Supporting Information

for

Pd(OAc)$_2$-catalyzed dehydrogenative C–H activation:

An expedient synthesis of uracil-annulated

$\beta$-carbolinones

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(1) General remarks

Melting points were measured in open capillaries and are uncorrected. The $^1$H and $^{13}$C NMR spectroscopic data were recorded in CDCl$_3$ with TMS as the internal standard (chemical shift in $\delta$) with a Bruker DPX-400 spectrometer. Data are reported as follows: chemical shifts, multiplicity (s = singlet, d = doublet, t = triplet, q = quartet, quintet = quintet, m = multiplet, brs = broad singlet), coupling constant (Hz). In case of 5f two peaks are missing at 40.13 ppm and at 139.86 ppm and in case of starting materials (except 4i and 4j) one peak is missing in the region ~138 ppm in $^{13}$C NMR (assumed from DEPT NMR of 4c) and equivalent carbons are marked in $^{13}$C spectra of 5f and 4a. HRMS (ESI) spectra were taken using Waters Xevo G2 QTof mass spectrometer. Silica gel [60-120 mesh (Rankem, India), (230–400 mesh (Spectrochem, India)] was used for the chromatographic separations. Silica gel G and GF 254 (CDH, India) was used for TLC. Petroleum ether refers to the fraction boiling between 60 and 80 °C.

(2) (a) General procedure$^1$ for the preparation of starting material (4):

To a stirred solution of 1 equiv. of indole-2-carboxylic acid 1 in 10 mL of dry benzene containing 2 drops of N, N-dimethylformamide, 1.5 equiv. of oxalyl chloride was added. The resulting solution was stirred at rt for 3 h, subsequently, the solvent was removed under reduced pressure and taken up in 5 mL of dry THF. In a separate flask, 1 equiv. of 5-amino uracil derivative 3 dissolved in 3 mL of dry THF was added to a suspension of 1.2 equiv. of NaH in 5 mL of dry THF at 0 °C. The solution was allowed to stir at 0 °C for 30 min and the preformed acid chloride 2 was added drop wise. The resulting solution was allowed to warm to rt and stirred for 2 h. The solution was quenched with water and extracted with DCM. The combined organic layer was washed with brine, dried over Na$_2$SO$_4$ and concentrated under reduced pressure. The crude residue was subjected to flash silica gel chromatography to give the corresponding amide 4.
(b) General procedure for the preparation of uracil annulated β-carbolinones (5):

In a flame dried round bottomed flask equipped with a magnetic bar, a mixture of 1 equiv. starting material (4), 5 mL dry DMF, 2 equiv. of AgOAc and 10 mol % Pd(OAc)$_2$ was taken and stirred at room temperature for 5 min. Then the reaction mixture was heated in an oil bath fixed at 90 °C for 8 h under air. Completion of the reaction was monitored by checking TLC. The reaction mixture was cooled to room temperature, diluted with water and 50 mL of EtOAc and passed through a celite pad. The ethyl acetate part was washed with H$_2$O (2 × 10 mL) and saturated NaCl (aq) (1 × 10 mL). The organic part was dried over Na$_2$SO$_4$, evaporated and purified by flash chromatography, using ethyl acetate/petroleum ether (2:8) as the eluent to afford product 5.
(3) Characterization data

(a) Characterization data of uracil annulated β-carbolinones (5):

1,3,5,7-tetramethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5a):

Colourless solid; mp: 190-192 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): δ 7.98 (d, $J = 8$ Hz, 1H), 7.61 (d, $J = 3.6$ Hz, 2H), 7.39 - 7.43 (m, 1H), 4.46 (s, 3H), 4.12 (s, 3H), 3.81 (s, 3H), 3.48 (s, 3H).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): δ 157.5, 155.0, 151.7, 139.8, 130.5, 129.4, 126.9, 123.7, 121.9, 120.4, 116.1, 113.6, 111.1, 46.7, 40.1, 37.2, 33.7.

HRMS calcd. for C$_{17}$H$_{16}$N$_4$O$_3$ [M + H]$^+$: 325.1301; found: 325.1298

1,3-Diethyl-5,7-dimethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5b):

White solid; mp: 242-244 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): δ 7.94 (d, $J = 8$ Hz, 1H), 7.60 (d, $J = 3.6$ Hz, 2H), 7.39 - 7.42 (m, 1H), 4.55 (q, $J = 6.8$ Hz, 2H), 4.45 (s, 3H), 4.12 (s, 3H), 4.12 (q, $J = 6.8$ Hz, 2H), 1.32 (t, $J = 7.2$ Hz, 3H), 1.03 (t, $J = 6.8$ Hz, 3H).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): δ 157.5, 155.5, 151.6, 140.9, 131.1, 129.5, 127.0, 123.6, 122.0, 120.2, 116.4, 113.4, 111.0, 46.6, 37.2, 33.7, 31.8, 13.1, 13.0.

HRMS calcd. for C$_{19}$H$_{20}$N$_4$O$_3$ [M + H]$^+$: 353.1614; found: 353.1616

5-Ethyl-1,3,7-trimethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5c):
White solid; mp: 212-214 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): $\delta$ 7.97 (d, $J = 8$ Hz, 1H), 7.60 (d, $J = 3.6$ Hz, 2H), 7.38 - 7.42 (m, 1H), 4.83 (q, $J = 6.8$ Hz, 2H), 4.46 (s, 3H), 3.80 (s, 3H), 3.49 (s, 3H), 1.47 (t, $J = 7.2$ Hz, 3H).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): $\delta$ 157.3, 155.0, 152.3, 140.9, 131.5, 130.8, 126.9, 123.8, 121.9, 120.3, 114.3, 112.9, 111.0, 41.4, 40.2, 31.9, 28.8, 15.2.

HRMS calcd. for C$_{18}$H$_{18}$N$_4$O$_3$ [M + H]$^+$: 339.1457; found: 339.1458

3,5-Diethyl-1,7-dimethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5d):

Colourless solid; mp: 182-184 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): $\delta$ 7.93 (d, $J = 8.4$ Hz, 1H), 7.59 (d, $J = 4$ Hz, 2H), 7.38 - 7.42 (m, 1H), 4.83 (q, $J = 6.8$ Hz, 2H), 4.55 (q, $J = 7.2$ Hz, 2H), 4.45 (s, 3H), 3.47 (s, 3H), 1.47 (t, $J = 6.8$ Hz, 3H), 1.02 (t, $J = 6.8$ Hz, 3H).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): $\delta$ 157.5, 155.2, 151.9, 140.9, 131.2, 129.5, 127.0, 123.5, 122.0, 120.2, 115.7, 113.4, 111.1, 46.5, 41.4, 31.9, 28.7, 15.2, 13.2.

HRMS calcd. for C$_{19}$H$_{20}$N$_4$O$_3$ [M + H]$^+$: 353.1614; found: 353.1618

3-Ethyl-1,5,7-trimethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5e):

Colourless solid; mp: 186-188 °C
$^1$H-NMR (400 MHz, CDCl$_3$, ppm): $\delta$ 7.93 (d, $J = 8.4$ Hz, 1H), 7.60 (d, $J = 4$ Hz, 2H), 7.38 - 7.42 (m, 1H), 4.55 (q, $J = 6.8$ Hz, 2H), 4.45 (s, 3H), 4.13 (s, 3H), 3.46 (s, 3H), 1.03 (t, $J = 6.8$ Hz, 3H).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): $\delta$ 158.0, 155.5, 152.0, 140.9, 131.1, 129.4, 127.0, 123.5, 122.0, 120.2, 116.2, 113.4, 111.1, 46.9, 33.6, 31.9, 28.6, 13.1.

HRMS calcd. for C$_{18}$H$_{18}$N$_4$O$_3$ [M + H]$^+$: 339.1457; found: 339.1457

7-Ethyl-1,3,5-trimethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5f):

White solid; mp: 188-190 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): $\delta$ 7.98 (d, $J = 8.4$ Hz, 1H), 7.57 - 7.64 (m, 2H), 7.38 - 7.42 (m, 1H), 5.03 (q, $J = 7.2$ Hz, 2H), 4.12 (s, 3H), 3.81 (s, 3H), 3.49 (s, 3H), 1.52 (t, $J = 7.2$ Hz, 3H).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): $\delta$ 158.2, 155.1, 152.5, 139.9, 130.8, 126.8, 123.9, 121.8, 120.4, 114.9, 113.0, 111.1, 40.1, 33.9, 28.7, 15.9.

HRMS calcd. for C$_{18}$H$_{18}$N$_4$O$_3$ [M + H]$^+$: 339.1457; found: 339.1460

5,7-Diethyl-1,3-dimethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5g):

White solid; mp: 165-167 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): $\delta$ 7.97 (d, $J = 8$ Hz, 1H), 7.56 - 7.64 (m, 2H), 7.37 - 7.41 (m, 1H), 5.02 (q, $J = 7.2$ Hz, 2H), 4.83 (q, $J = 6.8$ Hz, 2H), 3.80 (s, 3H), 3.49 (s, 3H), 1.52 (t, $J = 7.2$ Hz, 3H), 1.48 (t, $J = 6.8$ Hz, 3H).
$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): $\delta$ 157.3, 154.6, 152.4, 139.8, 130.9, 130.7, 126.8, 123.9, 121.8, 120.4, 114.3, 113.0, 111.1, 41.5, 40.2, 40.1, 28.8, 16.0, 15.2.

HRMS calcd. for C$_{19}$H$_{20}$N$_4$O$_3$ [M + H]$^+$/found: 353.1614

1,3,7-Triethyl-5-methyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5h):

White solid; mp: 172-174 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): $\delta$ 7.94 (d, $J = 8$ Hz, 1H), 7.56 - 7.63 (m, 2H), 7.37 - 7.41 (m, 1H), 5.01 (q, $J = 7.2$ Hz, 2H), 4.54 (q, $J = 6.8$ Hz, 2H), 4.13 (s, 3H), 4.12 (q, $J = 6.8$ Hz, 2H), 1.52 (t, $J = 7.2$ Hz, 3H), 1.31 (t, $J = 7.2$ Hz, 3H), 1.05 (t, $J = 6.8$ Hz, 3H).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): $\delta$ 157.5, 155.0, 151.7, 139.8, 130.5, 129.4, 126.9, 123.7, 121.9, 120.4, 116.4, 113.6, 111.1, 46.7, 40.7, 37.2, 33.7, 16.0, 13.1, 13.0.

HRMS calcd. for C$_{20}$H$_{22}$N$_4$O$_3$ [M + H]$^+$/found: 367.1768

7-Benzyl-1,3,5-triethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5i):

White solid; mp: 138-140 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): $\delta$ 7.88 (d, $J = 8$ Hz, 1H), 7.50 (d, $J = 8.4$ Hz, 1H), 7.44 (t, $J = 8.4$ Hz, 1H), 7.30 (t, $J = 7.2$ Hz, 1H), 7.15 - 7.24 (m, 5H), 6.23 (s, 2H), 4.76 (q, $J = 6.8$ Hz, 2H), 4.47 (q, $J = 6.8$ Hz, 2H), 4.07 (q, $J = 7.2$ Hz, 2H), 1.39 (t, $J = 6.8$ Hz, 3H), 1.25 (t, $J = 7.2$ Hz, 3H), 0.98 (t, $J = 6.8$ Hz, 3H).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): $\delta$ 157.0, 154.9, 151.6, 140.4, 137.4, 130.8, 129.4, 128.7, 127.6, 127.1, 127.0, 123.6, 122.1, 120.5, 116.2, 114.0, 111.9, 48.2, 46.8, 41.6, 37.3, 15.2, 13.1, 13.0.
HRMS calcd. for C_{28}H_{26}N_{4}O_{3} [M + H]^+: 443.2083; found: 443.2088

7-Benzyl-1,3-diethyl-5-methyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5j):

White solid; mp: 156-158 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): $\delta$ 7.98 (d, $J = 8.4$ Hz, 1H), 7.60 (d, $J = 8$ Hz, 1H), 7.54 (t, $J = 8$ Hz, 1H), 7.40 (t, $J = 7.6$ Hz, 1H), 7.24 - 7.42 (m, 5H), 6.27 (s, 2H), 4.57 (q, $J = 7.2$ Hz, 2H), 4.15 (s, 3H), 4.10 - 4.20 (m, 2H), 1.34 (t, $J = 7.2$ Hz, 3H), 1.08 (t, $J = 6.8$ Hz, 3H).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): $\delta$ 157.5, 155.2, 151.6, 140.4, 137.3, 130.6, 129.3, 128.8, 127.6, 127.2, 126.9, 123.7, 122.2, 120.5, 116.7, 114.0, 111.9, 48.2, 46.7, 37.2, 33.8, 13.1, 13.0.

HRMS calcd. for C_{25}H_{24}N_{4}O_{3} [M + H]^+: 429.1927; found: 429.1930

7-Butyl-1,3-diethyl-5-methyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5k):

White solid; mp: 115-117 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): $\delta$ 7.87 (d, $J = 8.4$ Hz, 1H), 7.48-7.55 (m, 2H), 7.31 - 7.33 (m, 1H), 4.86 (t, $J = 7.6$ Hz, 2H), 4.47 (q, $J = 6.8$ Hz, 2H), 4.05 (q, $J = 6.8$ Hz, 2H), 4.05 (s, 3H), 1.83 - 1.84 (m, 2H), 1.36 - 1.42 (m, 2H), 1.24 (t, $J = 7.2$ Hz, 3H), 0.97 (t, $J = 6.8$ Hz, 3H), 0.91 (t, $J = 7.2$ Hz, 3H).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): $\delta$ 157.5, 155.1, 151.7, 140.1, 130.7, 129.4, 126.8, 123.6, 121.8, 120.2, 116.4, 113.5, 111.3, 46.7, 44.9, 37.1, 33.7, 33.0, 20.2, 13.9, 13.1, 13.0.

HRMS calcd. for C_{22}H_{26}N_{4}O_{3} [M + H]^+: 395.2083; found: 395.2083
7-Butyl-1,3,5-triethyl-5,7-dihydro-1'H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5I):

White solid; mp: 162-164 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): δ 7.86 (d, $J$ = 8.4 Hz, 1H), 7.47 – 7.54 (m, 2H), 7.28 - 7.32 (m, 1H), 4.86 (t, $J$ = 7.6 Hz, 2H), 4.75 (q, $J$ = 6.8 Hz, 2H), 4.46 (q, $J$ = 6.8 Hz, 2H), 4.06 (q, $J$ = 7.2 Hz, 2H), 1.78 – 1.86 (m, 2H), 1.41 (t, $J$ = 6.8 Hz, 3H), 1.34-1.43 (m, 2H), 1.24 (t, $J$ = 7.2 Hz, 3H), 0.97 (t, $J$ = 6.8 Hz, 3H), 0.91 (t, $J$ = 7.2 Hz, 3H).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): δ 157.0, 154.7, 151.6, 140.1, 130.9, 129.5, 126.8, 121.8, 120.3, 115.9, 113.5, 111.3, 46.7, 44.9, 41.5, 37.2, 33.0, 20.2, 15.2, 13.9, 13.1, 13.0.

HRMS calcd. for C$_{23}$H$_{28}$N$_4$O$_3$ [M + H]$^+$: 409.2240; found: 409.2238

(b) Experimental characterization data of starting material (4):

$N$-(1,3-Dimethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-$N$-1-dimethyl-1'H-indole-2-carboxamide (4a):

Colourless gummy gel

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): δ 7.55 (d, $J$ = 7.6 Hz, 1H); 7.26-7.35 (m, 3H, indole C3-H and two ArH); 7.10 (t, $J$ = 7.2 Hz, 1H); 6.46 (s, 1H, uracil C6-H); 3.89 (s, 3H); 3.42 (s, 3H), 3.37 (s, 3H); 3.32 (s, 3H).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): δ 165.4, 160.7, 150.9, 141.1, 138.1, 131.9, 126.0, 123.7, 121.8, 120.2, 110.0, 105.3, 37.8, 37.3, 31.3, 28.5.

HRMS calcd. for C$_{17}$H$_{18}$N$_4$O$_3$ [M + H]$^+$: 327.1457; found: 327.1455
**N-(1,3-Diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-N,1-dimethyl-1H-indole-2-carboxamide (4b):**

White solid; mp: 135-137 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): δ 7.53 (d, $J = 7.2$ Hz, 1H); 7.33 (d, $J = 8.4$ Hz, 1H); 7.25-7.29 (m, 2H, indole C3-H and one ArH); 7.10 (t, $J = 7.2$ Hz, 1H), 6.43 (s, 1H, uracil C6-H); 3.91-3.92 (m, 2H, NCH$_2$CH$_3$), 3.88 (s, 3H); 3.71-3.77 (m, 2H, NCH$_2$CH$_3$); 3.35 (s, 3H); 1.23-1.31 (m, 3H, NCH$_2$CH$_3$); 1.08 (s, 3H, NCH$_2$CH$_3$).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): δ 165.4, 160.2, 149.9, 139.7, 137.9, 132.1, 126.1, 123.6, 121.6, 120.2, 109.9, 105.0, 45.1, 37.3, 37.1, 31.3, 14.3, 12.6.

HRMS calcd. for C$_{19}$H$_{22}$N$_4$O$_3$ [M + H]$^+$: 355.1770; found: 355.1771

**N-(1,3-Dimethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-N-ethyl-1-methyl-1H-indole-2-carboxamide (4c):**

White solid; mp: 154-156 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): δ 7.55 (d, $J = 7.6$ Hz, 1H); 7.34 (d, $J = 8.0$ Hz, 1H); 7.27-7.29 (m, 2H, one ArH and one indole C3-H), 7.10 (t, $J = 7.2$ Hz, 1H); 6.27 (s, 1H, uracil C6-H); 3.89 (s, 3H); 3.79 (brs, 2H, NCH$_2$CH$_3$); 3.40 (s, 3H); 3.27 (s, 3H); 1.23 (t, $J = 6.8$ Hz, 3H, NCH$_2$CH$_3$).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): δ 165.0, 161.0, 150.9, 141.1, 138.0, 132.5, 126.1, 123.5, 121.7, 120.1, 110.0, 104.7, 44.7, 37.4, 31.3, 28.5, 13.1.

HRMS calcd. for C$_{18}$H$_{20}$N$_4$O$_3$ [M + H]$^+$: 341.1614; found: 341.1619

**N-Ethyl-N-(3-ethyl-1-methyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-1-methyl-1H-indole-2-carboxamide (4d):**
Colourless gummy gel

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): δ 7.53 (d, $J = 7.6$ Hz, 1H); 7.33 (d, $J = 8$ Hz, 1H); 7.25-7.28 (m, 2H, one ArH and one indole C3-H); 7.09 (t, $J = 7.2$ Hz, 1H); 6.40 (s, 1H, uracil C6-H); 3.86-3.91 (m, 4H, one N-Me and 1H of NCH$_2$CH$_3$); 3.70-3.80 (m, 3H, 1H of NCH$_2$CH$_3$ and 2H of NCH$_2$CH$_3$); 3.26 (s, 3H, N-Me); 1.23 (t, $J = 6.8$ Hz, 6H, two NCH$_2$CH$_3$).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): δ 165.0, 160.9, 150.4, 140.3, 138.0, 132.5, 126.1, 123.5, 121.6, 120.1, 109.9, 104.8, 45.2, 44.4, 31.3, 28.5, 14.4, 13.2.

HRMS calcd. for C$_{19}$H$_{22}$N$_4$O$_3$ [M + H]$^+$: 355.1770; found: 355.1767

$N$-(3-Ethyl-1-methyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-$N$1,1-dimethyl-$H$-indole-2-carboxamide (4e):

Colourless gummy gel

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): δ 7.53 (d, $J = 8$ Hz, 1H); 7.33 (d, $J = 8$ Hz, 1H); 7.25-7.29 (m, 2H, one ArH and one indole C3-H); 7.10 (t, $J = 7.6$ Hz, 1H); 6.44 (s, 1H, uracil C6-H); 3.77 (s, 3H, N-Me); 3.77 (q, $J = 7.2$ Hz, 2H, NCH$_2$CH$_3$); 3.33 (s, 3H, N-Me); 3.27 (s, 3H, N-Me); 1.26 (t, $J = 7.2$ Hz, 3H, NCH$_2$CH$_3$).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): δ 165.5, 160.6, 150.4, 140.1, 138.0, 132.0, 126.1, 123.6, 121.7, 120.2, 110.0, 105.3, 45.2, 37.5, 31.3, 28.4, 14.3.

HRMS calcd. for C$_{18}$H$_{20}$N$_4$O$_3$ [M + H]$^+$: 341.1614; found: 341.1617

$N$-(1,3-Dimethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-1-ethyl-$N$-methyl-$H$-indole-2-carboxamide (4f):

Colourless gummy gel
$^1$H-NMR (400 MHz, CDCl$_3$, ppm): δ 7.57 (d, $J = 7.6$ Hz, 1H); 7.37 (d, $J = 8.4$ Hz, 1H); 7.28 (d, $J = 7.6$ Hz, 1H); 7.26 (s, 1H, indole C3-H); 7.11 (t, $J = 7.6$ Hz, 1H); 6.48 (s, 1H, uracil C6-H); 4.40 (q, $J = 7.2$ Hz, 2H, NCH$_2$CH$_3$) 3.39 (s, 3H, N-Me); 3.33 (s, 3H, N-Me); 3.31 (s, 3H, N-Me); 1.46 (t, $J = 7.2$ Hz, 3H, NCH$_2$CH$_3$).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): δ 165.4, 160.6, 150.9, 141.2, 137.0, 131.0, 126.3, 123.6, 121.9, 120.1, 110.2, 105.5, 39.7, 38.1, 37.4, 28.5, 15.6.

HRMS calcd. for C$_{18}$H$_{20}$N$_4$O$_3$ [M + H]$^+$: 341.1614; found: 341.1611

$N$(1,3-Dimethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-N,1-diethyl-1H-indole-2-carboxamide (4g):

Colourless solid mp: 85-87 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): δ 7.56 (d, $J = 6.8$ Hz, 1H); 7.36 (d, $J = 8.0$ Hz, 1H); 7.22-7.28 (m, 2H, one ArH and one indole C3-H ); 7.09 (t, $J = 7.2$ Hz, 1H); 6.45 (s, 1H, uracil C6-H); 4.38 (d, $J = 6.8$ Hz, 2H, NCH$_2$CH$_3$) 3.70-3.82 (m, 2H, NCH$_2$CH$_3$); 3.38 (s, 3H, N-Me); 3.30 (s, 3H, N-Me); 1.44-1.48 (m, 3H, NCH$_2$CH$_3$); 1.20-1.1.26 (m, 3H, NCH$_2$CH$_3$).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): δ 165.4, 160.5, 150.4, 140.1, 136.9, 131.1, 126.3, 123.6, 121.8, 120.1, 110.2, 105.5, 45.2, 39.7, 37.2, 28.4, 15.6, 14.3.

HRMS calcd. for C$_{19}$H$_{22}$N$_4$O$_3$ [M + H]$^+$: 355.1770; found: 355.1773
**N-(1,3-Diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-1-ethyl-N-methyl-1H-indole-2-carboxamide (4h):**

White solid; mp:126-128 °C

\[^1\text{H}-\text{NMR}\ (400 \text{ MHz, CDCl}_3, \text{ ppm}): \delta 7.53 (d, J = 7.2 \text{ Hz, } 1\text{H}); 7.36 (d, J = 8.0 \text{ Hz, } 1\text{H}); 7.22-7.28 (m, 2\text{H, one ArH and one indole C3-H}); 7.09 (t, J = 7.2 \text{ Hz, } 1\text{H}); 6.44 (s, 1\text{H, uracil C6-H}); 4.39 (q, J = 6.8 \text{ Hz, } 2\text{H, NCH}_2\text{CH}_3); 3.93 (d, J = 6.4 \text{ Hz, } 2\text{H, NCH}_2\text{CH}_3); 3.75 (d, J = 6.8 \text{ Hz, } 2\text{H, NCH}_2\text{CH}_3); 3.34 (s, 3\text{H, N-Me}); 1.45 (t, J = 7.2 \text{ Hz, } 3\text{H, NCH}_2\text{CH}_3); 1.22 (s, 3\text{H, NCH}_2\text{CH}_3); 1.12 (s, 3\text{H, NCH}_2\text{CH}_3).**

\[^{13}\text{C}-\text{NMR}\ (100 \text{ MHz, CDCl}_3, \text{ ppm}): \delta 165.4, 160.2, 150.0, 139.9, 136.8, 131.2, 126.4, 123.5, 121.8, 120.1, 110.1, 105.2, 45.1, 39.6, 37.3, 37.1, 15.6, 14.3, 12.7.**

HRMS calcd. for C\text{\textsubscript{20}H\textsubscript{24}N\textsubscript{4}O\textsubscript{3}} [M + H]^+: 369.1927; found: 369. 1923

**1-Benzyl-N-(1,3-diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-N-ethyl-1H-indole-2-carboxamide (4i):**

White solid; mp:132-134 °C

\[^1\text{H}-\text{NMR}\ (400 \text{ MHz, CDCl}_3, \text{ ppm}): \delta 7.51 (d, J = 8.4 \text{ Hz, } 2\text{H}); 7.22-7.34 (m, 7\text{H, six ArH and one indole C3-H}); 7.11 (t, J = 7.6 \text{ Hz, } 1\text{H}); 6.40 (s, 1\text{H, uracil C6-H}); 5.43 (s, 2\text{H}); 4.18 (s, 1\text{H, NCH}_2\text{CH}_3); 3.99 (q, J = 7.2 \text{ Hz, } 2\text{H, NCH}_2\text{CH}_3); 3.42 (s, 1\text{H, NCH}_2\text{CH}_3); 3.27 (s, 1\text{H, NCH}_2\text{CH}_3); 3.12 (s, 1\text{H, NCH}_2\text{CH}_3); 1.99 (t, J = 7.2 \text{ Hz, } 3\text{H, NCH}_2\text{CH}_3); 1.12 (t, J = 7.2 \text{ Hz, } 3\text{H, NCH}_2\text{CH}_3); 0.88 (s, 3\text{H, NCH}_2\text{CH}_3).**

\[^{13}\text{C}-\text{NMR}\ (100 \text{ MHz, CDCl}_3, \text{ ppm}): \delta 163.9, 160.0, 149.3, 140.2, 138.9, 137.1, 130.9, 128.2, 127.9, 127.2, 125.9, 123.3, 121.4, 119.8, 116.8, 109.3, 104.2, 46.4, 44.2, 41.8, 36.5, 13.7, 12.2, 11.9.**
HRMS calcd. for C_{26}H_{29}N_{4}O_{3} [M + H]^+: 444.2161; found: 444.2159

1-Benzyl-\textit{N}(1,3-diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-\textit{N}-methyl-1\textit{H}-indole-2-carboxamide (4j):

White solid; mp: 116-118 °C

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): $\delta$ 7.54 (d, $J = 7.6$ Hz, 1H); 7.48 (d, $J = 8.4$ Hz, 1H); 7.26-7.35 (m, 7H, six ArH and one indole C3-H); 7.13 (t, $J = 7.6$ Hz, 1H); 6.43 (s, 1H, uracil C6-H); 5.70 (s, 2H); 4.38 (s, 1H, NCH$_2$CH$_3$); 4.02 (q, $J = 6.8$ Hz, 2H, NCH$_2$CH$_3$); 3.44 (s, 1H, NCH$_2$CH$_3$); 3.26 (s, 3H, NCH$_3$); 1.22 (t, $J = 6.8$ Hz, 3H, NCH$_2$CH$_3$); 0.93 (s, 3H, NCH$_2$CH$_3$).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): $\delta$ 164.8, 160.3, 149.8, 140.1, 139.1, 137.6, 131.2, 128.7, 127.9, 127.7, 126.4, 123.9, 121.9, 120.4, 119.4, 110.0, 105.1, 47.1, 44.8, 36.9, 36.2, 14.2, 12.8.

HRMS calcd. for C_{26}H_{29}N_{4}O_{3} [M + H]^+: 431.2083; found: 431.2082

1-Butyl-\textit{N}(1,3-diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-\textit{N}-methyl-1\textit{H}-indole-2-carboxamide (4k):

Colourless gummy liquid

$^1$H-NMR (400 MHz, CDCl$_3$, ppm): $\delta$ 7.55 (d, $J = 7.2$ Hz, 1H); 7.37 (d, $J = 8.0$ Hz, 1H); 7.26 (d, $J = 8.0$ Hz, 1H); 7.20 (s, 1H, C3-H of indole); 7.11 (t, $J = 7.2$ Hz, 1H); 6.49 (s, 1H, C6-H of uracil); 4.35 (s, 2H, NCH$_2$CH$_3$); 3.98 (d, $J = 6.8$ Hz, 2H, may NCH$_2$CH$_2$ or NCH$_2$CH$_3$); 3.75 (d, $J = 6.8$ Hz, 2H, may NCH$_2$CH$_2$ or NCH$_2$CH$_3$); 3.36 (s, 3H, NCH$_3$); 1.85 (quintet, $J = 7.6$ Hz, 2H, NCH$_2$CH$_2$CH$_2$); 1.29-1.45 (m, 2H, -CH$_2$CH$_2$CH$_3$); 1.22 (s, 3H, NCH$_2$CH$_3$); 1.64 (s, 3H, NCH$_2$CH$_3$); 0.98 (t, $J = 7.6$ Hz, 3H, -CH$_2$CH$_3$).

$^{13}$C-NMR (100 MHz, CDCl$_3$, ppm): $\delta$ 165.3, 160.2, 150.0, 139.9, 137.3, 131.2, 126.3, 123.5, 121.8, 120.1, 110.3, 105.3, 45.1, 44.5, 37.2, 37.1, 32.5, 20.3, 14.3, 13.9, 12.7.
HRMS calcd. for C$_{22}$H$_{28}$N$_{4}$O$_{3}$ [M + H]$^+$: 397.2240; found: 397.2243

1-Butyl-\(N\)-(1,3-diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-\(N\)-ethyl-1\(H\)-indole-2-carboxamide (4I):

Colourless solid; mp: 82-84 °C

\(^1\)H-NMR (400 MHz, CDCl$_3$, ppm): δ 7.45 (d, \(J = 7.2\) Hz, 1H); 7.26 (t, \(J = 8.0\) Hz, 1H); 7.14-7.19 (m, 1H); 7.01 (t, \(J = 7.2\) Hz, 2H); 6.37 (s, 1H, C6-H of uracil); 4.25 (s, 2H, may NCH$_2$CH$_2$ or NCH$_2$CH$_3$); 3.89 (d, \(J = 6.4\) Hz, 3H, may NCH$_2$CH$_2$ or NCH$_2$CH$_3$); 3.67-3.72 (m, 3H, may NCH$_2$CH$_2$ or NCH$_2$CH$_3$); 1.76 (quintet, \(J = 7.6\) Hz, 2H, NCH$_2$CH$_2$CH$_2$); 1.21-1.28 (m, 2H, -CH$_2$CH$_2$CH$_3$); 1.16 (t, \(J = 7.2\) Hz, 6H, NCH$_2$CH$_3$); 1.01 (s, 3H, NCH$_2$CH$_3$); 0.89 (t, \(J = 7.6\) Hz, 3H, -CH$_2$CH$_3$).

\(^{13}\)C-NMR (100 MHz, CDCl$_3$, ppm): δ 164.9, 160.5, 150.0, 140.2, 137.2, 131.7, 126.3, 123.3, 121.7, 120.0, 110.2, 104.7, 45.0, 44.5, 44.0, 37.1, 32.5, 20.3, 14.3, 13.9, 13.2, 12.6.

HRMS calcd. for C$_{23}$H$_{30}$N$_{4}$O$_{3}$ [M + H]$^+$: 411.2396; found: 411.2391

\(N\)-(1,3-Diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-\(N\)-methyl-1\(H\)-indole-2-carboxamide (4m):

White solid; mp: 210-212 °C

\(^1\)H-NMR (400 MHz, CDCl$_3$, ppm): δ 9.52 (s, 1H, NH); 7.47 (d, \(J = 7.2\) Hz, 1H); 7.34 (d, \(J = 8.4\) Hz, 1H); 7.28 (s, 1H, C3-H of indole); 7.19 (t, \(J = 8.4\) Hz, 1H); 7.02 (t, \(J = 7.2\) Hz, 1H); 6.13 (s, 1H, C6-H of uracil); 4.00 (q, \(J = 6.8\) Hz, 2H, NCH$_2$CH$_3$); 3.77(q, \(J = 7.2\) Hz, 2H, NCH$_2$CH$_3$); 3.32 (s, 3H, NCH$_3$); 1.24 (t, \(J = 7.2\) Hz, 3H, NCH$_2$CH$_3$); 1.18 (t, \(J = 6.8\) Hz, 3H, NCH$_2$CH$_3$).

\(^{13}\)C-NMR (100 MHz, CDCl$_3$, ppm): δ 163.4, 160.2, 150.3, 141.2, 135.6, 129.1, 127.6, 124.7, 122.1, 120.5, 118.6, 111.9, 105.9, 45.2, 37.8, 37.2, 14.5, 12.8.
HRMS calcd. for C_{18}H_{20}N_{4}O_{3} [M + H]^+: 341.1614; found: 341.1618

(4) References:

(5) NMR (\(^1\)H and \(^{13}\)C) spectra of products (5):

1,3,5,7-Tetramethyl-5,7-dihydro-1\(^H\)-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3\(^H\))-trione (5a):
1,3-Diethyl-5,7-dimethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5b):
5-Ethyl-1,3,7-trimethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5c):
3,5-Diethyl-1,7-dimethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5d):
3-Ethyl-1,5,7-trimethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5e):
7-Ethyl-1,3,5-trimethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5f):
5,7-Diethyl-1,3-dimethyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5g):
1,3,7-Triethyl-5-methyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5h):
7-Benzyl-1,3,5-triethyl-5,7-dihydro-1\textit{H}-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3\textit{H})-trione (5i):
7-Benzy1-1,3-diethyl-5-methyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5j):
7-Butyl-1,3-diethyl-5-methyl-5,7-dihydro-1H-pyrimido[4',5':5,6]pyrido[3,4-b]indole-2,4,6(3H)-trione (5k):
7-Butyl-1,3,5-triethyl-5,7-dihydro-1\textit{H}-pyrimido[4',5':5,6]pyrido[3,4-\textit{b}]indole-2,4,6(3\textit{H})-trione (5l):
NMR (\(^1\)H and \(^{13}\)C) spectra of starting materials (4):

\(N\)-(1,3-Dimethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-\(N\),\(N\)-dimethyl-1\(H\)-indole-2-carboxamide (4a):
* equivalent carbon
$N$-(1,3-Diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-$N,1$-dimethyl-$1H$-indole-2-carboxamide (4b):
$N$-(1,3-Dimethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-$N$-ethyl-1-methyl-$1H$-indole-2-carboxamide (4c):
DEPT spectra of 4c:
$N$-Ethyl-$N$-(3-ethyl-1-methyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-1-methyl-1$H$-indole-2-carboxamide (4d):
$N$-(3-Ethyl-1-methyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-$N$,1-dimethyl-$1H$-indole-2-carboxamide (4e):
N-(1,3-Dimethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-1-ethyl-N-methyl-1H-indole-2-carboxamide (4f):
$N$-(1,3-Dimethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-$N,1$-diethyl-$1H$-indole-2-carboxamide (4g):
$N$-(1,3-Diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-1-ethyl-$N$-methyl-1$H$-indole-2-carboxamide (4h):
1-Benzyl-\(N\)(1,3-diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-\(N\)-ethyl-1\(H\)-indole-2-carboxamide (4i):
1-Benzyl-N-(1,3-diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-N-methyl-1H-indole-2-carboxamide (4j):
1-Butyl-N-(1,3-diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-N-methyl-1H-indole-2-carboxamide (4k):
1-Butyl-N-(1,3-diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-N-ethyl-1H-indole-2-carboxamide (4l):
$N$-(1,3-Diethyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidin-5-yl)-$N$-methyl-1$H$-indole-2-carboxamide (4m):