl_3$), δ: 1.34 (d, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 13.5 Hz); 1.77 (d, 3 H, Me, ^3J = 6.9 Hz); 2.10 (ddm, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = 13.5 Hz, $^3J_{3b,4a}$ = 13.2 Hz); 2.93 (ddd, 1 H, $H_aC(4)$, $^2J_{4a,4b}$ = $^3J_{4a,3b}$ = 13.2 Hz, $^3J_{4a,3a}$ = 3.5 Hz); 2.96—3.03 (m, 2 H, $H_2C(2)$); 3.25 (d, 1 H, $\underline{H}_a(CH_b)Ar$, ^2J = 12.8 Hz); 3.42 (d, 1 H, $\underline{H}_b(CH_a)Ar$, ^2J = 12.8 Hz); 3.83 and 3.84 (both s, 3 H each, 2 OMe); 4.71 (q, 1 H, C<u>H</u>Me, ^3J = 6.9 Hz); 4.87 (dd, 1 H, $H_bC(4)$, $^2J_{4b,4a}$ = 13.2 Hz, $^3J_{4b,3b}$ = 4.4 Hz); 5.70 (s, 1 H, H(11b)); 6.28 (m, 2 H, H(10), H(11)); 6.69 (m, 1 H, H(9)); 6.71—6.78 (m, 3 H, Ar).

The ^{1}H NMR spectrum of the second diastereomer of **2c**, which was obtained by subtraction of the spectrum recorded immediately after the dissolution of crystalline compound **2c** in $CDCl_3$ from the spectrum recorded within one week after the dissolution ($CDCl_3$), δ: 1.41 (d, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 13.3 Hz); 1.79 (d, 3 H, Me, ^3J = 6.9 Hz); 2.04 (ddddd, 1 H, $H_bC(3)$, $^2J_{3b,3a}$ = 13.3 Hz, $^3J_{3b,4a}$ = $^3J_{3b,2a}$ = 13.1 Hz, $^3J_{3b,4b}$ = $^3J_{3b,2b}$ =

= 4.6 Hz); 2.79 (dd, 1 H, $H_aC(2)$, $^2J_{2a,2b} = {}^3J_{2a,3b} = 13.1$ Hz); 2.90 (ddd, 1 H, $H_aC(4)$, $^2J_{4a,4b} = {}^3J_{4a,3b} = 13.1$ Hz, $^3J_{4a,3a} = 2.9$ Hz); 2.96 (d, 1 H, $H_bC(2)$, $^2J_{2b,2a} = 13.1$ Hz); 3.28 (d, 1 H, $\underline{H}_a(CH_b)Ar$, $^2J = 12.7$ Hz); 3.61 (d, 1 H, $\underline{H}_b(CH_a)Ar$, $^2J = 12.7$ Hz); 3.84 (s, 6 H, 2 OMe); 4.74 (q, 1 H, $C\underline{H}Me$, $^3J = 6.9$ Hz); 4.85 (dd, 1 H, $H_bC(4)$, $^2J_{4b,4a} = 13.1$ Hz, $^3J_{4b,3b} = 4.6$ Hz); 5.51 (s, 1 H, H(11b)); 6.25 (m, 1 H, H(11)); 6.30 (m, 1 H, H(10)); 6.67 (m, 1 H, H(9)); 6.70—6.80 (m, 3 H, Ar).

7-Benzyl-1-(3,4-dimethoxybenzyl)-1,3,4,11b-tetrahydro-2*H*-pyrrolo[2´,1´:3,4]pyrazino[1,2-*a*]pyrimidin-6(7*H*)-one (2d) was synthesized by the reaction of aldehyde ester **4c** with diamine **8a**. The yield was 87%, white crystals, m.p. 110—112 °C. Found (%): C, 71.97; H, 6.85; N, 10.03. $C_{26}H_{29}N_3O_3$. Calculated (%): C, 72.37; H, 6.77; N, 9.74. ^{1}H NMR ($CDCl_3$), δ: 1.18 (d, 1 H, $H_aC(3)$, $^2J_{3a,3b} = 13.2$); 2.04 (ddddd, 1 H, $H_bC(3)$, $^2J_{3b,3a} = {}^3J_{3b,2a} = {}^3J_{3b,4a} = 13.2$ Hz, $^3J_{3b,2b} = {}^3J_{3b,4b} = 4.9$ Hz); 2.43 (ddd, 1 H, $H^aC(4)$, $^2J_{4a,4b} = {}^3J_{4a,3b} = 13.2$ Hz, $^3J_{4a,3a} = 3.8$ Hz); 2.68 (dd, 1 H, $H^aC(2)$, $^2J_{2a,2b} = 13.8$ Hz, $^3J_{2a,3b} = 13.2$ Hz); 2.82 (d, 1 H, $H_bC(2)$, $^2J_{2b,2a} = 13.8$ Hz); 3.20 (d, 1 H, $\underline{H}_a(CH_b)Ar$, $^2J = 13.4$ Hz); 3.22 (dd, 1 H, $\underline{H}_a(CH_b)Ph$, $^2J = 13.4$ Hz, $^3J = 3.4$ Hz); 3.34 (d, 1 H, $\underline{H}_b(CH_a)Ar$, $^2J = 13.4$ Hz); 3.44 (dd, $\underline{H}_b(CH_a)Ph$, $^2J = 13.4$ Hz, $^3J = 3.4$ Hz); 3.81 and 3.83 (both s, 3 H each, 2 OMe); 4.56 (s, 1 H, H(11b)); 4.71 (dd, 1 H, $H_bC(4)$, $^2J_{4b,4a} =$ $= 13.2$ Hz, $^3J_{4b,3b} = 4.9$ Hz); 4.74 (t, 1 H, $C\underline{H}CH_2Ph$, $^3J = 3.4$ Hz); 6.06 (m, 1 H, H(11)); 6.32 (m, 1 H, H(10)); 6.57—6.76 (m, 6 H, H(9), Ar, $H_{Ph}(2)$, $H_{Ph}(6)$); 7.08—7.25 (m, 3 H, $H_{Ph}(3)$, $H_{Ph}(4)$, $H_{Ph}(5)$).

3,3-Dimethyl-1-(3,4-dimethoxybenzyl)-1,3,4,11b-tetrahydro-2*H*-pyrrolo[2´,1´:3,4]pyrazino[1,2-*a*]pyrimidin-6(7*H*)-one (2e) was synthesized by the reaction of aldehyde ester **4a** with diamine **8b**. The yield was 88%, white crystals, m.p. 64—66 °C. Found (%): C, 66.47; H, 7.32; N, 11.05. $C_{21}H_{27}N_3O_3 \cdot 1/2H_2O$. Calculated (%): C, 66.64; H, 7.46; N, 11.10. ^{1}H NMR ($CDCl_3$), δ: 0.85 and 1.07 (both s, 3 H each, 2 Me); 2.09 (d, 1 H, $H_aC(2)$, $^2J_{2a,2b} = 12.0$ Hz); 2.61 (d, 1 H, $H_aC(4)$, $^2J_{4a,4b} = 12.6$ Hz); 2.63 (d, 1 H, $H_bC(2)$, $^2J_{2b,2a} = 12.0$ Hz); 3.03 (d, 1 H, $\underline{H}_a(CH_b)Ar$, $^2J = 13.1$ Hz); 3.80 and 3.83 (both s, 3 H each, 2 OMe); 4.12 (d, 1 H, $\underline{H}_b(CH_a)Ar$, $^2J = 13.1$ Hz); 4.51 (d, 1 H, $H_bC(4)$, $^2J_{4b,4a} = 12.6$ Hz); 4.67 and 4.79 (both d, 1 H each, $H_aC(7)$ and $H_bC(7)$, $^2J = 17.2$ Hz); 4.86 (s, 1 H, H(11b)); 6.19 (m, 2 H, H(10), H(11)); 6.63 (m, 1 H, H(9)); 6.68—6.77 (m, 3 H, Ar).

Reaction of methyl α-(2-formyl-1*H*-pyrrol-1-yl)carboxylates (4) with *N*-substituted 1,4-diamines (9) (general procedure). A solution of methyl α-(2-formyl-1*H*-pyrrol-1-yl)carboxylate (**4**) (10 mmol) and diamine **9** (11 mmol) in xylene (30 mL) was refluxed for 7 h. The reaction mixture was concentrated to dryness, and the residue was refluxed with petroleum ether (b.p. 80—100 °C, 60 mL; 150 mL in the synthesis of **3b**). The hot solution was decanted and kept at room temperature for one day. The oil that formed was dissolved in propan-2-ol (5 mL), and the solution was kept at −10 °C overnight. The precipitate that formed was filtered off and recrystallized from propan-2-ol.

1-Benzyl-1,2,3,4,5,12b-hexahydropyrrolo[2´,1´:3,4]pyrazino[1,2-*a*][1,3]diazepin-7(8*H*)-one (3a) was synthesized by the reaction of aldehyde ester **4a** with diamine **9a**. The yield was 83%, white crystals, m.p. 75—77 °C. Found (%): C, 72.97; H, 7.28; N, 14.03. $C_{18}H_{21}N_3O$. Calculated (%): C, 73.19; H, 7.17; N, 14.23. ^{1}H NMR ($CDCl_3$), δ: 1.62—1.98 (m, 4 H, $H_2C(3)$, $H_2C(4)$); 2.80—3.04 (m, 2 H, $H_2C(2)$); 3.40 (m, 1 H, $H_aC(5)$); 3.62 and 3.72 (both d, 1 H each, $C\underline{H}_2Ph$, $^2J = 13.6$ Hz); 4.16 (m, 1 H, $H_bC(5)$); 4.59 and 4.68 (both d, 1 H each, $H_2C(8)$, $^2J = 17.4$ Hz); 5.61 (s, 1 H, H(12b)); 6.24 (m, 2 H, H(11), H(12)); 6.57 (m, 1 H, H(10)); 7.16—7.35 (m, 5 H, Ph).

1-(3,4-Dimethoxybenzyl)-1,2,3,4,5,12b-hexahydropyrrolo[2´,1´:3,4]pyrazino[1,2-*a*][1,3]diazepin-7(8*H*)-one (3b) was synthesized by the reaction of aldehyde ester **4a** with diamine **9b**. The yield was 85%, white crystals, m.p. 100—102 °C. Found (%): C, 64.12; H, 7.27; N, 11.30. $C_{20}H_{25}N_3O_3 \cdot H_2O$. Calculated (%): C, 64.32; H, 7.29; N, 11.25. ^{1}H NMR ($CDCl_3$), δ: 1.63—2.02 (m, 4 H, $H_2C(3)$, $H_2C(4)$); 2.79—3.07 (m, 2 H, $H_2C(2)$); 3.39 (m, 1 H, $H_aC(5)$); 3.56 and 3.67 (both d, 1 H each, $C\underline{H}_2Ar$, $^2J = 13.4$ Hz); 3.83 (s, 6 H, 2 OMe); 4.14 (m, 1 H, $H_bC(5)$); 4.58 and 4.66 (both d, 1 H each, $H_2C(8)$, $^2J = 17.4$ Hz); 5.60 (s, 1 H, H(12b)); 6.23 (m, 2 H, H(11), H(12)); 6.57 (m, 1 H, H(10)); 6.74—6.80 (m, 3 H, Ar).

1-Benzyl-8-methyl-1,2,3,4,5,12b-hexahydropyrrolo[2´,1´:3,4]pyrazino[1,2-*a*][1,3]diazepin-7(8*H*)-one (3c). The reaction of aldehyde ester **4b** with diamine **9a** afforded compound **3c** as a mixture of diastereomers (see Table 1). The yield was 78%, pale-yellow crystals, m.p. 91—93 °C. Found (%): C, 73.88; H, 7.43; N, 13.55. $C_{19}H_{23}N_3O$. Calculated (%): C, 73.76; H, 7.49; N, 13.58. ^{1}H NMR ($CDCl_3$), δ (the signals of the minor diastereomer are given in italic type): 1.55—1.92 (m), 1.66, *1.79* (d) (7 H, $H_2C(3)$, $H_2C(4)$, Me, $^3J = 7.0$ Hz); 2.79—3.02 (m, 2 H, $H_2C(2)$); 3.38 (m, 1 H, $H_aC(5)$); *3.62* and *3.70*, 3.78 and 3.93 (both d, 1 H each, $C\underline{H}_2Ph$, $^2J = 13.5$, 13.8 Hz); 4.16 (m, 1 H, $H_bC(5)$); *4.67*, 4.71 (q, 1 H, HC(8), $^3J = 7.0$ Hz); *5.54*, 5.56 (s, 1 H, H(12b)); 6.24 (m, 2 H, H(11), H(12)); 6.65 (m, 1 H, H(10)); 7.22—7.34 (m, 5 H, Ph).

Catalytic hydrogenation of pyrrolo[2,1-*c*]-1,3-diazacycloalkano[1,2-*a*]pyrazinones 1 and 2 (general procedure). The 10% Pd/C catalyst (0.4 g) was added to a solution of pyrrolo[2,1-*c*]-1,3-diazacycloalkano[1,2-*a*]pyrazinone **1** or **2** (10 mmol) in ethanol (40 mL). The hydrogenation of the reaction mixture was performed until the theoretical amount of hydrogen was consumed. Then the catalyst was filtered off, and the filtrate was concentrated to dryness.

2-(2-Ethylaminoethyl)-1,2-dihydropyrrolo[1,2-*a*]pyrazin-3(4*H*)-one (15a) was synthesized by the catalytic hydrogenation of compound **1e**. The yield was 97%, viscous pale-yellow oil. ^{1}H NMR ($CDCl_3$), δ: 1.08 (t, 3 H, Me, $^3J = 7.2$ Hz); 1.84 (br.s, 1 H, NH); 2.66 (q, 2 H, $C\underline{H}_2Me$; $^3J = 7.2$ Hz); 2.87 (t, 2 H, $C\underline{H}_2NH$, $^3J = 6.6$ Hz); 3.62 (t, 2 H, $C\underline{H}_2CH_2NH$, $^3J = 6.6$ Hz); 4.59 (s, 4 H, $H_2C(1)$, $H_2C(4)$); 5.96 (m, 1 H, H(8)); 6.20 (m, 1 H, H(7)); 6.58 (m, 1 H, H(6)).

Hydrooxalate, m.p. 139—142 °C. Found (%): C, 49.40; H, 6.83; N, 13.52. $C_{11}H_{17}N_3O \cdot C_2H_2O_4 \cdot H_2O$. Calculated (%): C, 49.52; H, 6.71; N, 13.33.

2-(3-Methylaminopropyl)-1,2-dihydropyrrolo[1,2-*a*]pyrazin-3(4*H*)-one (15b) was synthesized by the catalytic hydrogenation of compound **2a**. The yield was 96%, viscous yellow oil. ^{1}H NMR ($CDCl_3$), δ: 1.81 (m, 2 H, $C\underline{H}_2CH_2NH$); 2.08 (br.s, 2 H, NH); 2.41 (s, 3 H, Me); 2.59 (t, 2 H, $C\underline{H}_2NH$, $^3J = 6.8$ Hz); 3.58 (t, 2 H, $C\underline{H}_2CH_2CH_2NH$, $^3J = 7.0$ Hz); 4.52 and 4.59 (both s, 2 H each, $H_2C(1)$, $H_2C(4)$); 5.96 (m, 1 H, H(8)); 6.20 (m, 1 H, H(7)); 6.57 (m, 1 H, H(6)).

Hydrooxalate, m.p. 129—131 °C. Found (%): C, 46.98; H, 6.99; N, 12.59. $C_{11}H_{17}N_3O \cdot C_2H_2O_4 \cdot 2H_2O$. Calculated (%): C, 46.84; H, 6.95; N, 12.61.

2-(2-Aminoethyl)-1,2-dihydropyrrolo[1,2-*a*]pyrazin-3(4*H*)-one (16a) was synthesized by the catalytic hydrogenation of compound **1a**. The yield was 94%, viscous yellow oil. ^{1}H NMR

($CDCl_3$), δ: 1.54 (br.s, 2 H, NH_2); 2.94 (t, 2 H, $C\underline{H}_2NH_2$, ^3J = 6.5 Hz); 3.57 (t, 2 H, $C\underline{H}_2CH_2NH_2$, ^3J = 6.5 Hz); 4.57 and 4.60 (both s, 2 H each, $H_2C(1)$, $H_2C(4)$); 5.96 (m, 1 H, H(8)); 6.20 (m, 1 H, H(7)); 6.58 (m, 1 H, H(6)).

Hydrooxalate, m.p. 182—184 °C. Found (%): C, 49.00; H, 5.68; N, 15.69. $C_9H_{13}N_3O \cdot C_2H_2O_4$. Calculated (%): C, 49.07; H, 5.62; N, 15.61.

2-(3-Aminopropyl)-1,2-dihydropyrrolo[1,2-*a*]pyrazin-3(4*H*)-one (16b) was synthesized by the catalytic hydrogenation of compound **2b**. The yield was 94%, viscous yellow oil. ^{1}H NMR ($CDCl_3$), δ: 1.73 (m, 2 H, $C\underline{H}_2CH_2NH_2$); 1.95 (br.s, 2 H, NH_2); 2.69 (t, 2 H, $C\underline{H}_2NH_2$, ^3J = 6.6 Hz); 3.59 (t, 2 H, $C\underline{H}_2CH_2CH_2NH_2$, ^3J = 6.9 Hz); 4.50 and 4.58 (both s, 2 H each, $H_2C(1)$, $H_2C(4)$); 5.96 (m, 1 H, H(8)); 6.20 (m, 1 H, H(7)); 6.57 (m, 1 H, H(6)).

Hydrooxalate, m.p. 161—163 °C. Found (%): C, 51.01; H, 6.08; N, 14.85. $C_{10}H_{15}N_3O \cdot C_2H_2O_4$. Calculated (%): C, 50.88; H, 6.05; N, 14.83.

Reduction of pyrrolo[2,1-*c*]-1,3-diazacycloalkano[1,2-*a*]-pyrazinones 1 and 2 with lithium aluminum hydride (general procedure). A solution of pyrrolo[2,1-*c*]-1,3-diazacycloalkano[1,2-*a*]-pyrazinone **1** or **2** (15 mmol) in toluene (30 mL) was added dropwise with stirring to a suspension of lithium aluminum hydride (0.95 g, 25 mmol) in anhydrous diethyl ether (40 mL) and toluene (20 mL) at 25 °C. The reaction mixture was stirred at 40 °C for 4 h, and then a 20% NaOH solution (1 mL) and water (8 mL) were added. The solution was decanted from the precipitate, and the precipitate was washed with toluene and also decanted. The combined solutions were concentrated to dryness. The residue was distilled.

1-Ethyl-1,2,3,5,6,10b-hexahydroimidazo[1,2-*a*]pyrrolo-[2,1-*c*]pyrazine (17a) was synthesized by the reduction of compound **1e**. The yield was 80%, colorless oil, b.p. 122—124 °C (1.5 Torr), n_D^{20} = 1.5443. Found (%): C, 68.90; H, 9.26; N, 21.71. $C_{11}H_{17}N_3$. Calculated (%): C, 69.07; H, 8.96; N, 21.97. ^{1}H NMR (DMSO), δ: 1.01 (t, 3 H, Me, ^3J = 7.2 Hz); 2.16 (q, 2 H, $C\underline{H}_2Me$; ^3J = 7.2 Hz); 2.70—3.00 and 3.07—3.24 (both m, 3 H each, $H_2C(2)$, $H_2C(3)$, $H_2C(5)$); 3.73—3.95 (m, 2 H, $H_2C(6)$); 3.98 (s, 1 H, H(10b)); 5.93 (m, 1 H, H(10)); 5.99 (m, 1 H, H(9)); 6.67 (m, 1 H, H(8)).

1-Methyl-1,3,4,6,7,11b-hexahydro-2*H*-pyrrolo[2´,1´:3,4]-pyrazino[1,2-*a*]pyrimidine (17b) was synthesized by the reduction of compound **2a**. The yield was 82%, colorless oil, b.p. 105—108 °C (1 Torr), n_D^{20} = 1.5546. Found (%): C, 68.89; H, 8.94; N, 21.83. $C_{11}H_{17}N_3$. Calculated (%): C, 69.07; H, 8.96; N, 21.97. ^{1}H NMR (DMSO), δ: 1.50 (dm, 1 H, $H_aC(3)$, $^2J_{3a,3b}$ = 13.2 Hz); 2.34 (m, 1 H, $H_bC(3)$); 2.50 (s, 3 H, Me); 2.63—2.85 and 3.03—3.33 (both m, 2 H and 4 H, $H_2C(2)$, $H_2C(4)$, $H_2C(6)$); 4.14 (m, 2 H, $H_2C(7)$); 4.36 (s, 1 H, H(11b)); 6.08 (m, 1 H, H(11)); 6.27 (m, 1 H, H(10)); 6.86 (m, 1 H, H(9)).

Catalytic hydrogenation of pyrrolo[2,1-*c*]-1,3-diazacycloalkano[1,2-*a*]pyrazines 17 (general procedure). The 10% Pd/C catalyst (0.2 g) was added to a solution of pyrrolo[2,1-*c*]-1,3-diazacycloalkano[1,2-*a*]pyrazine **17** (10 mmol) in ethanol (40 mL). The hydrogenation of the reaction mixture was performed until the theoretical amount of hydrogen was consumed. Then the catalyst was filtered off, and the filtrate was concentrated to dryness. The residue was distilled.

2-(2-Ethylaminoethyl)-1,2,3,4-tetrahydropyrrolo[1,2-*a*]-pyrazine (19a) was synthesized by the catalytic hydrogenation of compound **17a**. The yield was 92%, colorless oil, b.p. 131—134 °C (1.5 Torr), n_D^{20} = 1.5242. Found (%): C, 67.88; H, 10.21; N, 21.55. $C_{11}H_{19}N_3$. Calculated (%): C, 68.35; H, 9.91; N, 21.74. ^{1}H NMR ($CDCl_3$), δ: 1.10 (t, 3 H, Me, ^3J = 7.2 Hz); 2.05 (br.s, 1 H, NH); 2.60—2.79 (m, 6 H, $C\underline{H}_2C\underline{H}_2NHC\underline{H}_2$); 2.84 (t, 2 H, $H_2C(3)$, ^3J = 5.6 Hz); 3.63 (s, 2 H, $H_2C(1)$); 3.98 (t, 2 H, $H_2C(4)$, ^3J = 5.6 Hz); 5.83 (m, 1 H, H(8)); 6.13 (m, 1 H, H(7)); 6.53 (m, 1 H, H(6)).

2-(3-Methylaminopropyl)-1,2,3,4-tetrahydropyrrolo[1,2-*a*]-pyrazine (19b) was synthesized by the catalytic hydrogenation of compound **17b**. The yield was 94%, colorless oil, b.p. 135—138 °C (1.5 Torr), n_D^{20} = 1.5285. Found (%): C, 67.97; H, 10.10; N, 21.87. $C_{11}H_{19}N_3$. Calculated (%): C, 68.35; H, 9.91; N, 21.74. ^{1}H NMR (DMSO), δ: 1.61 (m, 2 H, $C\underline{H}_2CH_2NH$); 2.25 (s, 3 H, Me); 2.48 (m, 4 H, $C\underline{H}_2CH_2C\underline{H}_2NH$); 2.72 (t, 2 H, $H_2C(3)$, ^3J = 5.5 Hz); 3.49 (s, 2 H, $H_2C(1)$); 3.89 (t, 2 H, $H_2C(4)$, ^3J = 5.5 Hz); 5.69 (m, 1 H, H(8)); 5.96 (m, 1 H, H(7)); 6.57 (m, 1 H, H(6)).

X-ray diffraction study. Single crystals of compounds **1c** and **2d** suitable for X-ray diffraction were obtained by recrystallization from ethanol. The low-temperature X-ray diffraction studies were carried out on SMART APEX II CCD (**1c**) and SMART APEX 1000 CCD (**2d**) diffractometers (Mo-Kα radiation, graphite monochromator, ω-scanning technique). The structures were

Table 2. Principal crystallographic data and the refinement statistics for compounds **1c** and **2d**

Parameter	**1c**	**2d**
Empirical formula	$C_{19}H_{25}N_3O_5$	$C_{26}H_{29}N_3O_3$
Molecular weight	375.42	431.52
T/K	100	120
Crystal system	Triclinic	Triclinic
Space group	$P\bar{1}$	$P\bar{1}$
Z	2	4
a/Å	7.1503(5)	9.4746(9)
b/Å	9.1065(7)	13.0247(12)
c/Å	14.9297(13)	18.6863(17)
α/deg	106.213(2)	81.435(4)
β/deg	102.096(3)	89.817(3)
γ/deg	92.722(2)	78.018(2)
V/Å^3	906.91(12)	2229.6(4)
d_{calc}/g cm^{-3}	1.375	1.286
μ/cm^{-1}	1	0.85
F(000)	400	920
$2\theta_{max}$/deg	60	56
Number of measured reflections	11907	28591
Number of independent reflections	5270	10720
Number of reflections with $I > 2\sigma(I)$	4438	4523
Number of refined parameters	255	581
R_1	0.0404	0.0565
wR_2	0.1125	0.1172
GOOF	1.002	1.000
Residual electron density (d_{max}/d_{min})/e · Å^{-3}	0.461/−0.250	0.414/−0.252

solved by direct methods and refined by the full-matrix least-squares method with anisotropic displacement parameters based on F^2_{hkl}. The hydrogen atoms were positioned geometrically and refined using a riding model, except for the hydrogen atoms of the water molecule in compound **1c**, which were located in difference Fourier maps and refined isotropically. Principal crystallographic data and the structure refinement statistics are given in Table 2. All calculations were carried out using the SHELXTL PLUS program package.[24]

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