A new method for the synthesis of *N*-substituted (4-oxo-4,5,6,7-tetrahydroindol-3-yl)acetic acids

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A new method for the synthesis of *N*-substituted 2-(4-oxo-4,5,6,7-tetrahydroindol-3-yl)acetic acids was developed. Alkylation of cyclic 1,3-diketones with 3,5,5,5-tetrachloropentan-2-one affords 1,4-diketones, which undergo cyclization with different primary amines into *N*-substituted 3-(2,2,2-trichloroethyl)-4,5,6,7-tetrahydroindol-4-ones. Acid hydrolysis of the latter gives the corresponding indol-3-ylacetic acids. The structures of the compounds obtained were confirmed by ^{1}H and ^{13}C NMR data.

Key words: cyclic 1,3-diketones, 3,5,5,5-tetrachloropentan-2-one, aliphatic and aromatic amines, cyclization, *N*-substituted 3-(2,2,2-trichloroethyl)-4,5,6,7-tetrahydroindol-4-ones, hydrolysis of the trichloroethyl group, *N*-substituted (4-oxo-4,5,6,7-tetrahydroindol-3-yl)acetic acids.

Indolylacetic acids are used as drugs[1] and plant growth regulators.[2] At present, an intensive search for compounds with potential biological activity is being carried out among partially hydrogenated heterocyclic compounds. Some partially hydrogenated indolylacetic acids have been tested as peroral hypoglycemic drugs, which is very important in the treatment of diabetes.[3]

In the present work, a new method for the synthesis of *N*-substituted (4-oxo-4,5,6,7-tetrahydroindol-3-yl)acetic acids is proposed.

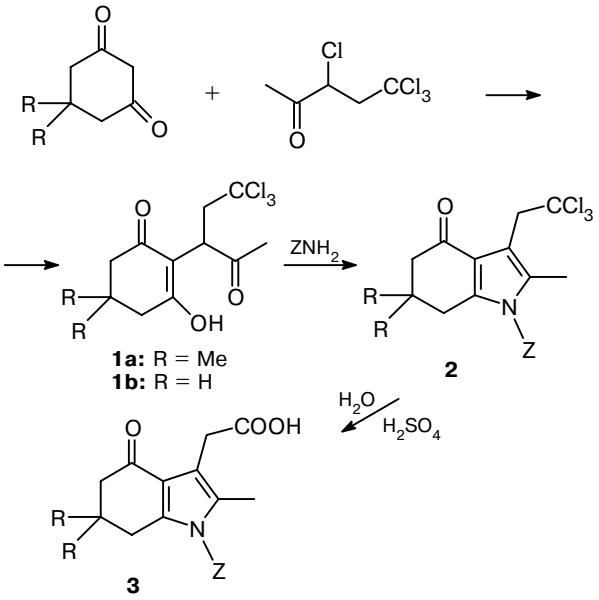
Results and Discussion

The first step of our three-step synthesis is based on a common alkylation of dimedone or cyclohexanedione with α-halogenocarbonyl compounds.[4—6] Easily accessible 3,5,5,5-tetrachloropentan-2-one (TCP) employed previously for constructing various heterocyclic systems[7,8] was used as the alkylating agent.

The resulting 1,4-diketones (**1**) were brought into reactions with aliphatic and aromatic primary amines or with 1,1-disubstituted hydrazines in boiling acetic acid for several hours. Cyclization gave the corresponding *N*-substituted 2-methyl-3-(2,2,2-trichloroethyl)tetrahydroindol-4-ones (**2**) in 60—85% yields.

Then the trichloromethyl group was hydrolyzed to the COOH group. It is known[9] that the hydrolysis of aliphatic trichloromethyl derivatives to carboxylic acids requires concentrated H_2SO_4 (≥94%). For this reason, we hydrolyzed trichloroethyl derivatives in a hot mixture of sulfuric acid with oleum to provide a 95% concentration of H_2SO_4. The corresponding indolylacetic acids (**3**) were obtained in 50—90% yields (Scheme 1).

Scheme 1



This method have some advantages over the published procedures,[3,10] providing little (if any) by-products, which hampers isolation of the target compounds in each step, and higher yields of the corresponding indolylacetic acids.

The structures of compounds **2** and **3** were confirmed by ^{1}H and ^{13}C NMR data and by elemental analysis (Tables 1—3).

In some compounds containing the 2,5-disubstituted benzene ring or a similar heterocyclic fragment, the CH_2X protons (X = CCl_3 or COOH) give two signals in

Published in Russian in *Izvestiya Akademii Nauk. Seriya Khimicheskaya,* No. 7, pp. 1201—1205, July, 2001.

1066-5285/01/5007-1259 $25.00

Table 1. Yields, melting points, and elemental analysis data for compounds **2** and **3**

Compound	Substituents		Yield (%)	M.p. /°C	Found / Calculated (%)				Molecular formula
	R	Z			C	H	N	Cl	
2a	CH_3	3-Cl-4-MeC_6H_3	156—157	85	55.57 55.45	4.81 4.89	3.39 3.23	32.24 32.74	$C_{20}H_{21}Cl_4NO$
2b	H	4-FC_6H_4	168—169	68	54.72 54.50	3.85 4.04	3.50 3.74	—	$C_{17}H_{15}Cl_3FNO$
2c	CH_3	4-$NO_2C_6H_4$	176—177	72	52.93 52.86	5.01 4.90	6.55 6.49	—	$C_{19}H_{21}Cl_3N_2O_3$
2d	CH_3	4-BrC_6H_4	168—169	82	49.48 49.22	4.13 4.13	3.30 3.02	22.87 22.94	$C_{19}H_{19}BrCl_3NO$[a]
2e	CH_3	2-Me-5-$NO_2C_6H_3$	179—180	78	54.10 54.13	4.66 4.77	6.15 6.28	—	$C_{20}H_{21}Cl_3N_2O_3$
2f	CH_3	2,4-$F_2C_6H_3$	180—182	70	53.95 54.24	4.09 4.31	3.50 3.33	—	$C_{19}H_{18}Cl_3F_2NO$
2g	CH_3	3-FC_6H_4	157—159	74	56.31 56.67	4.98 4.76	3.65 3.48	—	$C_{19}H_{19}Cl_3FNO$
2h	CH_3	2-Cl-5-$CF_3C_6H_3$	190—192	59	49.02 49.31	3.95 3.72	3.05 2.88	—	$C_{20}H_{18}Cl_4F_3NO$
2i	CH_3	Bu^n	137—138	56	55.91 55.98	6.70 6.63	3.75 3.84	—	$C_{17}H_{24}Cl_3NO$
2j	CH_3	4-$CF_3OC_6H_4$	142—143	81	50.98 51.25	4.35 4.09	2.74 2.99	—	$C_{20}H_{19}Cl_3F_3NO_2$
2k	CH_3	3-NO_2-4-FC_6H_3	180—181	73	50.47 50.74	4.73 4.48	6.41 6.23	—	$C_{19}H_{20}Cl_3FN_2O_2$
2l	CH_3	4-$H_2NSO_2C_6H_4$	242—244	83	49.07 49.16	4.64 4.56	5.86 6.04	22.50 22.93	$C_{19}H_{21}Cl_3N_2O_3$
2m	H	2-$NO_2C_6H_4$	162—163	53	50.89 50.83	3.57 3.76	6.89 6.97	—	$C_{17}H_{15}Cl_3N_2O_3$
2n	H	3-Pyridyl	180—181	60	53.77 53.73	4.23 4.23	7.64 7.83	29.55 29.74	$C_{16}H_{15}Cl_3N_2O$
2o	CH_3	$CH_2C_6H_5$	117—118	62	60.22 60.24	5.57 5.56	3.45 3.51	26.39 26.67	$C_{20}H_{22}Cl_3NO$
2p	CH_3	4-ClC_6H_4	187—188	70	54.48 54.44	4.47 4.57	2.97 3.34	33.61 33.82	$C_{19}H_{19}Cl_4NO$
2q	CH_3	*n*-C_5H_{12}	121—122	58	57.01 57.08	7.04 6.92	3.48 3.70	27.95 28.08	$C_{18}H_{26}Cl_3NO$
2r	CH_3	CH_2CH_2COOH	179—180	55	50.74 50.48	5.21 5.30	3.65 3.68	27.54 27.94	$C_{16}H_{20}Cl_3NO_3$
2s	CH_3	—CH_2CO_2Et	141—142	57	51.71 51.73	5.80 5.62	3.48 3.55	26.92 26.95	$C_{17}H_{22}Cl_3NO_3$
2t	CH_3	—NMe_2	155—156	64	51.30 51.22	5.74 6.02	8.11 7.97	29.97 30.24	$C_{15}H_{21}Cl_3N_2O$
2u	CH_3	3-Cyano-4,6-dimethyl-2-oxo-1,2-dihydro-1-pyridyl	191—192	34	55.41 55.46	4.89 4.88	9.32 9.24	23.34 23.39	$C_{21}H_{22}Cl_3N_3O_2$
2v	H	Morpholino	157—158	67	49.40 49.27	5.03 5.24	7.58 7.6	—	$C_{15}H_{19}Cl_3N_2O_2$
3a	CH_3	3-Cl-4-MeC_6H_3	153—155	53	67.03 66.75	6.38 6.16	4.29 3.89	9.87 9.85	$C_{20}H_{22}ClNO_3$
3b	H	4-FC_6H_4	157—158	74	67.97 67.76	5.60 5.35	4.49 4.65	—	$C_{17}H_{16}FNO_3$

(*to be continued*)

Table 1 (*continued*)

Compound	Substituents		Yield (%)	M.p. /°C	Found / Calculated (%)				Molecular formula
	R	Z			C	H	N	Cl	
3c	CH_3	4-$NO_2C_6H_4$	220—222	78	63.60 63.68	6.25 6.19	7.71 7.82	—	$C_{19}H_{22}N_2O_5$
3d	CH_3	4-BrC_6H_4	182—184	55	58.54 58.47	5.29 5.17	3.67 3.59	—	$C_{19}H_{20}BrNO_3$[b]
3e	CH_3	2-Me-5-$NO_2C_6H_3$	100—101	78	64.91 64.85	5.88 5.99	7.45 7.52	—	$C_{20}H_{22}N_2O_5$
3f	CH_3	2,4-$F_2C_6H_3$	103—105	64	65.46 65.70	5.35 5.51	3.85 4.03	—	$C_{19}H_{19}F_2NO_3$
3g	CH_3	3-FC_6H_4	179—181	53	69.01 69.29	6.45 6.12	4.53 4.25	—	$C_{19}H_{20}FNO_3$
3h	CH_3	2-Cl-5-$CF_3C_6H_3$	106—108	86	58.30 58.05	4.91 4.63	3.12 3.38	—	$C_{20}H_{19}ClF_3NO_3$
3i[c]	CH_3	Bu^n	109—110	63	69.88 70.07	8.73 8.65	4.70 4.81	—	$C_{17}H_{25}NO_3$

[a] Found (%): Br, 17.19. Calculated (%): 17.24.
[b] Found (%): Br, 20.50. Calculated (%): 20.47.
[c] *Cf.* Ref. 3: m.p. 120—122 °C.

Table 2. ^{1}H NMR data (DMSO-d_6) for compounds **2**

Compound	δ (*J*/Hz)				
	$C(CH_3)_2$ (s)	CH_2	CH_3 (s, 3 H)	CH_2CCl_3 (s, 2 H)	Z
2a	1.00 (6 H)	2.42 (s, 2 H); 2.25 (s, 2 H)	2.08	4.26	7.55 (d, 1 H); 7.45 (d, 1 H); 7.24 (dd, 1 H)
2b	—	2.52 (m, 2 H); 2.34 (m, 2 H); 1.99 (m, 2 H)	2.07	4.29	7.44 (m, 4 H)
2c	1.00 (6 H)	2.48 (s, 2 H); 2.27 (s, 2 H)	2.11	4.29	8.38 (d, 2 H); 7.72 (d, 2 H)
2d	1.00 (6 H)	2.43 (s, 2 H); 2.27 (s, 2 H)	2.08	4.29	7.76 (d, 2 H, *J* = 9); 7.35 (d, 2 H, *J* = 9)
2e	1.03 (3 H); 0.99 (3 H)	2.49—2.08 (m, 4 H)	2.04	4.32 (d, 1 H, *J* = 15) 4.30 (d, 1 H, *J* = 15)	8.34 (dd, 1 H, *J* = 8, 2.5); 8.13 (d, 1 H, *J* = 8); 7.78 (dd, 1 H, *J* = 8, 2.5)
2f	1.02 (3 H); 0.97 (3 H)	2.47—2.22 (m, 4 H)	2.04	4.28	7.54 (m, 2 H); 7.33 (m, 1 H)
2g	1.00 (6 H)	2.47 (s, 2 H); 2.26 (s, 2 H)	2.08	4.28	7.64 (m, 1 H); 7.42 (m, 2 H); 7.26 (m, 1 H)
2h	1.01 (3 H); 0.96 (3 H)	2.51—2.04 (m, 4 H)	1.97	4.28	8.10 (s, 1 H); 8.02 (s, 2 H)
2i	1.04 (6 H)	2.58 (s, 2 H); 2.18 (s, 2 H)	2.23	4.19	3.82 (t, 2 H, NCH_2, *J* = 8); 1.52 (m, 2 H, CH_2); 1.30 (m, 2 H, CH_2); 0.9 (t, 3 H, CH_3, *J* = 8)
2j	1.00 (6 H)	2.44 (s, 2 H); 2.27 (s, 2 H)	2.08	4.29	7.56 (s, 4 H)
2k	1.02 (6 H)	2.48 (s, 2 H); 2.27 (s, 2 H)	2.11	4.28	8.20 (m, 1 H); 7.90 (m, 1 H); 7.80 (m, 1 H)
2l	1.01 (6 H)	2.46 (s, 2 H); 2.27 (s, 2 H)	2.09	4.30	8.02 (d, 2 H, *J* = 7); 7.61 (d, 2 H, *J* = 7); 7.48 (s, 2 H, NH_2)

(*to be continued*)

Table 2 (*continued*)

Compound	δ (*J*/Hz)				
	$C(CH_3)_2$ (s)	CH_2	CH_3 (s, 3 H)	CH_2CCl_3 (s, 2 H)	Z
2m	—	2.50 (m, 2 H); 2.35 (m, 2 H); 2.00 (m, 2 H)	1.98	4.30	8.23 (d, 1 H); 7.95 (dd, 1 H); 7.85 (dd, 1 H); 7.70 (d, 1 H)
2n	—	2.55 (m, 2 H); 2.35 (m, 2 H); 2.00 (m, 2 H)	2.10	4.30	8.75 (d, 1 H); 8.65 (d, 1 H); 7.94 (d, 1 H); 7.60 (dd, 1 H)
2o	1.00 (6 H)	2.60 (s, 2 H); 2.21 (s, 2 H)	2.14	4.24	7.38—7.35 (m, 3 H); 6.94 (d, 2 H); 5.29 (s, 2 H)
2p	1.00 (6 H)	2.40 (s, 2 H); 2.25 (s, 2 H)	2.05	4.30	7.63 (dd, 2 H); 7.38 (dd, 2 H)
2q	1.04 (6 H)	2.68 (s, 2 H); 2.18 (s, 2 H)	2.24	4.20	3.84 (t, 2 H, *J* = 8); 1.57 (m, 2 H); 1.57 (m, 2 H); 0.86 (t, 3 H, *J* = 8)
2r	1.04 (6 H)	2.72 (s, 2 H); 2.18 (s, 2 H)	2.25	4.20	12.42 (s, 1 H); 4.10 (t, 2 H); 2.60 (t, 2 H)
2s	1.04 (6 H)	2.60 (s, 2 H); 2.21 (s, 2 H)	2.18	4.20	4.82 (s, 2 H); 4.15 (q, 2 H); 1.23 (t, 3 H)
2t	1.06 (6 H)	2.82 (s, 2 H); 2.18 (s, 2 H)	2.25	4.18	2.91 (s, 6 H, $N(CH_3)_2$)
2u	1.03 (3 H); 1.00 (3 H)	2.48 (s, 2 H); 2.29 (s, 2 H)	2.05	4.39 (d, 1 H, *J* = 15) 4.15 (d, 1 H, *J* = 15)	6.63 (s, 1 H); 2.48 (s, 3 H); 2.01 (s, 3 H)
2v	—	2.95 (m, 2 H); 2.29 (m, 2 H); 2.02 (m, 2 H)	2.29	4.18	3.78 (t, 2 H); 3.68 (t, 2 H); 3.32 (t, 2 H); 3.13 (t, 2 H)

Table 3. ^{1}H NMR data (DMSO-d_6) for compounds **3**

Compound	δ (*J*/Hz)					
	$C(CH_3)_2$ (s)	CH_2	CH_3 (s, 3 H)	$C\underline{H}_2COOH$ (s, 2 H)	COOH	Z
3a	1.00 (6 H)	2.44 (s, 2 H); 2.24 (s, 2 H)	1.92	3.70	11.85	7.53 (d, 1 H); 7.43 (d, 1 H); 7.22 (dd, 1 H)
3b	—	2.50 (s, 2 H); 2.31 (s, 2 H); 1.97 (s, 2 H)	1.91	3.64	12.00	7.43 (m, 4 H)
3c	0.96 (6 H)	2.47 (s, 2 H); 2.23 (s, 2 H)	1.96	3.65	11.96	8.38 (d, 2 H); 7.68 (d, 2 H)
3d	0.96 (6H)	2.41 (s, 2 H); 2.22 (s, 2 H)	1.92	3.65	11.9	7.76 (d, 2 H, *J* = 9); 7.34 (d, 2 H, *J* = 9)
3e	1.02 (3 H); 0.97 (3 H)	2.47—2.10 (m, 4 H)	1.80	3.69	11.84	8.23 (dd, 1 H, *J* = 8, 0.7); 8.10 (d, 1 H, *J* = 0.7); 7.77 (d, 1 H, *J* = 8)
3f	1.01 (3 H); 0.97 (3 H);	2.48—2.27 (m, 4 H)	1.88	3.64	11.97	7.62 (m, 2 H); 7.30 (m, 1 H)
3g	0.96 (6 H)	2.45 (s, 2 H); 2.22 (s, 2 H)	1.95	3.66	11.97	7.63 (m, 1 H); 7.38 (m, 2 H); 7.24 (m, 1 H)
3h	1.01 (3 H); 0.97 (3 H)	2.49—2.07 (m, 4 H)	1.83	3.67 (d, 1 H, *J* = 15); 3.65 (d, 1 H, *J* = 15)	12.58	8.07 (s, 1 H); 7.87 (s, 2 H)
3i	1.04 (6 H)	2.63 (s, 2 H); 2.18 (s, 2 H)	2.09	3.58	11.78	3.80 (t, 2 H, NCH_2); 1.54 (m, 2 H, CH_2); 1.30 (m, 2 H, CH_2); 0.9 (t, 3 H, CH_3)

the ^{1}H NMR spectra. Among the compounds obtained, the methylene protons are nonequivalent in **2e**, **3h**, and **2u**. In other cases, these protons manifest themselves as singlets. This cannot be explained by hindered rotation about the C—N (N—N) bond, which gives rise to a center of asymmetry in the molecule and changes the CH_2X group from enantiotopic to diastereotopic,[11] because the aforesaid splitting is not observed for many other compounds **2** and **3** containing different substituents in position 2. It should be noted that such an effect is absent in structurally related compounds **3e** and **2h**.

Thus, we developed a new method for the synthesis of *N*-substituted 3-(2,2,2-trichloroethyl)-4,5,6,7-tetrahydroindol-4-ones and *N*-substituted 2-(4-oxo-(4,5,6,7-tetrahydroindol-3-yl)acetic acids with potential biological activity.

Experimental

^{1}H and ^{13}C NMR spectra were recorded on Bruker WM-250 and Bruker AM-300 instruments in DMSO-d_6 with Me_4Si as the internal standard. The melting points were determined on a Boetius microscope stage and are uncorrected. Column chromatography was carried out on Aldrich silica gel (60—100 mesh).

2-(1-Acetyl-3,3,3-trichloropropyl)-3-hydroxy-5,5-dimethylcyclohex-2-en-1-one (1a). Potassium hydroxide (3.2 g, 58 mmol) and dimedone (8 g, 58 mmol) were dissolved successively in 35 mL of MeOH. 3,5,5,5-Tetrachloropentan-2-one (12.78 g, 58 mmol) was added dropwise with vigorous stirring. The solution became turbid, and a precipitate was formed within 10—15 min. The reaction mixture was stirred for 36 h and concentrated *in vacuo*. The residue was diluted with EtOH (80 mL) and brought to boiling. Water (~50 mL) and dilute HCl (pH 5—6) were added dropwise until the precipitate dissolved. Cooling gave compound **1a** (14.3 g, 79%), m.p. 191 °C. Found (%): C, 47.39; H, 5.13; Cl, 32.02. $C_{13}H_{17}Cl_3O_3$. Calculated (%): C, 47.66; H, 5.23; Cl, 32.46. ^{1}H NMR, δ: 11.1 (s, 1 H, OH); 4.05 (dd, 1 H, CH, J = 6.5, 3.0 Hz); 3.73 (dd, 1 H, $\underline{H}CHCCl_3$, J = 15.3, 3.0 Hz); 2.75 (dd, 1 H, $HC\underline{H}CCl_3$, J = 15.3, 6.5 Hz); 2.30 (s, 4 H, 2 CH_2); 1.92 (s, 3 H, $COCH_3$); 1.04 (s, 6 H, 2 CH_3).

2-(1-Acetyl-3,3,3-trichloropropyl)-3-hydroxycyclohex-2-en-1-one (1b). Potassium hydroxide (3.75 g, 67 mmol) and cyclohexane-1,3-dione (7.47 g, 66.6 mmol) were dissolved successively in 45 mL of MeOH. 3,5,5,5-Tetrachloropentan-2-one (14.93 g, 66.6 mmol) was added dropwise with vigorous stirring. The solution became turbid, and a precipitate was formed within 10—15 min. The reaction mixture was stirred for 96 h and worked up as described above. The yield of compound **1b** was 8.38 g (42%), m.p. 164—166 °C. Found (%): C, 44.48; H, 4.37; Cl, 35.40. $C_{11}H_{13}Cl_3O_3$. Calculated (%): C, 44.10; H, 4.37; Cl, 35.50. ^{1}H NMR, δ: 11.2 (s, 1 H, OH); 4.06 (dd, 1 H, CH, J = 6.7, 3.2 Hz); 3.69 (dd, 1 H, $\underline{H}CHCCl_3$, J = 15.3, 3.2 Hz); 2.77 (dd, 1 H, $HC\underline{H}CCl_3$, J = 15.3, 6.7 Hz); 2.40 (s, 4 H, 2 CH_2); 1.90 (s, 5 H, CH_2 + CH_3).

Synthesis of compounds 2 (general procedure). A mixture of ketone **1a** or **1b** (10 mmol) and the corresponding amine (10 mmol) or hydrazine was refluxed in 15 mL of glacial AcOH for 8 h. The solution was cooled and added dropwise to water (40 mL) with stirring. The precipitate that formed was filtered off and recrystallized from ethanol or aqueous ethanol.

3-(2,2,2-Trichloroethyl)-2,6,6-trimethyl-1-(4-methyl-3-chlorophenyl)-4,5,6,7-tetrahydro-1*H*-indol-4-one (2a). ^{13}C NMR, δ: 192.8 (C=O); 142.3 (C(1-Ar)); 136.5 (C(7a)); 135.2 (C(3-Ar)); 133.8 (C(2)); 132.0 (C(2-Ar)); 131.2 (C(4a)); 127.2 (C(6-Ar)); 126.5 (C(5-Ar)); 117.2 (C(4-Ar)); 110.5 (C(3)); 101.9 (CCl_3); 52.2 (C(5)); 48.3 (C(7)); 35.9 (C(6)); 34.8 ($\underline{C}H_2CCl_3$); 27.9 (2 CH_3); 19.3 (4-$\underline{C}H_3C_6H_4$); 11.6 (CH_3).

Synthesis of compounds 3 (general procedure). A mixture of trichloroethyl derivative **2** (5 mmol) and 93% H_2SO_4 (8 mL) was heated with stirring to 60—70 °C. Then oleum (~30%, 1 mL) was added, and heating was continued at 90—100 °C for 3—4 h until vigorous evolution of HCl ceased. The reaction mixture was cooled and slowly poured into 40 mL of cold water. The precipitate that formed was filtered off, dissolved in an excess of 20% KOH, and washed with ether (3×10 mL). Then the aqueous solution was acidified with dilute HCl, and the precipitate that formed was isolated, dried, and recrystallized from aqueous ethanol or chromatographed in ethyl acetate—hexane (3 : 1).

[2,6,6-Trimethyl-(4-methyl-3-chlorophenyl)-4-oxo-4,5,6,7-tetrahydro-1*H*-indol-3-yl)acetic acid (3a). ^{13}C NMR, δ: 193.0 (C=O); 172.4 (COOH); 141.7 (C(7a)); 136.1 (C(1-Ar)); 135.4 (C(3-Ar)); 133.7 (C(4a)); 131.9 (C(2-Ar)); 128.4 (C(2)); 127.6 (C(6-Ar)); 126.3 (C(5-Ar)); 116.7 (C(4-Ar)); 111.2 (C(3)); 52.0 (C(5)); 35.8 (C(7)); 34.9 (C(6)); 30.2 ($\underline{C}H_2COOH$); 28.0 (2 CH_3); 19.2 (4-$\underline{C}H_3C_6H_4$)); 9.7 (CH_3).

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Received September 18, 2000;
in revised form March 20, 2001