

SUPPLEMENTARY MATERIAL TO
**Scalable methodologies for the synthesis of novel
unsymmetrically substituted secondary amines**

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ANALYTICAL AND SPECTRAL DATA OF THE SYNTHESIZED COMPOUNDS

4-Methoxy-N-(naphthalen-1-ylmethylene)benzylamine (1a). Yield: 6.48 g, 47 %; m.p. 44–45 °C (ethanol); Anal. Calcd. for C₁₉H₁₇NO: C, 82.88; H 6.22; N, 5.09 %. Found: C, 83.07, H, 6.11, N, 5.01 %; ¹H-NMR (400 MHz, CDCl₃, δ / ppm): 3.82 (3H, s), 4.91 (2H, s), 6.93 (2H, d, J = 8.4 Hz), 7.35 (2H, d, J = 8.4 Hz), 7.48–7.65 (3H, m), 7.85–8.02 (3H, m), 8.95 (1H, d, J = 8.4 Hz), 9.04 (1H, s); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 55.4, 65.6, 114.1, 124.5, 125.4, 126.1, 127.3, 128.7, 129.1, 129.3, 131.2, 131.5, 131.7, 134.0, 158.8, 161.4; HRMS (ESI) Calcd. for C₁₉H₁₈NO: 276.1388 [M+H]⁺. Found: 276.1382.

2-Chloro-N-(naphthalen-1-ylmethylene)benzylamine (1b).¹ Off-white solid. Yield: 9.65 g, 69 %; m.p. 60–61 °C (ethanol); Anal. Calcd. for C₁₈H₁₄ClN: C, 77.28; H 5.04; N, 5.01 %. Found: C, 77.20, H, 5.12, N, 4.94 %; ¹H-NMR (400 MHz, CDCl₃, δ / ppm): 5.05 (2H, s), 7.21–7.32 (2H, m), 7.43 (1H, dd, J = 0.8 and 8.0 Hz), 7.51–7.65 (4H, m), 7.88–8.00 (3H, m), 8.99 (1H, d, J = 8.4 Hz), 9.08 (1H, s); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 63.1, 124.5, 125.4, 126.2, 127.1, 127.4, 128.4, 128.8, 129.3, 129.4, 129.8, 131.4, 131.5, 131.6, 133.5, 134.0, 162.9; HRMS (ESI) Calcd. for C₁₈H₁₅ClN: 280.0893 [M+H]⁺. Found: 280.0885.

2-Methyl-N-(naphthalen-1-ylmethylene)benzylamine (1c). Off-white solid. Yield: 7.36 g, 57%; m.p. 52–53 °C (ethanol); Anal. Calcd. for C₁₉H₁₇N: C, 87.99; H 6.61; N, 5.40 %. Found: C, 87.88, H, 6.21, N, 5.31 %; ¹H-NMR (400 MHz, CDCl₃, δ / ppm): 2.47 (3H, s), 4.96 (2H, s), 7.20–7.26 (3H, m), 7.36–7.42 (1H, m), 7.50–7.63 (3H, m), 7.87–7.98 (3H, m), 8.98 (1H, d, J = 8.4 Hz), 9.03 (1H, s); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 19.5, 63.9, 124.5, 125.4, 126.2, 126.3, 127.2, 127.3, 128.6, 128.8, 129.2, 130.3, 131.2, 131.5, 131.8, 134.0,

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136.3, 137.8, 161.7; HRMS (ESI) Calcd. for $C_{19}H_{18}N$: 260.1439 $[M+H]^+$. Found: 260.1446.

2,4-Dichloro-N-(naphthalen-1-ylmethylene)benzylamine (1d). Yellowish solid. Yield: 12.21 g, 78 %; m.p. 97–98 °C (methanol); Anal. Calcd. for $C_{18}H_{13}Cl_2N$: C, 68.81; H 4.17; N, 4.46 %. Found: C, 68.68, H, 4.25, N, 4.37 %; 1H -NMR (400 MHz, $CDCl_3$, δ / ppm): 4.98 (2H, *s*), 7.27 (1H, *dd*, $J = 2.0$ and 8.4 Hz), 7.44 (1H, *d*, $J = 2.0$ Hz), 7.47 (1H, *d*, $J = 8.4$ Hz), 7.51–7.65 (3H, *m*), 7.89–7.99 (3H, *m*), 8.98 (1H, *d*, $J = 8.4$ Hz), 9.07 (1H, *s*); ^{13}C -NMR (100 MHz, $CDCl_3$, δ / ppm): 62.6, 124.5, 125.4, 126.3, 127.4, 127.5, 128.8, 129.2, 129.5, 130.6, 131.4, 131.5, 131.6, 133.4, 134.0, 134.1, 136.1, 163.2; HRMS (ESI) Calcd. for $C_{18}H_{14}Cl_2N$: 314.0503 $[M+H]^+$. Found: 314.0498.

1-(1,3-Benzodioxol-5-yl)-N-(naphthalen-1-ylmethylene)methylamine (1e). Off-white solid. Yield: 10.29 g, 71 %; m.p. 63–64 °C (ethanol); Anal. Calcd. for $C_{19}H_{15}NO_2$: C, 78.87; H 5.23; N, 4.84 %. Found: C, 78.98, H, 5.31, N, 4.77 %; 1H -NMR (400 MHz, $CDCl_3$, δ / ppm): 4.86 (2H, *s*), 5.96 (2H, *s*), 6.82 (1H, *d*, $J = 8.0$ Hz), 6.88 (1H, *dd*, $J = 0.8$ and 8.0 Hz), 6.93 (1H, *d*, $J = 0.8$ Hz), 7.48–7.63 (3H, *m*), 7.86–7.98 (3H, *m*), 8.95 (1H, *d*, $J = 8.4$ Hz), 9.03 (1H, *s*); ^{13}C -NMR (100 MHz, $CDCl_3$, δ / ppm): 65.9, 101.1, 108.4, 108.8, 121.2, 124.4, 125.4, 126.2, 127.3, 128.8, 129.2, 131.3, 131.5, 131.6, 133.5, 134.0, 146.7, 147.9, 161.6; HRMS (ESI) Calcd. for $C_{19}H_{16}NO_2$: 290.1181 $[M+H]^+$. Found: 290.1183.

1-(4-Methoxybenzyl)-N-(naphthalen-1-yl)methylamine (2a). Yellowish oil. Yield: 5.31 g, 96 %; R_f 0.24 (hexane–ethyl acetate 3:1 v/v); Anal. Calcd. for $C_{19}H_{19}NO$: C, 82.28; H 6.90; N, 5.05 %. Found: C, 82.54, H, 6.71, N, 4.89 %; 1H -NMR (400 MHz, $CDCl_3$, δ / ppm): 3.82 (3H, *s*), 3.87 (2H, *s*), 4.24 (2H, *s*), 6.90 (2H, *d*, $J = 8.8$ Hz), 7.32 (2H, *d*, $J = 8.4$ Hz), 7.40–7.57 (4H, *m*), 7.79 (1H, *d*, $J = 8.0$ Hz), 7.87 (1H, *dd*, $J = 2.0$ and 7.6 Hz), 8.09 (1H, *d*, $J = 8.4$ Hz); ^{13}C -NMR (100 MHz, $CDCl_3$, δ / ppm): 50.9, 53.3, 55.4, 113.9, 123.9, 125.5, 125.7, 126.1, 126.2, 127.9, 128.8, 129.5, 132.0, 132.6, 134.0, 136.1, 158.8; HRMS (ESI) Calcd. for $C_{19}H_{20}NO$: 278.1545 $[M+H]^+$. Found: 278.1553. *Hydrochloride*: colorless solid, m.p. 191–192 °C (ethanol); 1H -NMR (400 MHz, $DMSO-d_6$, δ / ppm): 3.78 (3H, *s*), 4.24 (2H, *s*), 4.56 (2H, *s*), 7.00 (2H, *d*, $J = 8.8$ Hz), 7.51–7.65 (5H, *m*), 7.79 (1H, *d*, $J = 7.2$ Hz), 7.95–8.10 (3H, *m*), 9.89 (2H, *br s*); ^{13}C -NMR (100 MHz, $DMSO-d_6$, δ / ppm): 45.8, 49.7, 55.2, 113.9, 123.6, 123.7, 125.3, 126.2, 126.7, 128.1, 128.6, 129.1, 129.5, 131.1, 132.0, 133.2, 159.7.

1-(2-Chlorobenzyl)-N-(naphthalen-1-yl)methylamine (2b). Yellowish oil. Yield: 5.48 g, 97 %; R_f 0.24 (hexane–ethyl acetate 9:1 v/v); Anal. Calcd. for $C_{18}H_{16}ClN$: C, 76.72; H 5.72; N, 4.97 %. Found: C, 76.49, H, 5.53, N, 5.15 %; 1H -NMR (400 MHz, $CDCl_3$, δ / ppm): 4.04 (2H, *s*), 4.27 (2H, *s*), 7.20–7.30 (2H, *m*), 7.37–7.56 (6H, *m*), 7.79 (1H, *d*, $J = 8.0$ Hz), 7.88 (1H, *dd*, $J = 1.6$ and 8.4

Hz), 8.12 (1H, *dd*, $J = 1.2$ and 8.0 Hz); ^{13}C -NMR (100 MHz, CDCl_3 , δ / ppm): 51.0, 51.4, 123.9, 125.5, 125.7, 126.2, 126.3, 126.9, 128.0, 128.5, 128.8, 129.7, 130.5, 132.0, 134.0, 135.8, 137.7; HRMS (ESI) Calcd. for $\text{C}_{18}\text{H}_{17}\text{ClN}$: 282.1050 $[\text{M}+\text{H}]^+$. Found: 282.1055. *Hydrochloride*: colorless solid, m.p. 173–174 °C (ethanol); ^1H -NMR (400 MHz, $\text{DMSO}-d_6$, δ / ppm): 4.41 (2H, *s*), 4.74 (2H, *s*), 7.39–7.48 (2H, *m*), 7.49–7.67 (4H, *m*), 7.83–7.90 (2H, *m*), 8.02 (2H, *d*, $J = 7.6$ Hz), 8.19 (1H, *d*, $J = 8.0$ Hz), 10.02 (2H, *br s*); ^{13}C -NMR (100 MHz, $\text{DMSO}-d_6$, δ / ppm): 46.8, 47.0, 123.6, 125.3, 126.2, 126.8, 127.4, 127.7, 128.6, 129.4, 129.5, 129.6, 129.9, 130.7, 131.2, 132.0, 133.2, 133.6.

1-(2-Methylbenzyl)-N-(naphthalen-1-yl)methylamine (2c). Yellowish oil. Yield: 5.10 g, 97 %; R_f 0.29 (hexane–ethyl acetate 9:1 v/v); Anal. Calcd. for $\text{C}_{19}\text{H}_{19}\text{N}$: C, 87.31; H 7.33; N, 5.36 %. Found: C, 87.54, H, 7.14, N, 5.52 %; ^1H -NMR (400 MHz, CDCl_3 , δ / ppm): 2.35 (3H, *s*), 3.92 (2H, *s*), 4.31 (2H, *s*), 7.17–7.23 (3H, *m*), 7.35–7.41 (1H, *m*), 7.42–7.56 (4H, *m*), 7.79 (1H, *d*, $J = 8.0$ Hz), 7.88 (1H, *dd*, $J = 2.4$ and 7.2 Hz), 8.13 (1H, *dd*, $J = 2.4$ and 7.2 Hz); ^{13}C -NMR (100 MHz, CDCl_3 , δ / ppm): 19.1, 51.5, 51.7, 124.0, 125.5, 125.7, 126.0, 126.1, 126.2, 127.2, 127.9, 128.7, 128.8, 130.4, 132.1, 134.1, 136.1, 136.7, 138.4; HRMS (ESI) Calcd. for $\text{C}_{19}\text{H}_{20}\text{N}$: 262.1596 $[\text{M}+\text{H}]^+$. Found: 262.1590. *Hydrochloride*: colorless solid, m.p. 191–192 °C (ethanol); ^1H -NMR (400 MHz, $\text{DMSO}-d_6$, δ / ppm): 2.30 (3H, *s*), 4.26 (2H, *s*), 4.71 (2H, *s*), 7.21–7.33 (3H, *m*), 7.54–7.67 (4H, *m*), 7.88 (1H, *d*, $J = 6.8$ Hz), 7.99–8.05 (2H, *m*), 8.15–8.20 (1H, *m*), 9.90 (2H, *br s*); ^{13}C -NMR (100 MHz, $\text{DMSO}-d_6$, δ / ppm): 19.0, 46.5, 47.2, 123.6, 125.2, 125.9, 126.1, 126.7, 127.9, 128.6, 128.8, 129.3, 129.5, 130.4, 130.6, 131.2, 133.2, 137.5.

1-(2,4-Dichlorobenzyl)-N-(naphthalen-1-yl)methylamine (2d). Yellow solid. Yield: 5.98 g, 95 %; m.p. 69–70 °C; R_f 0.32 (hexane–ethyl acetate 9:1 v/v); Anal. Calcd. for $\text{C}_{18}\text{H}_{15}\text{Cl}_2\text{N}$: C, 68.37; H 4.78; N, 4.43 %. Found: C, 68.57, H, 4.61, N, 4.69 %; ^1H -NMR (400 MHz, CDCl_3 , δ / ppm): 3.98 (2H, *s*), 4.25 (2H, *s*), 7.23 (1H, *dd*, $J = 2.0$ and 8.4 Hz), 7.38–7.55 (6H, *m*), 7.78 (1H, *d*, $J = 8.0$ Hz), 7.87 (1H, *dd*, $J = 2.0$ and 8.0 Hz), 8.10 (1H, *d*, $J = 8.0$ Hz); ^{13}C -NMR (100 MHz, CDCl_3 , δ / ppm): 50.7, 51.1, 123.8, 125.5, 125.8, 126.2, 126.3, 127.2, 128.1, 128.9, 129.4, 131.1, 132.0, 133.4, 134.0, 134.6, 135.6, 136.5; HRMS (ESI) Calcd. for $\text{C}_{18}\text{H}_{16}\text{Cl}_2\text{N}$: 316.0660 $[\text{M}+\text{H}]^+$. Found: 316.0669. *Hydrochloride*: colorless solid, m.p. 224–225 °C (ethanol); ^1H -NMR (400 MHz, $\text{DMSO}-d_6$, δ / ppm): 4.40 (2H, *s*), 4.74 (2H, *s*), 7.53 (1H, *dd*, $J = 2.4$ and 8.4 Hz), 7.55–7.67 (3H, *m*), 7.70 (1H, *d*, $J = 2.4$ Hz), 7.87 (1H, *d*, $J = 7.2$ Hz), 7.91 (1H, *d*, $J = 8.4$ Hz), 8.01 (2H, *d*, $J = 8.0$ Hz), 8.21 (1H, *d*, $J = 8.4$ Hz), 10.10 (2H, *br s*); ^{13}C -NMR (100 MHz, $\text{DMSO}-d_6$, δ / ppm): 46.5, 46.8, 123.6, 125.3, 126.2, 126.8, 127.5, 127.8, 128.6, 128.9, 129.1, 129.4, 129.6, 131.2, 133.2, 133.3, 134.4, 134.7.

1-(1,3-Benzodioxol-5-ylmethyl)-N-(naphthalen-1-yl)methylamine (2e). Yellow oil. Yield: 5.52 g, 95 %; R_f 0.35 (hexane–ethyl acetate 3:1 v/v); Anal. Calcd. for $C_{19}H_{17}NO_2$: C, 78.33; H 5.88; N, 4.81 %. Found: C, 78.10, H, 6.06, N, 5.08 %; 1H -NMR (400 MHz, $CDCl_3$, δ / ppm): 3.83 (2H, s), 4.23 (2H, s), 5.95 (2H, s), 6.76–6.86 (2H, m), 6.92 (1H, s), 7.39–7.56 (4H, m), 7.78 (1H, d, $J = 8.0$ Hz), 7.87 (1H, dd, $J = 1.6$ and 8.4 Hz), 8.11 (1H, d, $J = 8.4$ Hz); ^{13}C -NMR (100 MHz, $CDCl_3$, δ / ppm): 50.8, 53.6, 101.0, 108.2, 108.9, 121.4, 123.9, 125.5, 125.7, 126.2, 126.3, 127.9, 128.8, 132.0, 134.0, 134.5, 136.0, 146.7, 147.9; HRMS (ESI) Calcd. for $C_{19}H_{18}NO_2$: 292.1338 $[M+H]^+$. Found: 292.1331. *Hydrochloride*: colorless solid. m.p. 219–220 °C (ethanol); 1H -NMR (400 MHz, $DMSO-d_6$, δ / ppm): 4.23 (2H, s), 4.55 (2H, s), 6.06 (2H, s), 6.98 (1H, d, $J = 8.0$ Hz), 7.12 (1H, dd, $J = 1.6$ and 8.0 Hz), 7.31 (1H, d, $J = 1.6$ Hz), 7.52–7.65 (3H, m), 7.79 (1H, d, $J = 7.2$ Hz), 7.96–8.03 (2H, m), 8.05–8.12 (1H, m), 9.87 (2H, br s); ^{13}C -NMR (100 MHz, $DMSO-d_6$, δ / ppm): 45.9, 50.1, 101.3, 108.3, 110.7, 123.6, 124.5, 125.3, 126.2, 126.7, 128.1, 128.7, 129.1, 129.5, 131.1, 133.2, 147.3, 147.7.

N-(4-Bromobenzylidene)-2-(1H-indol-3-yl)ethanamine (3a). Colorless solid. Yield: 8.66 g, 88 %; m.p. 164–165 °C (ethyl acetate); Anal. Calcd. for $C_{17}H_{15}BrN_2$: C, 62.40; H 4.62; N, 8.56 %. Found: C, 62.30, H, 4.55, N, 8.64 %; 1H -NMR (400 MHz, $DMSO-d_6$, δ / ppm): 3.03 (2H, t, $J = 7.2$ Hz), 3.86 (2H, t, $J = 7.2$ Hz), 6.96 (1H, t, $J = 7.6$ Hz), 7.06 (1H, t, $J = 7.6$ Hz), 7.14 (1H, d, $J = 2.0$ Hz), 7.33 (1H, d, $J = 8.0$ Hz), 7.56 (1H, d, $J = 8.0$ Hz), 7.63 (2H, d, $J = 8.4$ Hz), 7.67 (2H, d, $J = 8.4$ Hz), 8.26 (1H, s), 10.79 (1H, s); ^{13}C -NMR (100 MHz, $DMSO-d_6$, δ / ppm): 26.6, 61.4, 111.3, 112.2, 118.2, 118.5, 120.8, 122.8, 123.9, 127.3, 129.7, 131.7, 135.4, 136.2, 159.7; HRMS (ESI) Calcd. for $C_{17}H_{16}BrN_2$: 327.0497 $[M+H]^+$. Found: 327.0504.

N-(3,4-Dimethoxybenzylidene)-2-(1H-indol-3-yl)ethanamine (3b). Cream-colored solid. Yield: 6.67 g, 72 %; m.p. 101–102 °C (ethyl acetate); Anal. Calcd. for $C_{19}H_{20}N_2O_2$: C, 74.00; H, 6.54; N, 9.08 %. Found: C, 73.88, H, 6.62, N, 9.01 %; 1H -NMR (400 MHz, $CDCl_3$, δ / ppm): 3.17 (2H, t, $J = 7.2$ Hz), 3.91 (3H, s), 3.92 (3H, s), 3.93 (2H, t, $J = 7.2$ Hz), 6.86 (1H, d, $J = 8.4$ Hz), 7.00 (1H, d, $J = 2.4$ Hz), 7.08–7.23 (3H, m), 7.35 (1H, d, $J = 8.0$ Hz), 7.44 (1H, d, $J = 1.6$ Hz), 7.68 (1H, d, $J = 8.0$ Hz), 8.10 (1H, s), 8.12 (1H, br s); ^{13}C -NMR (100 MHz, $CDCl_3$, δ / ppm): 27.2, 56.1, 62.1, 108.9, 110.6, 111.2, 114.3, 119.1, 119.3, 122.0, 122.2, 123.1, 127.7, 129.7, 136.4, 149.5, 151.4, 161.0; HRMS (ESI) Calcd. for $C_{19}H_{21}N_2O_2$: 309.1603 $[M+H]^+$. Found: 309.1595.

2-(1H-Indol-3-yl)-N-(naphthalen-1-ylmethylene)ethanamine (3c). Cream-colored solid. Yield: 6.28 g, 70 %; m.p. 134–135 °C (lit.² m.p. 134–135 °C) (ethyl acetate); Anal. Calcd. for $C_{21}H_{18}N_2$: C, 84.53; H, 6.08; N, 9.39 %. Found: C, 84.41, H, 5.99, N, 9.47 %; 1H -NMR (400 MHz, $CDCl_3$, δ / ppm): 3.28 (2H, t, $J = 7.0$ Hz), 4.08 (2H, t, $J = 7.0$ Hz), 7.01 (1H, d, $J = 2.0$ Hz), 7.13–7.25 (2H, m),

7.36 (1H, *d*, *J* = 8.0 Hz), 7.48–7.58 (3H, *m*), 7.74 (1H, *d*, *J* = 8.0 Hz), 7.85–7.93 (3H, *m*), 7.99 (1H, *br s*), 8.59–8.65 (1H, *m*), 8.77 (1H, *s*); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 27.0, 62.8, 111.2, 114.0, 119.1, 119.4, 122.0, 122.5, 124.3, 125.4, 126.1, 127.0, 127.6, 128.2, 128.7, 130.9, 131.4, 132.0, 133.9, 136.4, 161.0; HRMS (ESI) Calcd. for C₂₁H₁₉N₂: 299.1548 [M+H]⁺. Found: 299.1557.

2-(1*H*-Indol-3-yl)-N-(2-thienylmethylene)ethanamine (**3d**). Cream-colored solid. Yield: 5.20 g, 68 %; mp 127–128 °C (lit.³ 133–134 °C) (ethyl acetate); Anal. Calcd. for C₁₅H₁₄N₂S: C, 70.83; H, 5.55; N, 11.01 %. Found: C, 70.98, H, 5.43, N, 10.93 %; ¹H-NMR (400 MHz, CDCl₃, δ / ppm): 3.16 (2H, *t*, *J* = 7.2 Hz), 3.90 (2H, *t*, *J* = 7.2 Hz), 7.00 (1H, *d*, *J* = 2.0 Hz), 7.05 (1H, *dd*, *J* = 3.6 and 5.2 Hz), 7.10–7.25 (3H, *m*), 7.35 (1H, *d*, *J* = 8.0 Hz), 7.39 (1H, *d*, *J* = 5.2 Hz), 7.67 (1H, *d*, *J* = 7.6 Hz), 8.03 (1H, *br s*), 8.22 (1H, *s*); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 26.9, 61.8, 111.2, 114.0, 119.1, 119.3, 122.0, 122.4, 127.4, 127.6, 128.8, 130.4, 136.3, 142.6, 154.8; HRMS (ESI) Calcd. for C₁₅H₁₅N₂S: 255.0956 [M+H]⁺. Found: 255.0948.

N-(4-Bromobenzyl)-2-(1*H*-indol-3-yl)ethanamine (**4a**). Colorless solid. Yield: 6.33 g, 96%; m.p. 80–81 °C (lit.⁴ 78–80 °C); Anal. Calcd. for C₁₇H₁₇BrN₂: C, 62.02; H, 5.20; N, 8.51 %. Found: C, 61.78, H, 5.38, N, 8.70 %; ¹H-NMR (400 MHz, CDCl₃, δ / ppm): 2.94–3.05 (4H, *m*), 3.76 (2H, *s*), 7.01 (1H, *d*, *J* = 2.4 Hz), 7.10–7.24 (4H, *m*), 7.33–7.37 (1H, *m*), 7.41 (2H, *d*, *J* = 8.4 Hz), 7.62 (1H, *d*, *J* = 8.0 Hz), 8.09 (1H, *br s*); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 25.9, 49.5, 53.3, 111.3, 114.1, 119.0, 119.5, 120.7, 122.0, 122.2, 127.6, 129.9, 131.5, 136.6, 139.6; HRMS (ESI) Calcd. for C₁₇H₁₈BrN₂: 329.0653 [M+H]⁺. Found: 329.0664.

N-(3,4-Dimethoxybenzyl)-2-(1*H*-indol-3-yl)ethanamine (**4b**). Light yellow oil. Yield: 5.88 g, 95 %; *R*_f 0.22 (ethyl acetate–methanol 4:1 v/v); Anal. Calcd. for C₁₉H₂₂N₂O₂: C, 73.52; H, 7.14; N, 9.03 %. Found: C, 73.32, H, 7.02, N, 8.86 %; ¹H-NMR (400 MHz, CDCl₃, δ / ppm): 2.95–3.04 (4H, *m*), 3.76 (2H, *s*), 3.82 (3H, *s*), 3.86 (3H, *s*), 6.77–6.84 (3H, *m*), 7.03 (1H, *d*, *J* = 2.0 Hz), 7.08–7.14 (1H, *m*), 7.16–7.22 (1H, *m*), 7.36 (1H, *d*, *J* = 8.0 Hz), 7.62 (1H, *d*, *J* = 8.0 Hz), 7.98 (1H, *br s*); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 25.9, 49.4, 53.8, 55.9, 56.0, 111.2, 111.3, 111.4, 114.0, 119.0, 119.3, 120.3, 122.1, 127.6, 133.2, 136.5, 148.1, 149.1; HRMS (ESI) Calcd. for C₁₉H₂₃N₂O₂: 311.1760 [M+H]⁺. Found: 311.1766. *Hydrochloride*: colorless solid. m.p. 252–254 °C; ¹H-NMR (400 MHz, DMSO-*d*₆, δ / ppm): 3.04–3.19 (4H, *m*), 3.75 (3H, *s*), 3.78 (3H, *s*), 4.10 (2H, *s*), 6.93–7.02 (2H, *m*), 7.04–7.11 (2H, *m*), 7.21 (1H, *d*, *J* = 2.4 Hz), 7.34–7.39 (2H, *m*), 7.57 (1H, *d*, *J* = 7.6 Hz), 9.56 (2H, *br s*); ¹³C-NMR (100 MHz, DMSO-*d*₆, δ / ppm): 21.5, 46.4, 49.6, 55.5, 55.6, 109.4, 111.4, 111.6, 113.9, 118.1, 118.3, 121.1, 122.6, 123.1, 124.2, 126.7, 136.2, 148.6, 149.1.

2-(1*H*-Indol-3-yl)-N-(naphthalen-1-ylmethyl)ethanamine (**4c**). Light yellow oil. Yield: 5.75 g, 96 %; *R*_f 0.31 (ethyl acetate); Anal. Calcd. for C₂₁H₂₀N₂: C, 83.96; H, 6.71; N, 9.33 %. Found: C, 84.20, H, 6.87, N, 9.14 %; ¹H-NMR (400

MHz, CDCl₃, δ / ppm): 3.06 (2H, *t*, J = 6.8 Hz), 3.14 (2H, *t*, J = 6.8 Hz), 4.27 (2H, *s*), 6.96 (1H, *d*, J = 2.0 Hz), 7.11–7.25 (2H, *m*), 7.34 (1H, *d*, J = 8.0 Hz), 7.37–7.51 (4H, *m*), 7.66 (1H, *dd*, J = 0.8 and 8.0 Hz), 7.76 (1H, *d*, J = 8.0 Hz), 7.86 (1H, *dd*, J = 2.4 and 7.2 Hz), 8.02 (1H, *dd*, J = 1.6 and 8.0 Hz), 8.08 (1H, *br s*); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 25.9, 50.0, 51.6, 111.3, 114.1, 119.0, 119.4, 122.1 (2 \times), 123.7, 125.5, 125.7, 126.0, 126.1, 127.6, 127.8, 128.8, 131.9, 133.9, 136.1, 136.5; HRMS (ESI) Calcd. for C₂₁H₁₈N₂: 301.1705 [M+H]⁺. Found: 301.1694. *Hydrochloride*: colorless solid. m.p. 234–235 °C (methanol); ¹H-NMR (400 MHz, DMSO-*d*₆, δ / ppm): 3.18–3.38 (4H, *m*), 4.70 (2H, *s*), 6.97–7.15 (2H, *m*), 7.23 (1H, *d*, J = 2.4 Hz), 7.38 (1H, *d*, J = 8.0 Hz), 7.53–7.70 (4H, *m*), 7.87 (1H, *d*, J = 6.8 Hz), 8.01 (2H, *d*, J = 7.6 Hz), 8.28 (1H, *d*, J = 8.4 Hz), 9.69 (2H, *br s*); ¹³C-NMR (100 MHz, DMSO-*d*₆, δ / ppm): 21.6, 46.6, 47.6, 109.5, 111.5, 118.2, 118.3, 121.1, 123.1, 123.7, 125.2, 126.1, 126.7, 126.8, 128.2, 128.5, 128.9, 129.4, 131.0, 133.2, 136.2.

2-(1H-Indol-3-yl)-N-(2-thienylmethyl)ethanamine (4d). Colorless solid. Yield: 4.99 g, 97 %; m.p. 63–64 °C; Anal. Calcd. for C₁₅H₁₆N₂S: C, 70.27; H, 6.29; N, 10.93 %. Found: C, 70.05, H, 6.48, N, 11.11 %; ¹H-NMR (400 MHz, CDCl₃, δ / ppm): 2.98–3.08 (4H, *m*), 4.03 (2H, *s*), 6.88–6.93 (1H, *m*), 6.94 (1H, *dd*, J = 3.6 and 5.2 Hz), 7.02 (1H, *d*, J = 2.0 Hz), 7.10–7.25 (3H, *m*), 7.35 (1H, *d*, J = 8.4 Hz), 7.63 (1H, *d*, J = 8.0 Hz), 8.11 (1H, *br s*); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 25.8, 48.5, 49.3, 111.3, 114.0, 119.0, 119.4, 122.1, 122.2, 124.4, 124.9, 126.7, 127.6, 136.5, 144.3; HRMS (ESI) Calcd. for C₁₅H₁₇N₂S: 257.1112 [M+H]⁺. Found: 257.1120.

1-(4-Bromobenzyl)-N-(pyridin-3-yl)methylamine (5a). Yellowish oil. Yield: 7.15 g, 86 %; R_f 0.39 (ethyl acetate–methanol 6:1 v/v); Anal. Calcd. for C₁₃H₁₃BrN₂: C, 56.34; H, 4.73; N, 10.11 %. Found: C, 56.56, H, 4.57, N, 9.92 %; ¹H-NMR (400 MHz, CDCl₃, δ / ppm): 3.76 (2H, *s*), 3.79 (2H, *s*), 7.18–7.29 (3H, *m*), 7.45 (2H, *d*, J = 8.0 Hz), 7.68 (1H, *d*, J = 7.6 Hz), 8.50 (1H, *dd*, J = 1.2 and 4.8 Hz), 8.56 (1H, *d*, J = 1.6 Hz); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 50.5, 52.6, 121.0, 123.6, 129.9, 131.7, 135.5, 135.9, 139.1, 148.735, 149.9; HRMS (ESI) Calcd. for C₁₃H₁₄BrN₂: 277.0340 [M+H]⁺. Found: 277.0331. *Dihydrochloride*: colorless solid, m.p. 201–202 °C (ethanol); ¹H-NMR (400 MHz, CD₃OD, δ / ppm): 4.41 (2H, *s*), 4.63 (2H, *s*), 7.54–7.60 (2H, *m*), 7.61–7.67 (2H, *m*), 8.17–8.24 (1H, *m*), 8.88–8.94 (1H, *m*), 8.98 (1H, *d*, J = 5.6 Hz), 9.20 (1H, *d*, J = 2.0 Hz); ¹³C-NMR (100 MHz, CD₃OD, δ / ppm): 48.3, 52.1, 125.1, 128.9, 131.3, 133.3, 133.4, 133.5, 143.7, 144.8, 149.9.

1-(4-Methoxybenzyl)-N-(pyridin-3-yl)methylamine (5b). Yellow oil. Yield: 5.60 g, 82 %; R_f 0.33 (ethyl acetate–methanol 6:1 v/v); Anal. Calcd. for C₁₄H₁₆N₂O: C, 73.66; H, 7.06; N, 12.27 %. Found: C, 73.89, H, 6.88, N, 12.09 %; ¹H-NMR (400 MHz, CDCl₃, δ / ppm): 3.73 (2H, *s*), 3.79 (5H, *br s*), 6.86 (2H, *d*, J = 8.8 Hz), 7.21–7.28 (3H, *m*), 7.68 (1H, *d*, J = 7.6 Hz), 8.48 (1H, *dd*, J = 1.6

and 4.8 Hz), 8.55 (1H, *d*, *J* = 2.0 Hz); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 50.5, 52.7, 55.4, 114.0, 123.5, 129.4, 132.2, 135.8, 135.9, 148.5, 149.9, 158.9; HRMS (ESI) Calcd. for C₁₄H₁₇N₂O: 229.1341 [M+H]⁺. Found: 229.1349. *Dihydrochloride*: colorless solid. m.p. 204–205 °C (ethanol); ¹H-NMR (400 MHz, DMSO-*d*₆, δ / ppm): 3.75 (3H, *s*), 4.14 (2H, *s*), 4.38 (2H, *s*), 6.95 (2H, *d*, *J* = 8.4 Hz), 7.54 (2H, *d*, *J* = 8.4 Hz), 8.06 (1H, *dd*, *J* = 5.6 and 8.0 Hz), 8.80 (1H, *d*, *J* = 8.0 Hz), 8.91 (1H, *dd*, *J* = 1.2 and 5.6 Hz), 9.13 (1H, *d*, *J* = 1.2 Hz), 10.32 (2H, *br s*); ¹³C-NMR (100 MHz, DMSO-*d*₆, δ / ppm): 46.1, 49.6, 55.2, 113.9, 123.6, 126.4, 131.8, 131.9, 142.3, 144.1, 147.0, 159.7.

1-(4-Biphenyl)-N-(pyridin-3-yl)methylamine (5c). Yellow oil. Yield 7.51 g, 91 %; *R*_f 0.66 (ethyl acetate–methanol 6:1 v/v); Anal. Calcd. for C₁₉H₁₈N₂: C, 83.18; H, 6.61; N, 10.21 %. Found: C, 83.41, H, 6.42, N, 10.40 %; ¹H-NMR (400 MHz, CDCl₃, δ / ppm): 3.85 (2H, *s*), 3.86 (2H, *s*), 7.24–7.29 (1H, *m*), 7.31–7.37 (1H, *m*), 7.39–7.47 (4H, *m*), 7.55–7.63 (4H, *m*), 7.70–7.75 (1H, *m*), 8.52 (1H, *dd*, *J* = 1.6 and 4.8 Hz), 8.60 (1H, *d*, *J* = 2.0 Hz); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 50.6, 53.0, 123.5, 127.2, 127.3, 128.7, 128.9, 135.8, 135.9, 139.2, 140.3, 141.1, 148.7, 149.9; HRMS (ESI) Calcd. for C₁₉H₁₉N₂: 275.1548 [M+H]⁺. Found: 275.1554. *Dihydrochloride*: colorless solid, m.p. 258–260 °C (methanol); ¹H-NMR (400 MHz, DMSO-*d*₆, δ / ppm): 4.26 (2H, *s*), 4.43 (2H, *s*), 7.35–7.41 (1H, *m*), 7.43–7.51 (2H, *m*), 7.65–7.75 (6H, *m*), 8.03 (1H, *dd*, *J* = 6.4 and 8.0 Hz), 8.77 (1H, *d*, *J* = 8.0 Hz), 8.91 (1H, *dd*, *J* = 1.2 and 5.6 Hz), 9.14 (1H, *d*, *J* = 1.2 Hz), 10.41 (2H, *s*); ¹³C-NMR (100 MHz, DMSO-*d*₆, δ / ppm): 46.4, 49.7, 126.2, 126.7, 126.8, 127.8, 129.0, 130.9, 131.1, 131.4, 139.4, 140.6, 142.9, 144.7, 146.3.

1-(Naphthalen-1-ylmethyl)-N-(pyridin-3-yl)methylamine (5d). Orange oil. Yield: 4.78 g, 64 %; *R*_f 0.37 (ethyl acetate–methanol 6:1 v/v); Anal. Calcd. for C₁₇H₁₆N₂: C, 82.22; H, 6.49; N, 11.28 %. Found: C, 82.00, H, 6.68, N, 11.07 %; ¹H-NMR (400 MHz, CDCl₃, δ / ppm): 3.91 (2H, *s*), 4.25 (2H, *s*), 7.23–7.29 (1H, *m*), 7.39–7.56 (4H, *m*), 7.73 (1H, *d*, *J* = 8.0 Hz), 7.78 (1H, *d*, *J* = 8.0 Hz), 7.86 (1H, *dd*, *J* = 1.6 and 7.6 Hz), 8.10 (1H, *d*, *J* = 8.0 Hz), 8.52 (1H, *dd*, *J* = 1.6 and 4.8 Hz), 8.62 (1H, *d*, *J* = 1.6 Hz); ¹³C-NMR (100 MHz, CDCl₃, δ / ppm): 51.1, 51.2, 123.5, 123.8, 125.5, 125.8, 126.2, 126.3, 128.1, 128.8, 131.9, 134.0, 135.5, 135.8, 136.0, 148.7, 149.9; HRMS (ESI) Calcd. for C₁₇H₁₇N₂: 249.1392 [M+H]⁺. Found: 249.1401. *Dihydrochloride*: Colorless solid. m.p. 236–237 °C (methanol); ¹H-NMR (400 MHz, DMSO-*d*₆, δ / ppm): 4.61 (2H, *s*), 4.74 (2H, *s*), 7.52–7.67 (3H, *m*), 7.85 (1H, *d*, *J* = 6.8 Hz), 7.96–8.07 (3H, *m*), 8.27 (1H, *d*, *J* = 8.4 Hz), 8.80 (1H, *d*, *J* = 8.0 Hz), 8.90 (1H, *d*, *J* = 5.6 Hz), 9.16 (1H, *s*), 10.22 (2H, *br s*); ¹³C-NMR (100 MHz, DMSO-*d*₆, δ / ppm): 47.0, 47.1, 123.9, 125.4, 126.4, 126.9, 128.0, 128.7, 129.3, 129.7, 131.1, 131.5, 133.3, 142.8, 144.7, 146.8.

1-(Pyridin-3-ylmethyl)-N-(2-thienyl)methylamine (5e). Orange oil. Yield: 3.85 g, 63 %; R_f 0.34 (ethyl acetate–methanol 6:1 v/v); Anal. Calcd. for $C_{11}H_{12}N_2S$: C, 64.67; H, 5.92; N, 13.71 %. Found: C, 64.90, H, 6.14, N, 13.48 %; 1H -NMR (400 MHz, $CDCl_3$, δ / ppm): 3.84 (2H, s), 4.00 (2H, s), 6.90–6.98 (2H, m), 7.20–7.29 (2H, m), 7.70 (1H, d, $J = 7.6$ Hz), 8.50 (1H dd, $J = 1.6$ and 4.8 Hz), 8.56 (1H, d, $J = 2.0$ Hz); ^{13}C -NMR (100 MHz, $CDCl_3$, δ / ppm): 47.7, 50.1, 123.5, 124.7, 125.2, 126.8, 135.4, 136.0, 143.8, 148.7, 149.9; HRMS (ESI) Calcd. for $C_{11}H_{13}N_2S$: 205.0799 $[M+H]^+$. Found: 205.0807. *Dihydrochloride*: colorless solid. m.p. 228–230 °C (dec) (ethanol); 1H -NMR (400 MHz, CD_3OD , δ / ppm): 4.63 (2H, s), 4.67 (2H, s), 7.12 (1H, dd, $J = 3.6$ and 5.2 Hz), 7.45 (1H, d, $J = 3.2$ Hz), 7.59 (1H, dd, $J = 0.8$ and 5.2 Hz), 8.21 (1H, dd, $J = 6.0$ and 8.0 Hz), 8.89 (1H, d, $J = 8.0$ Hz), 8.98 (1H, d, $J = 6.0$ Hz), 9.18 (1H, d, $J = 0.8$ Hz); ^{13}C -NMR (100 MHz, CD_3OD , δ / ppm): 46.6, 47.8, 128.8, 128.9, 129.8, 132.6, 132.7, 133.3, 143.7, 144.7, 149.9.

N-Benzyl-3-(1H-imidazol-1-yl)propylamine (6a). Flash column chromatography (silica gel, ethyl acetate–methanol 1:1 v/v) afforded a colorless oil. Yield: 1.46 g, 68 %; R_f 0.25 (ethyl acetate–methanol 1:1 v/v); Anal. Calcd. for $C_{13}H_{17}N_3$: C, 72.52; H, 7.96; N, 19.52 %. Found: C, 72.81, H, 8.28, N, 19.85 %; 1H -NMR (400 MHz, $CDCl_3$, δ / ppm): 1.87–1.97 (2H, m), 2.61 (2H, t, $J = 6.8$ Hz), 3.75 (2H, s), 4.04 (2H, t, $J = 6.8$ Hz), 6.87 (1H, t, $J = 1.2$ Hz), 7.03 (1H, s), 7.22–7.36 (5H, m), 7.43 (1H, s); ^{13}C -NMR (100 MHz, $CDCl_3$, δ / ppm): 31.5, 44.7, 45.8, 54.1, 119.0, 127.2, 128.2, 128.6, 129.5, 137.3, 140.3. *Dihydrochloride*: hygroscopic colorless crystals. Yield: 56 %; m.p. 144–145 °C (2-propanol); 1H -NMR (400 MHz, CD_3OD , δ / ppm): 2.32–2.43 (2H, m), 3.16 (2H, t, $J = 7.6$ Hz), 4.25 (2H, s), 4.44 (2H, t, $J = 7.2$ Hz), 7.43–7.50 (3H, m), 7.53–7.59 (2H, m), 7.62 (1H, t, $J = 1.6$ Hz), 7.75 (1H, t, $J = 1.6$ Hz), 9.09 (1H, s); ^{13}C -NMR (100 MHz, CD_3OD , δ / ppm): 27.9, 45.3, 47.5, 52.5, 121.4, 123.3, 130.3, 130.7, 131.1, 132.4, 136.7; HRMS (ESI) Calcd. for $C_{13}H_{18}N_3$: 216.1501 $[M+H]^+$.

3-(1H-Imidazol-1-yl)-N-(1-naphthalenylmethyl)propylamine (6b). Flash column chromatography (silica gel, ethyl acetate–methanol 3:1 v/v) afforded a yellowish oil. Yield: 1.35 g, 51 %; R_f 0.18 (ethyl acetate–methanol 3:1 v/v); Anal. Calcd. for $C_{17}H_{19}N_3$: C, 76.95; H, 7.22; N, 15.84 %. Found: C, 77.29, H, 6.91, N, 16.15 %; 1H -NMR (400 MHz, $CDCl_3$, δ / ppm): 1.89–2.00 (2H, m), 2.71 (2H, t, $J = 6.8$ Hz), 4.03 (2H, t, $J = 6.8$ Hz), 4.21 (2H, s), 6.84 (1H, s), 7.03 (1H, s), 7.38–7.45 (3H, m), 7.47–7.58 (2H, m), 7.75–7.83 (1H, m), 7.88 (1H, dd, $J = 1.6$ and 8.4 Hz), 8.13 (1H, d, $J = 8.4$ Hz); ^{13}C -NMR (100 MHz, $CDCl_3$, δ / ppm): 31.5, 44.7, 46.2, 51.9, 118.9, 123.8, 125.4, 125.8, 126.2, 126.3, 128.0, 128.9, 129.4, 131.8, 134.0, 135.8, 137.3; HRMS (ESI) Calcd. for $C_{17}H_{20}N_3$: 266.1657 $[M+H]^+$. Found: 266.1665. *Dihydrochloride*: hygroscopic colorless crystals. Yield: 61 %, m.p. 153–154 °C (ethanol); 1H -NMR (400 MHz, $DMSO-d_6$, δ / ppm): 2.29–2.43 (2H, m), 3.07 (2H, t, $J = 6.8$ Hz), 4.42 (2H, t, $J = 6.8$ Hz), 4.63

(2H, *s*), 7.52–7.66 (3H, *m*), 7.72 (1H, *t*, $J = 1.6$ Hz), 7.84–7.90 (2H, *m*), 7.96–8.03 (2H, *m*), 8.24 (1H, *d*, $J = 8.0$ Hz), 9.32 (1H, *s*), 9.83 (2H, *br s*); ^{13}C -NMR (100 MHz, $\text{DMSO-}d_6$, δ / ppm): 25.8, 43.9, 45.8, 46.7, 119.9, 122.0, 123.8, 125.3, 126.2, 126.8, 128.1, 128.6, 128.9, 129.4, 131.0, 133.2, 135.4.

3-(1H-Imidazol-1-yl)-N-(2-thienylmethyl)propylamine (6c). Flash column chromatography (silica gel, ethyl acetate–methanol 3:1 v/v) afforded a colorless oil. Yield: 1.15 g, 52 %; R_f 0.17 (ethyl acetate–methanol 3:1 v/v); Anal. Calcd. for $\text{C}_{11}\text{H}_{15}\text{N}_3\text{S}$: C, 59.69; H, 6.83; N, 18.99 %. Found: C, 60.02, H, 6.51, N, 19.29 %; ^1H -NMR (400 MHz, CDCl_3 , δ / ppm): 1.86–1.95 (2H, *m*), 2.62 (2H, *t*, $J = 6.8$ Hz), 3.94 (2H, *s*), 4.04 (2H, *t*, $J = 6.8$ Hz), 6.86–6.91 (2H, *m*), 6.93 (1H, *dd*, $J = 3.6$ and 4.8 Hz), 7.02 (1H, *t*, $J = 1.2$ Hz), 7.20 (1H, *dd*, $J = 1.2$ and 4.8 Hz), 7.43 (1H, *br s*); ^{13}C -NMR (100 MHz, CDCl_3 , δ / ppm): 31.4, 44.7, 45.4, 48.4, 118.9, 124.6, 125.0, 126.8, 129.4, 137.3, 144.0; HRMS (ESI) Calcd. for $\text{C}_{11}\text{H}_{16}\text{N}_3\text{S}$: 222.1065 $[\text{M}+\text{H}]^+$. Found: 222.1057.

N-(2,4-Dimethoxybenzyl)-3-(1H-imidazol-1-yl)propylamine (6d). Flash column chromatography (silica gel, ethyl acetate–methanol 1:1 v/v) afforded a colorless oil. Yield: 1.43 g, 52 %; R_f 0.18 (ethyl acetate–methanol 1:1 v/v); Anal. Calcd. for $\text{C}_{15}\text{H}_{21}\text{N}_3\text{O}_2$: C, 65.43; H, 7.69; N, 15.26 %. Found: C, 65.68, H, 8.03, N, 15.55 %; ^1H -NMR (400 MHz, CDCl_3 , δ / ppm): 1.86–1.95 (2H, *m*), 2.53 (2H, *t*, $J = 6.8$ Hz), 3.66 (2H, *s*), 3.78 (3H, *s*), 3.80 (3H, *s*), 4.01 (2H, *t*, $J = 6.8$ Hz), 6.38–6.49 (2H, *m*), 6.88 (1H, *s*), 7.02 (1H, *s*), 7.07 (1H, *d*, $J = 8.0$ Hz), 7.44 (1H, *s*); ^{13}C -NMR (100 MHz, CDCl_3 , δ / ppm): 31.4, 44.8, 45.4, 48.9, 55.4, 55.5, 98.7, 103.8, 119.0, 120.7, 129.4, 130.5, 137.3, 158.7, 160.3; HRMS (ESI) Calcd. for $\text{C}_{15}\text{H}_{22}\text{N}_3\text{O}_2$: 276.1712 $[\text{M}+\text{H}]^+$. Found: 276.1720.

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