

SYNTHESIS OF 3,4-DIHYDROPYRIDAZINO[1,6-*a*]BENZ-  
IMIDAZOLES<sup>#</sup>

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Abstract - 3,4-Dihydropyridazino[1,6-*a*]benzimidazoles (4a-e) have been prepared by catalytical hydrogenation of dialkyl (*E*)-2-(substituted 2-nitrophenylhydrazono)glutarates (1a-e) to the respective amines (2) followed by their base catalyzed cyclization in a tandem reaction proceeding via 4,5-dihydropyridazin-6(1H)-one intermediates (3).

## INTRODUCTION

The synthesis of several pyridazino[1,6-*a*]benzimidazoles from appropriate heterocyclic precursors by means of thermolysis,<sup>1</sup> cyclocondensation,<sup>2-4</sup> and photocyclization<sup>5,6</sup> has been reported. Furthermore, the preparation of both 1,2,3,4-tetrahydro<sup>7</sup> and 6,7,8,9-tetrahydro derivatives<sup>8,9</sup> has been described.

As part of a program on the synthesis of various *N*-heterocycles by reductive cyclization of ortho-nitroaryls<sup>10-13</sup> involved with reactions of different 2-nitrophenylhydrazono compounds we have out of them made

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<sup>#</sup> Dedicated to Professor Alan R. Katritzky on the occasion of his 65th birthday.

accessible 4*H*-pyrazolo[1,5-*a*]benzimidazoles,<sup>14</sup> (1,2,4-benzotriazin-3-yl)-acetic acid derivatives,<sup>15</sup> benzo[1,2-*b*:5,4-*b'*]bis(1*H*)-imidazo[1,2-*b*]pyrazoles),<sup>16</sup> and substituted 1,2-dihydro-1,2,5-benzotriazepines.<sup>17</sup>

We wish to report here the first synthesis of 3,4-dihydropyridazino[1,6-*a*]benzimidazoles by reductive cyclization of 2-nitrophenylhydrazones of dialkyl 2-oxoglutarates.

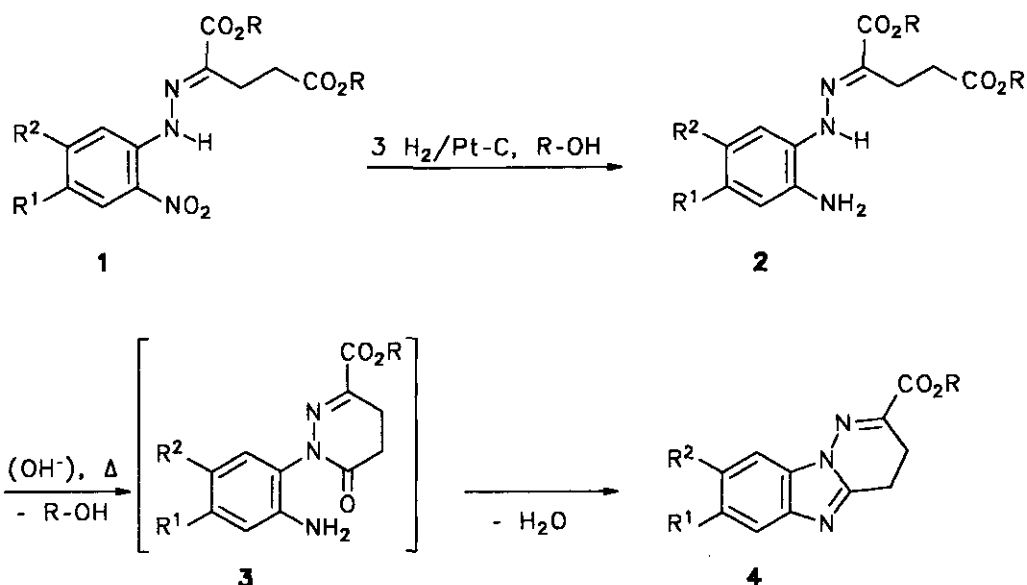
## RESULTS AND DISCUSSION

The requisite dimethyl 2-(substituted 2-nitrophenylhydrazono)glutarates (1*a-c*) and (5) were prepared in form of pure (*E*)-isomers from the corresponding substituted 2-nitrophenylhydrazines and dimethyl 2-oxoglutarate similar to the procedure of Schwesinger *et al.*, who recently reported on the tautomerism of such hydrazones.<sup>18</sup> Compounds (1*d-e*) were accessible by transesterification of the methyl esters (1*a-b*).

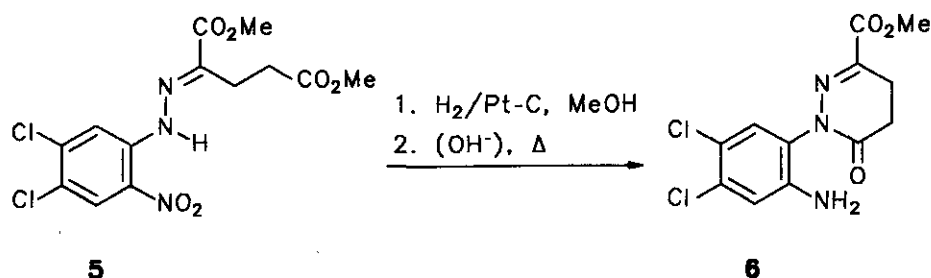
On hydrogenation over 3% Pt-C catalyst under standard conditions the (*E*)-nitrohydrazones (1) reacted to form the dialkyl (*E*)-2-(2-amino-4,5-substituted phenylhydrazono)glutarates (2), detectable on tlc plates by condensation with the 4-dimethylaminobenzaldehyde reagent yielding orange coloured azomethines. As examples, amines (2*a*) and (2*d*) have been isolated and characterized.

It was the aim of our investigation to compare the reaction behaviour of the nitrohydrazones (1) during catalytical hydrogenation with that of related compounds, which have been reductively cyclized under the same conditions. Hence, it seemed possible, that the amines (2) could either cyclize by addition to the azomethine double bond forming a 1,2,4-benzotriazine derivative<sup>15</sup> or by condensation with the 1-alkoxycarbonyl group yielding a 1,2,5-benzotriazepine compound.<sup>17</sup> However, none of these opportunities has been realized.

We have, therefore, tried to achieve cyclization by heating the amines



| 1 - 4 | R  | R <sup>1</sup> | R <sup>2</sup> |
|-------|----|----------------|----------------|
| a     | Me | H              | H              |
| b     | Me | Cl             | H              |
| c     | Me | H              | Cl             |
| d     | Et | H              | H              |
| e     | Et | Cl             | H              |



(2) in an acidic or basic medium. Whereas acidic catalysis did not influence (2), heating of the filtrated hydrogenation solutions under alkaline catalysis yielded the heterocycles (4) as first representatives of the 3,4-dihydropyridazino[1,6-a]benzimidazole system. Obviously, the reaction proceeds *via* the intermediate alkyl [1-(2-amino-4,5-substituted

phenyl)-4,5-dihydropyridazin-6(1*H*)-on-3-yl]carboxylates (3), which then undergo another cyclization to form (4). This suggestion is supported by the finding, that dimethyl (*E*)-2-(4,5-dichloro-2-nitrophenylhydrazono)glutarate (5) under the same conditions underwent only the first cyclization leading to the isolable pyridazine derivative (6).

Thus, the formation of the 3,4-dihydropyridazino[1,6-*a*]benzimidazole skeleton by a tandem reaction starting from (2) is in principle analogously to the synthesis of 4*H*-pyrazolo[1,5-*a*]benzimidazoles<sup>14</sup> from 2-nitrophenylhydrazones of  $\beta$ -ketocarboxylic acid esters and may be regarded as a ring-enlarged variant thereof.

Furthermore, we have been interested in comparing the reaction behaviour of the (*E*)- and (*Z*)-isomers of (1). Therefore, the thermodynamically more stable (*Z*)-isomers of (1) have been prepared by stirring an ethereal solution of a corresponding (*E*)-(1) compound after addition of concentrated hydrochloric acid. Completeness of isomerization was detected by tlc and proven by the clear downfield shift of the (*Z*)-NH signal in the <sup>1</sup>H-nmr spectra.<sup>17,18</sup> However, the (*Z*)-isomers of (1) on hydrogenation yielded only respective (*Z*)-2-aminophenylhydrazones as isomers of (2), which could neither be forced to cyclize by acidic nor basic catalysis.

In summary, we are now able to evaluate the behaviour of 2-nitrophenylhydrazones of dialkyl 2-oxodicarboxylates under the conditions of reductive cyclization. The 2-nitrophenylhydrazones of dialkyl mesoxalates can be cyclized to form 3-alkoxycarbonyl-4-hydroxy-1*H*-1,2,5-benzotriazepines.<sup>19</sup> Similarly, the (*Z*)-2-nitrophenylhydrazones of dialkyl 2-oxalacetates react to form 3-alkoxycarbonylmethylen-4-hydroxy-1,2-dihydro-1,2,5-benzotriazepines,<sup>17</sup> whereas the (*E*)-isomers yield uncyclized amines only. Finally, in contrast to their lower homologues, (*E*)-2-nitrophenylhydrazones of dialkyl 2-oxoglutarates (2) have been transformed to 3,4-dihydro-

pyridazino[1,6-a]benzimidazoles (4).

The financial support for this work by the Deutsche Forschungsgemeinschaft and the Fonds der Chemischen Industrie is gratefully acknowledged.

#### EXPERIMENTAL

Melting points were determined on a Boetius micro hotstage and are corrected. Ir spectra were measured on a Carl Zeiss Jena spectrophotometer Specord M80. Nmr spectra were recorded with a Varian Gemini 200 spectrometer at 199.975 MHz and a Varian 400 spectrometer at 399.952 MHz, resp., for  $^1\text{H}$  and with a Varian Gemini 200 spectrometer at 50.289 MHz for  $^{13}\text{C}$ . Chemical shifts for nmr signals are reported in ppm from tetramethylsilane. Mass spectra were collected at 70 eV with a Finnigan MAT 212 spectrometer at an ion source temperature of 200 °C. Elemental analyses were performed on a Heraeus CHN-O-RAPID analyzer.

**General procedure for the synthesis of dimethyl (*E*)-2-(substituted 2-nitrophenylhydrazono)glutarates (1a-c) and (5):** To a suspension of 10 mmol of the corresponding substituted phenylhydrazine in 10 ml of methanol were added 150 mg of acetic acid (for (1a) or 150 mg of concentrated hydrochloric acid (for (1b), (1c) and (5)). The resulting mixture was stirred for 1 h at 20 °C, became clear in between times and finally separated the hydrazones (1a-c) and (5) as crystals.

**Dimethyl (*E*)-2-(2-nitrophenylhydrazono)glutarate (1a):** Yield 2.24 g (72%) orange crystals, mp 108-110 °C (MeOH); lit.,<sup>18</sup> mp 109-111 °C. Ir (KBr):  $\nu=1700\text{ cm}^{-1}$  (CO).  $^1\text{H}$ -Nmr ( $\text{CDCl}_3$ ):  $\delta$  = 11.44 (s, 1H), 8.23-6.98 (m, 4H), 3.88 (s, 3H), 3.66 (s, 3H), 2.95 (t,  $J=6.5\text{ Hz}$ , 2H), 2.74 (t,  $J=6.5\text{ Hz}$ , 2H).

**Dimethyl (E)-2-(4-chloro-2-nitrophenylhydrazono)glutarate (1b):** Yield 2.34 g (68%), dark orange crystals, mp 104-106 °C (MeOH). Ir (KBr):  $\nu = 1725 \text{ cm}^{-1}$  (C=O).  $^1\text{H-Nmr}$  ( $\text{CDCl}_3$ ):  $\delta = 11.40$  (s, 1H), 8.18-7.49 (m, 3H), 3.88 (s, 3H), 3.68 (s, 3H), 2.92 (t,  $J=5.8 \text{ Hz}$ , 2H), 2.74 (t,  $J=5.8 \text{ Hz}$ , 2H). Anal. Calcd for  $\text{C}_{13}\text{H}_{14}\text{N}_3\text{O}_6\text{Cl}$ : C, 45.42; H, 4.10; N, 12.22; Cl, 10.31. Found: C, 45.23; H, 4.26; N, 12.17; Cl, 10.39.

**Dimethyl (E)-2-(5-chloro-2-nitrophenylhydrazono)glutarate (1c):** Yield 2.58 g (75%), yellow crystals, mp 99-101 °C (MeOH). Ir (KBr):  $\nu = 1720 \text{ cm}^{-1}$  (CO).  $^1\text{H-Nmr}$  ( $\text{CDCl}_3$ ):  $\delta = 11.47$  (s, 1H), 8.17-6.91 (m, 3H), 3.90 (s, 3H), 3.67 (s, 3H), 2.91 (t,  $J=5.9 \text{ Hz}$ , 2H), 2.78 (t,  $J=5.9 \text{ Hz}$ , 2H). Anal. Calcd for  $\text{C}_{13}\text{H}_{14}\text{N}_3\text{O}_6\text{Cl}$ : C, 45.52; H, 4.10; N, 12.22; Cl, 10.31. Found: C, 45.13; H, 4.31; N, 12.40; Cl, 10.26.

**Dimethyl (E)-2-(4,5-dichloro-2-nitrophenylhydrazono)glutarate (5):** Yield 2.46 g (65%), yellow crystals, mp 122-124 °C (MeOH). Ir (KBr):  $\nu = 1730, 1700 \text{ cm}^{-1}$  (CO).  $^1\text{H-Nmr}$  ( $\text{CDCl}_3$ ):  $\delta = 11.43$  (s, 1H), 8.32 (s, 1H), 8.13 (s, 1H), 3.91 (s, 3H), 3.67 (s, 3H), 2.90 (t,  $J=6.3 \text{ Hz}$ , 2H), 2.77 (t,  $J=6.3 \text{ Hz}$ , 2H). Anal. Calcd for  $\text{C}_{13}\text{H}_{13}\text{N}_3\text{O}_6\text{Cl}_2$ : C, 41.29; H, 3.46; N, 11.11; Cl, 18.75. Found: C, 41.12; H, 3.59; N, 10.99; Cl, 18.79.

**General procedure for the synthesis of diethyl (E)-2(substituted 2-nitrophenylhydrazono)glutarates (1d-e):** Sodium carbonate (1.06 g, 10 mmol) was added to a solution of the appropriate methyl precursor (1a) or (1b), resp., in 100 ml of absolute ethanol. After refluxing for 1 h, the mixture was filtered. The filtrate separated the hydrazones (1d-e) on standing as crystals.

**Diethyl (E)-2-(2-nitrophenylhydrazono)glutarate (1d):** Yield 1.44 g (85%), yellow crystals, mp 90-92 °C (EtOH). Ir (KBr):  $\nu = 1745, 1710 \text{ cm}^{-1}$  (CO).  $^1\text{H-Nmr}$  ( $\text{CDCl}_3$ ):  $\delta = 11.39$  (s, 1H), 8.18-6.96 (m, 4H), 4.23 (q,  $J=7.0 \text{ Hz}$ , 2H), 4.11 (q,  $J=7.0 \text{ Hz}$ , 2H), 2.93 (t,  $J=7.0 \text{ Hz}$ , 2H), 2.69 (t,  $J=7.0 \text{ Hz}$ , 2H), 1.37 (t,  $J=7.0 \text{ Hz}$ , 3H), 1.21 (t,  $J=7.0 \text{ Hz}$ , 3H). Anal. Calcd for  $\text{C}_{15}\text{H}_{19}\text{N}_3\text{O}_6$ : C, 58.82; H, 6.15; N, 11.03. Found: C, 58.65; H, 6.15; N, 11.03.

53.40; 5.68; N, 12.46. Found: C, 53.18; H, 5.89; N, 12.60.

**Diethyl (E)-2-(4-chloro-2-nitrophenylhydrazono)glutarate (1e):** Yield 1.38 g (74%), orange crystals, mp 69 -71 °C (EtOH). Ir (KBr):  $\nu$  = 1735, 1725  $\text{cm}^{-1}$  (CO).  $^1\text{H-Nmr}$  ( $\text{CDCl}_3$ ):  $\delta$  = 11.41 (s, 1H), 8.20-7.51 (m, 3H), 4.34 (q, J=7.1 Hz, 2H), 4.13 (q, J=7.1 Hz, 2H), 2.93 (t, J=6.3 Hz, 2H), 2.73 (t, J=6.3 Hz, 2H), 1.40 (t, J=7.1 Hz, 3H), 1.23 (t, J=7.1 Hz, 3H). Anal. Calcd for  $\text{C}_{15}\text{H}_{18}\text{N}_3\text{O}_6\text{Cl}$ : C, 48.45; H, 4.88; N, 11.30; Cl 9.94. Found: C, 48.70; H, 5.02; N, 11.13; Cl, 9.31.

**General procedure for the isomerization of (E)-hydrazones (1a-e) to the corresponding (Z)-isomers:** 1 mmol of concentrated hydrochloric acid was added to a solution of 1 mmol of the appropriate (E)-hydrazone (1a-e) in 50 ml of diethyl ether. After stirring for 6 h the solution separated the corresponding (Z)-hydrazone on slow evaporation of the solvent. A characteristic feature of all (Z)-isomers is the downfield shift of their NH signal in the  $^1\text{H-nmr}$  to values of about 13.8 ppm in  $\text{CDCl}_3$ , which has already been described elsewhere.<sup>18</sup>

**General procedure for the synthesis of dialkyl (E)-2-(substituted 2-aminophenylhydrazono)glutarates (2a-e):** The respective hydrazones (1) (3 mmol) in methanol (50 ml) (for methyl esters 1a-c) or in ethanol (50 ml) (for ethyl esters (1d) and (1e)) were hydrogenated over 3% Pt on carbon powder (100 mg) under normal pressure at 20 °C until no more hydrogen was consumed (uptake 200 ml). The catalyst was filtered off and the filtrates allowed to stand overnight in an open beaker. In the cases of (2a) and (2d) the products separated as crystalline solids and were characterized. (2b), (2c), and (2e) separated as oily liquids, pure according to tlc, which were not further characterized.

**Dimethyl (E)-2-(2-aminophenylhydrazono)glutarate (2a):** Yield 0.72 g (85%), pale yellow crystals, mp 80-82 °C (MeOH). Ir (KBr):  $\nu$  = 3410, 3350, 3300

(NH<sub>2</sub>), 1720, 1695 (CO). <sup>1</sup>H-Nmr (CDCl<sub>3</sub>): δ = 12.04 (s, 1H), 7.18-6.76 (m, 4H), 4.71 (br s, 2H), 3.82 (s, 3H), 3.69 (s, 3H), 2.87 (t, J=6.3 Hz, 2H), 2.70 (t, J=6.3 Hz, 2H). Anal. Calcd for C<sub>13</sub>H<sub>17</sub>N<sub>3</sub>O<sub>4</sub>: C, 55.90; H, 6.14; N, 15.05. Found: C, 56.21; H, 6.18; N, 15.29.

**Diethyl (E)-2-(2-aminophenylhydrazono)glutarate (2d):** Yield 0.76 g (82%), yellow crystals, mp 68-70 °C (cyclohexane). Ir (KBr): ν = 3400, 3350, 3280 (NH<sub>2</sub>), 1740, 1690 (CO). <sup>1</sup>H-Nmr (CDCl<sub>3</sub>): δ = 12.01 (s, 1H), 7.30-6.73 (m, 4H), 4.29 (q, J=7.0 Hz, 2H), 4.14 (q, J=7.0 Hz, 2H), 3.75 (br s, 2H), 2.87 (t, J=6.6 Hz, 2H), 2.71 (t, J=6.6 Hz, 2H), 1.35 (t, J=7.0 Hz, 3H), 1.25 (t, J=7.0 Hz, 3H). Anal. Calcd for C<sub>15</sub>H<sub>21</sub>N<sub>3</sub>O<sub>4</sub>: C, 58.62; H, 6.89; N, 13.67. Found: C, 58.47; H, 6.96; N, 13.55.

**General procedure for the synthesis of alkyl (substituted 3,4-dihydropyridazino[1,6-a]benzimidazol-2-yl)carboxylates (4a-e):** 0.2 ml of 10% aqueous NaOH were added to the filtered hydrogenation solution of a 3 mmol run for the respective amine (2). After refluxing for 1 min, the hot solution was diluted with 5 volumes water and afforded on standing overnight crystals of the crude pyridazino[1,6-a]benzimidazole (4).

**Methyl (3,4-dihydropyridazino[1,6-a]benzimidazol-2-yl)carboxylate (4a):** Yield 0.55 g (80%), colourless crystals, mp 136-138 °C (cyclohexane). Ir (KBr): ν = 1710 cm<sup>-1</sup> (CO). <sup>1</sup>H-Nmr (CDCl<sub>3</sub>): δ = 7.72-7.29 (m, 4H), 3.98 (s, 3H), 3.24 (t, J=8.2 Hz, 2H), 3.06 (t, J=8.2 Hz, 2H). <sup>13</sup>C-Nmr (CDCl<sub>3</sub>): δ = 163.8, 148.1, 143.6, 141.5, 132.6, 124.2, 123.9, 119.8, 109.1, 53.2, 21.5, 19.6. Ms (m/z, relative intensity) 229 (M<sup>+</sup>, 100), 197 (15), 169 (27), 142 (15). Anal. Calcd for C<sub>12</sub>H<sub>11</sub>N<sub>3</sub>O<sub>2</sub>: C, 62.87; H, 4.83; N, 18.33. Found: C, 62.64; H, 5.35; N, 18.14.

**Methyl (7-chloro-3,4-dihydropyridazino[1,6-a]benzimidazol-2-yl)carboxylate (4b):** Yield 0.40 g (50%), colourless crystals, mp 208-210 °C (MeOH). Ir (KBr): ν = 1740 cm<sup>-1</sup> (CO). <sup>1</sup>H-Nmr (CDCl<sub>3</sub>): δ = 7.65-7.29 (m, 3H), 3.96 (s, 3H), 3.23 (t, J=8.1 Hz, 2H), 3.06 (t, J=8.1 Hz, 2H). Ms (m/z, relative in-

tensity) 263 ( $M^+$ , 100), 231 (11), 203 (28), 177 (23). Anal. Calcd for  $C_{12}H_{10}N_3O_2Cl$ : C, 54.66; H, 3.82; N, 15.93; Cl, 13.45. Found: C, 54.72; H, 3.67; N, 15.60; Cl 13.79.

**Methyl (8-chloro-3,4-dihydropyridazino[1,6-a]benzimidazol-2-yl)carboxylate (4c):** Yield 0.52 g (65%), colourless crystals, mp 134-136 °C (cyclohexane) Ir (KBr):  $\nu = 1740\text{ cm}^{-1}$  (CO).  $^1H$ -Nmr ( $CDCl_3$ ):  $\delta = 7.65-7.19$  (m, 3H), 3.95 (s, 3H), 3.19 (t, J=8.0 Hz, 2H), 3.04 (t, J=8.0 Hz, 2H). Ms (m/z, relative intensity) 263 ( $M^+$ , 100), 232 (6), 203 (19), 177 (18). Anal. Calcd for  $C_{12}H_{10}N_3O_2Cl$ : C, 54.66; H, 3.82; N, 15.93; Cl 13.45. Found: C, 54.41; H, 3.86; N, 15.61; Cl, 13.29.

**Ethyl (3,4-dihydropyridazino[1,6-a]benzimidazol-2-yl)carboxylate (4d):** Yield 0.44 g (60%), colourless crystals, mp 94-96 °C (cyclohexane). Ir (KBr):  $\nu = 1705\text{ cm}^{-1}$  (CO).  $^1H$ -Nmr ( $CDCl_3$ ):  $\delta = 7.69-7.29$  (m, 4H), 4.43 (q, J=7.1 Hz, 2H), 3.24 (t, J=8.0 Hz, 2H), 3.06 (t, J=8.0 Hz, 2H), 1.43 (t, J=7.1 Hz, 3H).  $^{13}C$ -Nmr ( $CDCl_3$ ):  $\delta = 163.3, 148.4, 143.6, 141.5, 132.7, 124.1, 132.9, 119.8, 110.1, 62.6, 21.6, 19.6, 14.2$ . Ms (m/z, relative intensity) 243 ( $M^+$ , 100), 197 (25), 169 (27), 143 (12). Anal. Calcd for  $C_{13}H_{13}N_3O_2$ : C, 64.19; H, 5.39; N, 17.27. Found: C, 64.06; H, 5.36; N, 17.20.

**Ethyl (7-chloro-3,4-dihydropyridazino[1,6-a]benzimidazol-2-yl)carboxylate (4e):** Yield 0.44 g (52%), colourless crystals, mp 115-117 °C (cyclohexane) Ir (KBr):  $\nu = 1715\text{ cm}^{-1}$  (CO).  $^1H$ -Nmr ( $CDCl_3$ ):  $\delta = 7.65-7.30$  (m, 3H), 4.43 (q, J=7.1 Hz, 2H), 3.23 (t, J=8.0 Hz, 2H), 3.06 (t, J=8.0 Hz, 2H), 1.43 (t, J=7.1 Hz, 3H). Ms (m/z, relative intensity) 277 ( $M^+$ , 100), 231 (13), 203 (24), 178 (16). Anal. Calcd for  $C_{13}H_{12}N_3O_2$ : C, 56.22; H, 4.36; N, 15.13; Cl, 12.77. Found: C, 56.49; H, 4.56; N, 15.03; Cl, 13.13.

**Methyl [1-(2-amino-4,5-dichlorophenyl)-4,5-dihydropyridazin-6(1H)-on-3-yl]-carboxylate (6):** Nitrohydrazone (5) (3 mmol) was hydrogenated as described for hydrazones (1). The resulting hydrogenation solution was further

treated as reported for the synthesis of heterocycles (4) from amines (2). Compound (6) in contrast to (4) separated already during refluxing and showed no tendency for further cyclization under the conditions given. Yield: 0.75 g (79%), colourless crystals, mp 185-186 °C (MeOH). Ir (KBr):  $\nu = 3410, 3340, (\text{NH}_2), 1735, 1675 (\text{CO}) \text{ cm}^{-1}$ .  $^1\text{H-Nmr}$  ( $\text{CDCl}_3$ ):  $\delta = 7.28 (\text{s}, 1\text{H})$ , 6.91 (s, 1H), 4.58 (br s, 2H), 3.89 (s, 3H), 3.06 (t,  $J=8.0 \text{ Hz}$ , 2H), 2.76 (t,  $J=8.0 \text{ Hz}$ , 2H).  $^{13}\text{C-Nmr}$  ( $\text{DMSO-d}_6$ ):  $\delta = 171.4, 169.0, 151.3, 149.6, 136.8, 136.0, 131.0, 121.3, 120.9, 58.1, 32.0, 27.3$ . Ms ( $m/z$ , relative intensity) 315 ( $\text{M}^+$ , 75), 298 (18), 256 (48), 175 (100), 139 (75). Anal. Calcd for  $\text{C}_{12}\text{H}_{11}\text{N}_3\text{O}_3\text{Cl}_2$ : C, 45.59; H, 3.51; N, 13.29; Cl, 22.43. Found: C, 45.48; H, 3.71; N, 13.03; Cl, 22.73.

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Received, 14th July, 1993