Synthesis of Bicyclic Azacompounds (3-Dimethylcarbamoyloxyphenyl) Substituted as Acetylcholinesterase Inhibitors

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This paper describes the synthesis of some bicyclic 2-(3-dimethylcarbamoyloxyphenyl) substituted azaderivatives, obtained from 1,4- and 1,5-diketones, which were cyclized with ammonium acetate, methylamine and by reductive amination. Corresponding 3-substituted derivatives were instead prepared by reaction of 1,5-ketoesters with formamide. The carbamates were tested as *in vitro* acetylcholinesterase inhibitors.

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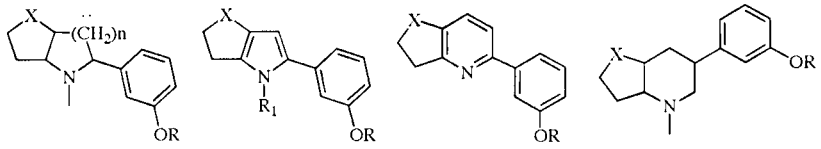
Although numerous central nervous system dysfunctions have been documentated in Alzheimer's disease, the cholinergic deficiency is the most consistent and has the greatest quantitative significance [1]. Several strategies have been adopted in an attempt to enhance cholinergic function [2], but to date the most effective approach has been the use of inhibitors of acetylcholinesterase which improve cholinergic transmission by reducing the enzymatic degradation of acetylcholine [3]. In addition to the inhibitors already approved by the Food & Drug Administration, including tacrine [4], donezepil [5] and, more recently, rivastigmine [6], other inhibitors are under investigation as potential drugs for Alzheimer's disease.

As a further development of our program directed to the search for new cholinesterase inhibitors [7-10], in this paper we report the synthesis and *in vitro* acetylcholinesterase inhibition of bicyclic azaderivatives 2- and 3-(3-dimethylcarbamoyloxyphenyl) substituted (Figure 1).

Compounds **4** were instead synthesized either by catalytic hydrogenation of 3-phenylquinoline followed by reduction with sodium in ethanol [16] or by catalytic hydrogenation of 5-keto aromatic nitriles [17].

Our synthetical pathways to compounds **1-3** are summarized in Scheme 1.

According to conventional methods described in the literature [18,19], 1-(4-morpholino)-1-cyclopentene (**5a**), 1-(4-morpholino)-1-cyclohexene (**5b**) and 1-(4-morpholino)-1-cycloheptene (**5c**) with α-bromo-3-methoxyacetophenone in refluxing toluene gave 1,4-diketones **6a-c**, while with 3-dimethylamino-1-(3-methoxyphenyl)propan-1-one at 120° afforded 1,5-diketones **7a-c**. Under the same conditions, 1-methyl-4-(4-morpholino)-1,2,5,6-tetrahydropyridine (**5d**) reacted with the above reported Mannich base to give 1,5-diketone **7d**, while with α-bromo-3-methoxyacetophenone afforded only dark tar material.



1 **2** **3** **4**

1,3: X = $-CH_2-$, $-(CH_2)_2-$, $-(CH_2)_3-$, $-N(CH_3)CH_2-$;
2,4: X = $-(CH_2)_2-$, $-(CH_2)_3-$; n = 1,2; R = $-CON(CH_3)_2$; R_1 = H, $-CH_3$.

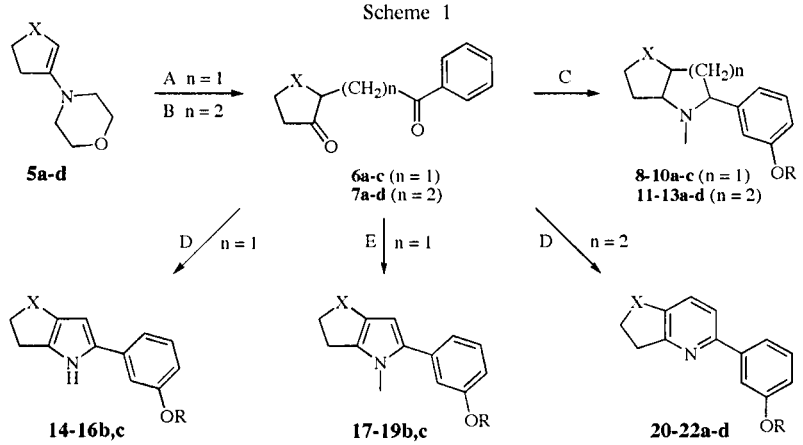
Figure 1

Several references related to bicyclic structures **2** and **3** are reported in the literature [11,12], but very few analogues of compounds **1** and **4**, prepared by different synthetic routes, have been described.

Particularly, compounds **1** (X = $-CH_2-$, n = 1 and X = $-(CH_2)_2-$, n = 1) were obtained by cyclization of 2-azaallyl anions with lithium diethylamide [13]. The decahydroquinolines (X = $-(CH_2)_2-$, n = 2) were prepared by reductive amination of pyrilium salts with amines and Raney nickel in an autoclave [14] or by catalytic hydrogenation of pyridinium salts [15].

Diketones **6** and **7** were subjected to reductive amination with 40% aqueous methylamine and sodium borohydride (to obtain compounds **8a-c** and **11b,d**) or with methylamine hydrochloride and sodium cyanoborohydride (to obtain compounds **11a** and **11c**). Only bicyclic derivatives **11b-d** afforded an isomeric mixture easily separated by column chromatography (see Experimental), while all the other perhydrocompounds were isolated as a predominant isomer. Isomers **11c-II** and **11c-III** were obtained in such a low yield that it wasn't possible to carry out the following steps.

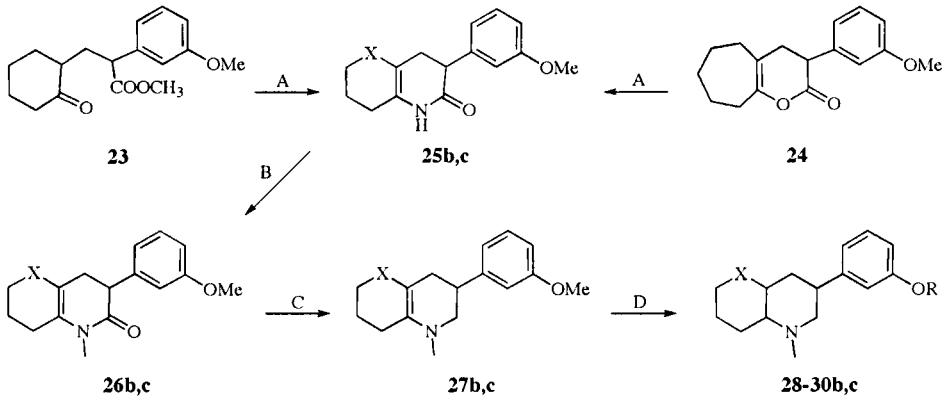
Scheme 1



	X		R
a	$-CH_2-$	8,11,14,17,20	$-CH_3$
b	$-(CH_2)_2-$	9,12,15,18,21	H
c	$-(CH_2)_3-$	10,13,16,19,22	$-CON(CH_3)_2$
d	$-N(CH_3)CH_2-$		

Reagents: A: α-bromo-3-methoxyacetophenone. B: 3-dimethylamino-1-(3-methoxyphenyl)propan-1-one. C: a) 40% aqueous CH_3NH_2, $NaBH_4$,dioxane or methanol, (CH_3NH_2.HCl, $NaBH_3CN$, methanol to obtain compounds **11a,c**); b) 48% HBr ; c) $(CH_3)_2NCOCl$, pyridine. D: a) CH_3COONH_4, refluxing ethanol; b) 48% HBr, (pyridinium chloride/180° to obtain compounds **15b,c** and **21b**); c) $(CH_3)_2NCOCl$, pyridine. E: a) 40% aqueous CH_3NH_2, methanol; b) 48% HBr ; c) $(CH_3)_2NCOCl$, pyridine.

Scheme 2



	X		R
b	$-CH_2-$	28	$-CH_3$
c	$-(CH_2)_2-$	29	H
		30	$-CON(CH_3)_2$

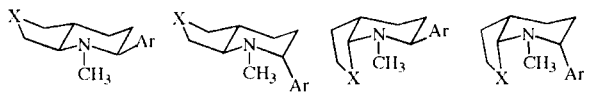
Reagents: A: $HCONH_2$/170°. B: CH_3I/NaH, DMF. C: $LiAlH_4$. D: a) HCO_2H, $HCONH_2$/170°; b) 48% HBr; c) $(CH_3)_2NCOCl$, pyridine.

Diketones **6b,c** by reaction with ammonium acetate in refluxing ethanol gave the bicyclic pyrroles **14b,c**, and, by treatment with 40% aqueous methylamine in methanol at room temperature, the corresponding 1-methylderivatives **17b,c**. Attempted cyclization of **6a** by the same procedure was unsuccessful. From all compounds **7** instead, the pyridoderivatives **20** were smoothly obtained.

Satisfactory demethylation of methoxy group in all compounds reported above was accomplished by heating at 125° in 48% hydrobromic acid, except compounds **15b,c** and **21b**, which were better demethylated by melting with pyridinium chloride.

Finally, all phenol derivatives **9**, **12**, **15**, **18** and **21** were carbamoylated by treatment with dimethylcarbamoyl chloride in anhydrous pyridine to give compounds **1-3**.

Methyl 2-(3-methoxyphenyl)-3-(2-oxocyclohexyl)-propanoate (**23**) and 3-(3-methoxyphenyl)-4,5,6,7,8,9-hexahydrocyclohepta[*b*]pyran-2(3*H*)-one (**24**), obtained



trans-2(e)- *trans*-2(a)- *cis*-2(e)- *cis*-2(a)-

11a, 11b-I, 11c-I **11b-II,11c-II, 11d-II** **11b-III, 11d-I** **11c-III**

Ar = 3-methoxyphenyl

	X		Isomer yield/total yield		
11a	$-CH_2-$	**11a**	1.00	**11c-I**	0.81
11b-I,II,III	$-(CH_2)_2-$	**11b-I**	0.19	**11c-II**	0.14
11c-I,II,III	$-(CH_2)_3-$	**11b-II**	0.40	**11c-III**	0.05
11d	$-N(CH_3)CH_2-$	**11b-III**	0.41	**11d-I**	0.50
				11d-II	0.50

Figure 2

respectively by heating enamines **5b** and **5c** at 120° with methyl 3-dimethylamino-2-(3-methoxyphenyl)-propanoate, served as starting materials for the syntheses of compounds **4** outlined in Scheme 2.

Compounds **23** and **24**, by heating in formamide at 170° afforded the 3-(3-methoxyphenyl)-3,4,5,6,7,8-hexahydro-2*H*-quinolin-2(1*H*)-one (**25b**) and 3-(3-methoxyphenyl)-1,3,4,5,6,7,8,9-octahydro-2*H*-cyclohepta-[*b*]pyridin-2-one (**25c**). Methylation of **25** with methyl iodide and sodium hydride in anhydrous dimethylformamide, followed by reduction of the resulting **26** with lithium aluminum hydride led to 3-(3-methoxyphenyl)-1-methyl-1,2,3,4,5,6,7,8-octahydroquinoline (**27b**) and 3-(3-methoxyphenyl)-1-methyl-2,3,4,5,6,7,8,9-octahydro-1*H*-cyclohepta[*b*]pyridine (**27c**). Hydrogenation of the olefinic bond of the cyclic enamines **27** was performed, in satisfactory yields, by formic acid in formamide at 170°.

The decahydroderivatives **28** were first demethylated with 48% hydrobromic acid, then carbamoylated with dimethylaminocarbamoyl chloride, in the same manner as compounds **1-3**, to afford the expected compounds **4**.

The *in vitro* assessment of acetylcholinesterase inhibition of the carbamoylderivatives was carried out according to the classic procedure of Ellmann [20], and the activities for rat cerebral cortex were expressed as IC_{50} values. Pyrrolo and pyrido derivatives **16**, **19** and **22** were not shown to possess activity ($IC_{50} > 10^{-5}$ M), while perhydroderivatives **10**, **13** and **30** displayed significant enzyme affinity (IC_{50} ranging from 0.24 μM for **10c** to 2.2 μM for **13b II**) and compound **10b** showed the strongest inhibitory effect with an IC_{50}= 0.07 μM. The synthesized compounds, all having three stereo centers, were evaluated *in vitro* as racemates. The most active of them were chosen for further investigation with the aim to achieve the separation of their stereoisomers by chiral preparative HPLC. Studies are in progress and the eventual pure enantiomers will be tested as acetylcholinesterase inhibitors.

EXPERIMENTAL

Melting points were taken on a Köfler hot stage apparatus and are uncorrected. The ^{1}H-nmr spectra were obtained on a Varian Gemini 200 MHz instrument; all values were reported in ppm (δ) and standard abbreviations were used (at = apparent triplet; b = broad; d = doublet; dd = doublet of doublets; m = multiplet; q = quadruplet; t = triplet; s = singlet); peak assignments were also based on ^{13}C-APT, ^{1}H-COSY and ^{13}C-^{1}H HETCOR nmr experiments; electron ionization mass spectra were recorded on a HP 59580 B spectrometer operating at 70 eV. Column chromatographic separations were accomplished on Merck silica gel (70-230 mesh) or Merck aluminum oxide 90. The purity of each compound was checked on silica gel C.Erba 60 F_{254} or Merck aluminum oxide 60 F_{254} (Type E) plates and spots were located by uv light. Sodium sulfate was used to dry organic solutions.

The syntheses of **5a** [21], **5b** [22], **5c** [23] and **5d** [24] have been reported elsewhere.

General Procedure for the Preparation of 1,4-diketones **6a-c**.

To a solution of 1-(4-morpholino)-1-cyclopentene (**5a**), 1-(4-morpholino)-1-cyclohexene (**5b**) or 1-(4-morpholino)-1-cycloheptene (**5c**) (0.1 mole) and 4-ethylmorpholine (12.7 g, 0.11 mole) in anhydrous toluene (150 ml) stirred and heated to reflux was added dropwise during 30 minutes a solution of α-bromo-3-methoxyacetophenone (25.2 g, 0.11 mole) [25] in toluene (100 ml). After addition was completed, stirring and heating were continued for 8 hours. After cooling, 10% hydrochloric acid (150 ml) was added and the heterogeneous mixture was stirred for 2 hours at room temperature. The layers were separated; the toluene was washed with water, shaken with charcoal, filtered and evaporated *in vacuo*. The residue was chromatographed on a silica gel column and eluted with a 1:2 v/v ethyl acetate/*n*-hexane mixture. Analytical samples were obtained by distillation.

2-[2-(3-Methoxyphenyl)-2-oxoethyl]-cyclopentanone (**6a**).

This compound was obtained from **5a** in 56% yield, bp 130-135°/0.005 mm; ^{1}H-nmr (deuteriochloroform): δ 7.52 (dd, 1H, H-6"), 7.45 (d, 1H, H-2"), 7.34 (t, 1H, H-5"), 7.07 (dd, 1H, H-4"), 3.83 (s, 3H, OCH_3), 3.50 (dd, 1H, H-1', J_{gem} = 18.1 Hz, $J_{1'-2}$ = 3.3 Hz), 3.01 (dd, 1H, H-1', J_{gem} = 18.1 Hz, $J_{1'-2}$ = 7.9 Hz), 2.62 (m, 1H, H-2), 2.40-1.50 (m, 6H, H-5, H-4 and H-3); ms: (m/z) 232 (M^+), 190, 150, 135.

Anal. Calcd. for $C_{14}H_{16}O_3$: C, 72.39; H, 6.94. Found: C, 72.66; H, 6.74.

2-[2-(3-Methoxyphenyl)-2-oxoethyl]-cyclohexanone (**6b**).

This compound was obtained from **5b** in 50% yield, bp 140-145°/0.005 mm; [lit [26] bp 178-182/0.15 mm]; ^{1}H-nmr (deuteriochloroform): δ 7.56 (dd, 1H, H-6"), 7.48 (dd, 1H, H-2"), 7.34 (t, 1H, H-5"), 7.08 (dd, 1H, H-4"), 3.83 (s, 3H, OCH_3), 3.56 (dd, 1H, H-1', J_{gem} = 17.6 Hz, $J_{1'-2}$ = 6.6 Hz), 3.12 (m, 1H, H-2), 2.65 (dd, 1H, H-1', J_{gem} = 17.6 Hz, $J_{1'-2}$ = 5.6 Hz), 2.30 - 1.30 (m, 8H, H-6, H-5, H-4 and H-3); ms: (m/z) 246 (M^+), 150, 135.

Anal. Calcd. for $C_{15}H_{18}O_3$: C, 73.14; H, 7.37. Found: C, 73.30; H, 7.29.

2-[2-(3-Methoxyphenyl)-2-oxoethyl]-cycloheptanone (**6c**).

This compound was obtained from **5c** in 39% yield, bp 140-145°/0.005 mm; ^{1}H-nmr (deuteriochloroform): δ 7.54 (dd, 1H, H-6"),

7.45 (d, 1H, H-2"), 7.34 (t, 1H, H-5"), 7.07 (dd, 1H, H-4"), 3.82 (s, 3H, OCH_3), 3.60 (dd, 1H, H-1', J_{gem} = 17.7 Hz, $J_{1'-2}$ = 8.3 Hz), 3.29 (m, 1H, H-2), 2.88 (dd, 1H, H-1', J_{gem} = 17.7 Hz, $J_{1'-2}$ = 4.8 Hz), 2.73 (dt, 1-H, H-7eq), 2.47 (m, 2H, H-7ax and H-6eq), 2.00 - 1.30 (m, 7H, H-6ax, H-5, H-4 and H-3); ms: (m/z) 260 (M^+), 150, 135.

Anal. Calcd. for $C_{16}H_{20}O_3$: C, 73.82; H, 7.74. Found: C, 74.00; H, 7.87.

General Procedure for the Preparation of 1,5-diketones **7a-d**.

A mixture of each compound **5** (0.1 mole) and 3-dimethylamino-1-(3-methoxyphenyl)propan-1-one (31.1 g, 0.15 mole) [27] was heated at 120° for 20 hours. After cooling, 10% hydrochloric acid (100 ml) was added and the mixture was stirred for 3 hours at room temperature, then extracted with ethyl acetate. In the case of **7d** the mixture was made alkaline with 19% ammonium hydroxide before extraction. The solvent was evaporated and the crude diketones **7** were purified by chromatography on a silica gel column by eluting with a 1:2 v/v ethyl acetate/*n*-hexane mixture for **7a-c** and 10% methanol/ethyl acetate for **7d**.

2-[3-(3-Methoxyphenyl)-3-oxopropyl]-cyclopentanone (**7a**).

This compound was obtained from **5a** in 61% yield, mp 45-46° (cyclohexane); ^{1}H-nmr (deuteriochloroform): δ 7.54 (d, 1H, H-6"), 7.49 (d, 1H, H-2"), 7.37 (t, 1H, H-5"), 7.09 (d, 1H, H-4"), 3.83 (s, 3H, OCH_3), 3.04 (t, 2H, H-2'), 2.40-1.40 (m, 9H, H-2 H-5, H-1', H-3 and H-4); ms: (m/z) 246 (M^+), 150, 135.

Anal. Calcd. for $C_{15}H_{18}O_3$: C, 73.14; H, 7.37. Found: C, 73.09; H, 7.25.

As by-product the 2,5-di-[3-(3-methoxyphenyl)-3-oxopropyl]-cyclopentanone was isolated in 11% yield, mp 85-87° (ethyl acetate); ^{1}H-nmr (deuteriochloroform): δ 7.51 (d, 2H, H-6"), 7.40 (m, 4H, H-2" and H-5"), 7.15 (d, 2H, H-4"), 3.76 (s, 6H, OCH_3), 3.03 (t, 4H, H-2'), 2.11 - 1.90 (m, 6H, H-1', H-2 and H-5), 1.65 - 1.37 (m, 4H, H-3 and H-4); ms: (m/z) 408 (M^+), 390, 258, 240, 150, 135.

Anal. Calcd. for $C_{25}H_{28}O_5$: C, 73.51; H, 6.91. Found: C, 73.48; H, 7.05.

2-[3-(3-Methoxyphenyl)-3-oxopropyl]-cyclohexanone (**7b**).

This compound was obtained from **5b** in 59% yield, mp 40-42° (*n*-hexane);^{1}H-nmr (deuteriochloroform): δ 7.53 (d, 1H, H-6"), 7.48 (s, 1H, H-2"), 7.32 (t, 1H, H-5"), 7.06 (d, 1H, H-4"), 3.83 (s, 3H, OCH_3), 3.00 (m, 2H, H-2'), 2.35 (m, 3H, H-2 and H-6), 2.06 (m, 3H, H-1', H-3 and H-5), 1.80 - 1.40 (m, 5H, H-1', H-4, H-3 and H-5); ms: (m/z) 260 (M^+), 163, 150.

Anal. Calcd. for $C_{16}H_{20}O_3$: C, 73.82; H, 7.74. Found: C, 73.60; H, 7.84.

2-[3-(3-Methoxyphenyl)-3-oxopropyl]-cycloheptanone (**7c**).

This compound was obtained from **5c** in 73% yield, bp 150-160°/0.005 mm; ^{1}H-nmr (deuteriochloroform): δ 7.50 (dd, 1H, H-6"), 7.45 (d, 1H, H-2"), 7.32 (t, 1H, H-5"), 7.06 (dd, 1H, H-4"), 3.82 (s, 3H, OCH_3), 2.90 (m, 2H, H-2'), 2.57 (m, 1H, H-2), 2.43 (m, 2H, H-7), 2.00 (m, 1H, H-1'), 1.90 - 1.10 (m, 9H, H-1', H-3, H-4, H-5 and H-6); ms: (m/z) 274 (M^+), 150.

Anal. Calcd. for $C_{17}H_{22}O_3$: C, 74.42; H, 8.08. Found: C, 74.61; H, 7.96.

3-[3-(3-Methoxyphenyl)-3-oxopropyl]-1-methyl-piperidin-4-one (**7d**).

This compound was obtained from **5d** in 85% yield, hydrochloride mp 200-203° (ethanol/diethyl ether); ^{1}H-nmr (base) (deuteriochloroform): δ 7.54 - 7.47 (m, 2H, H-2" and H-6"), 7.33 (t, 1H, H-5"), 7.09 (d, 1H, H-4"), 3.83 (s, 3H, OCH_3), 3.11 - 2.93 (m, 4H, H-2', H-2eq and H-6eq), 2.60 (m, 2H, H-3 and H-5eq), 2.36 (m, 5H, H-6ax, H-5ax and N-CH_3), 2.17 (m, 2H, H-2ax and H-1'), 1.69 (m, 1H, H-1'); ms: (m/z) 275 (M^+), 253, 126.

Anal. Calcd. for $C_{16}H_{22}ClNO_3$: C, 61.63; H, 7.11; N, 4.49. Found: C, 61.52; H, 7.22; N, 4.28.

General Procedure for the Preparation of Compounds **8** and **11** by reductive amination of diketones **6** and **7**.

Procedure A: to obtain **8a-c** and **11b,d**.

To a stirred and cooled solution of diketones **6a-c** or **7b,d** (0.05 mole) and methylamine (solution 40% in water, 20 ml) in methanol (150 ml) sodium borohydride (3.8 g, 0.1 mole) was added in several portions during 1 hour. The mixture was stirred overnight at room temperature. The solvent was evaporated under reduced pressure at no more than 40°. The residue was treated with 10% potassium hydroxide solution and extracted with diethyl ether. The organic layer was thoroughly extracted with 5% hydrochloric acid. The acidic phase was finally made alkaline with 10% potassium hydroxide solution then extracted with diethyl ether. After removal of the solvent, the residue was purified by column chromatography on alumina by eluting with a 1:4 v/v ethyl acetate/*n*-hexane mixture.

Procedure B: to obtain **11a,c**.

To a stirred solution of diketones **7a,c** (0.05 mole) and methylamine hydrochloride (5.4 g, 0.08 mole) in methanol (100 ml) sodium cyanoborohydride (3.8 g, 0.06 mole) was added in several portions. Stirring was continued until tlc on alumina (1:4 v/v ethly acetate/*n*-hexane mixture) indicated that the starting material had disappeared (about 6-7 days). Methanol was evaporated *in vacuo* and the residue was treated in the same manner as described for Procedure-A.

All compounds **8** and **11** are yellowish viscous oils, so analytical samples were yielded by carefully drying the products purified by chromatography.

trans-2(e)-(3-Methoxyphenyl)-1-methyl-octahydrocyclopenta-[*b*]pyrrole (**8a**).

This compound was obtained from **6a** in 24% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.19 (t, 1H, H-5'), 6.91 (dd, 1H, H-4'), 6.89 (d, 1H, H-2'), 6.75 (dd, 1H, H-6'), 3.78 (s, 3H, OCH_3), 3.12 (q, 1H, H-2ax, $J_{2ax-3ax}$ = 10.7 Hz, $J_{2ax-3eq}$ = 5.5 Hz), 2.82 (dd, 1H, H-6a, J_{6a-6ax} = 8.0 Hz, J_{6a-3a} = 6.0 Hz), 2.51 (m, 1H, H-3a), 2.25 (m, 1H, H-3eq), 2.09 (s, 3H, N-CH_3), 1.90 - 1.20 (m, 7H, H-4, H-5, H-6 and H-3ax); ^{13}C-nmr (deuteriochloroform): δ 159.9 (C-3'), 145.4 (C-1'), 129.4 (C-5'), 120.0 (C-6'), 113.1 (C-2'), 112.3 (C-4'), 72.6 (C-6a trans), 72.4 (C-2 bearing H-2ax), 55.4 (OCH_3), 44.1 (C-3a trans), 40.6 (C-3), 40.0 (N-CH_3), 34.0 (C-6), 33.1 (C-4), 24.6 (C-5); ms: (m/z) 231 (M^+), 229, 218, 202, 148, 124.

Anal. Calcd. for $C_{15}H_{21}NO$: C, 77.88; H, 9.15; N, 6.05. Found: C, 77.58; H, 9.02; N, 5.91.

trans-2(e)-(3-Methoxyphenyl)-1-methyl-octahydro-1*H*-indole (**8b**).

This compound was obtained from **6b** in 25% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.25 (t, 1H, H-5'), 7.02 (s, 1H, H-2'), 7.00 (d, 1H, H-4'), 6.78 (d, 1H, H-6'), 3.85 (s, 3H, OCH_3), 3.20 (dd, 1H, H-2ax, $J_{2ax-3ax}$ = 10.0 Hz, $J_{2ax-3eq}$ = 6.9 Hz), 2.35 (m, 2H, H-7a and H-3a), 2.13 (s, 3H, N-CH_3), 1.94 (m, 2H,

H-3eq and H-7eq), 1.80 - 1.15 (m, 8H, H-7ax, H-4, H-5, H-6 and H-3ax); ^{13}C-nmr (deuteriochloroform): δ 159.7 (C-3'), 147.5 (C-1'), 129.2 (C-5'), 119.8 (C-6'), 113.0 (C-2'), 111.4 (C-4'), 71.0 (C-7a trans), 65.3 (C-2 bearing H-2ax), 55.1 (OCH_3), 40.2 (N-CH_3), 38.5 (C-3), 37.2 (C-3a trans), 31.1 (C-7), 26.5 (C-4), 25.6 (C-5), 20.5 (C-6); ms: (m/z) 245 (M^+), 230, 202.

Anal. Calcd. for $C_{16}H_{23}NO$: C, 78.32; H, 9.45; N, 5.71. Found: C, 78.03; H, 9.25; N, 5.50.

trans-2(e)-(3-Methoxyphenyl)-1-methyldecahydrocyclohepta-[*b*]pyrrole (**8c**).

This compound was obtained from **6c** in 50% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.21 (t, 1H, H-5'), 6.92 (d, 1H, H-4'), 6.91 (s, 1H, H-2'), 6.74 (d, 1H, H-6'), 3.79 (s, 3H, OCH_3), 3.13 (q, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 10.8 Hz, $J_{2ax\text{-}3eq}$ = 5.6 Hz), 2.42 (m, 1H, H-8a), 2.28 (m, 1H, H-3a), 2.15 (m, 1H, H-3eq), 2.06 (s, 3H, N-CH_3), 1.90 –1.10 (m, 11H, H-8, H-4, H-5, H-6, H-7 and H-3ax); ^{13}C-nmr (deuteriochloroform): δ159.9 (C-3'), 148.0 (C-1'), 129.5 (C-5'), 120.4 (C-6'), 113.4 (C-2' and C-4'), 72.5 (C-2), 71.6 (C-8a trans), 55.6 (OCH_3), 42.3 (C-3a trans), 41.8 (N-CH_3), 39.1 (C-3), 32.5 (C-8), 31.6 (C-4), 29.6 (C-5), 28.3 (C-7), 26.1 (C-6); ms: (m/z) 259 (M^+), 244, 202.

Anal. Calcd. for $C_{17}H_{25}NO$: C, 78.71; H, 9.72; N, 5.40. Found: C, 78.94; H, 9.45; N, 5.33.

trans-2(e)-(3-Methoxyphenyl)-1-methyl-octahydro-1*H*-cyclopenta[*b*]pyridine (**11a**).

This compound was obtained from **7a** in 54% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.19 (t, 1H, H-5'), 6.89 (overlapped d and s, 2H, H-4' and H-2'), 6.75 (d, 1H, H-6'), 3.78 (s, 3H, OCH_3), 2.78 (dd, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 10.5 Hz, $J_{2ax\text{-}3eq}$ = 4.0 Hz), 1.96 (s, 3H, N-CH_3), 1.90 - 1.00 (m, 12H, H-7a, H-4a, H-3eq, H-4, H-5, H-6, H-7 and H-3ax); ^{13}C-nmr (deuteriochloroform): δ 159.6 (C-3'), 146.2 (C-1'), 129.1 (C-5'), 120.1 (C-6'), 113.1 (C-2'), 112.2 (C-4'), 72.8 (C-7a trans), 72.1 (C-2 bearing H-2ax), 55.1 (OCH_3), 44.2 (C-4a trans), 41.6 (N-CH_3), 36.4 (C-3), 30.2 (C-7), 29.7 (C-5), 29.5 (C-4), 20.5 (C-6); ms: (m/z) 245 (M^+).

Anal. Calcd. for $C_{16}H_{23}NO$: C, 78.32; H, 9.45; N, 5.71. Found: C, 78.24; H, 9.22; N, 5.44.

trans-2(e)-(3-Methoxyphenyl)-1-methyldecahydroquinoline (**11b-I**).

This compound was obtained from **7b** in 8.5% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.20 (t, 1H, H-5'), 6.94 (overlapped s and d, 2H, H-2' and H-4'), 6.74 (d, 1H, H-6'), 3.81 (s, 3H, OCH_3), 2.85 (dd, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 11.1 Hz, $J_{2ax\text{-}3eq}$ = 3.1 Hz), 2.12 (overlapped m, 3H, H-8a, H-3eq and H-4a), 1.89 (s, 3H, N-CH_3), 1.85 –1.15 (m, 11H, H-4, H-5, H-8, H-6, H-7 and H-3ax); ^{13}C-nmr (deuteriochloroform): δ 159.6 (C-3'), 148.9 (C-1'), 129.1 (C-5'), 119.8 (C-6'), 113.1 (C-2'), 111.3 (C-4'), 71.3 (C-8a trans), 63.1 (C-2 bearing H-2ax), 55.1 (OCH_3), 39.9 (N-CH_3), 37.4 (C-4a trans), 32.2 (C-5), 30.9 (C-3), 30.5 (C-8), 27.1 (C-4), 26.4 (C-6), 19.8 (C-7); ms: (m/z) 259 (M^+), 216.

Anal. Calcd. for $C_{17}H_{25}NO$: C, 78.71; H, 9.72; N, 5.40. Found: C, 78.75; H, 9.80; N, 5.32.

trans-2(a)-(3-Methoxyphenyl)-1-methyldecahydroquinoline (**11b-II**).

This compound was obtained from **7b** in 18% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.19 (t, 1H, H-5'), 6.90 (d, 1H, H-4'), 6.88 (s, 1H, H-2'), 6.73 (d, 1H, H-6'), 3.79 (s, 3H, OCH_3), 2.87 (dd, 1H, H-2eq, $J_{2eq\text{-}3eq}$ = 8.1 Hz, $J_{2eq\text{-}3ax}$ = 4.7 Hz), 2.15 (dt, 1H, H-8a), 1.97 (s, 3H, N-CH_3), 1.90 - 0.95 (m, 13H, H-4a, H-3, H-4, H-5, H-8, H-6 and H-7); ^{13}C-nmr (deuteriochloroform): δ 159.7 (C-3'), 147.9 (C-1'), 129.3 (C-5'), 119.9 (C-6'), 113.1 (C-2'), 112.0 (C-4'), 71.6 (C-8a trans), 69.9 (C-2 bearing H-2eq), 55.2 (OCH_3), 41.6 (C-4a trans), 39.9 (N-CH_3), 35.9 (C-3), 33.3 (C-5), 32.8 (C-4), 30.6 (C-8), 25.9 (C-6), 25.8 (C-7); ms: (m/z) 259 (M^+), 216.

Anal. Calcd. for $C_{17}H_{25}NO$: C, 78.71; H, 9.72; N, 5.40. Found: C, 78.55; H, 9.66; N, 5.55.

cis-2(e)-(3-Methoxyphenyl)-1-methyldecahydroquinoline (**11b-III**).

This compound was obtained from **7b** in 19% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.19 (t, 1H, H-5'), 6.88 (d, 1H, H-4'), 6.86 (s, 1H, H-2'), 6.77 (d, 1H, H-6'), 3.79 (s, 3H, OCH_3), 3.30 (dd, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 10.7 Hz, $J_{2ax\text{-}3eq}$ = 3.4 Hz), 2.88 (m, 1H H-8a), 2.13 (m, 1H, H-3eq), 2.04 (s, 3H, N-CH_3), 1.90 - 1.00 (m, 12H, H-4a, H-4, H-5, H-8, H-6, H-7 and H-3ax); ^{13}C-nmr (deuteriochloroform): δ 159.7 (C-3'), 146.8 (C-1'), 129.2 (C-5'), 120.1 (C-6'), 112.6 (C-2'), 112.5 (C-4'), 63.0 (C-8a cis), 62.3 (C-2 bearing H-2ax), 55.2 (OCH_3), 40.4 (N-CH_3), 36.7 (C-4a cis), 36.3 (C-3), 31.9 (C-5), 25.9 (C-8), 24.2 (C-6), 21.2 (C-4), 17.0 (C-7); ms: (m/z) 259 (M^+), 216.

Anal. Calcd. for $C_{17}H_{25}NO$: C, 78.71; H, 9.72; N, 5.40. Found: C, 78.68; H, 9.54; N, 5.61.

trans-2(e)-(3-Methoxyphenyl)-1-methyldecahydro-1*H*-cyclohepta[*b*]pyridine (**11c-I**).

This compound was obtained from **7c** in 30% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.20 (t, 1H, H-5'), 6.93 (overlapped d and s, 2H, H-4' and H-2'), 6.73 (d, 1H, H-6'), 3.80 (s, 3H, OCH_3), 2.84 (dd, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 9.7 Hz, $J_{2ax\text{-}3eq}$ = 2.7 Hz), 2.21 (m, 1H, H-9a), 2.07 (at, 1H, H-4a), 1.92 (s, 3H, N-CH_3), 1.80 - 1.20 (m, 14H, H-9, H-3, H-4, H-5, H-8, H-6 and H-7); ^{13}C-nmr (deuteriochloroform): δ 159.6 (C-3'), 148.3 (C-1'), 129.2 (C-5'), 119.8 (C-6'), 113.0 (C-2', C-4'), 71.5 (C-9a trans), 65.8 (C-2 bearing H-2ax), 55.2 (OCH_3), 40.4 (C-4a trans), 40.1 (N-CH_3), 33.4 (C-5), 32.9 (C-3), 32.0 (C-9), 30.1 (C-6), 29.5 (C-4), 28.6 (C-8), 21.6 (C-7); ms: (m/z) 273 (M^+), 216.

Anal. Calcd. for $C_{18}H_{27}NO$: C, 79.07; H, 9.95; N, 5.12. Found: C, 79.12; H, 9.79; N, 5.34.

trans-2(a)-(3-Methoxyphenyl)-1-methyldecahydro-1*H*-cyclohepta[*b*]pyridine (**11c-II**).

This compound was obtained from **7c** in 5% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.19 (t, 1H, H-5'), 6.91 (d, 1H, H-4'), 6.89 (s, 1H, H-2'), 6.72 (d, 1H, H-6'), 3.79 (s, 3H, OCH_3), 2.93 (dd, 1H, H-2eq, $J_{2eq\text{-}3eq}$ = 8.1 Hz, $J_{2eq\text{-}3ax}$ = 7.0 Hz), 2.05 (m, 1H, H-9a), 2.02 (s, 3H, N-CH_3), 1.78 (m, 1H, H-4a), 1.70 - 1.10 (m, 14H, H-9, H-3, H-4, H-5, H-8, H-6 and H-7); ms: (m/z) 273 (M^+), 216.

Anal. Calcd. for $C_{18}H_{27}NO$: C, 79.07; H, 9.95; N, 5.12. Found: C, 78.92; H, 9.74; N, 5.00.

cis-2(a)-(3-Methoxyphenyl)-1-methyldecahydro-1*H*-cyclohepta-[*b*]pyridine (**11c-III**).

This compound was obtained from **7c** in 2% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.19 (t, 1H, H-5'), 6.87 (d, 1H, H-4'), 6.85 (s, 1H, H-2'), 6.76 (d, 1H, H-6'), 3.79 (s, 3H, OCH_3), 3.15 (dd, 1H, H-2eq, $J_{2eq\text{-}3ax}$ = 6.8 Hz, $J_{2eq\text{-}3eq}$ = 6.2 Hz), 2.95 (q, 1H,

H-9a), 2.10 (m, 1H, H-4a), 2.03 (s, 3H, N-CH_3), 2.00 - 0.90 (m, 14H, H-3, H-4, H-5, H-6, H-7, H-8 and H-9); ms: (m/z) 273 (M^+), 216.

Anal. Calcd. for $C_{18}H_{27}NO$: C, 79.07; H, 9.95; N, 5.12. Found: C, 79.09; H, 10.02; N, 5.29.

cis-1,6-Dimethyl-2(e)-(3-methoxyphenyl)-decahydro[1,6]naphthyridine (**11d-I**).

This compound was obtained from **7d** in 8% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.18 (t, 1H, H-5'), 6.86 (d, 1H, H-4'), 6.84 (s, 1H, H-2'), 6.75 (d, 1H, H-6'), 3.78 (s, 3H, OCH_3), 3.27 (dd, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 10.7 Hz, $J_{2ax\text{-}3eq}$ = 3.1 Hz), 2.89 (m, 2H, H-5eq and H-8a), 2.69 (d, 1H, H-7eq, J_{gem} = 11.4 Hz), 2.19 (s, 3H, N*6*-CH_3), 2.10 (dd, 1H, H-7ax, J_{gem} = 11.4 Hz), 2.04 (s, 3H, N*1*-CH_3), 2.01 (m, 2H, H-4a and H-3eq), 1.86 (dd, 1H, H-5ax), 1.80 - 1.40 (m, 5H, H-4, H-8 and H-3ax); ^{13}C-nmr (deuteriochloroform): δ 159.6 (C-3'), 146.4 (C-1'), 129.1 (C-5'), 119.8 (C-6'), 112.6 (C-2'), 112.4 (C-4'), 62.1 (C-2 bearing H-2ax), 61.7 (C-8a cis), 60.8 (C-5), 55.8 (C-7), 55.1 (OCH_3), 46.6 (N*6*-CH_3), 40.1 (N*1*-CH_3), 36.9 (C-4a cis), 35.9 (C-3), 24.6 (C-8), 17.4 (C-4); ms: (m/z) 274 (M^+), 216.

Anal. Calcd. for $C_{17}H_{26}N_2O$: C, 74.41; H, 9.55; N, 10.21. Found: C, 74.66; H, 9.26; N, 10.08.

trans-1,6-Dimethyl-2(a)-(3-methoxyphenyl)-decahydro[1,6]-naphthyridine (**11d-II**).

This compound was obtained from **7d** in 8% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.19 (t, 1H, H-5'), 6.89 (d, 1H, H-4'), 6.87 (s, 1H, H-2'), 6.71 (d, 1H, H-6'), 3.79 (s, 3H, OCH_3), 2.82 (m, 2H, H-5eq and H-7eq), 2.66 (dd, 1H, H-2eq, $J_{2eq\text{-}3eq}$ = 7.8 Hz, $J_{2eq\text{-}3ax}$ = 1.9 Hz), 2.17 (s, 3H, N*6*-CH_3), 1.90 (overlapped m and s, 5H, H-8a, H-5ax and N*1*-CH_3), 1.58 (m, 7H, H-4a, H-3, H-4eq, H-7ax and H-8), 1.12 (m, 1H, H-4ax); ^{13}C-nmr (deuteriochloroform): δ 159.5 (C-3'), 147.1 (C-1'), 129.2 (C-5'), 119.7 (C-6'), 112.8 (C-2'), 112.0 (C-4'), 71.3 (C-8a trans), 67.7 (C-2 bearing H-2eq), 61.2 (C-5), 55.4 (C-7), 55.1 (OCH_3), 45.9 (N*6*-CH_3), 40.0 (C-4a trans), 39.5 (N*1*-CH_3), 35.3 (C-8), 30.2 (C-3), 29.2 (C-4); ms: (m/z) 274 (M^+), 243, 216.

Anal. Calcd. for $C_{17}H_{26}N_2O$: C, 74.41; H, 9.55; N, 10.21. Found: C, 74.34; H, 9.41; N, 9.98.

General Procedure for the Preparation of Compounds **14** and **20**.

A mixture of each compound **6** or **7** (0.02 mole) and ammonium acetate (7.7 g, 0.1 mole) in ethanol (100 ml) was refluxed for 3 hours. The solvent was removed under reduced pressure and the residue, made alkaline with diluted ammonia, was extracted with dichloromethane. The solvent was removed and the crude material was purified by chromatography on a silica gel column with a 1:2 v/v ethyl acetate/*n*-hexane mixture as eluant, then crystallized.

2-(3-Methoxyphenyl)-4,5,6,7-tetrahydro-1*H*-indole (**14b**).

This compound was obtained from **6b** in 72% yield, mp 125-127° (benzene); ^{1}H-nmr (deuteriochloroform): δ 7.95 (bs, 1H, NH), 7.23 (t, 1H, H-5'), 6.98 (d, 1H, H-4'), 6.94 (s, 1H, H-2'), 6.68 (d, 1H, H-6'), 6.26 (s, 1H, H-3), 3.81 (s, 3H, OCH_3), 2.62 (t, 2H, H-7), 2.52 (t, 2H, H-4), 1.81 (m, 4H, H-6 and H-5); ms: (m/z) 227 (M^+).

Anal. Calcd. for $C_{15}H_{17}NO$: C, 79.26; H, 7.54; N, 6.16. Found: C, 79.52; H, 7.29, N, 6.02.

2-(3-Methoxyphenyl)-1,4,5,6,7,8-hexahydrocyclohepta[*b*]pyrrole (**14c**).

This compound was obtained from **6c** in 70% yield, mp 118-120° (benzene); ^{1}H-nmr (deuteriochloroform): δ 8.06 (bs, 1H, NH), 7.23 (t, 1H, H-5'), 6.98 (d, 1H, H-4'), 6.95 (s, 1H, H-2'), 6.69 (d, 1H, H-6'), 6.28 (s, 1H, H-3), 3.83 (s, 3H, OCH_3), 2.71 (at, 2H, H-8), 2.61 (at, 2H, H-4), 1.70 (m, 6H, H-7, H-5 and H-6); ms: (m/z) 241 (M^+).

Anal. Calcd. for $C_{16}H_{19}NO$: C, 79.63; H, 7.94; N, 5.80. Found: C, 79.38; H, 7.79; N, 6.04.

2-(3-Methoxyphenyl)-6,7-dihydro-5*H*-cyclopenta[*b*]pyridine (**20a**).

This compound was obtained from **7a** in 62% yield, mp 74-75° (ethyl acetate/*n*-hexane); ^{1}H-nmr (deuteriochloroform): δ 7.51 - 7.39 (m, 4H, H-4, H-3, H-2' and H-6'), 7.33 (t, 1H, H-5'), 6.91 (dd, 1H, H-4'), 3.85 (s, 3H, OCH_3), 3.06 (t, 2H, H-7), 2.96 (t, 2H, H-5), 2.13 (q, 2H, H-6); ms: (m/z) 225 (M^+), 195.

Anal. Calcd. for $C_{15}H_{15}NO$: C, 79.97; H, 6.71; N, 6.22. Found: C, 79.78; H, 6.99; N, 6.44.

2-(3-Methoxyphenyl)-5,6,7,8-tetrahydroquinoline (**20b**).

This compound was obtained from **7b** in 57% yield, mp 63-64° (ethyl acetate/*n*-hexane); ^{1}H-nmr (deuteriochloroform): δ 7.50 - 7.46 (m, 2H, H-2 and H-6'), 7.40 (d, 2H, H-4 and H-3), 7.33 (t, 1H, H-5'), 6.90 (dd, 1H, H-4'), 3.86 (s, 3H, OCH_3), 2.98 (t, 2H, H-8), 2.77 (t, 2H, H-5), 1.86 (m, 4H, H-6 and H-7); ms: (m/z) 239 (M^+), 209.

Anal. Calcd. for $C_{16}H_{17}NO$: C, 80.30; H, 7.16; N, 5.85. Found: C, 80.48; H, 6.96; N, 5.70.

2-(3-Methoxyphenyl)-6,7,8,9-tetrahydro-5*H*-cyclohepta[*b*]pyridine (**20c**).

This compound was obtained from **7c** in 50% yield, mp 79-80° (ethyl acetate/*n*-hexane); ^{1}H-nmr (deuteriochloroform): δ 7.55 - 7.49 (m, 2H, H-2' and H-6'), 7.40 (s, 2H, H-3 and H-4), 7.33 (t, 1H, H-5'), 6.90 (dd, 1H, H-4'), 3.86 (s, 3H, OCH_3), 3.10 (m, 2H, H-9), 2.78 (m, 2H, H-5), 1.88 - 1.64 (m, 6H, H-6, H-7 and H-8); ms: (m/z) 253 (M^+), 223.

Anal. Calcd. for $C_{17}H_{19}NO$: C, 80.60; H, 7.56; N, 5.53. Found: C, 80.44; H, 7.84; N, 5.38.

2-(3-Methoxyphenyl)-6-methyl-6,7,8,9-tetrahydro[1,6]naphthyridine (**20d**).

This compound was obtained from **7d** in 64% yield, mp 81-83° (ethyl acetate/*n*-hexane); ^{1}H-nmr (deuteriochloroform): δ 7.52 - 7.33 (m, 5H, H-2', H-5', H-6', H-3 and H-4), 6.91 (dd, 1H, H-4'), 3.86 (s, 3H, OCH_3), 3.60 (s, 2H, H-5), 3.13 (t, 2H, H-7), 2.80 (t, 2H, H-8), 2.48 (s, 3H, N-CH_3); ms: (m/z) 254 (M^+), 239, 210.

Anal. Calcd. for $C_{16}H_{18}N_2O$: C, 75.56; H, 7.13; N, 11.02. Found: C, 75.70; H, 7.34; N, 11.02.

2-(3-Methoxyphenyl)-1-methyl-4,5,6,7-tetrahydro-1*H*-indole (**17b**) and 2-(3-methoxyphenyl)-1-methyl-1,4,5,6,7,8-hexahydrocyclohepta[*b*]pyrrole (**17c**).

A mixture of compound **6b** or **6c** (0.02 mole) and methylamine (40% solution in water, 20 ml) in methanol (80 ml) was stirred for 6 hours at room temperature. The solvent was removed and the residue chromatographed on a silica gel column using a 1:3 v/v ethyl acetate/*n*-hexane mixture as eluant.

Compound **17b** was obtained from **6b** in 62% yield as a viscous oil; ^{1}H-nmr (deuteriochloroform): δ 7.27 (t, 1H, H-5'), 6.95 (d, 1H, H-4'), 6.91 (s, 1H, H-2'), 6.80 (d, 1H, H-6'), 6.02 (s, 1H, H-3), 3.81 (s, 3H, OCH_3), 3.47 (s, 3H, N-CH_3), 2.56 (two partially overlapped t, 4H, H-7 and H-4), 1.86 (m, 4H, H-6 and H-5); ms: (m/z) 241 (M^+), 213, 135.

Anal. Calcd. for $C_{16}H_{19}NO$: C, 79.63; H, 7.94; N, 5.80. Found: C, 79.84; H, 7.80; N, 5.62.

Compound **17c** was obtained from **6c** in 70% yield as a viscous oil; ^{1}H-nmr (deuteriochloroform): δ 7.27 (t, 1H, H-5'), 6.93 (d, 1H, H-4'), 6.89 (s, 1H, H-2'), 6.79 (d, 1H, H-6'), 5.99 (s, 1H, H-3), 3.81 (s, 3H, OCH_3), 3.50 (s, 3H, N-CH_3), 2.73 (at, 2H, H-8), 2.60 (at, 2H, H-4), 1.79 (m, 6H, H-7, H-5 and H-6); ms: (m/z) 255 (M^+).

Anal. Calcd. for $C_{17}H_{21}NO$: C, 79.96; H, 8.29; N, 5.49. Found: C, 79.80; H, 8.50; N, 5.32.

Methyl 3-(2-oxocyclohexyl)-2-(3-methoxyphenyl)propanoate (**23**) and 3-(3-methoxyphenyl)-4,5,6,7,8,9-hexahydrocyclohepta[*b*]pyran-2(3*H*)-one (**24**).

A mixture of enamine **5b** or **5c** (0.1 mole) and methyl 3-dimethylamino-2-(3-methoxyphenyl)propanoate (obtained in 74% yield according to a previously described procedure [28]: bp 115-120°/0.005 mm) (28.4 g, 0.12 mole) was heated at 120° for 20 hours. The reaction mixture was worked in the same manner as described for the preparation and purification of compounds **7**.

Compound **23** was obtained from **5b** in 66% yield, bp 155-160°/0.005 mm; ^{1}H-nmr (deuteriochloroform): δ 7.18 (t, 1H, H-5'), 6.86 - 6.76 (m, 3H, H-6', H-4' and H-2'), 3.77 (s, 3H, aromatic OCH_3), 3.69 (m, 1H, H-2), 3.62 (s, 3H, OCH_3), 2.32 - 1.39 (m, 11H, H-3, H-1", H-3", H-4", H-6" and H-5"), ms: (m/z) 290 (M^+), 258, 230, 192.

Anal. Calcd. for $C_{17}H_{22}O_4$: C, 70.32; H, 7.64. Found: C, 70.18; H, 7.47.

Compound **24** was obtained from **5c** in 83% yield, bp 150-155°/0.005 mm; ^{1}H-nmr (deuteriochloroform): δ 7.20 (t, 1H, H-5'), 6.82 - 6.77 (m, 3H, H-6', H-2' and H-4'), 3.78 (s, 3H, OCH_3), 3.62 (d, 2H, H-4), 3.55 (m, 1H, H-3), 2.44 - 2.07 (m, 4H, H-9 and H-5), 1.78 (m, 4H, H-6 and H-8), 1.30 (m, 2H, H-7); ms: (m/z) 272 (M^+), 244, 192, 148.

Anal. Calcd. for $C_{17}H_{20}O_3$: C, 74.97; H, 7.40. Found: C, 74.68; H, 7.19.

3-(3-Methoxyphenyl)-3,4,5,6,7,8-hexahydroquinolin-2(1*H*)-one (**25b**) and 3-(3-methoxyphenyl)-1,3,4,5,6,7,8,9-octahydro-2*H*-cyclohepta[*b*]pyridin-2-one (**25c**).

A mixture of compound **23** or **24** (0.05 mole) in formamide (80 ml) was heated at 170° for 4 hours. After cooling, water was added to the mixture and the resulting suspension was extracted with ethyl acetate. The solvent was removed and the crude product crystallized.

Compound **25b** was obtained from **23** in 60% yield, mp 138-140° (ethyl acetate); ^{1}H-nmr (deuteriochloroform): δ 7.23 (d, 1H, H-5'), 6.86 - 6.77 (m, 3H, H-6', H-4' and H-2'), 3.77 (s, 3H, OCH_3), 3.67 (t, 1H, H-3), 2.47 (d, 2H, H-4), 2.08 (bs, 4H, H-8 and H-5), 1.65 (m, 4H, H-6 and H-7); ms: (m/z) 257 (M^+), 148.

Anal. Calcd. for $C_{16}H_{19}NO_2$: C, 74.68; H, 7.44; N, 5.44. Found: C, 74.80; H, 7.32; N, 5.52.

Compound **25c** was obtained from **24** in 53% yield, mp 166-168° (ethyl acetate); ^{1}H-nmr (deuteriochloroform): δ 7.28 (t, 1H, H-5'), 6.85 - 6.79 (m, 3H, H-6', H-2' and H-4'), 3.82 (s, 3H, OCH_3), 3.64 (t, 1H, H-3'), 2.22 - 1.34 (m, 12H, H-4, H-5, H-9, H-6, H-8 and H-7); ms: (m/z) 271 (M^+), 164, 148.

Anal. Calcd. for $C_{17}H_{21}NO_2$: C, 75.24; H, 7.80; N, 5.16. Found: C, 75.13; H, 7.68; N, 4.99.

3-(3-Methoxyphenyl)-1-methyl-3,4,5,6,7,8-hexahydroquinolin-2(1*H*)-one (**26b**) and 3-(3-methoxyphenyl)-1-methyl-1,3,4,5,6,7,8,9-octahydro-2*H*-cyclohepta-[*b*]pyridin 2-one (**26c**).

To a stirred suspension of sodium hydride (1.4 g of 50% oil dispersion, 0.03 mole) in anhydrous dimethylformamide (50 ml) each compound **25** (0.02 mole) was added in several portions. After stirring for 1 hour at room temperature, a solution of methyl iodide (2.5 ml, 0.04 mole) in dimethylformamide (10 ml) was added dropwise, the mixture stirred for 2 hours at room temperature, then poured into water and extracted with ethyl acetate. The solvent was removed and the crude product crystallized.

Compound **26b** was obtained from **25b** in 62% yield, mp 68-70° (ethyl acetate/*n*-hexane); ^{1}H-nmr (deuteriochloroform): δ 7.19 (t, 1H, H-5'), 6.81 - 6.74 (m, 3H, H-6', H-4' and H-2'), 3.76 (s, 3H, OCH_3), 3.64 (t, 1H, H-3), 3.09 (s, 3H, N-CH_3), 2.40 (m, 2H, H-4), 2.18 (m, 2H, H-5), 2.05 (m, 2H, H-8), 1.72 - 1.60 (m, 4H, H-6 and H-7); ms: (m/z) 271 (M^+), 148.

Anal. Calcd. for $C_{17}H_{21}NO_2$: C, 75.24; H, 7.80; N, 5.16. Found C, 75.31; H, 7.65; N, 5.00.

Compound **26c** was obtained from **25c** in 64% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.18 (t, 1H, H-5'), 6.77 - 6.73 (m, 3H, H-2', H-4' and H-6'), 3.75 (s, 3H, OCH_3), 3.57 (t, 1H, H-3), 3.08 (s, 3H, N-CH_3), 2.49 (t, 2H, H-4), 2.37 (m, 2H, H-5), 2.17 (t, 2H, H-9), 1.69 - 1.48 (m, 6H, H-8, H-6 and H-7); ms: (m/z) 285 (M^+), 257, 178, 148.

Anal. Calcd. for $C_{18}H_{23}NO_2$: C, 75.75; H, 8.12; N, 4.91. Found: 75.92; H, 8.30; N, 4.74.

3-(3-Methoxyphenyl)-1-methyl-1,2,3,4,5,6,7,8-octahydroquinoline (**27b**) and 3-(3-methoxyphenyl)-1-methyl-2,3,4,5,6,7,8,9-octahydro-1*H*-cyclohepta[*b*]pyridine (**27c**).

To a stirred suspension of lithium aluminum hydride (2.3 g, 0.06 mole) in anhydrous diethyl ether (100 ml), a suspension of each compound **26** (0.05 mole) in anhydrous diethyl ether (50 ml) was added slowly at room temperature. The mixture was refluxed for 2 hours, then cooled in an ice-water bath and carefully treated with water (10 ml). The precipitate hydroxides were removed by filtration and rinsed with diethyl ether. The crude material resulting from the solvent evaporation was purified by chromatography on alumina column with a 1:5 v/v ethyl acetate/*n*-hexane mixture as eluant.

Compound **27b** was obtained from **26b** in 84% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.22 (t, 1H, H-5'), 6.82 - 6.73 (m, 3H, H-6', H-4' and H-2'), 3.78 (s, 3H, OCH_3), 3.07-2.83 (m, 2H, H-2eq and H-3), 2.57 (s, 1H, N-CH_3), 2.08 - 1.45 (m, 11H, H-2ax, H-4, H-5, H-6, H-7 and H-8); ^{13}C-nmr (deuteriochloroform): δ 159.5 (C-3'), 146.3 (C-1'), 136.8 (C-8a), 129.2 (C-5'), 119.3 (C-6'), 112.8 (C-2'), 111.2 (C-4'), 107.7 (C-4a), 58.4 (C-2), 54.9 (OCH_3), 39.2 (C-3), 38.3 (N-CH_3), 36.1 (C-4), 29.8 and 26.3 (C-5 and C-8), 23.5 and 22.8 (C-6 and C-7); ms: (m/z) 257 (M^+), 134.

Anal. Calcd. for $C_{17}H_{23}NO$: C, 79.33; H, 9.01; N, 5.44. Found: C, 79.18; H, 9.22; N, 5.24.

Compound **27c** was obtained from **26c** in 91% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.22 (t, 1H, H-5'), 6.79 - 6.71 (m, 3H, H-2', H-4' and H-6'), 3.77 (s, 3H, OCH_3), 2.91 (m, 2H, H-2eq and H-3), 2.57 (s, 3H, N-CH_3), 2.22 - 1.48 (m, 13H, m, H-2ax, H-4, H-5, H-6, H-7, H-8 and H-9); ^{13}C-nmr (deuteriochloroform): δ 159.5 (C-3'), 146.4 (C-1'), 129.2 (C-5'), 128.1 (C-9a), 119.5 (C-6'), 117.7 (C-4a), 113.0 (C-2'), 111.1 (C-4'), 58.0 (C-2), 55.0 (OCH_3), 40.1 (C-3), 38.0 (C-4), 35.4 (N-CH_3), 33.6 (C-5), 32.4 (C-9), 31.4, 27.0, and 26.6 (C-6, C-7 and C-8); ms: (m/z) 271 (M^+), 257, 244.

Anal. Calcd. for $C_{18}H_{25}NO$: C, 79.66; H, 9.29; N, 5.16. Found: C, 79.53; H, 9.05; N, 5.30.

trans-3-(3-Methoxyphenyl)-1-methyldecahydroquinoline (**28b**) and *cis*-3-(3-methoxyphenyl)-1-methyldecahydro-1*H*-cyclohepta[*b*]pyridine (**28c**).

A solution of each compound **27** (0.02 mole) in 1:1 v/v formamide/99% formic acid (40 ml) was heated at 170° for 30 minutes. After cooling, water was added and the mixture, made alkaline with 10% sodium hydroxide solution, and was extracted with ethyl acetate. Crude compounds **28** were purified by chromatography on alumina using a 1:5 v/v ethyl acetate/*n*-hexane mixture as eluant.

Compound **28b** was obtained from **27b** in 57% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.20 (t, 1H, H-5'), 6.80 (d, 1H, H-6'), 6.75 (s, 1H, H-2'), 6.72 (d, 1H, H-4'), 3.78 (s, 3H, OCH_3), 2.95 (t, 2H, H-2eq and H-3), 2.25 (s, 3H, N-CH_3), 2.16 (m, 2H, H-8a and H-2ax), 1.81 (dd, 2H, H-4eq and H-8eq), 1.66 - 1.08 (m, 9H, H-4ax, H-4a, H-8ax, H-5, H-6 and H-7); ^{13}C-nmr (deuteriochloroform): δ 159.5 (C-3'), 146.0 (C-1'), 129.2 (C-5'), 119.5 (C-6'), 113.1 (C-2'), 111.2 (C-4'), 68.6 (C-8a *trans*), 64.6 (C-2), 55.1 (OCH_3), 42.7 (C-3), 42.6 (N-CH_3), 41.8 (C-4a), 39.2 (C-4), 32.8 (C-5), 30.3 (C-8), 25.9 (C-6), 25.6 (C-7); ms: (m/z) 260 (M^++1), 152.

Anal. Calcd. for $C_{17}H_{25}NO$: C, 78.71; H, 9.72; N, 5.40. Found: C, 78.61; H, 9.65; N, 5.43.

Compound **28c** was obtained from **27c** in 55% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.19 (t, 1H, H-5'), 6.79 - 6.71 (m, 3H, H-2', H-4' and H-6'), 3.77 (s, 3H, OCH_3), 2.91 (m, 2H, H-2eq and H-3), 2.20 (s, 3H, N-CH_3), 2.10 - 1.62 (m, 9H, H-2ax, H-9a, H-4, H-4a, H-5 and H-9), 1.38 - 1.22 (m, 6H, H-6, H-7 and H-8); ^{13}C-nmr (deuteriochloroform): δ 159.5 (C-3'), 146.3 (C-1'), 129.2 (C-5'), 119.4 (C-6'), 113.1 (C-2'), 111.2 (C-4'), 64.9 (C-2), 64.8 (C-9a), 55.1 (OCH_3), 43.2 (N-CH_3 or C-3), 40.3 (C-4), 39.8 (C-3 or N-CH_3), 38.3 (C-4a), 32.3 (C-5), 30.4 (C-9), 29.9 (C-6), 29.2 (C-7), 21.9 (C-8); ms: (m/z) 273 (M^+), 216.

Anal. Calcd. for $C_{18}H_{27}NO$: C, 79.07; H, 9.95; N, 5.12. Found: C, 79.13; H, 9.76; N, 5.19.

General Procedure for the Preparation of Phenol derivatives **9**, **12**, **15**, **18**, **21** and **29**.

Procedure A: to obtain **9**, **12**, **18**, **21a,c** and **29**.

A solution of the appropriate *O*-methylderivative (2 g) in 48% hydrobromic acid (25 ml) was heated at 125° for 3 hours. The hydrobromic acid was evaporated at reduced pressure and the residue, made alkaline with aqueous potassium carbonate solution, was extracted with ethyl acetate. Removal of the solvent gave a product which was crystallized or purified over silica gel column eluting by an appropriate ethyl acetate/*n*-hexane mixture.

Procedure B: to obtain **15** and **21b**.

Compounds **14b,c** or **20b** (2 g) and excess of pyridinium chloride (20 g) were mixed and heated at 180° for 1 hour. The hot syrup was poured into water, the mixture was made alkaline with ammonium hydroxide and extracted with diethyl ether. Removal of the solvent gave a residue which was crystallized (**15b**) or chromatographed over silica gel eluting by 1:2 v/v ethyl acetate/*n*-hexane mixture (**15c** and **21b**).

2-(3-Hydroxyphenyl)-1-methyl-octahydrocyclopenta[*b*]pyrrole (**9a**).

This compound was obtained from **8a** in 95% yield as a viscous oil; ^{1}H-nmr (deuteriochloroform): δ 7.14 (t, 1H, H-5'), 6.86 (overlapped d and s, 2H, H-2' and H-4'), 6.68 (d, 1H, H-6'), 3.80 (bs, 1H, OH), 3.10 (q, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 10.6 Hz, $J_{2ax\text{-}3eq}$ = 5.5 Hz), 2.82 (dd, 1H, H-6a), 2.51 (m, 1H, H-3a), 2.25 (m, 1H, H-3eq), 2.08 (s, 3H, N-CH_3), 1.80 - 1.20 (m, 7H, H-4, H-5, H-6 and H-3ax); ms: (m/z) 217 (M^+), 188, 157, 124.

Anal. Calcd. for $C_{14}H_{19}NO$: C, 77.38; H, 8.81; N, 6.45. Found: C, 77.27; H, 8.95; N, 6.31.

2-(3-Hydroxyphenyl)-1-methyl-octahydro-1*H*-indole (**9b**).

This compound was obtained from **8b** in 75% yield as a viscous oil; ^{1}H-nmr (deuteriochloroform): δ 7.15 (t, 1H, H-5'), 6.90 (s, 1H, H-2'), 6.88 (d, 1H, H-4'), 6.66 (d, 1H, H-6'), 3.55 (bs, 1H, OH), 3.11 (dd, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 10.0 Hz, $J_{2ax\text{-}3eq}$ = 6.9 Hz), 2.33 (m, 2H, H-7a and H-3a), 2.07 (s, 3H, N-CH_3), 1.91 (m, 2H, H-3eq and H-7eq), 1.70 - 1.10 (m, 8H, H-7ax, H-4, H-5, H-6 and H-3ax), ms: (m/z) 231 (M^+), 188,157.

Anal. Calcd. for $C_{15}H_{21}NO$: C, 77.88; H, 9.15; N, 6.05. Found: 77.75; H, 9.23; N, 5.87.

2-(3-Hydroxyphenyl)-1-methyldecahydrocyclohepta[*b*]pyrrole (**9c**).

This compound was obtained from **8c** in 95% yield as a viscous oil; ^{1}H-nmr (deuteriochloroform): δ 7.14 (t, 1H, H-5'), 6.83 (overlapped d and s, 2H, H-4' and H-2'), 6.68 (d, 1H, H-6'), 3.80 (bs, 1H, OH), 3.12 (q, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 10.8 Hz, $J_{2ax\text{-}3eq}$ = 5.5 Hz), 2.45 (m, 1H, H-8a), 2.35 (m, 1H, H-3a), 2.28 (m, 1H, H-3eq), 2.08 (s, 3H, N-CH_3), 1.90 - 1.10 (m, 11H, H-8, H-4, H-5, H-6, H-7 and H-3ax); ms: (m/z) 245 (M^+), 188.

Anal. Calcd. for $C_{16}H_{23}NO$: C, 78.32; H, 9.45; N, 5.71. Found: C, 78.19; H, 9.59; N, 5.48.

2-(3-Hydroxyphenyl)-1-methyl-octahydro-1*H*-cyclopenta-[*b*]pyridine (**12a**).

This compound was obtained from **11a** in 70% yield, mp 143-145° (ethyl acetate); ^{1}H-nmr (deuteriochloroform): δ 7.11 (t, 1H, H-5'), 6.90 (s, 1H, H-2'), 6.81 (d, 1H, H-4'), 6.71 (d, 1H, H-6'), 6.51 (s, 1H, OH), 2.78 (dd, 1H, H-2, $J_{2\text{-}3ax}$ = 8.9 Hz, $J_{2\text{-}3eq}$ = 5.7 Hz), 1.99 (s, 3H, N-CH_3), 1.95 - 1.00 (m, 12H, H-7a, H-3, H-4a, H-4, H-5, H-6 and H-7); ms: (m/z) 231 (M^+).

Anal. Calcd. for $C_{15}H_{21}NO$: C, 77.88; H, 9.15; N, 6.05. Found: C, 77.98; H, 9.26; N, 6.30.

trans-2(e)-(3-Hydroxyphenyl)-1-methyldecahydroquinoline (**12b-I**).

This compound was obtained from **11b-I** in 68% yield as hydrobromide, mp 144-146° (methanol/water); ^{1}H-nmr (deuteriochloroform): δ 7.14 (t, 1H, H-5'), 6.89 (overlapped s and d, 2H, H-2' and H-4'), 6.67 (d, 1H, H-6'), 2.84 (dd, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 11.0 Hz, $J_{2ax\text{-}3eq}$ = 2.8 Hz), 2.15 (overlapped m, 3H, H-8a, H-4a and H-3eq), 1.90 (s, 3H, N-CH_3), 1.85 - 1.15 (m, 11H, H-4, H-8, H-5, H-6, H-7 and H-3ax), ms: (m/z) 245 (M^+), 202.

Anal. Calcd. for $C_{16}H_{24}BrNO$: C, 58.90; H, 7.41; N, 4.29. Found: C, 59.08; H, 7.24; N 3.99.

trans-2(a)-(3-Hydroxyphenyl)-1-methyldecahydroquinoline (**12b-II**).

This compound was obtained from **11b-II** in 65% yield as hydrobromide, mp 265-267° (methanol/water); ^{1}H-nmr (deuteriochloroform): δ 7.13 (t, 1H, H-5'), 6.90 (s, 1H, H-2'), 6.80 (d, 1H, H-4'), 6.71 (d, 1H, H-6'), 4.41 (s, 1H, OH), 2.90 (dd, 1H, H-2eq, $J_{2eq\text{-}3eq}$ = 8.9 Hz, $J_{2eq\text{-}3ax}$ = 5.3 Hz), 2.16 (m, 1H, H-8a), 2.01 (s, 3H, N-CH_3), 1.90 - 0.90 (m, 13H, H-4a, H-3, H-4, H-8, H-5, H-6 and H-7); ms: (m/z) 245 (M^+), 202, 152.

Anal. Calcd. for $C_{16}H_{24}BrNO$: C, 58.90; H, 7.41; N, 4.29. Found: C, 58.88; H, 7.30; N 4.08.

cis-2(e)-(3-Hydroxyphenyl)-1-methyldecahydroquinoline (**12b-III**).

This compound was obtained from **11b-III** in 65% yield as hydrobromide, mp 284-286° (methanol/water); ^{1}H-nmr (deuteriochloroform): δ 7.13 (t, 1H, H-5'), 6.93 (s, 1H, H-2'), 6.73 (two partially overlapped d, 2H, H-4' and H-6'), 5.17 (s, 1H, OH), 3.31 (dd, 1H, H-2ax, $J_{2ax-3ax}$ = 10.2 Hz, $J_{2ax-3eq}$ = 3.6 Hz), 2.92 (m, 1H, H-8a), 2.14 (m, 1H, H-3eq), 2.04 (s, 3H, N-CH_3), 2.00 - 1.00 (m, 12H, H-4, H-4a, H-5, H-8, H-6, H-7 and H-3ax); ms: (m/z) 245 (M^+), 202.

Anal. Calcd. for $C_{16}H_{24}BrNO$: C, 58.90; H, 7.41; N, 4.29. Found: C, 58.86; H, 7.29; N 4.02.

trans-2(e)-(3-Hydroxyphenyl)-1-methyldecahydro-1*H*-cyclohepta[*b*]pyridine (**12c-I**).

This compound was obtained from **11c-I** in 90% yield as a viscous oil; ^{1}H-nmr (deuteriochloroform): δ 7.13 (t, 1H, H-5'), 6.87 (overlapped d and s, 2H, H-4' and H-2'), 6.68 (d, 1H, H-6'), 3.00 (bs, 1H, OH), 2.84 (dd, 1H, H-2ax, $J_{2ax-3ax}$ = 9.9 Hz, $J_{2ax-3eq}$ = 3.3 Hz), 2.22 (m, 1H, H-9a), 2.04 (at, 1H, H-4a), 1.92 (s, 3H, N-CH_3), 1.85 - 1.20 (m, 14H, H-9, H-3, H-4, H-5, H-8, H-6 and H-7).); ms: (m/z) 259 (M^+), 202.

Anal. Calcd. for $C_{17}H_{25}NO$: C, 78.72; H, 9.71; N, 5.40. Found: C, 78.69; H, 9.85; N, 5.20.

cis-1,6-Dimethyl-2(e)-(3-hydroxyphenyl)-decahydro[1,6]naphthyridine (**12d-I**).

This compound was obtained from **11d-I** in 79% yield, mp 108-110° (methanol/water); ^{1}H-nmr (deuteriochloroform): δ 7.11 (t, 1H, H-5'), 6.82 (s, 1H, H-2'), 6.77 (d, 1H, H-4'), 6.68 (d, 1H, H-6'), 4.43 (s, 1H, OH), 3.24 (dd, 1H, H-2ax, $J_{2ax-3ax}$ = 10.4 Hz, $J_{2ax-3eq}$ = 3.1 Hz), 2.96 (d, 1H, H-5eq, J_{gem} = 10.2 Hz), 2.87 (dt, 1H, H-8a), 2.73 (d, 1H, H-7eq, J_{gem} = 11.5 Hz), 2.19 (s, 3H, N*6*-CH_3), 2.08 (d, 1H, H-7ax, J_{gem} = 11.5 Hz), 2.02 (s, 3H, N*1*-CH_3), 2.00 (m, 2H, H-4a and H-3eq), 1.87 (d, 1H, H-5ax, J_{gem} = 10.2 Hz), 1.80 - 1.40 (m, 5H, H-4, H-8 and H-3ax); ms: (m/z) 260 (M^+), 245, 229, 202.

Anal. Calcd. for $C_{16}H_{24}N_2O$: C, 73.81; H, 9.29; N, 10.76. Found: C, 73.99; H, 9.21; N, 10.57.

trans-1,6-Dimethyl-2(a)-(3-hydroxyphenyl)-decahydro[1,6]-naphthyridine (**12d-II**).

This compound was obtained from **11d-II** in 68% yield, mp 193-195° (methanol/water); ^{1}H-nmr (deuteriochloroform): δ 7.11 (t, 1H, H-5'), 6.91 (s, 1H, H-2'), 6.73 (d, 1H, H-4'), 6.67 (d, 1H, H-6'), 4.25 (bs, 1H, OH), 2.86 (m, 3H, H-5eq, H-7eq and H-2eq), 2.25 (s, 3H, N*6*-CH_3), 2.00 (overlapped s and m, 5H, N*1*-CH_3, H-8a and H-5ax), 1.65 (m, 7H, H-4a, H-3, H-4eq, H-7ax and H-8), 1.12 (m, 1H, H-4ax); ms: (m/z) 260 (M^+), 229, 216, 125.

Anal. Calcd. for $C_{16}H_{24}N_2O$: C, 73.81; H, 9.29; N, 10.76. Found: C, 73.90; H, 9.05; N, 10.88.

2-(3-Hydroxyphenyl)- 4,5,6,7-tetrahydro-1*H*-indole (**15b**).

This compound was obtained from **14b** in 91% yield, mp 139-141° (benzene); ^{1}H-nmr (DMSO-d_6): δ 10.6 (s, 1H, NH), 7.13 (t, 1H, H-5'), 6.99 (d, 1H, H-4'), 6.89 (s, 1H, H-2'), 6.50 (d, 1H, H-6'), 6.10 (s, 1H, H-3), 2.53 (partially under DMSO signal, at, 2H, H-7), 2.39 (at, 2H, H-4), 1.69 (m, 4H, H-5 and H-6); ms: (m/z) 213 (M^+).

Anal. Calcd. for $C_{14}H_{15}NO$: C, 78.84; H, 7.09; N, 6.57. Found: C, 78.68; H, 7.15; N, 6.33.

2-(3-Hydroxyphenyl)-1,4,5,6,7,8-hexahydrocyclohepta[*b*]-pyrrole (**15c**).

This compound was obtained from **14c** in 74% yield as a viscous oil; ^{1}H-nmr (deuteriochloroform): δ 8.10 (s, 1H, NH), 7.16 (t, 1H, H-5'), 6.94 (d, 1H, H-4'), 6.86 (s, 1H, H-2'), 6.59 (d, 1H, H-6'), 6.25 (s, 1H, H-3), 4.80 (bs, 1H, OH), 2.69 (at, 2H, H-8), 2.57 (at, 2H, H-4), 1.70 (m, 6H, H-7, H-5 and H-6); ms: (m/z) 227 (M^+).

Anal. Calcd. for $C_{15}H_{17}NO$: C, 79.26; H, 7.54; N, 6.16. Found: C, 79.17; H, 7.72; N, 6.01.

2-(3-Hydroxyphenyl)-1-methyl-4,5,6,7-tetrahydro-1*H*-indole (**18b**).

This compound was obtained from **17b** in 93% yield as an oil; ^{1}H-nmr (deuteriochloroform) : δ 7.22 (t, 1H, H-5'), 6.93 (d, 1H, H-4'), 6.82 (s, 1H, H-2'), 6.73 (d, 1H, H-6'), 6.00 (s, 1H, H-3), 4.85 (bs, 1H, OH), 3.46 (s, 3H, N-CH_3), 2.55 (partially overlapped triplets, 4H, H-7 and H-4), 1.85 (m, 4H, H-6 and H-5); ms: (m/z) 227 (M^+), 199.

Anal. Calcd. for $C_{15}H_{17}NO$: C, 79.26; H, 7.54; N, 6.16. Found: C, 79.08; H, 7.37; N, 6.45.

2-(3-Hydroxyphenyl)-1-methyl-1,4,5,6,7,8-hexahydrocyclohepta[*b*]pyrrole (**18c**).

This compound was obtained from **17c** in 70% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 8.00 (s, 1H, OH), 7.22 (t, 1H, H-5'), 6.92 (d, 1H, H-4'), 6.83 (s, 1H, H-2'), 6.71 (d, 1H, H-6'), 5.99 (s, 1H, H-3), 3.50 (s, 3H, N-CH_3), 2.74 (at, 2H, H-8), 2.61 (at, 2H, H-4), 1.80 (m, 4H, H-6 and H-5); ms: (m/z) 241 (M^+).

Anal. Calcd. for $C_{16}H_{19}NO$: C, 79.63; H, 7.94; N, 5.80. Found: C, 79.52; H, 7.80; N, 5.59.

2-(3-Hydroxyphenyl)-6,7-dihydro-5*H*-cyclopenta[*b*]pyridine (**21a**).

This compound was obtained from **20a** in 88% yield, mp 164-166° (ethyl acetate); ^{1}H-nmr (deuteriochloroform): δ 7.57-7.19 (m, 5H, H-2', H-6', H-3 , H-4 and H-5'), 6.79 (dd, 1H, H-4'), 3.07 (t, 2H, H-7), 2.93 (t, 2H, H-5), 2.12 (q, 2H, H-6); ms: (m/z) 211 (M^+), 183.

Anal. Calcd. for $C_{14}H_{13}NO$: C, 79.59; H, 6.20; N, 6.63. Found: C, 79.45; H, 5.98; N, 6.84.

2-(3-Hydroxyphenyl)-5,6,7,8-tetrahydroquinoline (**21b**).

This compound was obtained from **20b** in 90% yield as hydrobromide, mp 290-292° (water); ^{1}H-nmr (DMSO-d_6): δ 10.00 (bs, 1H, OH), 8.19 (d, 1H, H-4 or H-3), 7.93 (d, 1H, H-3 or H-4), 7.38 (t, 1H, H-5'), 7.36 - 7.28 (d, 2H, H-2' and H-6'), 7.01 (d, 1H, H-4'), 3.07 (t, 2H, H-8), 2.88 (t, 2H, H-5), 1.84 (m, 4H, H-6 and H-7); ms: (m/z) 225 (M^+), 197.

Anal. Calcd. for $C_{15}H_{16}BrNO$: C, 58.84; H, 5.27; N, 4.57. Found: C, 58.79; H, 5.38; N, 4.30.

2-(3-Hydroxyphenyl)-6,7,8,9-tetrahydro-5*H*-cyclohepta[*b*]-pyridine (**21c**).

This compound was obtained from **20c** in 90% yield, mp 239-241° (methanol/water); ^{1}H-nmr (DMSO-d_6): δ 8.30 (d, 1H, H-4 or H-3), 7.90 (d, 1H, H-3 or H-4), 7.39 - 7.29 (m, 3H, H-2', H-5' and H-6'), 7.01 (dd, 1H, H-4'), 3.27 (m, 2H, H-9), 2.97 (m, 2H, H-5), 1.84 - 1.67 (m, 6H, H-6, H-7 and H-8); ms: (m/z) 239 (M^+), 224, 210.

Anal. Calcd. for $C_{16}H_{17}NO$: C, 80.30; H, 7.16; N, 5.85. Found: C, 80.03; H, 7.45; N, 6.04.

2-(3-Hydroxyphenyl)-6-methyl-6,7,8,9-tetrahydro[1,6]-naphthyridine (**21d**).

This compound was obtained from **20d** in 91% yield, mp 182-184° (methanol/water); ^{1}H-nmr (DMSO-d_6): δ 10.16 (bs, 1H, OH), 7.85 (s, 2H, H-3 and H-4), 7.43 (d, 2H, H-2' and H-6'), 7.28 (t, 1H, H-5'), 6.86 (d, 1H, H-4'), 4.59 - 4.47 (m, 2H, H-5), 3.66 (bs, 1H, H-7), 3.46 (bs, 1H, H-8), 2.99 (s, 3H, N-CH_3); ms: (m/z) 240 (M^+), 197.

Anal. Calcd. for $C_{15}H_{16}N_2O$: C, 74.97; H, 6.71; N, 11.66. Found C, 74.85; H, 6.58; N, 11.42.

trans-3-(3-Hydroxyphenyl)-1-methyldecahydroquinoline (**29b**).

This compound was obtained from **28b** in 83% yield, mp 183-185° (ethyl acetate); ^{1}H-nmr (deuteriochloroform): δ 7.19 (t, 1H, H-5'), 6.76 - 6.71 (m, 3H, H-2', H-6' and H-4'), 3.09 (m, 2H, H-2eq and H-3), 2.27 (s, 3H, N-CH_3), 2.18 (m, 2H, H-2ax and H-8a), 1.84 - 1.02 (m, 11H, H-4, H-4a, H-8, H-5, H-6 and H-7); ms: (m/z) 245 (M^+), 202.

Anal. Calcd. for $C_{16}H_{23}NO$: C, 78.32; H, 9.45; N, 5.71. Found: C, 78.44; H, 9.58; N, 5.56.

cis-3-(3-Hydroxyphenyl)-1-methyldecahydro-1*H*-cyclohepta[*b*]-pyridine (**29c**).

This compound was obtained from **28c** in 98% yield, mp 142-143° (ethyl acetate); ^{1}H-nmr (deuteriochloroform): δ 9.50 (bs, 1H, OH), 7.19 (t, 1H, H-5'), 6.77 - 6.71 (m, 3H, H-2', H-4' and H-6'), 3.11 (d, 1H, H-2eq $J_{ax\text{-}eq}$=11.6), 2.93 (m, 1H, H-3), 2.24 (s, 3H, N-CH_3), 2.18 - 1.60 (m, 9H, H-2ax, H-9a, H-4, H-4a, H-9 and H-5), 1.50 - 1.18 (m, 6H, H-6, H-8 and H-7); ms: (m/z) 259 (M^+), 202.

Anal. Calcd. for $C_{17}H_{25}NO$: C, 78.72; H, 9.71; N, 5.40. Found: C, 78.98; H, 9.63; N, 5.23.

General Procedure for the Preparation of *N,N*-dimethylcarbamoyloxy-derivatives **10**, **13**, **16**, **19**, **22** and **30**.

To a stirred solution of the appropriate phenolderivative (0.01 mole) in anhydrous pyridine (30 ml) cooled at 0°, *N,N*-dimethylcarbamoyl chloride (1.6 g, 0.015 mole) was added dropwise. The mixture was stirred overnight at room temperature, then evaporated to dryness under reduced pressure at no more than 40°. After addition of water, the mixture was extracted with ethyl acetate, the solvent removed and the residue chromatographed on alumina column by eluting with a 1:3 v/v ethyl acetate/*n*-hexane mixture. Several carbamoylderivatives are viscous oils, so analytical samples were yielded by carefully drying the products purified by chromatography. Solid carbamates were instead purified by crystallization.

2-(3-Dimethylcarbamoyloxyphenyl)-1-methyloctahydrocyclopenta[*b*]pyrrole (**10a**).

This compound was obtained from **9a** in 75% yield as a viscous oil; ^{1}H-nmr (deuteriochloroform): δ 7.25 (t, 1H, H-5'), 7.15 (d, 1H, H-4'), 7.07 (s, 1H, H-2'), 6.97 (d, 1H, H-6'), 3.13 (q, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 10.9 Hz, $J_{2ax\text{-}3eq}$ = 5.5 Hz), 3.07 (s, 3H, carbamic N-CH_3), 2.98 (s, 3H, carbamic N-CH_3), 2.82 (dd, 1H, H-6a, $J_{6a\text{-}6ax}$ = 8.0 Hz, $J_{6a\text{-}3a}$ = 6.0 Hz), 2.51 (m, 1H, H-3a), 2.25 (m, 1H, H-3eq), 2.07 (s, 3H, N-CH_3), 1.80 - 1.20 (m, 7H, H-4, H-5, H-6 and H-3ax); ms: (m/z) 288 (M^+), 259, 124.

Anal. Calcd. for $C_{17}H_{24}N_2O_2$: C, 70.80; H, 8.39; N, 9.71. Found: C, 70.67; H, 8.19; N, 9.54.

2-(3-Dimethylcarbamoyloxyphenyl)-1-methyloctahydro-1*H*-indole (**10b**).

This compound was obtained from **9b** in 51% yield as a viscous oil; ^{1}H-nmr (deuteriochloroform): δ 7.27 (t, 1H, H-5'), 7.19 (d, 1H, H-4'), 7.08 (s, 1H, H-2'), 6.95 (d, 1H, H-6'), 3.15 (dd, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 9.8 Hz, $J_{2ax\text{-}3eq}$ = 6.8 Hz), 3.09 (s, 3H, carbamic N-CH_3), 2.99 (s, 3H, carbamic N-CH_3), 2.31 (m, 2H, H-7a and H-3a), 2.06 (s, 3H, N-CH_3), 1.89 (m, 2H, H-3eq and H-7eq), 1.70 - 1.10 (m, 8H, H-7ax, H-4, H-5, H-6 and H-3ax); ms: (m/z) 302 (M^+).

Anal. Calcd. for $C_{18}H_{26}N_2O_2$: C, 71.49; H, 8.67; N, 9.26. Found: C, 71.26; H, 8.44; N, 9.11.

2-(3-Dimethylcarbamoyloxyphenyl)-1-methyldecahydrocyclohepta[*b*]pyrrole (**10c**).

This compound was obtained from **9c** in 82% yield as a viscous oil; ^{1}H-nmr (deuteriochloroform): δ 7.28 (t, 1H, H-5'), 7.14 (d, 1H, H-4'), 7.11 (s, 1H, H-2'), 6.99 (d, 1H, H-6'), 3.17 (q, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 10.8 Hz, $J_{2ax\text{-}3eq}$ = 5.5 Hz), 3.10 (s, 3H, carbamic N-CH_3), 3.01 (s, 3H, carbamic N-CH_3), 2.43 (m, 1H, H-8a), 2.30 (m, 1H, H-3a), 2.18 (m, 1H, H-3eq), 2.07 (s, 3H, N-CH_3), 1.90 - 1.10 (m, 11H, H-8, H-4, H-5, H-6, H-7 and H-3ax); ms: (m/z) 316 (M^+), 259.

Anal. Calcd. for $C_{19}H_{28}N_2O_2$: C, 72.12; H, 8.92; N, 8.85. Found: C, 72.25; H, 8.90; N, 8.59.

2-(3-Dimethylcarbamoyloxyphenyl)-1-methyloctahydro-1*H*-cyclopenta[*b*]pyridine (**13a**).

This compound was obtained from **12a** in 77% yield, mp 46-48° (diethyl ether/*n*-hexane); ^{1}H-nmr (deuteriochloroform): δ 7.25 (t, 1H, H-5'), 7.12 (d, 1H, H-4'), 7.10 (s, 1H, H-2'), 6.98 (d, 1H, H-6'), 3.07 (s, 3H, carbamic N-CH_3), 2.98 (s, 3H, carbamic N-CH_3), 2.80 (dd, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 11.0 Hz, $J_{2ax\text{-}3eq}$ = 3.4 Hz), 1.97 (s, 3H, N-CH_3), 1.95 - 1.00 (m, 12H, H-7a, H-3, H-4a, H-4, H-5, H-6 and H-7); ms: (m/z) 302 (M^+), 273, 138.

Anal. Calcd. for $C_{18}H_{26}N_2O_2$: C, 71.49; H, 8.67; N, 9.26. Found: C, 71.62; H, 8.44; N, 9.41.

trans-2(e)-(3-Dimethylcarbamoyloxyphenyl)-1-methyldecahydroquinoline (**13b-I**).

This compound was obtained from **12b-I** in 58% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.25 (t, 1H, H-5'), 7.15 (d, 1H, H-4'), 7.07 (s, 1H, H-2'), 6.95 (d, 1H, H-6'), 3.09 (s, 3H, carbamic N-CH_3), 2.98 (s, 3H, carbamic N-CH_3), 2.86 (dd, 1H, H-2ax, $J_{2ax\text{-}3ax}$ = 10.9 Hz, $J_{2ax\text{-}3eq}$ = 2.8 Hz), 2.15 (overlapped m, 3H, H-8a, H-4a and H-3eq), 1.89 (s, 3H, N-CH_3), 1.85 - 1.15 (m, 11H, H-4, H-8, H-5, H-6, H-7 and H-3ax); ms (m/z) 316 (M^+), 273.

Anal. Calcd. for $C_{19}H_{28}N_2O_2$: C, 72.12; H, 8.92; N, 8.85. Found: C, 72.00; H, 9.12; N, 9.10.

trans-2(a)-(3-Dimethylcarbamoyloxyphenyl)-1-methyldecahydroquinoline (**13b-II**).

This compound was obtained from **12b-II** in 56% yield, mp 37-39° (diethyl ether/*n*-hexane); ^{1}H-nmr (deuteriochloroform): δ 7.26 (t, 1H, H-5'), 7.11 (d, 1H, H-4'), 7.08 (s, 1H, H-2'), 6.96 (d, 1H, H-6'), 3.08 (s, 3H, carbamic N-CH_3), 3.00 (s, 3H, carbamic N-CH_3), 2.87 (dd, 1H, H-2eq, $J_{2eq\text{-}3ax}$ = 8.9 Hz, $J_{2eq\text{-}3eq}$ = 4.9 Hz), 2.15 (ad, 1H, H-8a), 1.99 (s, 3H, N-CH_3), 1.90 - 0.90 (m, 13H, H-4a, H-3, H-4, H-8, H-5, H-6 and H-7); ms: (m/z) 316 (M^+), 273, 152.

Anal. Calcd. for $C_{19}H_{28}N_2O_2$: C, 72.12; H, 8.92; N, 8.85. Found: C, 71.98; H, 8.85; N, 8.63.

cis-2(e)-(3-Dimethylcarbamoyloxyphenyl)-1-methyldecahydroquinoline (**13b-III**).

This compound was obtained from **12b-III** in 59% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.24 (t, 1H, H-5'), 7.09 (d, 1H, H-4'), 7.07 (s, 1H, H-2'), 6.96 (d, 1H, H-6'), 3.31 (dd, 1H, H-2ax, $J_{2ax-3ax}$ = 11.1 Hz, $J_{2ax-3eq}$ = 3.2 Hz), 3.06 (s, 3H, carbamic N-CH_3), 2.97 (s, 3H, carbamic N-CH_3), 2.86 (dt, 1H, H-8a), 2.05 (m, 1H, H-3eq), 2.03 (s, 3H, N-CH_3), 2.00 - 1.00 (m, 12H, H-4a, H-4, H-5, H-8, H-6, H-7 and H-3ax); ms: (m/z) 316 (M^+), 273, 152.

Anal. Calcd. for $C_{19}H_{28}N_2O_2$: C, 72.12; H, 8.92; N, 8.85. Found: C, 72.31; H, 8.83; N, 9.09.

trans-2(e)-(3-Dimethylcarbamoyloxyphenyl)-1-methyldecahydro-1*H*-cyclohepta[*b*]pyridine (**13c-I**).

This compound was obtained from **12c-I** in 71% yield, mp 68-69° (diethyl ether/petroleum ether); ^{1}H-nmr (deuteriochloroform): δ 7.25 (t, 1H, H-5'), 7.15 (d, 1H, H-4'), 7.08 (s, 1H, H-2'), 6.94 (d, 1H, H-6'), 3.08 (s, 3H, carbamic N-CH_3), 2.99 (s, 3H, carbamic N-CH_3), 2.85 (dd, 1H, H-2ax, $J_{2ax-3ax}$ = 9.8 Hz, $J_{2ax-3eq}$ = 3.5 Hz), 2.22 (m, 1H, H-9a), 2.04 (m, 1H, H-4a), 1.92 (s, 3H, N-CH_3), 1.85 - 1.20 (m, 14H, H-9, H-3, H-4, H-5, H-8, H-6 and H-7); ms: (m/z) 330 (M^+).

Anal. Calcd. for $C_{20}H_{30}N_2O_2$: C, 72.69; H, 9.15; N, 8.48. Found: C, 72.82; H, 9.28; N, 8.20.

cis-1,6-Dimethyl-2(e)-(3-dimethylcarbamoyloxyphenyl)decahydro[1,6]naphthyridine (**13d-I**).

This compound was obtained from **12d-I** in 60% yield, mp 71-73° (ethyl acetate/*n*-hexane); ^{1}H-nmr (deuteriochloroform): δ 7.24 (t, 1H, H-5'), 7.10 (d, 1H, H-4'), 7.04 (s, 1H, H-2'), 6.96 (d, 1H, H-6'), 3.29 (dd, 1H, H-2ax, $J_{2ax-3ax}$ = 11.0 Hz, $J_{2ax-3eq}$ = 2.9 Hz), 3.07 (s, 3H, carbamic N-CH_3), 2.97 (s, 3H, carbamic N-CH_3), 2.91 (d, 1H, H-5eq), 2.85 (m, 1H, H-8a), 2.69 (d, 1H, H-7eq, J_{gem} = 11.4 Hz), 2.18 (s, 3H, N6-CH_3), 2.08 (dd, 1H, H-7ax, J_{gem} = 11.4 Hz), 2.03 (s, 3H, N1-CH_3), 1.92 (m, partially under N1-CH_3 signal, 2H, H-4a and H-3eq), 1.90 - 1.40 (m, 6H, H-5ax, H-4, H-8 and H-3ax); ms: (m/z) 331 (M^+), 316, 273.

Anal. Calcd. for $C_{19}H_{29}N_3O_2$: C, 68.85; H, 8.82; N, 12.68. Found: C, 68.61; H, 8.63; N, 12.48.

trans-1,6-Dimethyl-2(a)-(3-dimethylcarbamoyloxyphenyl)decahydro[1,6]naphthyridine (**13d-II**).

This compound was obtained from **12d-II** in 62% yield, mp 80-82° (ethyl acetate/*n*-hexane); ^{1}H-nmr (deuteriochloroform): δ 7.25 (t, 1H, H-5'), 7.10 (d, 1H, H-4'), 7.07 (s, 1H, H-2'), 6.95 (d, 1H, H-6'), 3.07 (s, 3H, carbamic N-CH_3), 2.98 (s, 3H, carbamic N-CH_3), 2.89 (m, 2H, H-5eq and H-7eq), 2.71 (dd, 1H, H-2eq, $J_{2eq-3eq}$ = 7.4 Hz, $J_{2eq-3ax}$ = 1.6 Hz), 2.23 (s, 3H, N6-CH_3), 2.00 (overlapped m and s, 5H, H-8a, H-5ax and N1-CH_3), 1.62 (m, 7H, H-4a, H-3, H-4eq, H-7ax and H-8), 1.12 (m, 1H, H-4ax); ms: (m/z) 331 (M^+), 316, 273.

Anal. Calcd. for $C_{19}H_{29}N_3O_2$: C, 68.85; H, 8.82; N, 12.68. Found: C, 69.05; H, 8.98; N, 12.41.

2-(3-Dimethylcarbamoyloxyphenyl)- 4,5,6,7-tetrahydro-1*H*-indole (**16b**).

This compound was obtained from **15b** in 43% yield, mp 178-180° (ethyl acetate); ^{1}H-nmr (deuteriochloroform): δ 7.97 (bs, 1H, NH), 7.28 (t, 1H, H-5'), 7.21 (d, 1H, H-4'), 7.15 (s, 1H, H-2'), 6.87 (d, 1H, H-6'), 6.24 (s, 1H, H-3), 3.09 (s, 3H, carbamic N-CH_3), 3.01 (s, 3H, carbamic N-CH_3), 2.59 (t, 2H, H-7), 2.51 (t, 2H, H-4), 1.78 (m, 4H, H-5 and H-6); ms: (m/z) 284 (M^+), 256.

Anal. Calcd. for $C_{17}H_{20}N_2O_2$: C, 71.81; H, 7.09; N, 9.85. Found: C, 71.67; H, 7.11; N, 9.62.

2-(3-Dimethylcarbamoyloxyphenyl)-1,4,5,6,7,8-hexahydrocyclohepta[*b*]pyrrole (**16c**).

This compound was obtained from **15c** in 76% yield, mp 183-185° (ethyl acetate); ^{1}H-nmr (deuteriochloroform): δ 8.20 (bs, 1H, NH), 7.25 (t, 1H, H-5'), 7.19 (d, 1H, H-4'), 7.12 (s, 1H, H-2'), 6.85 (d, 1H, H-6'), 6.24 (s, 1H, H-3), 3.10 (s, 3H, carbamic N-CH_3), 3.01 (s, 3H, carbamic N-CH_3), 2.64 (at, 2H, H-8), 2.56 (at, 2H, H-4), 1.70 (m, 6H, H-5, H-6 and H-7); ms: (m/z) 298 (M^+), 269.

Anal. Calcd. for $C_{18}H_{22}N_2O_2$: C, 72.46; H, 7.43; N, 9.39. Found: C, 72.63; H, 7.58; N, 9.17.

2-(3-Dimethylcarbamoyloxyphenyl)-1-methyl-4,5,6,7-tetrahydro-1*H*-indole (**19b**).

This compound was obtained from **18b** in 46% yield, mp 99-101° (ethyl acetate/*n*-hexane); ^{1}H-nmr (deuteriochloroform): δ 7.33 (t, 1H, H-5'), 7.19 (d, 1H, H-4'), 7.11 (s, 1H, H-2'), 7.01 (d, 1H, H-6'), 6.02 (s, 1H, H-3), 3.47 (s, 3H, N-CH_3), 3.10 (s, 3H, carbamic N-CH_3), 3.00 (s, 3H, carbamic N-CH_3), 2.58 (t, 2H, H-7), 2.52 (t, 2H, H-4), 1.80 (m, 4H, H-5 and H-6), ms: (m/z) 298 (M^+), 270, 241, 198.

Anal. Calcd. for $C_{18}H_{22}N_2O_2$: C, 72.46; H, 7.43; N, 9.39. Found: C, 72.21; H, 7.56; N, 9.15.

2-(3-Dimethylcarbamoyloxyphenyl)-1-methyl-1,4,5,6,7,8-hexahydrocyclohepta[*b*]pyrrole (**19c**).

This compound was obtained from **18c** in 52% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.32 (t, 1H, H-5'), 7.15 (d, 1H, H-4'), 7.07 (s, 1H, H-2'), 6.99 (d, 1H, H-6'), 5.98 (s, 1H, H-3), 3.49 (s, 3H, N-CH_3), 3.09 (s, 3H, carbamic N-CH_3), 3.01(s, 3H, carbamic N-CH_3), 2.71 (at, 2H, H-8), 2.58 (at, 2H, H-4), 1.75 (m, 6H, H-5, H-6 and H-7); ms: (m/z) 312 (M^+), 273.

Anal. Calcd. for $C_{19}H_{24}N_2O_2$: C, 73.05; H, 7.74; N, 8.97. Found: C, 72.97; H, 7.68; N, 8.80.

2-(3-Dimethylcarbamoyloxyphenyl)-6,7-dihydro-5*H*-cyclopenta[*b*]pyridine (**22a**).

This compound was obtained from **21a** in 52% yield, mp 92-93° (ethyl acetate); ^{1}H-nmr (deuteriochloroform): δ 7.74 (d, 2H, H-6' and H-2'), 7.54 - 7.36 (m, 3H, H-5', H-4 and H-3), 7.10 (dd, 1H, H-4'), 3.09 (s, 3H, N-CH_3), 3.05 (t, 2H, H-7), 3.00 (s, 3H, N-CH_3), 2.93 (t, 2H, H-5), 2.13 (m, 2H, H-6); ms: (m/z) 282, 238.

Anal. Calcd. for $C_{17}H_{18}N_2O_2$: C, 72.32; H, 6.43; N, 9.92. Found: C, 72.55; H, 6.35; N, 10.00.

2-(3-Dimethylcarbamoyloxyphenyl)-5,6,7,8-tetrahydroquinoline (**22b**).

This compound was obtained from **21b** in 66% yield, mp 104-105° (ethyl acetate); ^{1}H-nmr (deuteriochloroform): δ 7.77 - 7.71 (m, 2H, H-2' and H-6'), 7.43 - 7.35 (m, 3H, H-4, H-3 and H-5'), 7.11 (dd, 1H, H-4'), 3.08 (s, 3H, N-CH_3), 2.99 (s, 3H, N-CH_3), 2.94 (m, 2H, H-8), 2.75 (t, 2H, H-5), 1.83 (m, 4H, H-6 and H-7); ms: (m/z) 296 (M^+), 252.

Anal. Calcd. for $C_{18}H_{20}N_2O_2$: C, 72.95; H, 6.80; N, 9.45. Found: C, 72.76; H, 7.03; N, 9.18.

2-(3-Dimethylcarbamoyloxyphenyl)-6,7,8,9-tetrahydro-5*H*-cyclohepta[*b*]pyridine (**22c**).

This compound was obtained from **21c** in 52% yield, mp 132-134° (ethyl acetate / *n*-hexane); ^{1}H-nmr (deuteriochloroform): δ 7.79 - 7.74 (m, 2H, H-2' and H-6'), 7.43 - 7.24 (t, 3H, H-3, H-4 and H-5'), 7.11 (dd, 1H, H-4'), 3.10 (m, 5H, H-9 and N-CH_3), 3.00 (s, 3H, N-CH_3), 2.77 (m, 2H, H-5), 1.87 - 1.65 (m, 6H, H-6, H-7 and H-8); ms: (m/z) 310 (M^+), 265.

Anal. Calcd. for $C_{19}H_{22}N_2O_2$: C, 73.52; H, 7.14; N, 9.03. Found C, 73.36; H, 7.32; N, 8.88.

2-(3-Dimethylcarbamoyloxyphenyl)-6-methyl-6,7,8,9-tetrahydro[1,6]naphthyridine (**22d**).

This compound was obtained from **21d** in 61% yield, mp 126-128° (ethyl acetate); ^{1}H-nmr (deuteriochloroform): δ 7.77 - 7.70 (dd, 2H, H-2' and H-6'), 7.47-7.32 (m, 3H, H-3, H-4 and H-5'), 7.15 - 7.09 (dd, 1H, H-4'), 3.60 (s, 2H, H-5), 3.13 - 3.08 (m, 2H, H-7), 3.09 (s, 3H, carbamic N-CH_3), 2.99 (s, 3H, carbamic N-CH_3), 2.80 (t, 2H, H-8), 2.48 (s, 3H, N6-CH_3); ms: (m/z) 311 (M^+), 296, 210.

Anal. Calcd. for $C_{18}H_{21}N_3O_2$: C, 69.43; H, 6.80; N, 13.49. Found: C, 69.22; H, 6.69; N, 13.65.

trans-3-(3-Dimethylcarbamoyloxyphenyl)-1-methyldecahydroquinoline (**30b**).

This compound was obtained from **29b** in 60% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.23 (t, 1H, H-5'), 7.07 - 6.94 (m, 3H, H-2', H-4' and H-6'), 3.09 (s, 3H, N-CH_3), 3.01 (m, 5H, carbamic N-CH_3, H-2eq and H-3), 2.28 (s, 3H, carbamic N-CH_3), 2.14 (t, 2H, H-2ax and H-8a), 1.86 - 1.08 (m, 11H, H-4, H-4a, H-8, H-5, H-6 and H-7), ms: (m/z) 316 (M^+), 273.

Anal. Calcd. for $C_{19}H_{28}N_2O_2$: C, 72.12; H, 8.92; N, 8.85. Found: C, 71.96; H, 8.70; N, 8.98.

cis-3-(3-Dimethylcarbamoyloxyphenyl)-1-methyldecahydro-1*H*-cyclohepta[*b*]pyridine (**30c**).

This compound was obtained from **29c** in 50% yield as an oil; ^{1}H-nmr (deuteriochloroform): δ 7.24 (t, 1H, H-5'), 7.02 (d, 1H, H-4'), 6.94 - 6.90 (m, 2H, H-2' and H-6'), 3.07 (s, 3H, carbamic N-CH_3), 2.98 (s, 3H, carbamic N-CH_3), 2.91 (m, 2H, H-2eq and H-3), 2.19 (s, 3H, N-CH_3), 2.09 - 1.10 (m, 15H, H-2ax, H-9a, H-4, H-4a, H-9, H-5, H-6, H-8 and H-7); ms: (m/z) 330 (M^+), 273.

Anal. Calcd. for $C_{20}H_{30}N_2O_2$: C, 72.69; H, 9.15; N, 8.48. Found C, 72.83; H, 9.35; N, 8.21.

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