Supporting Information

Highly Substituted Pyrrolidinones And Pyridones By Novel MCRs

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General. Proton and Carbon NMR spectra were obtained in CDCl₃ or in d₆-DMSO and determined with a Mercury 400 spectrometer. Chemical shifts are expressed in ppm (δ) with respect to TMS as an internal standard. Electrospray ionisation mass spectra (ESI) were obtained using a MSD (Hewlett-Packards HPLC 1100 driven electrospray MS instrument). Products were purified by column or wave chromatography with silica gel and a mixture of petrol ether and ethyl acetate as eluent. The purity was determined utilizing a Hewlett-Packard LC 1100 system using a YMC column, 2 mm x 50 mm, 2 µm ODSA, detection wave length at 220 and 254 nm; flow: 0,6 mL/min., with a 6 min. gradient from 90 % H₂O to 10 % H₂O (0,5% CH₃COOH) vs. CH₃CN. Preparative HPLC was performed on a Labomatic System, using a Grom Sil 120 ODS-4 HE column (10 µM, 100 x 30 mm) with a 30 min. methanol/water gradient (30-100% MeOH) with single wavelength detection at 254 nm, flow 50 mL/min. C, H, N, O Elementar analysis was performed at a microanalytic laboratory Ilse Beetz, Kronach, Germany. IR spectra were recorded on a Nexus FT-IR apparatus from Thermo-Nicolet. The Melting point was determined in open capillaries on an electrothermal I.A. 9100 melting point apparatus.

Experimental Procedures and analytical data for the described compounds

General procedure for the preparation of Ugi products for Pyrrolidinones and Pyridones:

To 1 mmol aldehyde in 2 mL MeOH, 1 mmol amine is added and it is stirred for 5 min. Then, 1 eq pyridinium hydrochloride is added in case of sodium salt of β-keto aldehydes. After that, 1 mmol phosphono acetic acid and 1 mmol isocyanide are added. The reaction mixture is stirred for a night at room temperature.

For the work up, the solvent is evaporated and 3 mL DCM and 2 mL water are added. The aqueous layer is extracted with 1 mL DCM. The organic layers are combined and washed twice with 2 mL citric acid (5%), sat. NaHCO₃ solution and brine. The organic layer was dried over Na₂SO₄ and concentrated in vacuum. The product could be crystallized or otherwise purified by column or wave chromatography or preparative HPLC.

{[Benzyl-(1-cyclohexylcarbamoyl-2-hydroxy-2-phenyl-vinyl)-carbamoyl]-methyl}-phosphonic acid diethyl ester (**1**): scheduled quantity 3 mmol gave 1204 mg (76%) C₂₈H₃₇N₂O₆P MW: 528.59 g/mol; HPLC-MS (ESI): t_R = 3.86 min, m/z = 529 [M+H]⁺, 551 [M+Na]⁺.

General procedure of Wittig-Horner-Emmons reaction for the preparation of Pyrrolidinones and Pyridones:

To the obtained Ugi product in 40 mL THF under N₂, 4.5 eq LiCl were added. The mixture was stirred at 0°C until the complete dissolution of LiCl. Then 10 eq Et₃N were added. The reaction mixture was stirred at 0°C for 30 min and then for 12 h at room temperature.

For the work up, the solution was filtered on SiO₂ that was next washed with ethyl acetate and methanol. The solvent is concentrated in vacuum. The product could be crystallized in ethyl acetate or otherwise purified by column, wave chromatography or preparative HPLC.

1-Benzyl-6-oxo-4-phenyl-1,2,3,6-tetrahydro-pyridine-2-carboxylic acid *tert*-butylamide (**2**): scheduled quantity 1 mmol gave 341 mg (94 %) C₂₃H₂₆N₂O MW: 362.48 g/mol; **HPLC-MS (ESI)**: t_R = 3.55 min, m/z = 363 [M+H]⁺, 385 [M+Na]⁺; **¹H-NMR (CDCl₃, 400 MHz)**: δ = 1.12 (s, 9H, C(CH₃)₃), 2.86–2.93 (q, 1H, CHCH₂C), 3.38–3.42 (d, 1H, CHCH₂C), 4.03–4.05 (d, 1H, CHCH₂C), 4.46–4.49 (d, 1H, PhCH₂N), 4.95–4.99 (d, 1H, PhCH₂N), 5.60 (br, 1H, NH), 6.30 (d, 1H, C=CHCO), 7.32–7.48 (m, 10H, ar-CH); **¹³C-NMR (CDCl₃, 100 MHz)**: δ = 28.26 (C(CH₃)₃), 30.27 (CHCH₂C), 50.04 (PhCH₂N), 51.43 (C(CH₃)₃), 59.69 (CHCH₂C); 118.80 (C=CHCO), 126.07 (ar-CH), 128.65 (ar-CH), 128.74 (ar-CH), 129.05 (ar-CH), 129.75 (ar-CH), 137.00 (ar-C), 137.10 (ar-C), 149.64 (ar-C), 164.57 (CO), 169.27 (CO).

General procedure for the preparation of Passerini products:

To 1 mmol aldehyde in 4 mL Et₂O and 1 eq pyridinium hydrochloride is added in case of sodium salt of β-keto aldehydes. After that, 1 mmol phosphono acetic acid and 1 mmol isocyanide are added. The reaction mixture is stirred for a night at room temperature.

For the work up, the solvent is evaporated and 3 mL DCM and 2 mL water are added. The aqueous layer is extracted with 1 mL DCM. The organic layers are combined and washed twice with 2 mL citric acid (5%), sat. NaHCO₃ solution and brine. The organic layer was dried over Na₂SO₄ and concentrated in vacuum. The product is purified by column chromatography on silica gel with a mixture of ethyl acetate and petrol ether (4:1, v/v) as eluent.

(Diethoxy-phosphoryl)-acetic acid 1-*tert*-butylcarbamoyl-4,4-dimethyl-3-oxo-pentyl ester (P-3CR-precursor for compound 3): scheduled quantity 3 mmol gave 586 mg (48 %) C₁₈H₃₄NO₇P MW 407.45 g/mol; **HPLC-MS (ESI)**: t_R = 3.54 min; m/z = 408 [M+H]⁺; 430 [M+Na]⁺; **¹H-NMR (CDCl₃, 400 MHz)**: δ = 1.10 (s, 9H, CH₃), 1.33 (m, 15 H, CH₃), 2.87–3.16 (m, 4 H, CH₂), 4.13 (m, 4 H, CH₂), 5.48 (m, 1H, CH), 6.89 (s, 1H, NH); **¹³C-NMR (CDCl₃, 100 MHz)**: δ = 16.20 (CH₃), 26.10 (C(CH₃)₃), 28.54 (C(CH₃)₃), 33.69, 34.99, 38.54, 44.00 (quart. C), 51.53 (quart. C), 62.81 and 63.16 (PCH₂), 70.66 (CH), 131.67, 135.39, 164.17 and 164.23 (PCH₂CO), 168.27 (CO), 211.53 (CO).

(Diethoxy-phosphoryl)-acetic acid 3-biphenyl-4-yl-1-*tert*-butylcarbamoyl-3-oxo-propyl ester (P-3CR-precursor for compound 4): scheduled quantity 3 mmol gave 543 mg (36 %) C₂₆H₃₄NO₇P MW 503.54 g/mol; **HPLC-MS (ESI)**: t_R = 3.59 min; m/z = 504 [M+H]⁺; 526 [M+Na]⁺; **¹H-NMR (CDCl₃, 400 MHz)**: δ = 1.3 (t, 6H, CH₃), 1.4 (s, 9 H, CH₃), 3.0 (m, 4 H, CH₂), 4.1 (s, 4 H, CH₂), 5.7 (d, 1H, CH), 5.9 (s, 1H, NH), 7.0 – 8.0 (m, 9 H, ar); **¹³C-NMR (CDCl₃, 100 MHz)**: δ = 16.43 (CH₃), 29.02 (C(CH₃)₃), 52.38 (quart. C), 64.27 (CH₂), 71.68 (CH), 126.12 – 140.35 (C-ar and CH-ar) 164.67 (CO), 168.53 (CO), 190.29 (CO).

General procedure for the HWE:

To the obtained Passerini product is diluted in 40mL THF under N₂, 4,5eq LiCl were added. The mixture was stirred at 0° until the complete dissolution of LiCl. Then 10 eq Et₃N were added. The reaction mixture was stirred at 0° for 30 min and then for 12 h at room temperature.

For the work up, the solution was filtered on SiO₂ that was next washed with ethyl acetate and methanol.

The solvent is concentrated in vacuum. The product could be crystallized in ethyl acetate or otherwise purified by column, wave chromatography or preparative HPLC.

5,5-Dimethyl-4-oxo-hex-2-enoic acid *tert*-butylamide (**3**): scheduled quantity 3 mmol gave 597 mg (94 %) C₁₂H₂₁NO₂ MW 211.31 g/mol; **HPLC-MS (ESI)**: t_R = 3,215 min; m/z =230 [M+18+H]⁺ ; 252 [M+18+Na]⁺.

4-Biphenyl-4-yl-4-oxo-but-2-enoic acid *tert*-butylamide (**4**): scheduled quantity 3 mmol gave 793 mg (86 %) C₂₀H₂₁NO₂; MW 307.40 g/mol; **HPLC-MS (ESI-TOF)**: t_R = 3,934 min; m/z =308 [M+H]⁺ ; 330 [M+Na]⁺; **¹H-NMR** (CDCl₃, 400 MHz): δ = 2,1 (s, 9H, CH₃), 6,9 – 8,0 (m, 11 H, CH and ar-CH); **¹³C-NMR** (CDCl₃, 100 MHz): δ =29 (CH₃), 128 - 130 (CH and ar-CH).

Analytical data for compounds described in table 1:

1-Benzyl-5-oxo-3-phenyl-2,5-dihydro-1*H*-pyrrole-2-carboxylic acid cyclohexyl amide (**5**): scheduled quantity 3 mmol gave 274 mg (24 %) C₂₄H₂₆N₂O₂ MW: 374.49 g/mol; **M.p.** 211.8°C-213.7°C; **R_f**: 0.84 (PE/EA, 1/4, v/v); **HPLC-MS (ESI)**: t_R = 3.61 min, m/z = 375 [M+H]⁺, 397 [M+Na]⁺; **¹H-NMR** (CDCl₃, 400 MHz): δ = 0.79-1.69 (m, 10H, 5xCH₂-cyclohexyl), 3.49-3.57 (m, 1H, CH-cyclohexyl), 4.00-4.04 (d, 1H, ²J_{H,H}=14.77Hz, 1H of NCH₂Ph), 4.85 (s, 1H, CHC=CHCO), 5.07-5.11 (d, 1H, ²J_{H,H}=14.80Hz, 1H of NCH₂Ph), 5.92-5.94 (d, 1H, ³J_{NH,CH}=8.30Hz, NH), 6.39 (s, 1H, C=CHCO), 7.19-7.30 (m, 5H, 5xCH-Ar), 7.49-7.52 (t, 2H, 2xCH-Ar); **¹³C-NMR** (CDCl₃, 100 MHz): δ = 24.61 (CH₂-cyclohexyl), 24.71 (CH₂-cyclohexyl), 25.17 (CH₂-cyclohexyl), 32.36 (CH₂-cyclohexyl), 32.62 (CH₂-cyclohexyl), 44.85 (NCH₂Ph), 48.55 (CH-cyclohexyl), 67.53 (CHC=CHCO), 120.49 (C=CHCO), 126.96 (CH-ar), 127.79 (CH-ar), 128.41 (CH-ar), 128.75 (CH-ar), 128.78 (CH-ar), 130.34 (CH-ar), 130.52 (CH-ar), 136.35, 155.71 (C=CHCO), 165.24 (CO), 171.21 (CO).

1-[2,3-Methoxy]-phenylethyl-5-oxo-3-phenyl-2,5-dihydro-1*H*-pyrrol-2-carboxylic acid cyclohexyl amide (**6**): scheduled quantity 3 mmol gave 459 mg (34 %) C₂₇H₃₂N₂O₄ MW: 448.57 g/mol; **HPLC-MS (ESI)**: t_R = 3.54 min, m/z = 449 [M+H]⁺, 471 [M+Na]⁺; **¹H-NMR** (CDCl₃, 400 MHz): δ = 0.83-1.80 (m, 10H, 5 x CH₂-cyclohexyl), 2.81-2.97 (m, 2H, NCH₂CH₂Ph), 3.30 (m, 1H, NCH₂CH₂Ph), 3.61 (m, 1H, CH-cyclohexyl), 3.81 (s, 6H, 2 x Ph-OCH₃), 4.07 (m, 1H, NCH₂CH₂Ph), 4.93 (s, 1H, COCH(N;C)), 5.91 (d, 1H, ³J_{NH,CH}: 8.19 Hz, NH), 6.39 (s, 1H, C=CHC=O), 6.76 (m, 3H, ar-CH), 7.37 (m, 3H, ar-CH), 7.55 (m, 2H, ar-CH); **¹³C-NMR** (CDCl₃, 100 MHz): δ = 24.60 (CH₂-cyclohexyl), 24.70 (CH₂-cyclohexyl), 25.13 (CH₂-cyclohexyl), 32.63 (CH₂-cyclohexyl), 32.68 (CH₂-cyclohexyl), 33.90 (Ph-OCH₃), 42.68 (CH₂), 48.56 (CH-cyclohexyl), 55.74 (CH), 68.47 (CH, COCH(C;N)), 111.24 (CH), 111.68 (CH), 120.59 (CH, C=CHC=O), 120.84 (CH, C=CHC=O), 126.84 (ar-CH), 128.81 (ar-CH), 130.30 (ar-CH), 130.50 (ar-CH), 130.58 (ar-CH), 147.60 (ar-C), 148.87 (ar-C), 155.30 (C=CHC=O), 165.54 (C=O), 171.14 (C=O).

3-(4-Methoxy-phenyl)-1-(3-methyl-butyl)-5-oxo-2,5-dihydro-1*H*-pyrrole-2-carboxylic acid allylamide (**7**): scheduled quantity 1 mmol gave 61 mg (18 %) C₂₀H₂₆N₂O₅ MW: 342.44 g/mol; **M.p.** 131°C; **HPLC-MS (ESI)**: t_R = 3.38 min, m/z = 343 [M+H]⁺, 365 [M+Na]⁺, 707 [2xM+Na]⁺; **¹H-NMR** (CDCl₃, 400 MHz): δ = 0.91 (d, 6H, 2 x CH₃), 1.54 (m, 3H, CH₂, CH), 3.04 (m, 1H, CH), 3.77-3.87 (m, 6H, PhOCH₃, CH₂, CH), 4.96 (m, 2H, CH=CH₂), 5.12 (s, 1H, COCH(C;N)), 5.66 (m, 1H, CH=CH₂), 6.26 (s, 1H, C=CHCO), 6.71 (t, 1H, NH), 6.89 (m, 2H, Ar-CH), 7.61 (m, 2H, ar-CH); **¹³C-NMR** (CDCl₃, 100 MHz): δ = 22.14 (CH₃),

22.48 (CH₃), 25.68 (CH(CH₃)₂), 36.81 (CH₂), 39.44 (CH₂), 41.72 (CH₂), 55.23 (OCH₃), 67.89 (COCHN), 114.18 (ar-CH), 116.18 (=CH₂), 118.39 (CH=CH₂), 122.99 (ar-CH), 128.60 (ar-CH), 133.39 (=CHCO), 154.70 (ar-C), 161.31 (C=CHCO), 167.01 (CO), 171.62 (CO).

Elementar analysis:

calc:	C	70.150%	H	7.650%	N	8.18%	O	14.02%
found:	C	69.535%	H	7.655%	N	8.15%	O	13.55%
diff.:	C	0.615%	H	0.005%	N	0.03%	O	0.47%

1-(3-Methyl-butyl)-5-oxo-3-phenyl-2,5-dihydro-1*H*-pyrrole-2-carboxylic acid cyclohexylamide (**8**): scheduled quantity 3 mmol gave 175 mg (16 %) C₂₂H₃₀N₂O₂ MW: 354.50 g/mol; **M.p.** 204°C; **HPLC-MS (ESI)**: t_R = 3.64 min, m/z = 355 [M+H]⁺, 377 [M+Na]⁺, 731 [2x M+Na]⁺; **¹H-NMR (CDCl₃, 400 MHz)**: δ = 0.92 (d, 6H, 2 x CH₃), 1.02-1.82 (12 H, 5 x CH₂-cyclohexyl, CH₂), 3.05 (m, 1H, CH₂), 3.64 (m, 1H, CH-cyclohexyl), 3.84 (m, 1H, CH₂), 5.10 (s, 1H, CH), 6.00 (d, 1H, ³J_{NH,CH}: 7.67 Hz, NH), 6.40 (s, 1H, CH), 7.38 (m, 3H, ar-CH), 7.62 (m, 2H, ar-CH); **¹³C-NMR (CDCl₃, 100 MHz)**: δ = 22.18 (CH₃), 22.55 (CH₃), 24.67 (CH₂-cyclohexyl), 24.78 (CH₂-cyclohexyl), 25.17 (CH₂-cyclohexyl), 25.94 ((CH₃)₂CH), 32.39 (CH₂-cyclohexyl), 32.72 (CH₂-cyclohexyl), 36.88 (CH₂), 39.51 (CH₂), 48.56 (CH-cyclohexyl), 68.14 (C=OCH(C,N)), 120.87 (C=CHCO), 126.92 (ar-CH), 128.83 (ar-CH), 130.45 (ar-CH), 155.18 (C=CHCO), 165.70 (C=O), 171.16 (C=O).

Elementar analysis:

calc:	C	74.540%	H	8.53%	N	7.90%	O	9.030%
found:	C	74.485%	H	8.47%	N	7.91%	O	9.025%
diff.:	C	0.055%	H	0.06%	N	0.01%	O	0.005%

1-(3,3-Dimethyl-butyl)-3-(4-methoxy-phenyl)-5-oxo-2,5-dihydro-1*H*-pyrrole-2-carboxylic acid allylamide (**9**): scheduled quantity 1 mmol gave 271 mg (76 %) C₂₁H₂₈N₂O₃ MW: 356.47 g/mol; **HPLC-MS (ESI)** t_R = 3.51 min, m/z = 357 [M+H]⁺, 379 [M+Na]⁺; **¹H-NMR (CDCl₃, 400 MHz)**: δ = 0.94 (s, 9H, ^tBu), 1.44-1.57 (m, 2H, CH₂C(CH₃)₃), 3.03-3.10 (m, 1H, CH₂), 3.74-3.79 (m, 1H, CH₂), 3.83 (s, 1H, OCH₃), 4.93-5.02 (m, 2H, CH=CH₂), 5.13 (s, 1H, COCHN), 5.60-5.70 (m, 1H, CH=CH₂), 6.13 (br, 1H, NH), 6.31 (s, 1H, C=CHCO), 6.91 (d, 2H, ar-CH), 7.60 (d, 2H, ar-CH); **¹³C-NMR (CDCl₃, 100 MHz)**: δ = 29.23 (C(CH₃)₃), 29.85 (C(CH₃)₃), 37.87 (CH₂C(CH₃)₃), 41.31 (CH₂NHCO), 41.75 (NCH₂), 55.36 (OCH₃), 67.86 (COCHN), 114.31 (ar-CH), 116.66 (CH=CH₂), 118.62 (CH=CH₂), 122.97 (ar-C), 128.69 (ar-CH), 133.21 (CCHCO), 154.88 (C=CHCO), 161.47 (ar-C), 167.00 (CO), 171.59 (CO).

5-Oxo-3-phenyl-1-propyl-2,5-dihydro-1*H*-pyrrole-2-carboxylic acid *tert*-butylamide (**10**): scheduled quantity 1 mmol gave 50 mg (25 %) C₁₈H₂₄N₂O₂ MW: 300.40 g/mol; **R_f**: 0.69 (PE/EA, 1/4, v/v); **HPLC-MS (ESI)**: t_R = 3.41 min, m/z = 301 [M+H]⁺, 323 [M+Na]⁺; **¹H-NMR (CDCl₃, 400 MHz)**: δ = 0.85-0.89 (t, 3H, NCH₂CH₂CH₃), 1.11 (s, 9H, T_{Bu}), 1.54-1.67 (m, 2H, NCH₂CH₂CH₃), 3.01-3.08 (m, 1H, 1H of NCH₂CH₂CH₃), 3.65-3.73 (d, 1H, 1H of NCH₂CH₂CH₃), 4.91 (s, 1H, CHC=CHCO), 5.26 (s, 1H, NH), 6.39 (s, 1H, C=CHCO), 7.34-7.35 (m, 3H, 3xCH-ar), 7.55-7.56 (m, 2H, 2xCH-ar); **¹³C-NMR (CDCl₃, 100 MHz)**: δ = 11.29 (CH₃), 21.58 (CH₂), 28.28 (CH₂), 51.60 (-C-(CH₃)₃), 69.38 (CHC=CHCO), 120.84 (C=CHCO), 126.86-130.54 (5xCH-ar), 155.16 (C=CHCO), 165.88 (CO), 171.31 (CO).

2-[[3-(4-Morpholin-4-yl-phenyl)-5-oxo-1-pentyl-2,5-dihydro-1*H*-pyrrol-2-carbonyl]-amino]-3-phenyl-propionic acid methylester (**11**): scheduled quantity 1 mmol gave 121 mg (23 %) C₃₀H₃₇N₃O₅ MW: 519.64; **HPLC-MS (ESI)**: t_R = 3.67 min, m/z = 520 [M+H]⁺, 542 [M+Na]⁺; **¹H-NMR (CDCl₃, 400 MHz)**: δ = 0.79 (t, 3H, CH₃), 1.16 (m, 6H, CH₂), 1.45 (t, 2H, NCH₂(CH₂)₃CH₃), 2.87 (t, 2H, CH₂), 2.96 (t, 2H, CH₂), 3.17 (m, 4H, CH₂), 3.49 (d, 1H, CHCH₂Ph), 3.60 (d, 1H, PhCH₂CH), 3.78 (s, 3H, CH₃OCO), 5.00 (s, 1H, COCHN), 6.17 (s, 1H, C=CHCO), 6.62 (d, 1H, ar-CH), 6.71 (d, 1H, ar-CH), 6.80 (m, 2H, ar-CH), 6.99 (m, 2H, ar-CH), 7.20 (d, 1H, ar-CH), 7.36 (d, 1H, ar-CH), 7.46 (d, 1H, ar-CH); **¹³C-NMR (CDCl₃, 100 MHz)**: δ = 13.88 (CH₃), 22.21 (CH₂), 28.49 (CH₂), 28.82 (CH₂), 37.03 (PhCH₂), 40.78 (NCH₂), 47.87 (CH₂), 52.33 (CH₃OCO), 52.25 (C=CHCO), 66.51 (CH₂), 67.48 (COCHNH), 114.31 (COCHN), 117.42 (ar-CH), 121.16 (ar-CH), 128.22 (ar-CH), 128.91 (ar-CH), 135.33 (ar-C), 152.37 (ar-C), 154.11 (C=CHCO), 167.18 (ar-C), 171.08 (CO), 171.76 (CO).

[3-(2-Cyclohexylcarbamoyl-5-oxo-3-phenyl-2,5-dihydro-pyrrol-1-yl)-propyl]-carbamic acid *tert*-butyl ester (**12**): scheduled quantity 3 mmol gave 347 mg (26 %) C₂₅H₃₅N₃O₄ MW: 441.56; **M.p.** 202°C; **HPLC-MS**

(ESI): $t_R = 3.56$ min, $m/z = 442$ $[M+H]^+$, 464 $[M+Na]^+$; **1H -NMR (CDCl₃, 400 MHz):** $\delta = 0.85$ -1.40 (m, 5H, 5 x CH₂-cyclohexyl), 1.52 (s, 9H, 3 x CH₃, ^{*t*}Bu), 1.54-1.79 (m, 7H, 5 x CH₂-cyclohexyl, CH₂), 3.00 (m, 1H, CH₂), 3.35 (m, 2 x 1H, CH₂), 3.62 (m, 1H, CH-cyclohexyl), 3.82 (m, 1H, CH₂), 5.13 (s, 1H, COCH(N;C)), 5.31 (br, 1H, NH), 6.08 (d, 1H, ³J_{CH,NH}: 8.53 Hz, NH), 6.40 (s, 1H, C=CHC=O), 7.34 (m, 3H, ar-CH), 7.60 (m, 2H, ar-CH); **^{13}C -NMR (CDCl₃, 100 MHz):** $\delta = 24.64$ (CH₂-cyclohexyl), 24.72 (CH₂-cyclohexyl), 25.16 (CH₂-cyclohexyl), 28.35 (C(CH₃)₃), 32.42 (CH₂), 32.60 (CH₂), 38.33 (CH₂), 48.62 (CH-cyclohexyl), 68.28 (COCH(C;N)), 79.06 (C(CH₃)₃), 120.60 (C=CHC=O), 126.86 (ar-CH), 128.80 (ar-CH), 130.28 (ar-C), 130.63 (ar-CH), 155.50 (C=CHC=O), 156.00 (C=O), 165.42 (C=O), 171.85 (C=O).

Elementar analysis:

calc:	C	68.000%	H	7.990%	N	9.52%	O	14.490%
found:	C	68.045%	H	7.955%	N	9.47%	O	14.425%
diff.:	C	0.045%	H	0.035%	N	0.05%	O	0.065%

3-(3,4-Dimethoxy-phenyl)-1-(3-methyl-butyl)-5-oxo-2,5-dihydro-1*H*-pyrrole-2-carboxylic acid allylamide (**13**): scheduled quantity 1 mmol gave 268.37 mg (78 %) C₂₁H₂₈N₂O₄ MW: 372.47 g/mol; **M.p.** 170.7°C-171.6°C; **R_f**: 0.29 (PE/EA, 1/4, v/v); **HPLC-MS (ESI):** $t_R = 3.34$ min, $m/z = 373.1$ $[M+H]^+$, 395.1 $[M+Na]^+$; **1H -NMR (CDCl₃, 400 MHz):** $\delta = 0.86$ -0.88 (d, 6H, 2xCH₃), 1.42-1.56 (m, 1H, CH(CH₃)₂), 2.96-3.03 (m, 2H, NCH₂CH₂CH(CH₃)₂), 3.71-3.76 (m, 2H, NCH₂CH₂CH(CH₃)₂), 3.78-3.82 (m, 2H, H₂C=CHCH₂NH), 3.84-3.85 (d, 6H, 2xOCH₃), 4.87-4.96 (m, 2H, =CH₂), 5.07 (m, 2H, CHC=CHCO), 5.56-5.66 (m, 1H, H₂C=CHCH₂NH), 6.25 (s, 1H, C=CHCO), 6.29-6.32 (t, 1H, NH), 6.80-6.82 (d, 1H, 1xCH-ar), 7.12-7.13 (d, 1H, 1xCH-ar), 7.21-7.25 (m, 1H, 1xCH-ar); **^{13}C -NMR (CDCl₃, 100 MHz):** $\delta = 22.19$ (CH₃), 22.56 (CH₃), 25.95 (CH(CH₃)₂), 36.87 (CH₂), 39.55 (CH₂), 41.78 (CH₂), 55.89 (OCH₃), 55.95 (OCH₃), 67.99 (CHC=CHCO), 109.75 (1xCH-ar), 110.99 (1xCH-ar), 116.53 (=CH₂), 118.66 (H₂C=CHCH₂NH), 120.46 (C=CHCO), 123.15 (1xCH-ar), 133.25, 148.98, 151.06, 154.84, 167.12 (CO), 171.53 (CO); **IR:** ? (cm⁻¹) = 3284 (m), 3076 (w), 2957 (w), 2871 (w), 1689 (s), 1651 (s), 1599 (w), 1519 (w), 1218 (m), 801 (w).

{[1-(3-Methyl-butyl)-5-oxo-3-phenyl-2,5-dihydro-1*H*-pyrrole-2-carbonyl]-amino}-acetic acid methyl ester (**14**): scheduled quantity 1 mmol gave 24 mg (7 %) C₁₉H₂₄N₂O₄ MW: 344.41 g/mol; **HPLC-MS (ESI):** $t_R = 3.31$ min, $m/z = 345$ $[M+H]^+$, 367 $[M+Na]^+$; **1H -NMR (CDCl₃, 400 MHz):** $\delta = 0.85$ (d, 6H, 2x CH₃), 1.52-1.42 (m, 3H, CH₃), 3.18-3.12 (m, 1H, C13), 3.56 (s, 3H, O-CH₃), 3.74-3.69 (m, 1H, 1H CO-CH₂-NH), 3.90-3.84 (m, 1H, N-CH₂), 4.03-3.97 (m, 1H, 1H CO-CH₂-NH), 5.14 (s, 1H, CH), 6.39 (s, 1H, CH), 7.62-7.34 (m, NH+5H CH-ar); **^{13}C -NMR (CDCl₃, 100 MHz):** $\delta = 22.13$ (CH₃), 22.59 (CH₃), 25.86 (CH(CH₃)₂), 36.87 (N-CH₂-CH₂-CH-), 39.30 (N-CH₂-), 171.5 (CO), 41.04 (OCO-CH₂-NH-), 52.13 (O-CH₃), 67.49, 120.97, 127.04 (CH ar.), 128.85 (CH ar), 130.37 (CH ar), 130.51, 154.753, 167.5 (CO); 169.56 (CO).

1-Cyclopropyl-3-naphthalen-2-yl-5-oxo-2,5-dihydro-1*H*-pyrrole-2-carboxylic acid (pyridin-3-ylmethyl)-amide (**15**): scheduled quantity 1 mmol gave 310 mg (81 %) C₂₄H₂₁N₃O₂ MW: 383.45 g/mol; **HPLC-MS (ESI):** $t_R = 2.96$ min, $m/z = 384$ $[M+H]^+$, 406 $[M+Na]^+$; **1H -NMR (d₆-DMSO, 400 MHz):** $\delta = 0.59$ -0.71 (m, 2H, CH₂-Cyclopropyl), 0.73-0.79 (m, 2H, CH₂-cyclopropyl), 3.15 (s, 1H, CH-cyclopropyl), 4.21-4.33 (m, 2H, CH₂-Py), 5.41 (s, 1H, CHC=CHCO), 6.68 (s, 1H, C=CHCO), 6.94-6.97 (q, 1H, 2xCH-ar), 7.30-7.32 (d, 1H, 1xCH-ar), 7.55-7.57 (t, 2H, 2xCH-ar), 7.77-7.81 (m, 2H, 2xCH-ar), 7.90-7.92 (m, 2H, 2xCH-ar), 8.20 (s, 1H, 1xCH-ar), 8.29-8.36 (m, 2H, 2xCH-ar), 9.28-9.31 (t, 1H, 1xCH-ar); **^{13}C -NMR (d₆-DMSO, 100 MHz):** $\delta = 3.55$ (CH₂-cyclopropyl), 5.77 (CH₂-cyclopropyl), 22.93 (CH-cyclopropyl), 66.37 (CHC=CHCO), 122.02 (C=CHCO), 123.82 (1xCH-ar), 125.73 (1xCH-ar), 126.33 (1xCH-ar), 126.77 (1xCH-ar), 127.13 (1xCH-ar), 127.85 (1xCH-ar), 132.00 (1xCH-ar), 132.85 (1xCH-ar), 134.43 (1xCH-ar), 147.33 (1xCH-ar), 147.91 (1xCH-ar), 153.82 (C=CHCO), 166.17 (CO), 171.47 (CO).

1-Benzyl-3-biphenyl-4-yl-5-oxo-2,5-dihydro-1*H*-pyrrole-2-carboxylic acid *tert*-butylamide (**16**): scheduled quantity 1 mmol gave 325 mg (95 %) C₂₈H₂₈N₂O₂ MW: 424.54 g/mol; **R_f**: 0.72 (PE/EA, 1/4, v/v); **HPLC-MS (ESI):** $t_R = 3.96$ min, $m/z = 425.2$ $[M+H]^+$, 447.2 $[M+Na]^+$; **1H -NMR (CDCl₃, 400 MHz):** $\delta = 1.07$ (s, 9H, T_{BU}), 4.11-4.15 (d, 1H, ²J=14.86Hz, 1H of NCH₂Ph), 4.77 (s, 1H, CHC=CHCO), 5.00-5.04 (d, 1H, ²J=14.86Hz, 1H of NCH₂Ph), 5.35 (s, 1H, NH), 6.44 (s, 1H, C=CHCO), 7.18-7.38 (m, 9H, 9xCH-ar), 7.50-7.59 (m, 5H, 5xCH-ar); **^{13}C -NMR (CDCl₃, 100 MHz):** $\delta = 28.21$ (T_{BU}), 45.11 (NCH₂Ph), 51.57 (C-(CH₃)₃), 68.69 (CHC=CHCO), 120.28 (C=CHCO), 126.92-128.84 (14xCH-ar), 129.24, 136.54, 139.71, 143.13, 155.29, 165.38 (CO), 171.25 (CO).

3-(3,4-Dimethoxy-phenyl)-1-(3-methyl-butyl)-5-oxo-4-phenyl-2,5-dihydro-1*H*-pyrrole-2-carboxylic acid allylamide (**17**): scheduled quantity 1 mmol gave 91 mg (20 %) $C_{27}H_{32}N_2O_4$ MW: 448.57 g/mol; **R_f**: 0.80 (PE/EA, 1/4, v/v); **HPLC-MS (ESI)**: t_R = 3.72 min, m/z = 449.2 $[M+H]^+$, 471.2 $[M+Na]^+$; **¹H-NMR (CDCl₃, 400 MHz)**: δ = 0.83-0.85 (d, 6H, 2xCH₃), 1.40-1.55 (m, 1H, CH(CH₃)₂), 3.00-3.08 (m, 1H, 1H of NCH₂CH₂CH(CH₃)₂), 3.29-3.38 (m, 1H, 1H of NCH₂CH₂CH(CH₃)₂), 3.44 (s, 3H, OCH₃), 3.70-3.74 (m, 1H, 1H of NCH₂CH₂CH(CH₃)₂), 3.78 (s, 3H, OCH₃), 3.87-3.96 (m, 1H, 1H of NCH₂CH₂CH(CH₃)₂), 4.99 (s, 1H, CHC=CHCO), 5.02-5.04 (m, 2H, =CH₂), 5.53-5.71 (m, 1H, H₂C=CHCH₂NH), 6.67-6.70 (m, 2H, 2xCH-ar), 6.83-6.85 (m, 1H, 1xCH-ar), 7.19-7.26 (m, 3H, 3xCH-ar), 7.31-7.36 (m, 2H, 2xCH-ar); **¹³C-NMR (CDCl₃, 100 MHz)**: δ = 22.23 (CH₃), 22.33 (CH₃), 22.42, 22.61, 25.90 (CH(CH₃)₂), 26.12, 36.88, 37.35 (CH₂), 38.18 (CH₂), 40.04, 41.70, 42.80 (CH₂), 55.37 (OCH₃), 55.72 (OCH₃), 67.38 (CHC=CHCO), 110.72 (1xCH-ar), 110.81 (1xCH-ar), 111.74 (1xCH-ar), 111.95 (1xCH-ar), 116.39 (=CH₂), 117.27 (H₂C=CHCH₂NH), 121.59 (1xCH-ar), 121.90 (1xCH-ar), 123.07 (1xCH-ar), 128.26 (1xCH-ar), 129.62 (1xCH-ar), 129.76 (1xCH-ar), 130.77 (1xCH-ar), 132.31 (1xCH-ar), 132.66 (1xCH-ar), 133.32 (1xCH-ar), 148.34, 148.46, 149.65, 149.88, 166.99, 169.04, 170.06 (CO), 171.24 (CO).

{[1-Allyl-4-(3-methoxy-phenyl)-6-oxo-1,2,3,6-tetrahydro-pyridine-2-carbonyl]-amino}-acetic acid methyl ester (**18**): scheduled quantity 7 mmol gave 135 mg (12 %) $C_{19}H_{22}N_2O_5$ MW: 358.39 g/mol; **HPLC-MS (ESI)**: t_R = 3.15 min, m/z = 359.1 $[M+H]^+$, 381.1 $[M+Na]^+$; **¹H-NMR (CDCl₃, 400 MHz)**: δ = 2.97-3.03 (dd, 1H, ²J=8.60Hz, ³J=17.99Hz; 1H of CHCH₂C=CHCO), 3.39-3.44 (d, 1H, ²J=17.21Hz 1H of CHCH₂C=CHCO), 3.50-3.56 (dd, 2H, ²J=7.82Hz, ³J=15.65Hz, COCH₂NH), 3.63 (s, 3H, COOCH₃), 3.78 (s, 3H, OCH₃), 3.93-3.94 (d, 1H, ²J=5.47Hz, 1H of NCH₂CH=CH₂), 4.06-4.07 (d, 1H, ²J=5.47Hz, 1H of NCH₂CH=CH₂), 4.22-4.24 (d, 1H, ²J=7.28Hz, CHCH₂C=CHCO), 4.86-4.91 (dd, 1H, ²J=4.69Hz, ³J=15.65Hz, NH), 5.24-5.28 (t, 2H, =CH₂), 5.78-5.90 (m, 1H, =CH), 6.27 (s, 1H, C=CHCO), 6.89-7.06 (m, 3H, CH-ar), 7.25-7.44 (m, 3H, CH-ar); **¹³C-NMR (CDCl₃, 100 MHz)**: δ = 28.73 (CHCH₂C=CHCO), 41.26 (NCH₂CH=CH₂), 48.17 (COCH₂NHCO), 52.25 (COOCH₃), 55.26 (OCH₃), 57.78 (CHCH₂C=CHCO), 111.47 (CH-ar), 115.61 (C=CHCO), 118.58 (=CH₂), 118.93 (=CH), 129.16 (CH-ar), 129.70 (CH-ar), 132.35 (CH-ar), 138.13, 148.64, 159.76, 169.71 (CO), 171.13 (CO).

1-(3-Ethoxy-phenyl)-4-(2-methoxy-phenyl)-6-oxo-1,2,3,6-tetrahydro-pyridine-2-carboxylic acid *tert*-butylamide (**19**): scheduled quantity 1 mmol gave 59 mg (14 %) $C_{25}H_{30}N_2O_4$ MW: 422.52 g/mol; **HPLC-MS (ESI)**: t_R = 3.74 min, m/z = 423.1 $[M+H]^+$, 445.1 $[M+Na]^+$; **¹H-NMR (CDCl₃, 400 MHz)**: δ = 1.24 (s, 9H, T_{BU}), 1.32-1.36 (t, 3H, OCH₂CH₃), 3.16-3.22 (ddd, 1H, ²J=1.81Hz, ³J=7.24Hz, ⁴J=17.20Hz, 1H of CHCH₂C=CHCO), 3.32-3.37 (dd, 1H, ²J=2.71Hz, ³J=18.11Hz, 1H of CHCH₂C=CHCO), 3.77 (s, 3H, OCH₃), 3.93-3.98 (q, 2H, OCH₂CH₃), 4.37-4.39 (dd, 1H, ²J=1.81Hz, ³J=6.33Hz, CHCH₂C=CHCO), 5.98 (s, 1H, NH), 6.19 (s, 1H, ²J=1.81Hz, C=CHCO), 6.71-6.73 (d, 1H, 1xCH-ar), 6.84-6.92 (m, 4H, 4xCH-ar), 7.17-7.30 (m, 3H, 3xCH-ar); **¹³C-NMR (CDCl₃, 100 MHz)**: δ = 14.76 (OCH₂CH₃), 28.54 (T_{BU}), 31.95 (CHCH₂C=CHCO), 51.78 (-C-(CH₃)₃), 55.30 (OCH₃), 63.52 (CHCH₂C=CHCO), 63.58 (OCH₂CH₃), 111.07 (1xCH-ar), 111.32 (1xCH-ar), 112.83 (1xCH-ar), 116.58 (C=CHCO), 120.78 (1xCH-ar), 122.23 (1xCH-ar), 127.36, 128.86-130.63 (3xCH-ar), 142.57, 150.55, 156.95, 164.03, 172.04 (CO).

1-Allyl-4-(3-methoxy-phenyl)-6-oxo-1,2,3,6-tetrahydro-pyridine-2-carboxylic acid allylamide (**20**): scheduled quantity 4 mmol gave 370 mg (64 %) $C_{19}H_{22}N_2O_3$ MW: 326.39 g/mol; **R_f**: 0.57 (EA); **HPLC-MS (ESI)**: t_R = 3.27 min, m/z = 337.15 $[M+H]^+$, 349.1 $[M+Na]^+$; **¹H-NMR (CDCl₃, 400 MHz)**: δ = 2.90-2.97 (ddd, 1H, ²J=2.34Hz, ³J=7.82Hz, ⁴J=17.21Hz, 1H of CHCH₂C=CHCO), 3.37-3.45 (m, 1H, 1H of CHCH₂C=CHCO), 3.73 (s, 3H, OCH₃), 3.78-3.85 (m, 2H, NHCH₂CH=CH₂), 4.13-4.15 (d, 1H, ²J=7.04Hz, CHCH₂C=CHCO), 4.76-4.81 (dd, 1H, ²J=4.69Hz, ³J=15.65Hz, 1H of NCH₂CH=CH₂), 4.98-5.02 (t, 2H, =CH₂), 5.18-5.22 (t, 2H, =CH₂), 5.63-5.73 (m, 1H, =CH), 5.75-5.83 (m, 1H, =CH), 6.19 (d, 1H, ²J=1.56Hz, C=CHCO), 6.59 (s, 1H, NH), 6.85-7.00 (m, 3H, 3xCH-ar), 7.19-7.24 (m, 1H, 1xCH-ar); **¹³C-NMR (CDCl₃, 100 MHz)**: δ = 28.93 (CHCH₂C=CHCO), 42.01 (CH₂), 48.34 (CH₂), 55.28 (OCH₃), 58.04 (CHCH₂C=CHCO), 111.58 (C=CHCO), 115.51 (=CH₂), 116.29 (=CH), 118.61 (=CH₂), 118.77 (=CH), 118.90, 129.77 (CH-ar), 132.32 (CH-ar), 133.42 (CH-ar), 138.16, 149.13, 159.79, 164.27 (CO), 170.26 (CO).

1-Benzyl-4-naphthalen-2-yl-6-oxo-1,2,3,6-tetrahydro-pyridine-2-carboxylic acid *tert*-butylamide (**21**): scheduled quantity 1 mmol gave 73 mg (17 %) $C_{27}H_{28}N_2O_2$ MW: 412.54 g/mol; **HPLC-MS (ESI)**: t_R = 3.92 min; m/z = 413 $[M+H]^+$, 435 $[M+Na]^+$; **1H -NMR ($CDCl_3$, 400 MHz)**: δ = 1.05 (s, 9H, $C(CH_3)_3$), 2.93-2.89 (m, 1H, CH), 3.51 (d, 1H, CH), 4.03 (d, 1H, CH), 4.44 (d, 1H, CH), 4.95 (d, 1H, CH), 5.57 (s, 1H, NH), 6.39 (d, 1H, CH), 7.35-7.23 (m, 5CH-ar), 7.43-7.41 (d, 2H, CH-ar), 7.54-7.52 (d, 1H, CH-ar), 7.85-7.73 (m, 4H, CH-ar); **^{13}C -NMR ($CDCl_3$, 100 MHz)**: δ = 28.2 ($C(\underline{CH_3})_3$), 30.1 ($\underline{CH_2}$), 50.0 (CH_2), 51.4 ($\underline{C}(\underline{CH_3})_3$), 59.7, 118.9, 123.2 (CH-ar), 129.0-126.2 (11xCH-ar), 132.9, 133.7 (C-ar), 134.1 (C-ar), 136.9 (C-ar), 149.1 (C-ar), 164.6 (CO), 169.3 (CO).

1-Benzyl-4-(2-methoxy-phenyl)-6-oxo-1,2,3,6-tetrahydro-pyridine-2-carboxylic acid *tert*-butylamide (**22**): scheduled quantity 1 mmol gave 259.4 mg (66 %) $C_{24}H_{28}N_2O_3$ MW: 392.50 g/mol; **R_f**: 0.74 (PE/EA, 1/4, v/v); **HPLC-MS (ESI)**: t_R = 3.73 min, m/z = 393.1 $[M+H]^+$, 415.1 $[M+Na]^+$; **1H -NMR ($CDCl_3$, 400 MHz)**: δ = 1.16 (s, 9H, $C(CH_3)_3$), 2.95-3.02 (ddd, 1H, $^2J=2.34$ Hz, $^3J=7.04$ Hz, $^4J=17.21$ Hz, 1H of $CHCH_2C=CHCO$), 3.21-3.26 (dd, 1H, $^2J=1.56$ Hz, $^3J=17.21$ Hz, 1H of $CHCH_2C=CHCO$), 3.78 (s, 3H, OCH_3), 3.95-3.97 (dd, 1H, $^2J=1.56$ Hz, $^3J=7.82$ Hz, $CHCH_2C=CHCO$), 4.36-4.40 (d, 1H, $^2J=14.86$ Hz, 1H of NCH_2Ph), 4.98-5.01 (d, 1H, $^2J=14.08$ Hz, 1H of NCH_2Ph), 5.62 (s, 1H, NH), 6.16 (s, 1H, $^2J=2.34$ Hz, $C=CHCO$), 6.85-6.94 (m, 2H, 2xCH-ar), 7.15-7.17 (m, 1H, 1xCH-ar), 7.25-7.39 (m, 6H, 6xCH-ar); **^{13}C -NMR ($CDCl_3$, 100 MHz)**: δ = 28.30 ($C(\underline{CH_3})_3$), 31.71 ($CHCH_2C=CHCO$), 49.76 (NCH_2Ph), 51.29 ($\underline{C}-(CH_3)_3$), 55.18 (OCH_3), 59.93 ($CHCH_2C=CHCO$), 110.95 ($C=CHCO$), 120.64-121.35 (2xCH-ar), 127.48, 127.88-130.43 (5xCH-ar), 137.10, 149.68, 156.82, 164.37 (CO), 169.31 (CO); **IR**: ? (cm^{-1}) = 3323 (m), 3068 (w), 2968 (m), 1659 (s), 1615 (m), 1547 (m), 1436 (m), 1366 (m), 1239 (m), 800 (w), 745 (m), 694 (m).

2-Benzyl-3-oxo-1,2,3,5,6,7,8,8a-octahydro-isoquinoline-1-carboxylic acid *tert*-butylamide (**23**): scheduled quantity 1 mmol gave 204 mg (27 %) $C_{21}H_{28}N_2O_2$ MW: 340.47 g/mol; **HPLC-MS (ESI)**: t_R = 3.60 min, m/z = 341 $[M+H]^+$, 363 $[M+Na]^+$; **1H -NMR ($CDCl_3$, 400 MHz)**: δ = 1.21 (s, 9H, $C(\underline{CH_3})_3$), 1.37 (m, 1H, CH), 1.49 (m, 1H, CH_2), 1.61 (d, 1H, CH_2), 1.94 (m, 1H, CH), 2.12 (m, 1H, CH), 2.37 (d, 1H, CH_2), 2.73 (d, 1H, CH_2), 3.81 (t, 1H, CH), 4.02 (d, 1H, $PhCH_2N$), 5.19 (d, 1H, $PhCH_2N$), 5.33 (d, 1H, CH), 5.55 (d, 1H, CH), 5.72 (s, 1H, NH), 7.26-7.31 (m, 5H, ar-CH); **^{13}C -NMR ($CDCl_3$, 100 MHz)**: δ = 24.58 (CH_2), 24.68 (CH_2), 25.93 (CH_2), 27.97 (CH_2), 28.40 ($C(\underline{CH_3})_3$), 28.62 ($C(\underline{CH_3})_3$), 29.74 (CH_2), 32.75 (CH), 35.37 (CH_2), 35.40 (CH_2), 42.65 (CH), 48.39 ($PhCH_2N$), 49.37 ($PhCH_2N$), 51.43 ($\underline{C}(\underline{CH_3})_3$), 51.66 ($\underline{C}(\underline{CH_3})_3$), 64.33 ($COCHN$), 65.03 ($COCHN$), 115.12 ($C=CH$), 117.73 ($C=CH$), 127.53 (ar-CH), 127.81 (ar-CH), 128.34 (ar-CH), 128.62 (ar-CH), 128.65 (ar-CH), 128.74 (ar-CH), 136.71 (ar-C), 137.00 (ar-C), 153.82 ($\underline{C}=CHCO$), 159.46 (qC), 163.87 (CO), 163.99 (CO), 168.45 (CO). The signals in the ^{13}C -nmr are doubled as a consequence of the mixture of diastereomers.

1-Benzyl-4-biphenyl-4-yl-6-oxo-1,2,3,6-tetrahydro-pyridine-2-carboxylic acid *tert*-butylamide (**24**): scheduled quantity 1 mmol gave 44 mg (14 %) $C_{29}H_{30}N_2O_2$ MW: 438.57 g/mol; **R_f**: 0.82 (PE/EA, 1/4, v/v); **HPLC-MS (ESI)**: t_R = 3.91 min, m/z = 439.1 $[M+H]^+$, 461.1 $[M+Na]^+$; **1H -NMR ($CDCl_3$, 400 MHz)**: δ = 1.06 (s, 9H, T_{BU}), 2.80-2.87 (ddd, 1H, $^2J=2.34$ Hz, $^3J=7.04$ Hz, $^4J=17.21$ Hz, 1H of $CHCH_2C=CHCO$), 3.35-3.39 (dd, 1H, $^2J=1.56$ Hz, $^3J=17.21$ Hz, 1H of $CHCH_2C=CHCO$), 3.97-3.99 (dd, 1H, $^2J=2.34$ Hz, $^3J=7.82$ Hz, $CHCH_2C=CHCO$), 4.40-4.43 (d, 1H, $^2J=14.86$ Hz, 1H of NCH_2Ph), 4.88-4.91 (d, 1H, $^2J=14.86$ Hz, 1H of NCH_2Ph), 5.54 (s, 1H, NH), 6.28 (s, 1H, $^2J=2.34$ Hz, $C=CHCO$), 7.23-7.38 (m, 8H, 8xCH-ar), 7.46-7.55 (m, 6H, 6xCH-ar); **^{13}C -NMR ($CDCl_3$, 100 MHz)**: δ = 28.25 ($C(CH_3)_3$), 30.13 ($CHCH_2C=CHCO$), 50.05 (NCH_2Ph), 51.43 ($\underline{C}-(CH_3)_3$), 59.69 ($CHCH_2C=CHCO$), 118.57 ($C=CHCO$), 126.63-128.06 (9xCH-ar), 128.63-129.04 (5xCH-ar), 135.82, 137.01, 139.97, 142.57, 149.04, 164.55 (CO), 169.28 (CO); **IR**: ? (cm^{-1}) = 3293 (s), 3060 (w), 2974 (m), 1673 (s), 1646 (s), 1600 (s), 1581 (m), 1544 (s), 1449 (s), 1429 (s), 1359 (m), 836 (s), 763 (s).

4-(3-Methoxy-phenyl)-6-oxo-1-phenyl-1,2,3,6-tetrahydro-pyridine-2-carboxylic acid *tert*-butylamide (**25**): scheduled quantity 0.19 mmol gave 50 mg (70 %) $C_{23}H_{26}N_2O_3$ MW: 378.47 g/mol; **HPLC-MS (ESI)**: t_R = 3.61 min, m/z = 379.1 $[M+H]^+$, 401.1 $[M+Na]^+$; **1H -NMR ($CDCl_3$, 400 MHz)**: δ = 1.25 (s, 9H, T_{BU}), 3.09-3.16 (ddd, 1H, $^2J=2.34$ Hz, $^3J=7.04$ Hz, $^4J=17.21$ Hz, 1H of $CHCH_2C=CHCO$), 3.50-3.55 (dd, 1H, $^2J=2.34$ Hz, $^3J=17.21$ Hz, 1H of $CHCH_2C=CHCO$), 3.82 (s, 3H, $\underline{OCH_3}$), 4.50-4.52 (dd, 1H, $^2J=2.34$ Hz, $^3J=6.26$ Hz, $CHCH_2C=CHCO$), 6.02 (s, 1H, NH), 6.37 (d, 1H, $^2J=1.56$ Hz, $C=CHCO$), 6.85-7.13 (m, 3H,

3xCH-ar), 7.25-7.43 (m, 6H, 6xCH-ar); ¹³C-NMR (CDCl₃, 100 MHz): δ = 28.43 (C(CH₃)₃), 30.34 (CHCH₂C=CHCO), 51.82 (-C-(CH₃)₃), 55.27 (OCH₃), 62.94 (CHCH₂C=CHCO), 111.58 (1xCH-Ar), 115.62 (C=CHCO), 118.69 (1xCH-ar), 119.62 (1xCH-ar), 124.66 (1xCH-ar), 126.46 (1xCH-ar), 127.83-129.80 (5xCH-ar), 138.29, 141.24, 150.12, 159.79, 164.06 (CO), 169.37 (CO).

4-(3-Methoxy-phenyl)-6-oxo-1-pyridin-2-ylmethyl-1,2,3,6-tetrahydro-pyridine-2-carboxylic acid *tert*-butylamide (**26**): scheduled quantity 1 mmol gave 59 mg (24 %) C₂₃H₂₇N₃O₃ MW: 393.49 g/mol; R_f: 0.84 (DCM/MeOH, 9/1, v/v); HPLC-MS (ESI): t_R = 3.22 min, m/z = 394.2 [M+H]⁺, 416.2 [M+Na]⁺; ¹H-NMR (CDCl₃, 400 MHz): δ = 1.26 (s, 9H, C(CH₃)₃), 2.92-2.96 (d, 1H, ²J=16.4Hz, 1H of CHCH₂C=CHCO), 3.48-3.52 (d, 1H, ²J=17.2Hz, 1H of CHCH₂C=CHCO), 3.79 (s, 3H, OCH₃), 4.2 (s, 1H, CHCH₂C=CHCO), 4.76 (s, 2H, NCH₂Py), 6.22 (s, 1H, C=CHCO), 6.90-7.07 (m, 2H, 2xCH-Py), 7.20-7.35 (m, 3H, 3xCH-ar), 7.68-7.78 (m, 2H, 2xCH-Py), 8.49(s, 1H, 1xCH-Ar); ¹³C-NMR (CDCl₃, 100 MHz): δ = 28.49 (C(CH₃)₃), 30.56 (CHCH₂C=CHCO), 51.39 (-C-(CH₃)₃), 53.01 (NCH₂Py), 55.23 (OCH₃), 61.30 (CHCH₂C=CHCO), 111.53 (C=CHCO), 115.27 (1xCH-ar), 118.72 (1xCH-ar), 119.09 (1xCH-ar), 122.43 (1xCH-ar), 122.49 (1xCH-ar), 129.65 (1xCH-ar), 137.13 (1xCH-ar), 138.79, 148.55, 149.75, 156.78, 159.68, 165.07 (CO), 169.56 (CO).

1-Benzyl-6-oxo-3,4-diphenyl-1,2,3,6-tetrahydro-pyridine-2-carboxylic acid *tert*-butylamide (**27**): scheduled quantity 1 mmol gave 20 mg (10 %) C₂₉H₃₀N₂O₂ MW: 438.57 g/mol; HPLC-MS (ESI): t_R = 3.98 min, m/z = 439 [M+H]⁺, 461 [M+Na]⁺; ¹H-NMR (CDCl₃, 400 MHz): δ = 1.04 (s, 9H, (CH₃)₃), 3.89 (s, 1H, CH), 4.27-4.23 (d, 1H, CH), 4.48-4.51 (d, 1H, CH), 4.80 (s, 1H, CH), 5.49 (s, 1H, NH), 6.53 (s, 1H, C=CHCO), 7.00-7.36 (m, 15H, ar-CH); ¹³C-NMR (CDCl₃, 100 MHz): δ = 28.18 ((CH₃)₃), 45.32, 50.88 (NCH₂), 51.43 (C(CH₃)₃), 68.35, 120.42, 126.56-129.70 (ar-CH), 135.91, 136.59, 138.10, 150.68, 164.49 (CO), 168.75 (CO).

X-ray Experiments

X-ray single crystal structure determination of compound **1**: *Crystal data*: C₂₈H₃₇N₂O₆P, M_r = 528.57, colourless crystal, 0.10 × 0.18 × 0.89 mm³, monoclinic, space group P2₁/n (No. 14), a = 12.1059(1), b = 10.3873(1), c = 21.7121(3) Å, β = 99.1995(5)°, V = 2695.13(5) Å³, Z = 4, ρ_{calcd} = 1.303 g cm⁻³, F(000) = 1128, m_{Mo-Kα} = 0.147 mm⁻¹, T = 173 K. *Data collection*: Nonius MACH3 κ-axis CCD-diffractometer, Mo-K_α radiation (λ = 0.71073 Å), unit cell parameters by least-squares for 5180 reflections. A total of 36131 reflections were integrated^{a)} and scaled^{b)}. After merging R_{int} = 0.039) 4940 [4160: I₀>2σ(I₀)] independent reflections remained and all were used to refine 482 parameters. *Structure solution and refinement*: The structure was solved by a combination of direct methods^{c)} and difference-Fourier syntheses^{d)}. All non-hydrogen atoms were refined with anisotropic displacement parameters. All hydrogen atoms were found and refined with individual isotropic displacement parameters. Full-matrix least-squares refinements were carried out by minimizing S_w(F_o²-F_c²)² and converged with R1 = 0.0384 [I₀>2σ(I₀)], wR2 = 0.0973 [all data], GOF = 1.028 and shift/error < 0.001.

X-ray single crystal structure determination of compound **2**: *Crystal data*: C₂₃H₂₆N₂O₂, M_r = 362.46, colourless crystal, 0.10 × 0.10 × 0.15 mm³, orthorhombic, space group Pna2₁ (No. 33), a = 11.0801(1), b = 9.6590(1), c = 18.7896(3) Å, V = 2010.91(4) Å³, Z = 4, ρ_{calcd} = 1.197 g cm⁻³, F(000) = 776, m_{Mo-Kα} = 0.077 mm⁻¹, T = 173 K. *Data collection*: Nonius MACH3 κ-axis CCD-diffractometer, Mo-K_α radiation (λ = 0.71073 Å), unit cell parameters by least-squares for 2050 reflections. A total of 25584 reflections were integrated^{a)} and scaled^{b)}. After merging R_{int} = 0.060) 3417 [3166: I₀>2σ(I₀)] independent reflections remained and all were used to refine 348 parameters. *Structure solution and refinement*: The structure was solved by a combination of direct methods^{c)} and difference-Fourier syntheses^{d)}. All non-hydrogen atoms were refined with anisotropic displacement parameters. All hydrogen atoms were found and refined with individual isotropic displacement parameters. Full-matrix least-squares refinements were carried out by minimizing S_w(F_o²-F_c²)² and converged with R1 = 0.0526 [I₀>2σ(I₀)], wR2 = 0.0888 [all data], GOF = 1.194 and shift/error < 0.001. As indicated by Flack's parameter ε = -0.1(15) the correct enantiomere could not be assigned reliably.

X-ray single crystal structure determination of compound **3**: *Crystal data*: $C_{12}H_{21}NO_2$, $M_r = 211.30$, colourless crystal, $0.38 \times 0.43 \times 0.89 \text{ mm}^3$, monoclinic, space group $P2_1/c$ (No. 14), $a = 11.8205(2)$, $b = 11.7100(2)$, $c = 10.0468(2) \text{ \AA}$, $\beta = 110.3970(7)^\circ$, $V = 1303.46(4) \text{ \AA}^3$, $Z = 4$, $\rho_{\text{calcd}} = 1.077 \text{ g cm}^{-3}$, $F(000) = 464$, $\mu_{\text{Mo-K}\alpha} = 0.072 \text{ mm}^{-1}$, $T = 173 \text{ K}$. *Data collection*: Nonius MACH3 κ -axis CCD-diffractometer, Mo-K α radiation ($I = 0.71073 \text{ \AA}$), unit cell parameters by least-squares for 2508 reflections. A total of 20268 reflections were integrated^{a)} and scaled^{b)}. After merging ($R_{\text{int}} = 0.035$) 2378 [$I_o > 2s(I_o)$] independent reflections remained and all were used to refine 220 parameters. *Structure solution and refinement*: The structure was solved by a combination of direct methods^{c)} and difference-Fourier syntheses^{d)}. All non-hydrogen atoms were refined with anisotropic displacement parameters. All hydrogen atoms were found and refined with individual isotropic displacement parameters. Full-matrix least-squares refinements were carried out by minimizing $S_w(F_o^2 - F_c^2)^2$ and converged with $R1 = 0.0406$ [$I_o > 2s(I_o)$], $wR2 = 0.1082$ [all data], $GOF = 1.040$ and shift/error < 0.001.

X-ray single crystal structure determination of compound **4**: *Crystal data*: $C_{20}H_{21}NO_2$, $M_r = 307.38$, colourless crystal, $0.10 \times 0.28 \times 0.66 \text{ mm}^3$, orthorhombic, space group $Pbca$ (No. 61), $a = 9.1359(1)$, $b = 29.9776(4)$, $c = 12.5566(1) \text{ \AA}$, $V = 3438.91(7) \text{ \AA}^3$, $Z = 8$, $\rho_{\text{calcd}} = 1.187 \text{ g cm}^{-3}$, $F(000) = 1312$, $\mu_{\text{Mo-K}\alpha} = 0.076 \text{ mm}^{-1}$, $T = 173 \text{ K}$. *Data collection*: Nonius MACH3 κ -axis CCD-diffractometer, Mo-K α radiation ($I = 0.71073 \text{ \AA}$), unit cell parameters by least-squares for 3578 reflections. A total of 58875 reflections were integrated^{a)} and scaled^{b)}. After merging ($R_{\text{int}} = 0.047$) 3136 [$I_o > 2s(I_o)$] independent reflections remained and all were used to refine 292 parameters. *Structure solution and refinement*: The structure was solved by a combination of direct methods^{c)} and difference-Fourier syntheses^{d)}. All non-hydrogen atoms were refined with anisotropic displacement parameters. All hydrogen atoms were found and refined with individual isotropic displacement parameters. Full-matrix least-squares refinements were carried out by minimizing $S_w(F_o^2 - F_c^2)^2$ and converged with $R1 = 0.0595$ [$I_o > 2s(I_o)$], $wR2 = 0.1655$ [all data], $GOF = 1.023$ and shift/error < 0.001.

Graphics

Thermal ellipsoids in all ORTEP-style representations of the molecular structures of **1**, **2**, **3**, and **4** in the solid state are drawn at the 50% probability level.^{e)}

Deposit

Crystallographic data (excluding structure factors) for the structures reported in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication no. CCDC-**WWWWW (1)**, **XXXXXX (2)**, **YYYYYY (3)**, **ZZZZZZ (4)**. Copies of the data can be obtained free of charge on application to CCDC, 12 Union Road, Cambridge CB2 1EZ, UK [Fax: (internat.) +44-1223/336-033; E-mail: deposit@ccdc.cam.ac.uk].

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