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Supporting Information

$B(C_6F_5)_3$-Catalyzed Tandem Protonation/Deuteration and Reduction of in situ-formed Enamines

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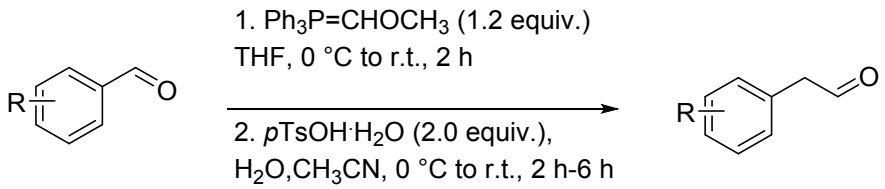
1. General Information.

Unless otherwise noted, materials were purchased from commercial suppliers and used without further purification. Flash column chromatography was performed using 200–300 mesh silica gel or basic aluminum oxide. ^{1}H and ^{13}C NMR spectra were recorded on a VARIAN-400 (400 MHz) or Bruker AV-400 (400 MHz) NMR spectrometer. Chemical shifts (δ) are reported in ppm from the resonance of tetramethyl silane as the internal standard (TMS: 0.00 ppm). Data are reported as follows: chemical shift, multiplicity (s, singlet; d, doublet; t, triplet; q, quartet; m, multiplet), coupling constants (herz) and integration. High-resolution mass spectra were obtained with a MICROTOF-10454 Premier LC HR mass spectrometer.

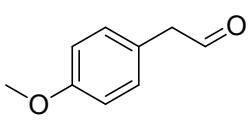
Materials. Unless otherwise noted, commercial reagents were purchased from Adamas, Energy-Chemical and other commercial suppliers and were used as received. (*E*)-1-Styrylpyrrolidine, [1] 2-(*o*-Tolyl)acetaldehyde (**1b**), 2-(*p*-Tolyl)acetaldehyde (**1d**),[2] 2-(*m*-Tolyl)acetaldehyde (**1c**), 2-(Naphthalen-1-yl)acetaldehyde (**1j**),[3] and 2-(4-Hydroxyphenyl)acetaldehyde (**1h**)[4] were prepared according to the literature procedure.

2. General Procedure and Spectral Data of Aldehydes.



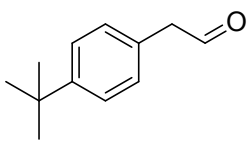
To a solution of (methoxymethyl)triphenylphosphonium chloride (1.54 g, 4.5 mmol) in THF (6 mL) were added sodium bis(trimethylsilyl)amide (2.0 M in THF, 2.25 mL, 4.5 mmol) dropwise at 0 ºC. The resulting mixture was stirred at 0 ºC for 0.5 h. A solution of arylacetylaldehyde (3.0 mmol) in THF (6 mL) was added dropwise at 0 ºC and stirred at room temperature for 2 h. The solvent was removed under vaccum. To the crude mixture were added H_2O (1.5 mL), and CH_3CN (6 mL). Then, *p*TsOH • H_2O (1.1413 g, 6.0 mmol) was added at 0 ºC, and the resulting mixture was stirred at room temperature for 2-6 h. Then the resulting mixture was concentrated under reduced pressure and then purified by flash column chromatography.[5]

2-(4-Methoxyphenyl)acetaldehyde (1f)



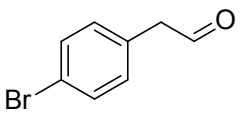
The typical procedure was applied to 2-(4-methoxyphenyl)acetaldehyde (408.5 mg, 3.0 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 20/1) of the crude product afforded the title compound as a yellow oil (239.9 mg, 53 %); R_f = 0.20 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.70 (t, J = 2.8 Hz, 1H), 7.12 (d, J = 7.9 Hz, 2H), 6.90 (d, J = 8.2 Hz, 2H), 3.79 (s, 3H), 3.61 (t, J = 1.9 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 199.6, 158.8, 130.6, 123.6, 114.3, 55.1, 49.5. The physical and spectral data were consistent with those previously reported.[6]

2-(4-(*tert*-Butyl)phenyl)acetaldehyde (1e)



The typical procedure was applied to 2-(4-*tert*-butylphenyl)acetaldehyde (444.6 mg, 3.0 mmol) for 3 h. Silica gel chromatography (eluent: PE/EtOAc = 50/1) of the crude product afforded the title compound as a yellow oil (116.3 mg, 22 %); R_f = 0.80 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.75 (t, J = 2.4 Hz, 1H), 7.40 (d, J = 8.1 Hz, 2H), 7.16 (d, J = 8.1 Hz, 2H), 3.68-3.66 (m, 2H), 1.32 (s, 9H); ^{13}C NMR (100 MHz, CDCl3) δ 199.7, 150.4, 129.3, 128.8, 126.0, 50.1, 34.5, 31.3. The physical and spectral data were consistent with those previously reported.[6]

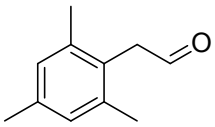
2-(4-Bromophenyl)acetaldehyde (1g)



The typical procedure was applied to 2-(4-bromophenyl)acetaldehyde (555.1 mg, 3.0 mmol) for 2 h. Silica gel chromatography (eluent: PE/EtOAc = 30/1) of the crude product afforded the title compound as a yellow solid (265.5 mg, 44 %); R_f = 0.20 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.73 (t, J = 1.9 Hz, 1H), 7.49 (d, J = 7.9 Hz, 2H), 7.09 (d, J = 8.0 Hz, 2H), 3.67 (d, J = 2.1 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 198.6, 132.1, 131.3, 130.7, 121.5, 49.8. The physical and spectral data were consistent with those previously reported.[7]

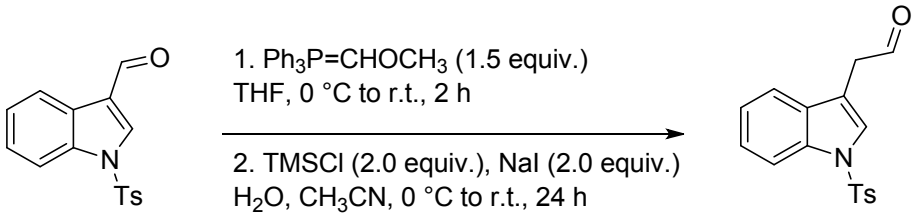
2-Mesitylacetaldehyde (1i)

The typical procedure was applied to 2-mesitylacetaldehyde (486.7 mg, 3.0 mmol) for 3 h. Silica gel chromatography (eluent: PE/EtOAc = 50/1) of the crude product afforded the title

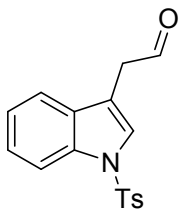


compound as a white solid (207.6 mg, 43 %); R_f = 0.80 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.66 (t, J = 2.8 Hz, 1H), 6.91 (s, 2H), 2.28 (d, J = 2.4 Hz, 3H), 2.25 (s, 6H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 199.1, 137.2, 137.0, 129.2, 126.2, 44.8, 20.9, 20.4. The physical and spectral data were consistent with those previously reported.[8]

2-(1-Tosyl-1*H*-indol-3-yl)acetaldehyde (1p)



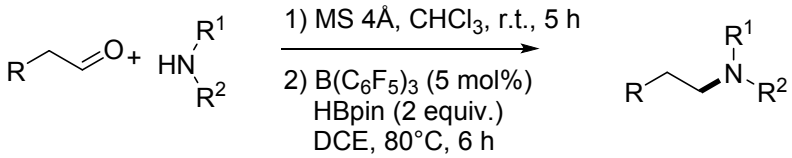
1-Tosyl-1*H*-indole-3-carbaldehyde was synthesized according to the literature.[9] To a solution of (methoxymethyl)triphenylphosphonium chloride (1.5426 g, 4.5 mmol) in THF (6 mL) was added sodium bis(trimethylsilyl)amide (2.0 M in THF, 2.25 mL, 4.5 mmol) dropwise at 0 ºC. The resulting mixture was stirred at 0 ºC for 0.5 h. A solution of 1-tosyl-1*H*-indole-3-carbaldehyde (0.8980 g, 3.0 mmol) in THF (6 mL) was added dropwise at 0 ºC, and stirred at room temperature for 2 h. The solvent was removed under vaccum. To the crude mixture was added H_2O (1.5 mL), and CH_3CN (6 mL). Then, TMSCl (0.76 mL, 6 mmol, 2.0 equiv.) and NaI (0.8993 g, 6 mmol, 2.0 equiv.) was added at 0 ºC, and the resulting mixture was stirred at room temperature for 24 h. Then the resulting mixture was concentrated under reduced pressure and then purified by flash column chromatography (eluent: PE/EtOAc = 10/1) to afford the title compound as a yellow solid (336.5 mg, 36 %).[5]



R_f = 0.30 (PE/EtOAc = 4/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.74 (q, J = 2.5 Hz, 1H), 8.01 (d, J = 8.4 Hz, 1H), 7.79-7.77 (m, 2H), 7.58 (s, 1H), 7.42 (d, J = 7.4 Hz, 1H), 7.36 (t, J = 7.8 Hz, 1H), 7.27-7.22 (m, 3H), 3.75 (s, 2H), 2.35 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 197.9, 145.1, 135.2, 135.1, 130.4, 130.0, 126.9, 125.2, 124.0, 123.4, 119.2, 113.8, 112.9, 39.9, 21.6. The physical and spectral data were consistent with those previously reported.[10]

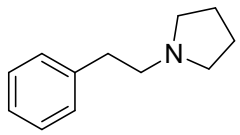
3. General Procedure and Spectral Data of Products.

3.1 General Procedure for Synthesis of Tertial Amines.



In a Schlenk tube were placed secondary amine (0.24 mmol, 1.2 equiv.), arylacetaldehyde (0.2 mmol), MS 4Å (135 mg) and CH_3Cl (0.4 mL). The resulting mixture was stirred at room temperature for 5 h (0 ºC, 0.5 h for morpholine), and then filtered and the solvent was concentrated under reduced pressure. The correspond enamine was directly used without further purification. To a Schlenk tube were placed the crude enamine, HBpin (51.2 mg, 0.4 mmol, 2.0 equiv.), $B(C_6F_5)_3$ (5.1 mg, 0.01 mmol), and DCE (0.8 mL). The resulting mixture was stirred at 80 ºC for 6 h, and then allowed to room temperature. The solution was concentrated in *vacuo* before purifying by column chromatography.

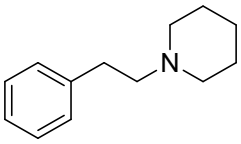
1-Phenethylpyrrolidine (3aa)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and pyrrolidine (17.1 mg, 0.24 mmol) for 6 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a colorless oil (29.1 mg, 83%); R_f = 0.3 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.29-7.24 (m, 2H), 7.21-7.17 (m, 3H), 2.92-2.88 (m, 2H), 2.81-2.77 (m, 2H), 2.71-2.68 (m, 4H), 1.89-1.82 (m, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 140.5, 128.6, 128.3, 126.0, 58.4, 54.2, 35.8, 23.4. The physical and spectral data were consistent with those previously reported.[11]

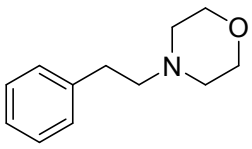
1-Phenethylpiperidine (3ab)

The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and piperidine (20.4 mg, 0.24 mmol) for 6 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a white solid



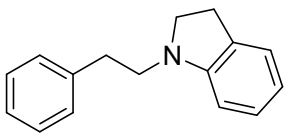
(30.0 mg, 79%); R_f = 0.4 (PE/EtOAc/MeOH = 40/10/1);,^{1}H NMR (400 MHz, $CDCl_3$) δ 7.39-7.34 (m, 2H), 7.30-7.26 (m, 3H), 2.92-2.88 (m, 2H), 2.66-2.62 (m, 2H), 2.56 (bs, 4H), 1.74-1.68 (m, 4H), 1.57-1.51 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 140.4, 128.6, 128.3, 125.9, 61.3, 54.4, 33.5, 25.8, 24.3. The physical and spectral data were consistent with those previously reported.[11]

4-Phenethylmorpholine (3ac)



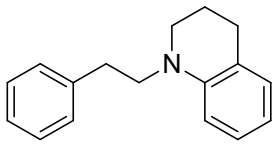
The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 6 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a white solid (34.2 mg, 89%); R_f = 0.5 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31-7.27 (m, 2H), 7.22-7.19 (m, 3H), 3.75 (t, J = 4.7 Hz, 4H), 2.83-2.79 (m, 2H), 2.60-2.58 (m, 2H), 2.54 (t, J = 4.8 Hz, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 140.0, 128.6, 128.3, 126.0, 66.9, 60.8, 53.6, 33.2. The physical and spectral data were consistent with those previously reported.[11]

1-Phenethylindoline (3ad)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and indoline (28.6 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 80/1) of the crude product afforded the title compound as a yellow oil (30.8 mg, 69%); R_f = 0.7 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.37-7.27 (m, 5H), 7.15-7.09 (m, 2H), 6.70 (q, J = 7.3 Hz, 1H), 6.54 (t, J = 7.3 Hz, 1H), 3.47-3.35 (m, 4H), 3.02 (q, J = 7.9 Hz, 2H), 2.94 (q, J = 7.5 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 152.1, 139.9, 130.0, 128.7, 128.5, 127.3, 126.2, 124.4, 117.4, 106.8, 53.0, 50.9, 33.5, 28.6. The physical and spectral data were consistent with those previously reported.[12]

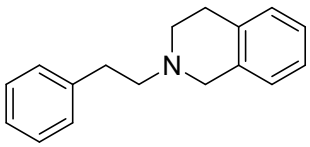
1-Phenethyl-1,2,3,4-tetrahydroquinoline (3ae) (Gram-scale)



In a Schlenk tube were placed 1,2,3,4-tetrahydroquinoline (**2e**) (961.2 mg, 9.6 mmol, 1.2 equiv.), phenylacetaldehyde (127.9 mg, 8.0 mmol),

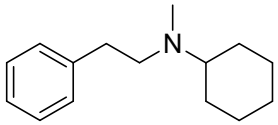
MS 4Å (5.4 g) and CH_3Cl (16 mL). The resulting mixture was stirred at room temperature for 5 h, and then filtered and the solvent was concentrated under reduced pressure. The correspond enamine was directly used without further purification. To a Schlenk tube was placed the crude enamine, HBpin (2.0476 g, 16 mmol, 2.0 equiv.), $B(C_6F_5)_3$ (204.8 mg, 0.4 mmol), and DCE (32 mL). The resulting mixture was stirred at 80 ºC for 6 h, and then allowed to room temperature. The solution was concentrated in *vacuo* before purifying by column chromatography. Silica gel chromatography of the crude product afforded the title compound as a yellow oil (1.3217 g, 70%); R_f = 0.8 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.35-7.32 (m, 2H), 7.27-7.25 (m, 3H), 7.11 (t, J = 7.8 Hz, 1H), 6.98 (d, J = 7.4 Hz, 1H), 6.69 (d, J = 8.2 Hz, 1H), 6.61 (t, J = 7.3 Hz, 1H), 3.51 (t, J = 7.8 Hz, 2H), 3.23 (t, J = 5.6 Hz, 2H), 2.90 (t, J =7.7 Hz, 2H), 2.77 (t, J = 6.4 Hz, 2H), 1.93 (p, J = 5.9 Hz, 2H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 144.7, 139.9, 129.2, 128.8, 128.4, 127.1, 126.1, 122.3, 115.5, 110.3, 53.3, 49.5, 32.4, 28.1, 22.1. The physical and spectral data were consistent with those previously reported.[13]

2-Phenethyl-1,2,3,4-tetrahydroisoquinoline (3af)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and 1,2,3,4-tetrahydroisoquinoline (32.0 mg, 0.24 mmol) for 6 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (27.9 mg, 59%); R_f = 0.3 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.25-7.11 (m, 5H), 7.07-7.01 (m, 3H), 6.98-6.94 (m, 1H), 3.65 (s, 2H), 2.87-2.82 (m, 4H), 2.77-2.66 (m, 4H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 140.3, 134.6, 134.2, 128.7, 128.6, 128.4, 126.6, 126.1, 126.0, 125.6, 60.3, 56.0, 50.9, 33.9, 29.0. The physical and spectral data were consistent with those previously reported.[14]

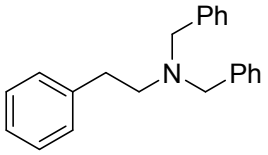
***N*-Methyl-*N*-phenethylcyclohexanamine (3ag)**



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and *N*-methylcyclohexanamine (27.2 mg, 0.24 mmol) for 6 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (35.8 mg, 82%);

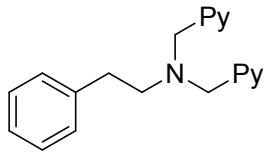
R_f = 0.2 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31-7.26 (m, 2H), 7.22-7.18 (m, 3H), 2.80-2.76 (m, 2H), 2.73-2.68 (m, 2H), 2.48-2.36 (m, 1H), 2.36 (s, 3H), 1.85-1.75 (m, 4H), 1.66-1.61 (m, 1H), 1.30-1.18 (m, 5H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 140.6, 128.7, 128.3, 125.9, 62.6, 55.8, 37.7, 34.5, 28.5, 26.3, 25.9. The physical and spectral data were consistent with those previously reported.[15]

_N_,_N_-Dibenzyl-2-phenylethanamine (3ah)



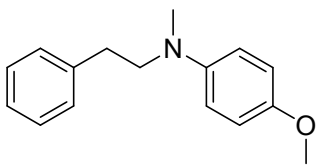
The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and dibenzylamine (47.3 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 50/1) of the crude product afforded the title compound as a yellow oil (41.1 mg, 68%); R_f = 0.7 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.25-7.18 (m, 9H), 7.16-7.14 (m, 3H), 7.12-7.07 (m, 1H), 7.00 (d, J = 7.4 Hz, 2H) 3.56 (s, 4H), 2.76-2.72 (m, 2H), 2.65-2.61 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 140.5, 139.7, 128.8, 128.7, 128.2, 128.1, 126.8, 125.8, 58.1, 55.1, 33.5. The physical and spectral data were consistent with those previously reported.[16]

2-Phenyl-_N_,_N_-bis(pyridin-2-ylmethyl)ethanamine (3ai)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and bis(pyridin-2-ylmethyl)amine (47.8 mg, 0.24 mmol) for 6 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (41.9 mg, 69%); R_f = 0.2 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.51 (d, J = 4.5 Hz, 2H), 7.58 (t, J = 7.6 Hz, 2H), 7.38 (d, J = 7.9 Hz, 2H), 7.26-7.22 (m, 2H), 7.19-7.18 (m, 1H), 7.14-7.09 (m, 4H), 3.89 (s, 4H), 2.87-2.81 (m, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 159.6, 148.8, 140.3, 136.3, 128.8, 128.1, 125.8, 122.7, 121.8, 60.2, 56.0, 33.4. HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{20}H_{22}N_3$: 304.1808, found 304.1813.

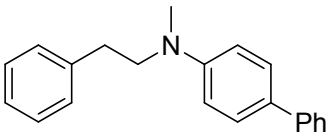
4-Methoxy-_N_-methyl-_N_-phenethylaniline (3aj)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and 4-methoxy-_N_-methylaniline (32.9 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 50/1)

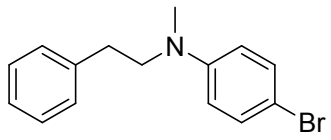
of the crude product afforded the title compound as a gray solid (29.1 mg, 60%); R_f = 0.5 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.37-7.31 (m, 2H), 7.28-7.22 (m, 3H), 6.92-6.88 (m, 2H), 6.79-6.76 (m, 2H), 3.81 (s, 3H), 3.56-3.50 (m, 2H), 2.90-2.84 (m, 5H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 151.5, 143.8, 139.9, 128.7, 128.4, 126.1, 114.8, 114.3, 55.8, 55.7, 39.0, 32.6. HRMS (ESI) *m*/*z* $[M + H]^+$ calcd for $C_{16}H_{20}NO$: 242.1539, found 242.1543.

_N_-Methyl-_N_-phenethyl-[1,1'-biphenyl]-4-amine (3ak)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and *N*-methyl-[1,1'-biphenyl]-4-amine (44.0 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 80/1) of the crude product afforded the title compound as a white solid (37.1 mg, 65%); R_f= 0.7 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.48 (d, *J* = 7.4 Hz, 2H), 7.45- 7.42 (m, 2H), 7.31 (t, *J* = 7.6 Hz, 2H), 7.24-7.21 (m, 2H), 7.19-7.11 (m, 4H), 6.69-6.73 (m, 2H), 3.53-3.49 (m, 2H), 2.83 (s, 3H), 2.81-2.77 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 148.1, 141.1, 139.7, 128.8 (two carbons are overlapped), 128.6, 128.5, 127.8, 126.21, 126.19, 125.9, 112.3, 54.7, 38.5, 32.9. HRMS (ESI) *m*/*z* $[M + H]^+$ calcd for $C_{21}H_{22}N$: 288.1747, found 288.1749.

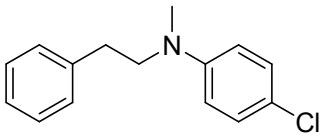
4-Bromo-_N_-methyl-_N_-phenethylaniline (3al)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and 4-bromo-*N*-methylaniline (44.7 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 100/1) of the crude product afforded the title compound as a yellow solid (45.1 mg, 78%); R_f= 0.7 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.32-7.28 (m, 4H), 7.26-7.22 (m, 1H), 7.19 (d, *J* = 7.6 Hz, 2H), 6.59-6.57 (m, 2H), 3.54 (t, *J* = 7.7 Hz, 2H), 2.84 (s, 3H), 2.84-2.81 (m, 2H).; ^{13}C NMR (100 MHz, $CDCl_3$) δ 147.7, 139.4, 131.8, 128.7, 128.5, 126.3, 113.6, 107.9, 54.7, 38.6, 32.7. HRMS (ESI) *m*/*z* $[M + H]^+$ calcd for $C_{15}H_{17}BrN$: 290.0539, found 290.0543.

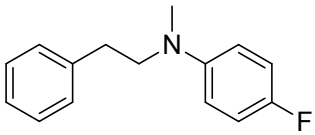
4-Chloro-_N_-methyl-_N_-phenethylaniline (3am)

The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and 4-chloro-*N*-methylaniline (34.0 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 200/1) of the crude product afforded the title compound as a yellow solid (31.6



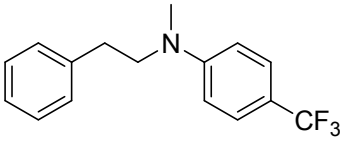
mg, 64%); R_f = 0.7 (PE/EtOAc = 20/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.41-7.37 (m, 2H), 7.31 (d, J = 6.3 Hz, 1H), 7.28-7.25 (m, 4H), 6.70 (d, J = 9.0 Hz, 2H), 3.64-3.60 (m, 2H), 2.93-2.89 (m, 5H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 147.3, 139.5, 129.0, 128.7, 128.5, 126.3, 120.8, 113.1, 54.7, 38.6, 32.7. HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{15}H_{17}ClN$: 246.1044, found 246.1048.

4-Fluoro-*N*-methyl-*N*-phenethylaniline (3an)



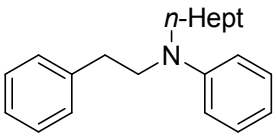
The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and 4-fluoro-*N*-methylaniline (30.0 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 200/1) of the crude product afforded the title compound as a yellow solid (30.6 mg, 67%); R_f = 0.6 (PE/EtOAc = 20/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.32 (t, J = 7.2 Hz, 2H), 7.26-7.21 (m, 3H), 7.00-6.95 (m, 2H), 6.69-6.66 (m, 2H), 3.56-3.52 (m, 2H), 2.90-2.82 (m, 5H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 155.2 (d, $^1J_{C\text{-}F}$ = 237.1 Hz), 145.6, 139.7, 128.7, 128.5, 126.2, 115.6 (d, $^2J_{C\text{-}F}$ = 21.9 Hz), 113.3 (d, $^3J_{C\text{-}F}$ = 5.5 Hz), 55.4, 38.9, 32.7. HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{15}H_{17}FN$: 230.1340, found 230.1345.

***N*-Methyl-*N*-phenethyl-4-(trifluoromethyl)aniline (3ao)**



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and *N*-methyl-4-(trifluoromethyl)aniline (42.0 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 50/1) of the crude product afforded the title compound as a yellow oil (42.5 mg, 76%); R_f = 0.4 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.46 (d, J = 8.6 Hz, 2H), 7.31 (t, J = 7.3 Hz, 2H), 7.25-7.18 (m, 3H), 6.69 (d, J = 8.8 Hz, 2H), 3.61 (t, J = 7.6 Hz, 2H), 2.89 (s, 3H), 2.86 (t, J = 7.6 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 150.7, 139.2, 128.8, 128.6, 126.5 (q, $^3J_{C\text{-}F}$ = 3.8 Hz), 127.3 (q, $^1J_{C\text{-}F}$ = 237.8 Hz), 117.2 (q, $^2J_{C\text{-}F}$ = 32.5 Hz), 110.9, 54.4, 38.6, 32.9. The physical and spectral data were consistent with those previously reported.[17]

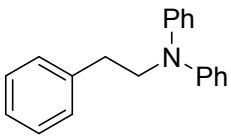
***N*-Heptyl-*N*-phenethylaniline (3ap)**



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and *N*-heptylaniline (45.9 mg, 0.24 mmol) for 6 h. Silica

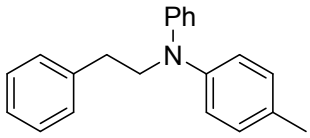
gel chromatography (eluent: PE/EtOAc = 400/1) of the crude product afforded the title compound as a yellow oil (46.0 mg, 84%); R_f = 0.8 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.26-7.22 (m, 2H), 7.19-7.13 (m, 5H), 6.64 (d, J = 8.2 Hz, 2H), 6.59 (t, J = 7.1 Hz, 1H), 3.46-3.42 (m, 2H), 3.16-3.12 (m, 2H), 2.81-2.77 (m, 2H), 1.51-1.44 (m, 2H), 1.25-1.18 (m, 8H), 0.81 (t, J = 6.7 Hz, 3H).; ^{13}C NMR (100 MHz, $CDCl_3$) δ 147.7, 139.8, 129.3, 128.7, 128.5, 126.2, 115.4, 111.7, 52.9, 51.2, 33.5, 31.9, 29.2, 27.3, 27.1, 22.6, 14.1. HRMS (ESI) *m*/*z* $[M + H]^+$ calcd for $C_{21}H_{30}N$: 296.2373, found 296.2371.

_N_-Phenethyl-_N_-phenylaniline (3aq)



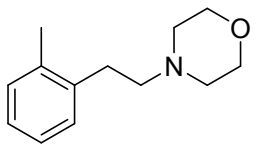
The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and diphenylamine (40.6 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 200/1) of the crude product afforded the title compound as a yellow oil (36.9 mg, 68%); R_f = 0.8 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.24-7.15 (m, 6H), 7.13-7.10 (m, 3H), 6.91-6.85 (m, 6H), 3.87-3.83 (m, 2H), 2.90-2.86 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 147.7, 139.4, 129.3, 128.8, 128.5, 126.3, 121.2, 120.9, 53.9, 33.6. The physical and spectral data were consistent with those previously reported.[13]

4-Methyl-_N_-phenethyl-_N_-phenylaniline (3ar)



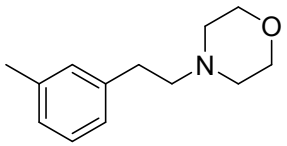
The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and 4-methyl-*N*-phenylaniline (44.0 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 80/1) of the crude product afforded the title compound as a yellow solid (40.1 mg, 70%); R_f = 0.8 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.35 (t, J = 7.2 Hz, 2H), 7.29-7.24 (m, 5H), 7.17 (d, J = 8.0 Hz, 2H), 7.01 (d, J = 8.0 Hz, 2H), 6.95-6.89 (m, 3H), 3.97-3.93 (m, 2H), 3.02-2.98 (m, 2H), 2.38 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 148.1, 145.0, 139.5, 132.2, 130.0, 129.1, 128.8, 128.5, 126.2, 123.3, 119.6, 118.3, 54.0, 33.6, 20.7. HRMS (ESI) *m*/*z* $[M + H]^+$ calcd for $C_{21}H_{22}N$: 288.1747, found 288.1744.

4-(2-Methylphenethyl)morpholine (3bc)



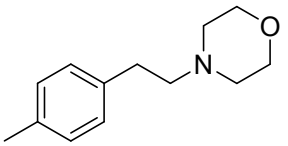
The typical procedure was applied to 2-(*o*-tolyl)acetaldehyde (26.8 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 10 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (28.1 mg, 68%); R_f = 0.2 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.18 (t, J = 7.6 Hz, 1H), 7.03-7.00 (m, 3H), 3.75 (t, J = 4.7 Hz, 4H), 2.80-2.75 (m, 2H), 2.61-2.57 (m, 2H), 2.53 (t, J = 4.7 Hz, 4H), 2.33 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 139.9, 137.9, 129.4, 128.3, 126.8, 125.6, 66.9, 60.9, 53.6, 33.1, 21.3. The physical and spectral data were consistent with those previously reported.[18]

4-(3-Methylphenethyl)morpholine (3cc)



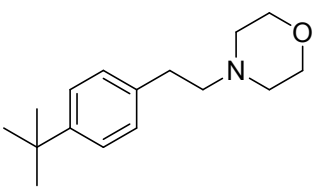
The typical procedure was applied to 2-(*m*-tolyl)acetaldehyde (26.8 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 10 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (30.8 mg, 75%); R_f = 0.2 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.16-7.11 (m, 4H), 3.76 (t, J = 4.7 Hz, 4H), 2.83-2.79 (m, 2H), 2.56-2.52 (m, 6H), 2.33 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 138.1, 135.9, 130.2, 129.2, 126.2, 126.0, 66.9, 59.6, 53.7, 30.4, 19.3. The physical and spectral data were consistent with those previously reported.[18]

4-(4-Methylphenethyl)morpholine (3dc)



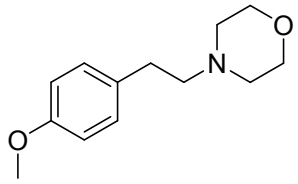
The typical procedure was applied to 2-(*m*-tolyl)acetaldehyde (26.8 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 10 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow solid (29.5 mg, 72%); R_f = 0.2 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.10 (s, 4H), 3.75 (t, J = 4.6 Hz, 4H), 2.80-2.75 (m, 2H), 2.60-2.56 (m, 2H), 2.53 (t, J = 4.7 Hz, 4H), 2.32 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 136.9, 135.5, 129.0, 128.5, 66.9, 61.0, 53.7, 32.8, 21.0. The physical and spectral data were consistent with those previously reported.[18]

4-(4-(*tert*-Butyl)phenethyl)morpholine (3ec)



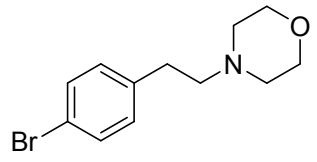
The typical procedure was applied to 2-(4-(*tert*-Butyl)phenyl)acetaldehyde (35.3 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 10 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow solid (27.8 mg, 56%); R_f = 0.3 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.32 (d, J = 8.0 Hz, 2H), 7.15 (d, J = 8.0 Hz, 2H), 3.76 (t, J = 4.7 Hz, 4H), 2.81-2.77 (m, 2H), 2.62-2.58 (m, 2H), 2.54 (t, J = 4.8 Hz, 4H), 1.31 (s, 9H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 145.0, 136.9, 128.4, 125.3, 67.0, 60.9, 53.7, 34.4, 32.7, 31.4. HRMS (ESI) *m*/*z* $[M + H]^+$ calcd for $C_{16}H_{26}NO$: 248.2009, found 248.2008.

4-(4-Methoxyphenethyl)morpholine (3fc)



The typical procedure was applied to 2-(4-methoxyphenyl)acetaldehyde (30.3 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 10 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow solid (33.1 mg, 75%); R_f = 0.3 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.12 (d, J = 8.2 Hz, 2H), 6.83 (d, J = 8.4 Hz, 2H), 3.78 (s, 3H), 3.74 (t, J = 4.7 Hz, 4H), 2.77-2.73 (m, 2H), 2.58-2.54 (m, 2H), 2.52 (t, J = 4.7 Hz, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.9, 132.1, 129.5, 113.8, 66.9, 61.1, 55.2, 53.6, 32.3. The physical and spectral data were consistent with those previously reported.[18]

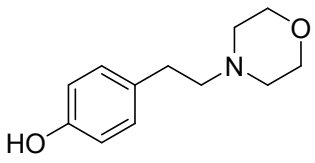
4-(4-Bromophenethyl)morpholine (3gc)



The typical procedure was applied to 2-(4-bromophenyl)acetaldehyde (39.8 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 10 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a white solid (29.6 mg, 59%); R_f = 0.2 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.39 (d, J = 7.8 Hz, 2H), 7.07 (d, J = 7.9 Hz, 2H), 3.72 (bs, 4H), 2.76-2.72

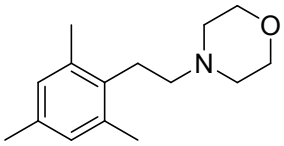
(m, 2H), 2.57-2.53 (m, 2H), 2.49 (bs, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 139.0, 131.4, 130.4, 119.8, 66.8, 60.3, 53.5, 32.5. The physical and spectral data were consistent with those previously reported.[19]

4-(2-Morpholinoethyl)phenol (3hc)



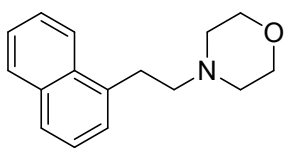
The typical procedure was applied to 2-(4-hydroxyphenyl)acetaldehyde (27.6 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 10 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 4/4/1) of the crude product afforded the title compound as a yellow solid (22.0 mg, 53%); R_f = 0.1 (PE/EtOAc/MeOH = 10/20/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.05 (d, J = 8.1 Hz, 2H), 6.74 (d, J = 8.3 Hz, 2H), 3.76 (t, J = 4.6 Hz, 4H), 2.78-2.73 (m, 2H), 2.61-2.56 (m, 6H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 154.3, 131.5, 129.7, 115.4, 66.7, 61.1, 53.6, 32.1. The physical and spectral data were consistent with those previously reported.[20]

4-(2,4,6-Trimethylphenethyl)morpholine (3ic)



The typical procedure was applied to 2-mesitylacetaldehyde (32.4 mg, 0.2 mmol) and morpholine (32.4 mg, 0.24 mmol) for 10 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (28.4 mg, 61%); R_f = 0.3 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 6.85 (s, 2H), 3.78 (t, J = 4.7 Hz, 4H), 2.85-2.79 (m, 2H), 2.58 (t, J = 4.7 Hz, 4H), 2.44-2.40 (m, 2H), 2.32 (s, 6H), 2.25 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 136.2, 135.4, 133.1, 128.9, 66.9, 57.8, 53.6, 26.3, 20.7, 19.7. HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{15}H_{24}NO$: 234.1852, found 234.1854.

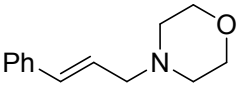
4-(2-(Naphthalen-1-yl)ethyl)morpholine (3jc)



The typical procedure was applied to 2-(naphthalen-1-yl)acetaldehyde (34.1 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 10 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (30.6 mg, 63%); R_f = 0.3 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.06 (d, J = 8.3 Hz, 1H),

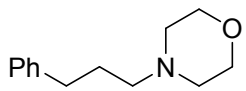
7.87 (d, J = 8.0 Hz, 1H), 7.74 (d, J = 8.0 Hz, 1H), 7.51 (td, J = 14.8, 6.8 Hz, 2H), 7.43-7.36 (m, 2H), 3.80 (t, J = 4.7 Hz, 4H), 3.32-3.28 (m, 2H), 2.75-2.71 (m, 2H), 2.61 (t, J = 4.6 Hz, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 136.1, 133.8, 131.8, 128.8, 126.9, 126.5, 125.9, 125.52, 125.47, 123.5, 66.9, 59.9, 53.7, 30.3. HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{16}H_{20}NO$: 242.1539, found 242.1542.

(*E*)-4-Cinnamylmorpholine (3kc)



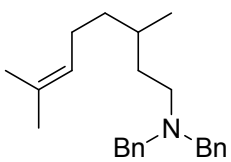
The typical procedure was applied to cinnamaldehyde (26.4 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 6 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (23.1 mg, 53 %); R_f = 0.2 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.44-7.41 (m, 2H), 7.39-7.32 (m, 2H), 7.31-7.27 (m, 1H), 6.46 (dd, J = 15.9, 4.9 Hz, 1H), 6.35-6.27 (m, 1H), 3.79 (t, J = 5.1 Hz, 4H), 3.20 (d, J = 6.0 Hz, 2H), 2.55 (bs, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 136.7, 133.5, 128.5, 127.5, 126.3, 125.8, 66.9, 61.4, 53.6. The physical and spectral data were consistent with those previously reported.[21]

4-(3-Phenylpropyl)morpholine (3lc)



The typical procedure was applied to 3-phenylpropanal (26.8 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 6 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (24.3 mg, 59%); R_f = 0.2 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.36-7.32 (m, 2H), 7.26-7.23 (m, 3H), 3.79-3.77 (m, 4H), 2.73-2.69 (m, 2H), 2.50 (t, J = 4.7 Hz, 4H), 2.45-2.41 (m, 2H), 1.89 (p, J = 7.7 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 142.0, 128.34, 128.28, 125.8, 66.9, 58.3, 53.6, 33.6, 28.2. The physical and spectral data were consistent with those previously reported.[22]

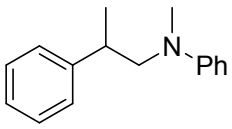
***N,N*-Dibenzyl-3,7-dimethyloct-6-en-1-amine (3mh)**



The typical procedure was applied to 3,7-dimethyloct-6-enal (30.8 mg, 0.2 mmol) and dibenzylamine (47.3 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 200/1) of the crude product afforded the title compound as a white oil (61.4 mg, 91%); R_f = 0.8 (PE/EtOAc = 10/1); ^{1}H NMR (400

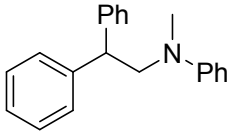
MHz, $CDCl_3$) δ 7.29-7.27 (m, 4H), 7.23-7.19 (m, 4H), 7.15-7.11 (m, 2H), 4.97 (t, J = 7.0 Hz, 1H), 3.52-3.40 (m, 4H), 2.34 (t, J = 7.3 Hz, 2H), 1.93-1.76 (m, 2H), 1.59 (s, 3H), 1.49 (s, 3H), 1.47-1.35 (m, 2H), 1.26-1.11 (m, 2H), 1.04-0.95 (m, 1H), 0.67 (d, J = 6.5 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 139.9, 130.9, 128.8, 128.1, 126.7, 124.9, 58.3, 51.3, 37.1, 34.0, 30.4, 25.7, 25.5, 19.6, 17.6. The physical and spectral data were consistent with those previously reported.[23]

_N_-Methyl-_N_-(2-phenylpropyl)aniline (3ns)



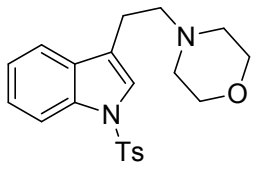
The typical procedure was applied to 2-phenylpropanal (26.8 mg, 0.2 mmol) and *N*-methylaniline (25.7 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 50/1) of the crude product afforded the title compound as a yellow oil (33.7 mg, 75%); R_f = 0.8 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.23-7.19 (m, 2H), 7.17-7.11 (m, 5H), 6.62-6.57 (m, 3H), 3.41 (dd, J = 14.6, 7.6 Hz, 1H), 3.30 (dd, J = 14.6, 7.1 Hz, 1H), 3.16-3.07 (m, 1H), 2.66 (s, 3H), 1.22 (d, J = 7.0 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 148.9, 145.1, 129.1, 128.4, 127.2, 126.3, 115.7, 111.7, 60.9, 39.5, 38.2, 18.8. The physical and spectral data were consistent with those previously reported.[24]

_N_-(2,2-Diphenylethyl)-_N_-methylaniline (3os)



The typical procedure was applied to 2,2-diphenylacetaldehyde (39.2 mg, 0.2 mmol) and *N*-methylaniline (25.7 mg, 0.24 mmol) for 6 h. Silica gel chromatography (eluent: PE/EtOAc = 200/1) of the crude product afforded the title compound as a yellow oil (36.8 mg, 64%); R_f = 0.7 (PE/EtOAc = 20/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.25-7.11 (m, 12H), 6.62 (t, J = 7.2 Hz, 1H), 6.56 (d, J = 8.2 Hz, 2H), 4.31 (t, J = 7.4 Hz, 1H), 3.92 (d, J = 7.4 Hz, 2H), 2.48 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 148.6, 143.1, 129.2, 128.5, 128.2, 126.5, 115.9, 111.9, 58.6, 48.6, 39.5. HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{21}H_{22}N$: 288.1747, found 288.1752.

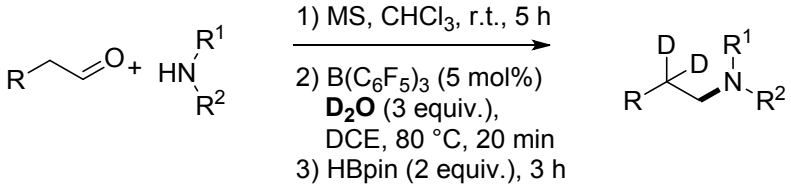
4-(2-(1-Tosyl-1*H*-indol-3-yl)ethyl)morpholine (3pc)



The typical procedure was applied to 2-(1-tosyl-1*H*-indol-3-yl)acetaldehyde (62.7 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24

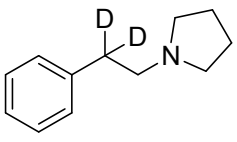
mmol) for 10 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (58.6 mg, 76%); R_f = 0.3 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.02 (d, J = 8.3 Hz, 1H), 7.79 (d, J = 7.9 Hz, 2H), 7.53 (d, J = 7.8 Hz, 1H), 7.44 (s, 1H), 7.36 (t, J = 7.7 Hz, 1H), 7.31-7.30 (m, 1H), 7.28-7.24 (m, 2H), 3.79 (t, J = 4.7 Hz, 4H), 2.92-2.89 (m, 2H), 2.74-2.70 (m, 2H), 2.58 (t, J = 4.6 Hz, 4H), 2.38 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 144.7, 135.3, 135.1, 130.9, 129.8, 126.7, 124.6, 123.0, 120.7, 119.3, 113.7, 66.9, 58.1, 53.6, 22.3, 21.5. HRMS (ESI) m/z [M + H]$^+$ calcd for $C_{21}H_{25}N_2O_3S$: 385.1580, found 385.1578.

3.3 General Procedure for *β*-Deuteration of Amines.



In a Schlenk tube were placed secondary amine (0.24 mmol), arylacetaldehyde (0.2 mmol), MS 4Å (135 mg) and CH_3Cl (0.4 mL). The resulting mixture was stirred at room temperature for 5 h (0 ºC, 0.5 h for morpholine), and then filtered and the solvent was concentrated under reduced pressure. The correspond enamine was directly used without further purification. In the glove box, to a Schlenk tube were placed the crude enamine, D_2O (12.0 mg, 0.6 mmol), $B(C_6F_5)_3$ (5.1 mg, 0.01 mmol), and DCE (0.8 mL). The resulting mixture was stirred at 80 ºC for 20 min, and then allowed to room temperature. HBpin (51.2 mg, 0.4 mmol, 2.0 equiv.) was added under N_2. The resulting mixture was stirred at 80 ºC for 3 h, and then allowed to room temperature. The solution was concentrated in vacuo before purifying by column chromatography. The deuterium incorporation is detected from ^{1}H NMR.

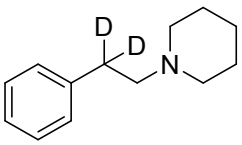
1-(2-Phenylethyl-2,2-*d*$_2$)pyrrolidine (3aa-*d*)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and pyrrolidine (17.1 mg, 0.24 mmol) for 3 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a colorless oil (29.4 mg, 84%, 173%D); R_f = 0.3

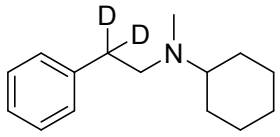
(PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.30-7.26 (m, 2H), 7.22-7.17 (m, 3H), 2.84-2.80 (m, 0.28H), 2.70 (d, J = 6.7 Hz, 2H), 2.60-2.57 (m, 4H), 1.85-1.77 (m, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 140.3, 128.6, 128.3, 126.0, 58.2 (m), 54.1, 35.1 (m, labeled), 23.4. HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{12}H_{16}D_2N$: 178.1559, found 178.1564.

1-(2-Phenylethyl-2,2-*d*$_2$)piperidine (3ab-*d*)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and piperidine (20.4 mg, 0.24 mmol) for 3 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (37.9 mg, 72%, 169%D); R_f = 0.3 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.36-7.32 (m, 2H), 7.27-7.23 (m, 3H), 2.86 (m, 0.30H), 2.63 (d, J = 7.0 Hz, 2H), 2.54 (s, 4H), 1.71-1.66 (m, 4H), 1.53 (q, J = 6.0 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 140.4, 128.6, 128.3, 125.9, 61.1 (m), 54.4, 32.9 (m, labeled), 25.8, 24.3. HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{13}H_{18}D_2NO$: 192.1716, found 192.1714.

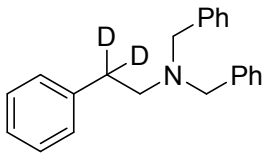
***N*-Methyl-*N*-(2-phenylethyl-2,2-*d*$_2$)cyclohexanamine (3ag-*d*)**



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and *N*-methylcyclohexanamine (27.2 mg, 0.24 mmol) for 3 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (33.9 mg, 78%, 140%D); R_f= 0.3 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.22-7.17 (m, 2H), 7.13-7.09 (m, 3H), 2.70-2.64 (m, 0.60H), 2.62 (d, J = 5.7 Hz, 2H), 2.40-2.34 (m, 1H), 2.36 (s, 3H), 1.79-1.67 (m, 4H), 1.57-1.52 (m, 1H), 1.22-1.12 (m, 5H).; ^{13}C NMR (100 MHz, $CDCl_3$) δ 140.5, 128.6, 128.3, 125.9, 62.6, 55.7 (m), 37.7, 28.5, 26.3, 25.9. (Labeled C was not detected due to low scan times). HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{15}H_{22}D_2N$: 220.2029, found 220.2029.

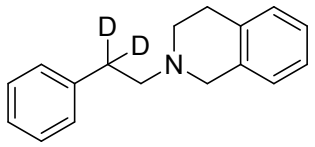
***N*,*N*-Dibenzyl-2-phenylethan-1-amine-2,2-*d*$_2$ (3ah-*d*)**

The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and dibenzylamine (47.3 mg, 0.24 mmol) for 3 h. Silica gel chromatography (eluent: PE/EtOAc =



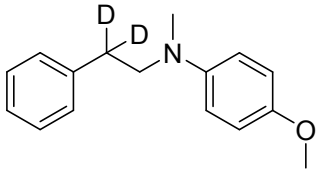
200/1) of the crude product afforded the title compound as a yellow oil (35.9 mg, 60%, 173%D); R_f = 0.7 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.25-7.06 (m, 13H), 6.99 (d, J = 7.3 Hz, 2H) 3.56 (s, 4H), 2.73-2.69 (m, 0.27H), 2.62 (d, J = 5.5 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 140.5, 139.7, 128.8, 128.7, 128.2, 128.1, 126.8, 125.8, 58.2, 55.0 (m). (Labeled C was not detected due to low scan times). HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{22}H_{22}D_2N$: 304.2029, found 304.2032.

2-(2-Phenylethyl-2,2-*d*$_2$)-1,2,3,4-tetrahydroisoquinoline (3af-*d*)



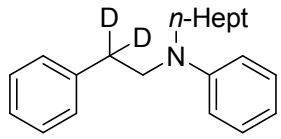
The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and 1,2,3,4-tetrahydroisoquinoline (32.0 mg, 0.24 mmol) for 3 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (28.0 mg, 59%, 142%D); R_f = 0.3 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.38 (t, J = 7.3 Hz, 2H), 7.33-7.27 (m, 3H), 7.21-7.20 (m, 3H), 7.13-7.12 (m, 1H), 3.81 (s, 2H), 3.04-2.97 (m, 2.58H), 2.90 (t, J = 6.0 Hz, 2H), 2.86 (d, J = 6.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 140.2, 134.5, 134.1, 128.7, 128.6, 128.4, 126.6, 126.2, 126.1, 125.6, 60.0 (m), 55.9, 50.9, 33.6 (m, labeled), 28.9. HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{17}H_{18}D_2N$: 240.1716, found 240.1711.

4-Methoxy-*N*-methyl-*N*-(2-phenylethyl-2,2-*d*$_2$)aniline (3aj-*d*)



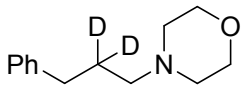
The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and 4-methoxy-*N*-methylaniline (32.9 mg, 0.24 mmol) for 3 h. Silica gel chromatography (eluent: PE/EtOAc = 80/1) of the crude product afforded the title compound as a yellow solid (33.5 mg, 69 %, 151%D); R_f = 0.5 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31-7.27 (m, 2H), 7.24-7.18 (m, 3H), 6.86 (d, J = 9.1 Hz, 2H), 6.73 (d, J = 9.1 Hz, 2H), 3.77 (s, 3H), 3.48 (d, J = 7.1 Hz, 2H), 2.85 (s, 3H), 2.81-2.77 (m, 0.49H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 151.6, 143.7, 139.8, 128.7, 128.5, 126.1, 114.8, 114.5, 55.81 (m), 55.76, 39.1, 32.4 (m, labeled). HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{16}H_{18}D_2NO$: 244.1665, found 244.1668.

***N*-Heptyl-*N*-(2-phenylethyl-2,2-d_2)aniline (3ap-*d*)**



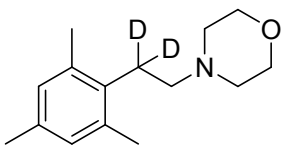
The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and *N*-heptylaniline (45.9 mg, 0.24 mmol) for 3 h. Silica gel chromatography (eluent: PE/EtOAc = 200/1) of the crude product afforded the title compound as a yellow oil (39.4 mg, 70%, 162%D); R_f = 0.8 (PE/EtOAc = 10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.35-7.31 (m, 2H), 7.28-7.22 (m, 5H), 6.74 (d, J = 8.2 Hz, 2H), 6.69 (t, J = 7.2 Hz, 1H), 3.53 (d, J = 6.8 Hz, 2H), 3.25-3.21 (m, 2H), 2.89-2.85 (m, 0.38H), 1.62-1.54 (m, 2H), 1.34-1.28 (m, 8H), 0.91 (t, J = 6.6 Hz, 3H).; ^{13}C NMR (100 MHz, $CDCl_3$) δ 147.7, 139.7, 129.3, 128.7, 128.5, 126.2, 115.4, 111.7, 52.8 (m), 51.2, 31.9, 29.2, 27.3, 27.1, 22.6, 14.1. (Labeled C was not detected due to low scan times). HRMS (ESI) *m*/*z* $[M + H]^+$ calcd for $C_{21}H_{28}D_2N$: 298.2498, found 298.2494.

4-(3-Phenylpropyl-2,2-d_2)morpholine (3lc-*d*)



The typical procedure was applied to 3-phenylpropanal (26.8 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 3 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (19.0 mg, 46%, 164%D); R_f = 0.4 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.30-7.26 (m, 2H), 7.19-7.15 (m, 3H), 3.72 (t, J = 4.7 Hz, 4H), 2.77-2.73 (m, 0.36H), 2.64 (d, J = 7.0 Hz, 2H), 2.44 (bs, 4H), 2.36 (d, J = 6.6 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 142.0, 128.35, 128.30, 125.8, 66.9, 58.2 (m), 53.6, 33.4 (m), 27.5 (m, labeled). HRMS (ESI) *m*/*z* $[M + H]^+$ calcd for $C_{13}H_{18}D_2NO$: 208.1665, found 208.1665.

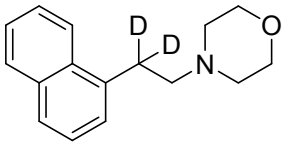
4-(2-Mesitylethyl-2,2-d_2)morpholine (3ic-*d*)



The typical procedure was applied to 2-mesitylacetaldehyde (32.4 mg, 0.2 mmol) and morpholine (32.4 mg, 0.24 mmol) for 3 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (26.1 mg, 56%, 170%D); R_f = 0.2 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 6.84 (s, 2H), 3.78 (t, J = 4.7 Hz, 4H), 2.83-2.79 (m, 0.30H), 2.57 (t, J = 4.7 Hz, 4H), 2.42 (d, J = 6.4 Hz, 2H), 2.31 (s, 6H), 2.25 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 136.2, 135.4, 133.1, 128.9, 66.9, 57.7

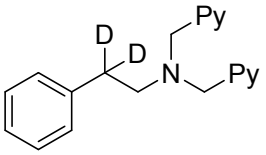
(m), 53.6, 20.7, 19.7 (labeled C was not detected due to low scan times). HRMS (ESI) *m*/*z* [M + H]$^+$ calcd for $C_{15}H_{22}D_2NO$: 236.1978, found 236.1980.

4-(2-(Naphthalen-1-yl)ethyl-2,2-*d*$_2$)morpholine (3jc-*d*)



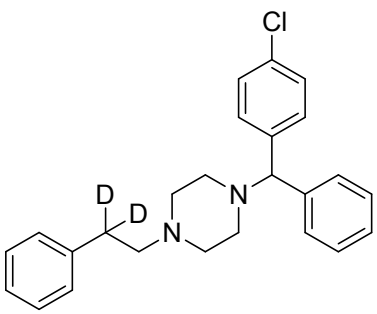
The typical procedure was applied to 2-(naphthalen-1-yl)acetaldehyde (34.1 mg, 0.2 mmol) and morpholine (20.9 mg, 0.24 mmol) for 3 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (36.2 mg, 75 %, 175%D); R_f = 0.4 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.06 (d, *J* = 8.2 Hz, 1H), 7.86 (d, *J* = 7.2 Hz, 1H), 7.73 (d, *J* = 8.1 Hz, 1H), 7.55-7.47 (m, 2H), 7.43-7.35 (m, 2H), 3.80 (t, *J* = 4.7 Hz, 4H), 3.31-3.25 (m, 0.25H), 2.72 (d, *J* = 6.9 Hz, 2H), 2.61 (t, *J* = 4.6 Hz, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 136.1, 133.8, 131.8, 128.8, 126.9, 126.5, 125.9, 125.6, 125.5, 123.6, 67.0, 59.9 (m), 53.7. (labeled C was not detected due to low scan times). HRMS (ESI) *m*/*z* [M + H]$^+$ calcd for $C_{16}H_{18}D_2NO$: 244.1665, found 244.1670.

2-Phenyl-*N*,*N*-bis(pyridin-2-ylmethyl)ethan-1-amine-2,2-*d*$_2$ (3ai-*d*)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and bis(pyridin-2-ylmethyl)amine (47.8 mg, 0.24 mmol) for 3 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (36.5 mg, 60%, 171%D); R_f = 0.2 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.53 (d, *J* = 4.9 Hz, 2H), 7.61 (t, *J* = 7.7 Hz, 2H), 7.40 (d, *J* = 7.9 Hz, 2H), 7.29-7.25 (m, 2H), 7.22-7.20 (m, 1H), 7.17-7.11 (m, 4H), 3.91 (s, 4H), 2.84-2.83 (m, 2.29H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 159.5, 148.7, 140.2, 136.4, 128.8, 128.1, 125.8, 122.7, 121.8, 60.1, 55.8 (m). (labeled C was not detected due to low scan times). HRMS (ESI) *m*/*z* [M + H]$^+$ calcd for $C_{20}H_{22}N_3$: 306.1934, found 306.1927.

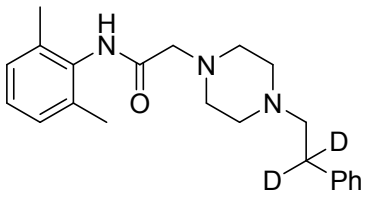
1-((4-Chlorophenyl)(phenyl)methyl)-4-(2-phenylethyl-2,2-*d*$_2$)piperazine (3at-*d*)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and 1-((4-chlorophenyl)(phenyl)methyl)piperazine (68.8 mg, 0.24 mmol)

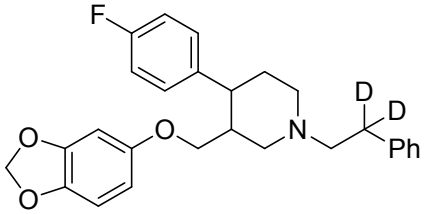
for 3 h. Silica gel chromatography (eluent: PE/EtOAc = 10/1) of the crude product afforded the title compound as a yellow oil (42.6 mg, 54%, 142%D); R_f = 0.7 (PE/EtOAc = 1/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.39-7.36 (m, 4H), 7.30-7.24 (m, 6H), 7.21-7.18 (m, 4H), 4.22 (s, 1H), 2.80-2.76 (m, 0.54H), 2.62-2.45 (m, 10H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 142.1, 141.3, 140.1, 132.5, 129.1, 128.63 (two carbons are overlapped), 128.57, 128.4, 127.8, 127.1, 126.1, 75.4, 60.2 (m), 53.3, 51.6, 33.0 (m, labeled). HRMS (ESI) *m*/*z* [M + H]$^+$ calcd for $C_{25}H_{26}D_2ClN_2$: 393.2061, found 393.2063.

_N_-(2,6-Dimethylphenyl)-2-(4-(2-phenylethyl-2,2-d_2)piperazin-1-yl)acetamide (3au-*d*)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and *N*-(2,6-dimethylphenyl)-2-(piperazin-1-yl)acetamide (59.4 mg, 0.24 mmol) for 3 h. Basic aluminum oxide chromatography (eluent: PE/EtOAc/MeOH = 100/10/1) of the crude product afforded the title compound as a yellow oil (40.4 mg, 57%, 163%D); R_f = 0.4 (PE/EtOAc/MeOH = 40/10/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.68 (bs, 1H), 7.32-7.28 (m, 2H), 7.22-7.20 (m, 3H), 7.11-7.07 (m, 3H), 3.23 (s, 2H), 2.83-2.77 (m, 4.37H), 2.65-2.62 (m, 6H), 2.24 (s, 6H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 168.5, 139.9, 134.9, 133.6, 128.6, 128.4, 128.3, 127.2, 126.1, 61.6, 60.2 (m), 53.7, 53.3, 18.6. (labeled C was not detected due to low scan times). HRMS (ESI) *m*/*z* [M + H]$^+$ calcd for $C_{27}H_{27}D_2FNO_3$: 436.2252, found 436.2248.

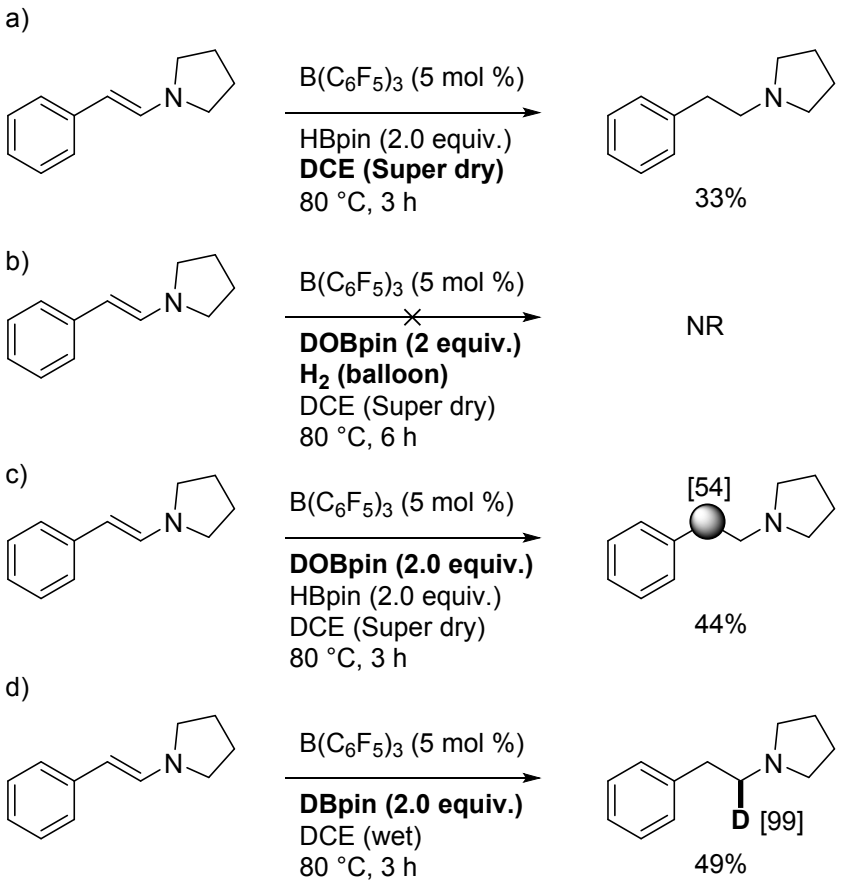
3-((Benzo[*d*][1,3]dioxol-5-yloxy)methyl)-4-(4-fluorophenyl)-1-(2-phenylethyl-2,2-d_2)piperidine (3av-*d*)



The typical procedure was applied to 2-phenylacetaldehyde (24.0 mg, 0.2 mmol) and 3-((benzo[*d*][1,3]dioxol-5-yloxy)methyl)-4-(4-fluorophenyl)piperidine (87.8 mg, 0.24 mmol) for 3 h. Silica gel chromatography (eluent: PE/EtOAc = 10/1) of the crude product afforded the title compound as a yellow oil (60.1 mg, 69%, 158%D); R_f = 0.6 (PE/EtOAc = 1/1); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.39 (t, *J* = 7.4 Hz, 2H), 7.34-7.31 (m, 3H), 7.26 (t, *J* = 6.8 Hz, 2H), 7.06 (t, *J* = 8.4 Hz, 2H), 6.71 (d, *J* = 8.4 Hz, 1H), 6.44 (d, *J* = 2.7 Hz, 1H), 6.22 (dd, *J* = 8.4, 2.6 Hz, 1H), 5.96 (s, 2H), 3.67 (dd, *J* = 9.3, 2.9 Hz,

1H), 3.54 (t, J = 8.1 Hz, 1H), 3.46 (d, J = 11.1, 1H), 3.24 (d, J = 11.2 Hz, 1H), 2.96-2.93 (m, 0.42H), 2.77 (s, 2H), 2.59 (td, J = 11.4, 4.5 Hz, 1H), 2.37-2.31 (m, 1H), 2.28-2.19 (m, 2H), 2.06-1.92 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 161.5 (d, $^1J_{C\text{-}F}$ = 244.5 Hz), 154.3, 148.1, 141.5, 140.1, 139.5 (d, $^4J_{C\text{-}F}$ = 3.2 Hz), 128.8 (d, $^3J_{C\text{-}F}$ = 7.7 Hz), 128.7 (two carbons are overlapped), 128.4 (two carbons are overlapped), 126.1, 115.4 (d, $^2J_{C\text{-}F}$ = 21.1 Hz), 107.8, 105.5, 101.0, 97.9, 69.5, 60.7 (m), 57.4, 54.0, 44.0, 42.0, 34.1.(labeled C was not detected due to low scan times). HRMS (ESI) m/z $[M + H]^+$ calcd for $C_{22}H_{28}D_2N_3O$: 354.2509, found 354.2507.

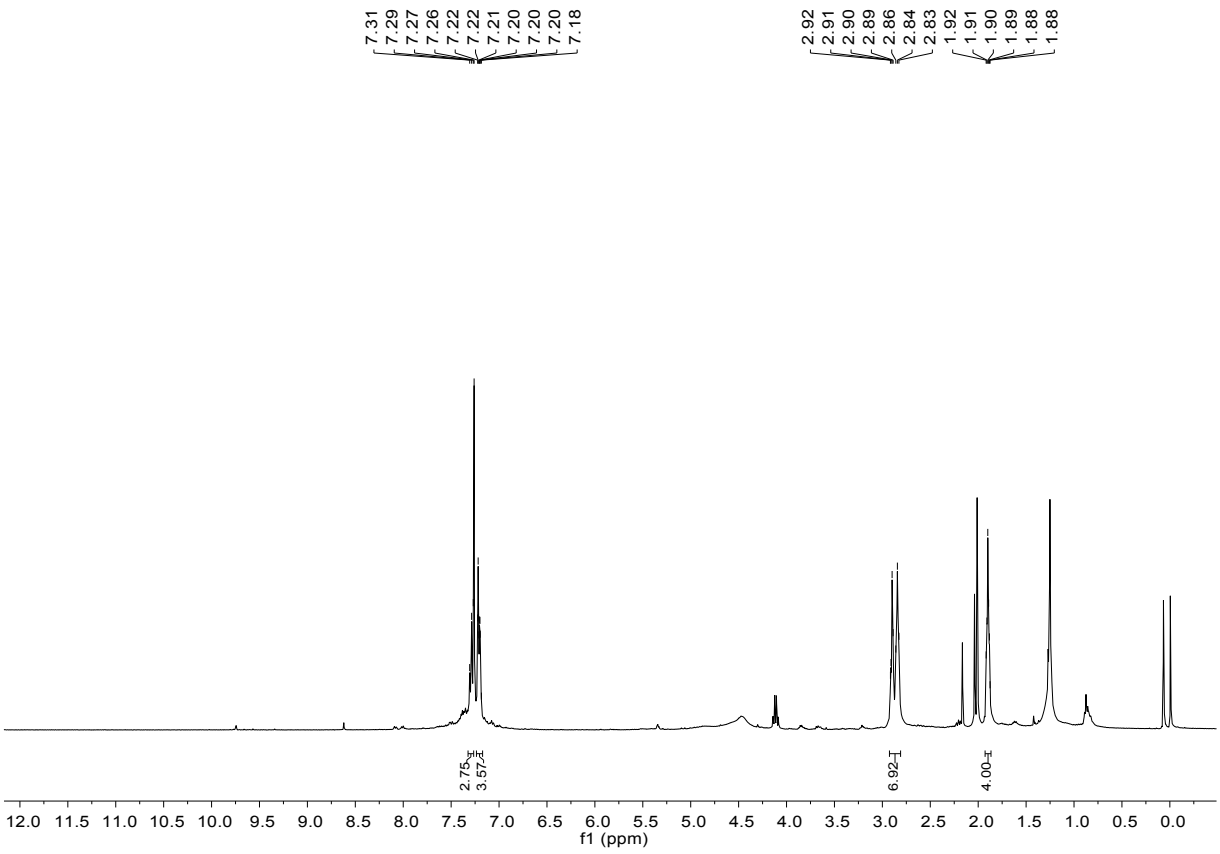
3.4 General Procedure for Control Experiments.



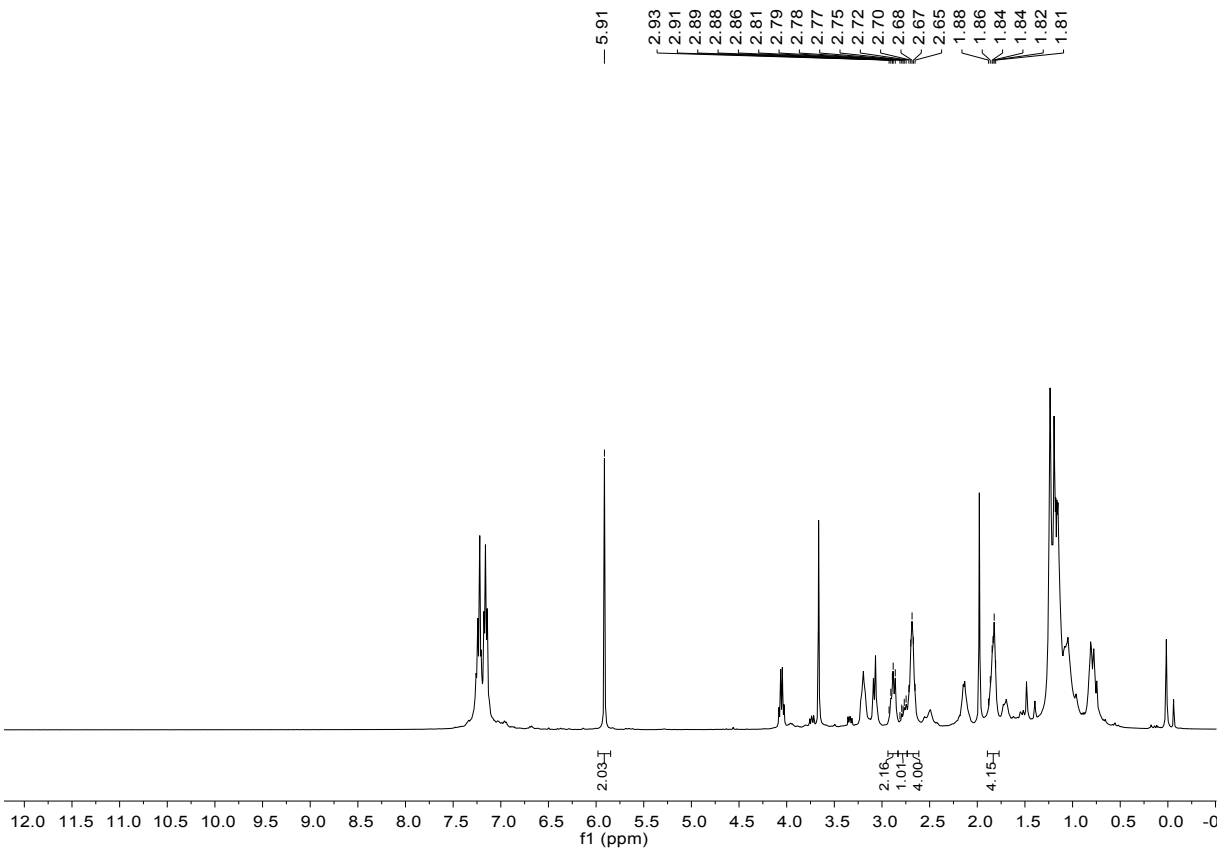
a) The typical procedure of synthesis of amine was applied to (*E*)-1-styrylpyrrolidine (35.1 mg, 0.2 mmol), $B(C_6F_5)_3$ (5.1 mg, 0.01 mol), HBpin (51.2 mg, 0.4 mmol, 2.0 equiv.) and super dry DCE (0.8 mL). The crude yield was detected from ^{1}H NMR using 1,1,2,2-tetrachloroethane as an internal standard. (NMR yield: 33%).

b) In the glove box, in an over-dried Schlenk tube were placed D_2O (8.0 mg, 0.4 mmol, 2.0 equiv.), and HBpin (51.2 mg, 0.4 mmol, 2.0 equiv.) and super dry DCE (0.8 mL) to generated DOBpin *in situ*. The resulting mixture was stirred at room temperature for 2 h, and then the purified (*E*)-1-styrylpyrrolidine (35.1 mg, 0.2 mmol), $B(C_6F_5)_3$ (5.1 mg, 0.01 mol) were added. The resulting mixture was stirred with a H_2 balloon at 80 ºC for 3 h, and then allowed to room temperature. The solution was concentrated in *vacuo.* The crude yield and deuterium incorporation were detected from ^1H NMR using 1,1,2,2-tetrachloroethane as an internal standard. (NMR yield: 0%).

c) In the glove box, in an over-dried Schlenk tube were placed D_2O (8.0 mg, 0.4 mmol, 2.0 equiv.), and HBpin (51.2 mg, 0.4 mmol, 2.0 equiv.) and super dry DCE (0.8 mL) to generated DOBpin *in situ*. The resulting mixture was stirred at room temperature for 2 h, and then the purified (*E*)-1-styrylpyrrolidine (35.1 mg, 0.2 mmol), $B(C_6F_5)_3$ (5.1 mg, 0.01 mol) and HBpin (51.2 mg, 0.4 mmol, 2.0 equiv.) were added. The resulting mixture was stirred at 80 ºC for 3 h, and then allowed to room temperature. The solution was concentrated in *vacuo* before purifying by column chromatography. Silica gel chromatography (eluent: PE/EtOAc/MeOH = 40/10/1) of the crude product afforded the title compound as a yellow oil (15.4 mg, 44%, 108%D).



d) In the glove box, in an over-dried Schlenk tube were placed $NaBD_4$ (0.0502 g, 1.2 mmol, 3.0 equiv.), pinacol (0.0473 g, 0.4 mmol, 2.0 equiv.) and super dry DCE (0.4 mL). A solution of iodine (0.1600 g, 0.63 mmol, 1.6 equiv.) in super dry DCE (0.4 mL) was added dropwise. The resulting mixture was stirred at room temperature for 2 h. Then the purified (*E*)-1-styrylpyrrolidine (35.1 mg, 0.2 mmol), $B(C_6F_5)_3$ (5.1 mg, 0.01 mol), H_2O (7.2 mg, 0.4 mmol, 2.0 equiv.) were added. The resulting mixture was stirred at 80 ºC for 3 h, and then allowed to room temperature. The solution was concentrated in *vacuo*. The crude yield and deuterium incorporation were detected from ^1H NMR using 1,1,2,2-tetrachloroethane as an internal standard. (NMR yield: 49%, 99%D).


5.91
2.93
2.91
2.89
2.88
2.86
2.81
2.79
2.78
2.77
2.75
2.72
2.70
2.68
2.67
2.65
1.88
1.86
1.84
1.84
1.82
1.81
2.03
2.16
1.01
4.00
4.15
12.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

Reference:

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^{1}H and ^{13}C NMR Spectra:

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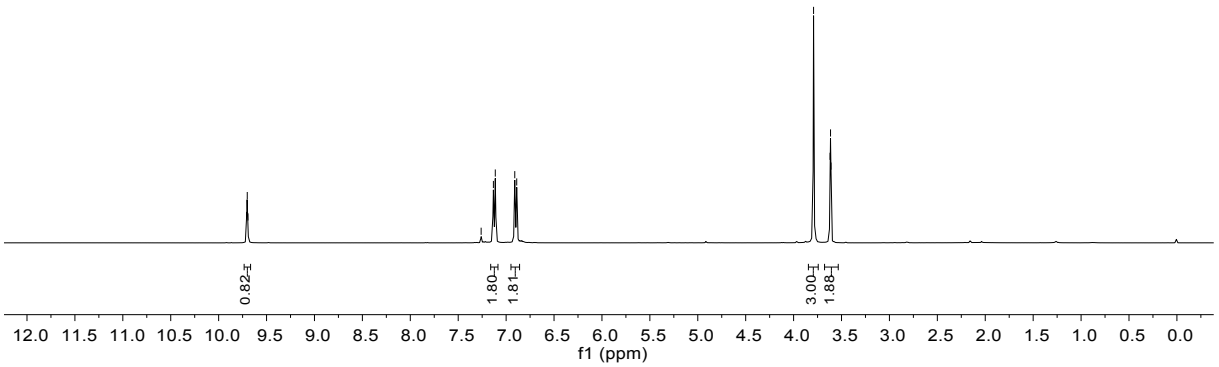

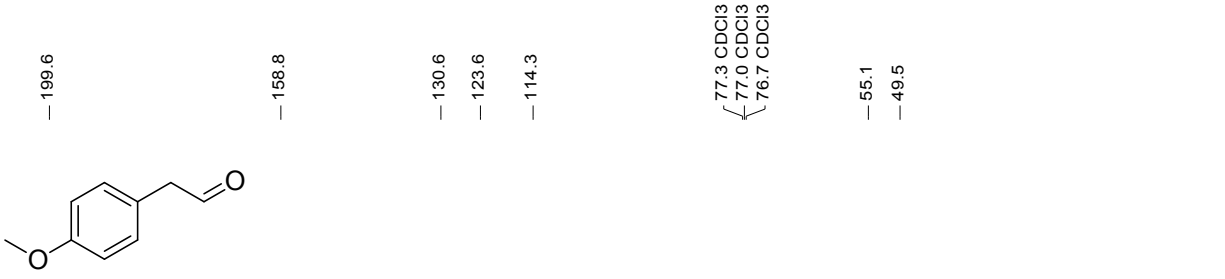

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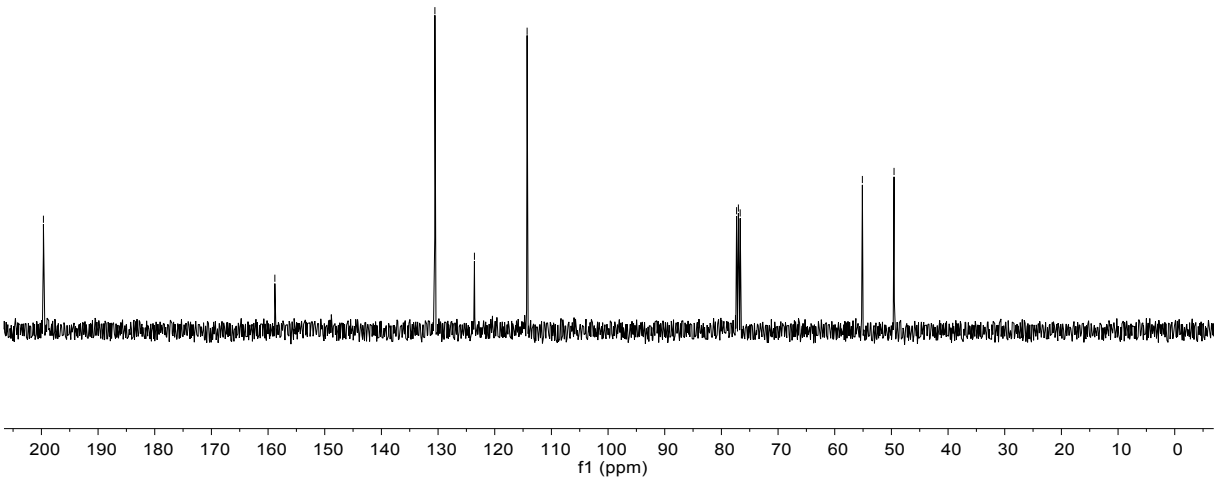

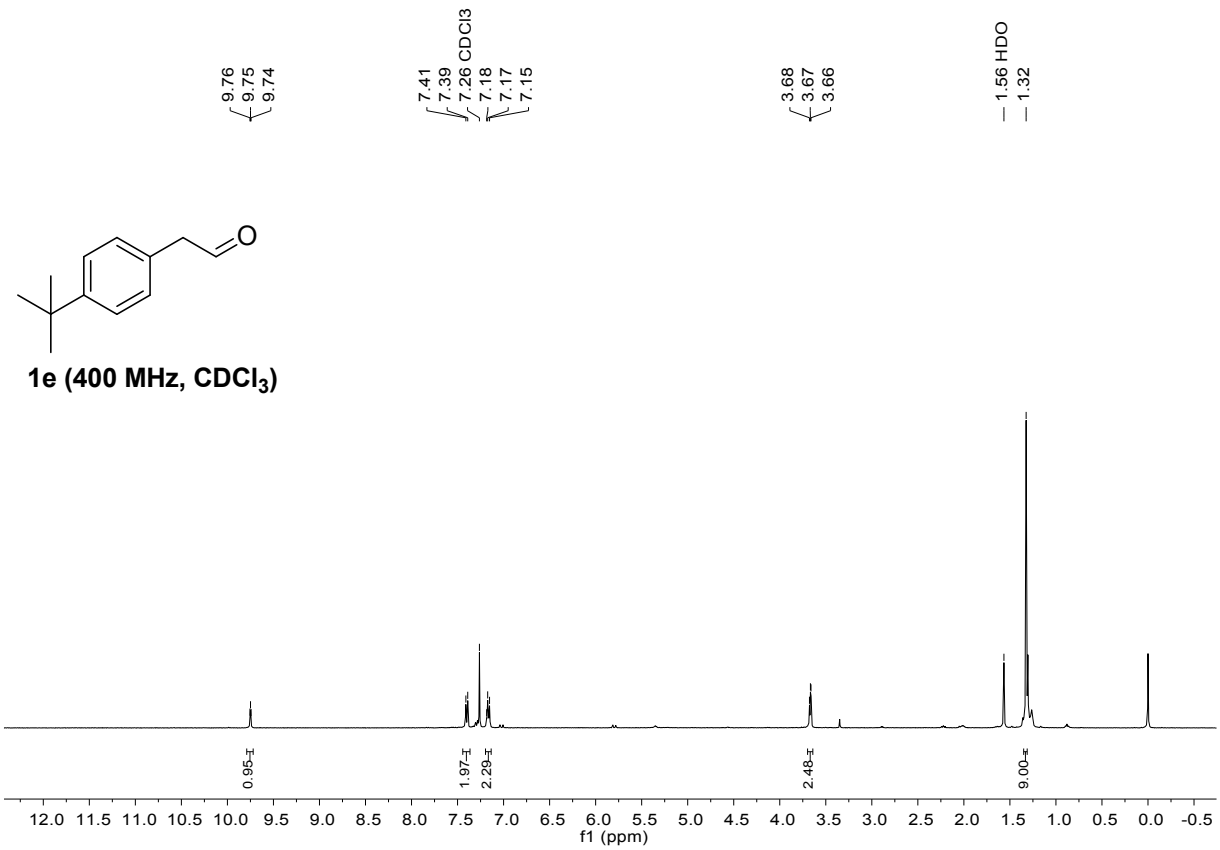

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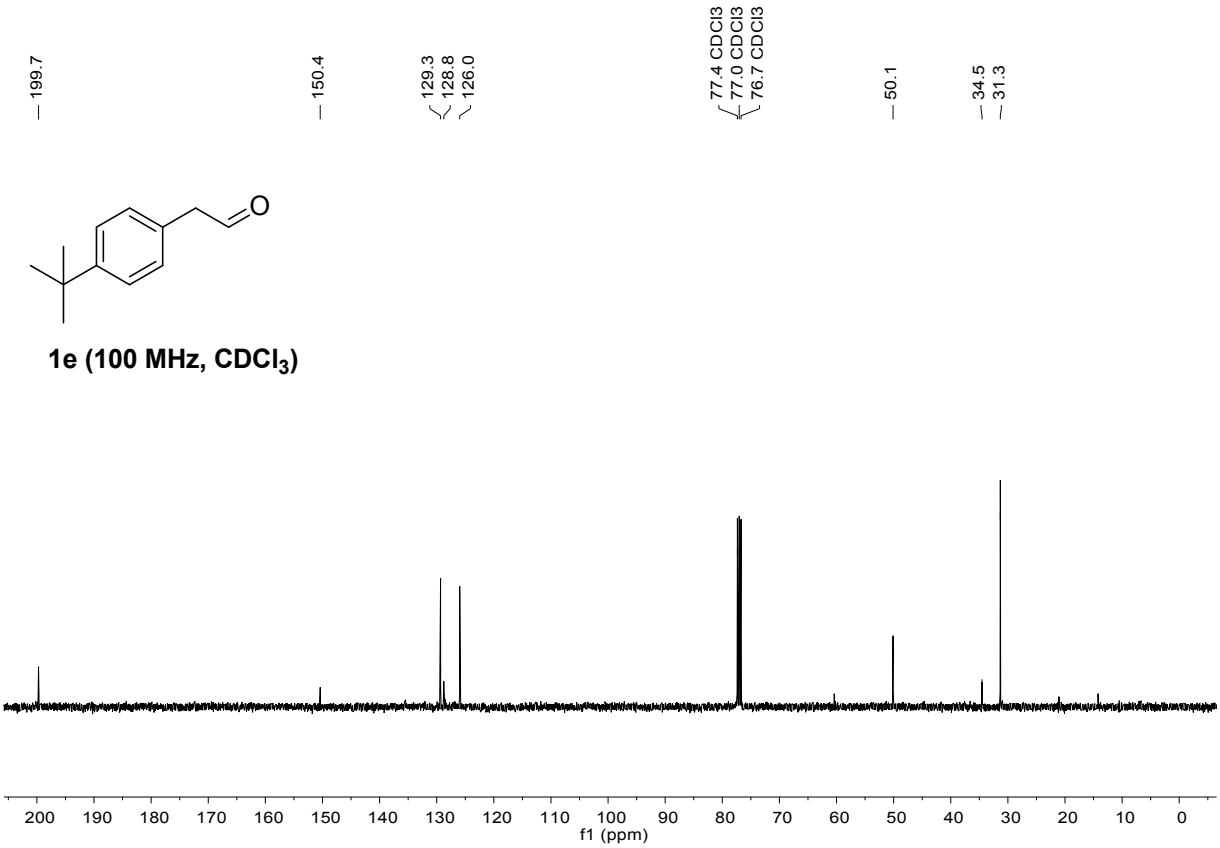

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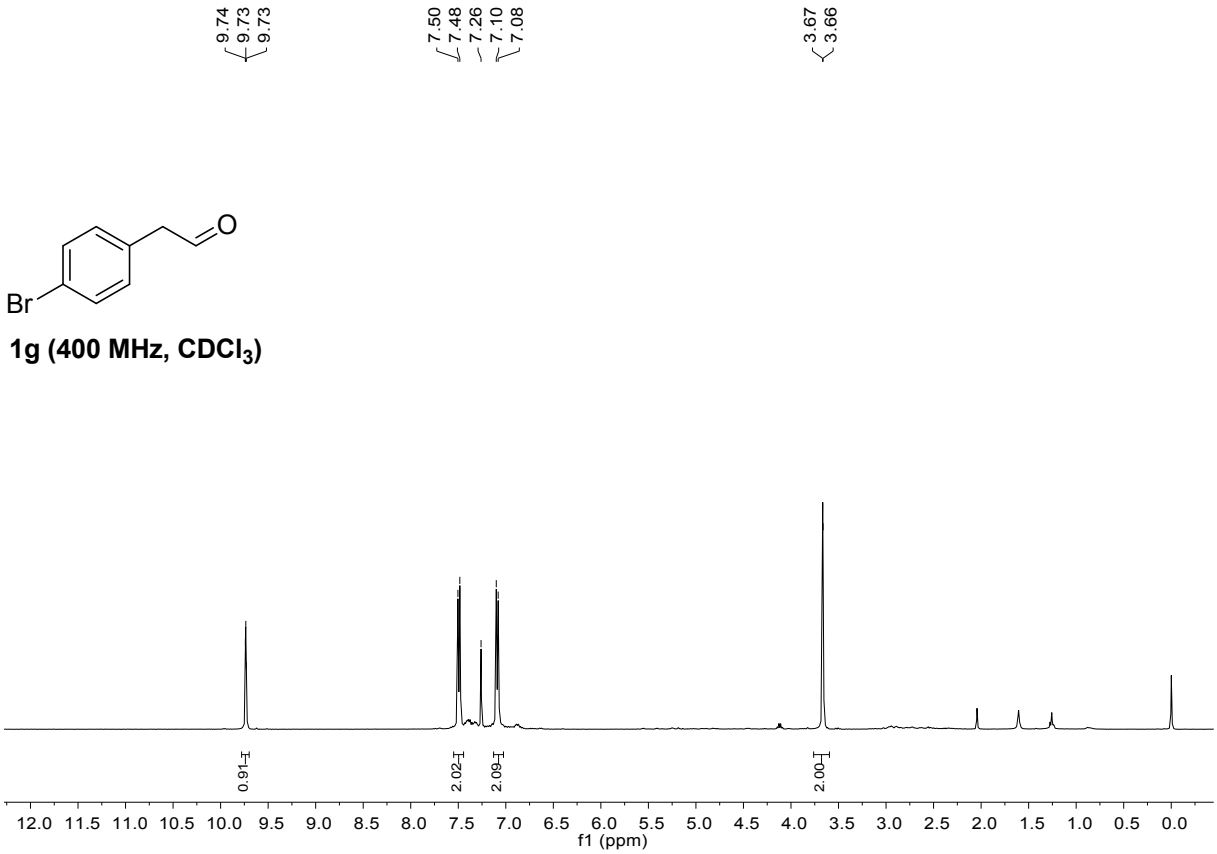

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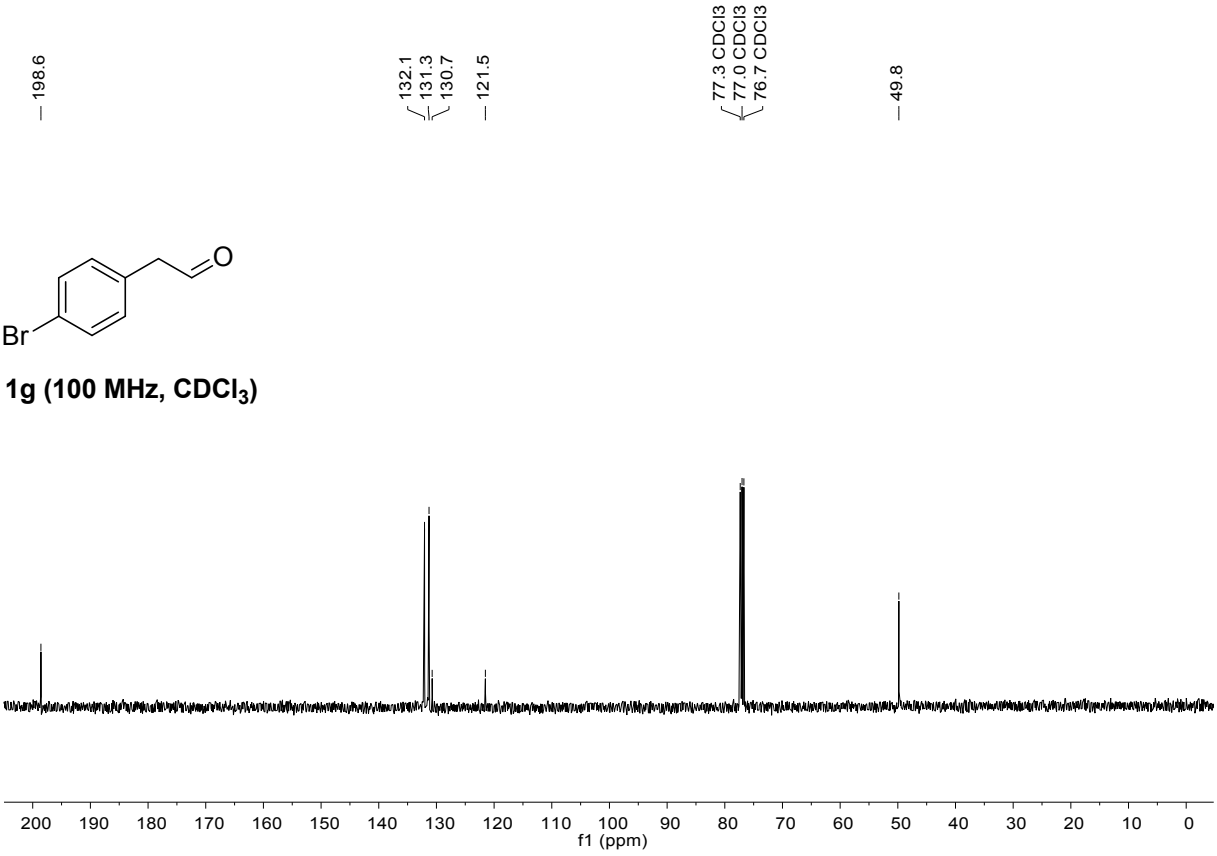

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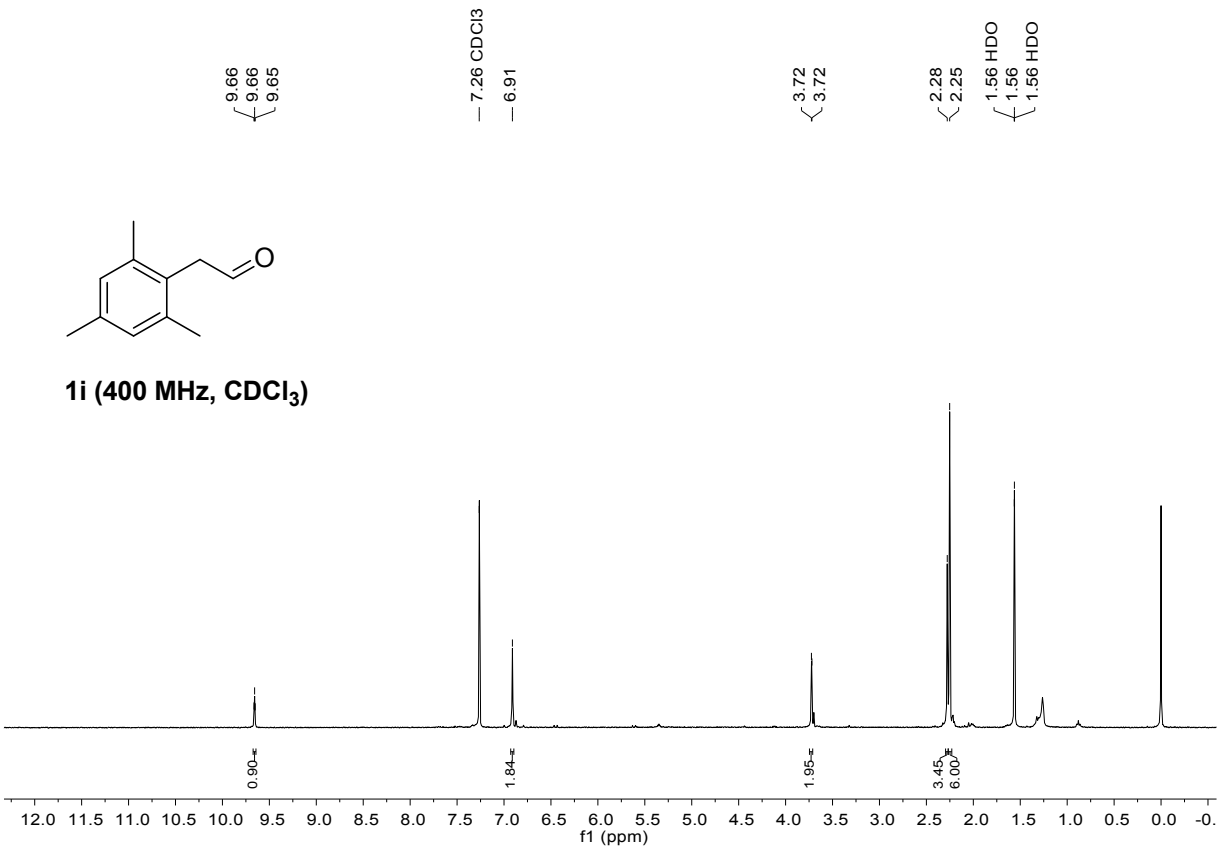
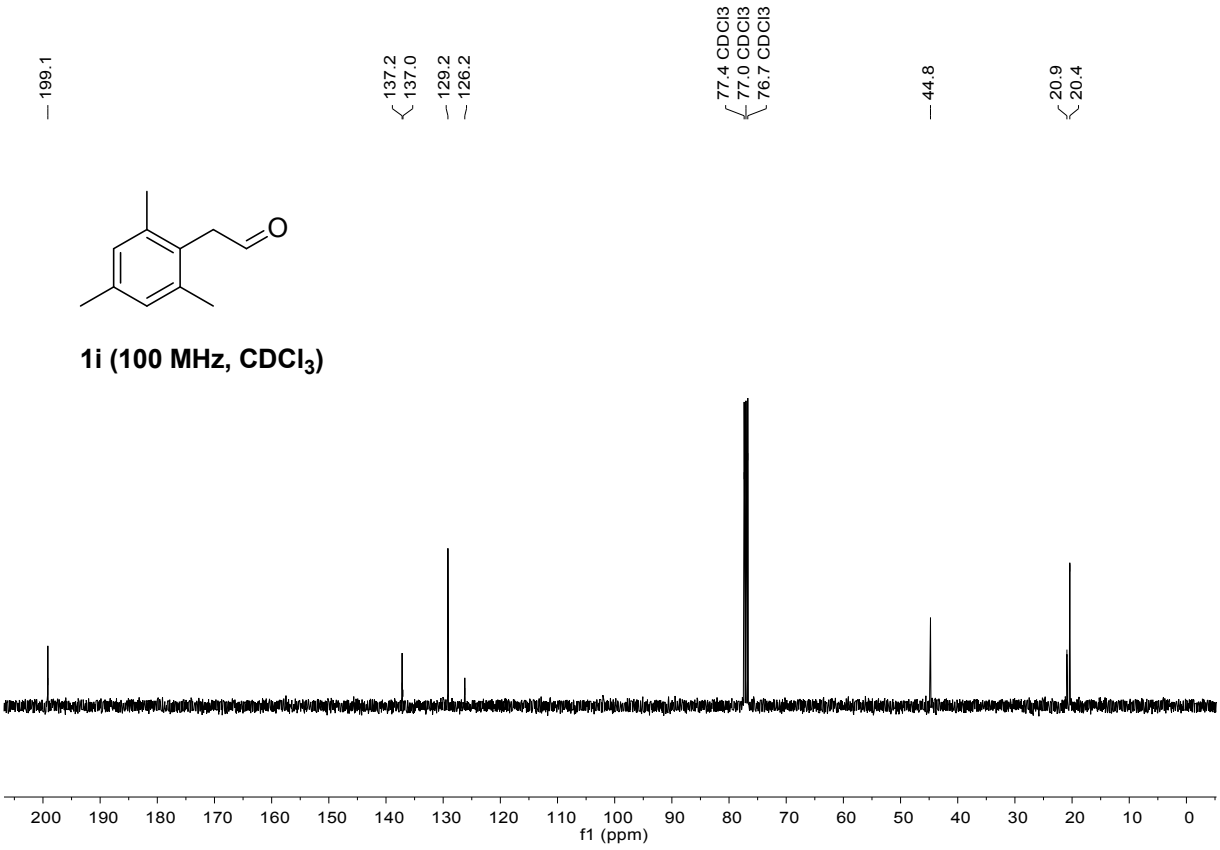


1i (400 MHz, $CDCl_3$)



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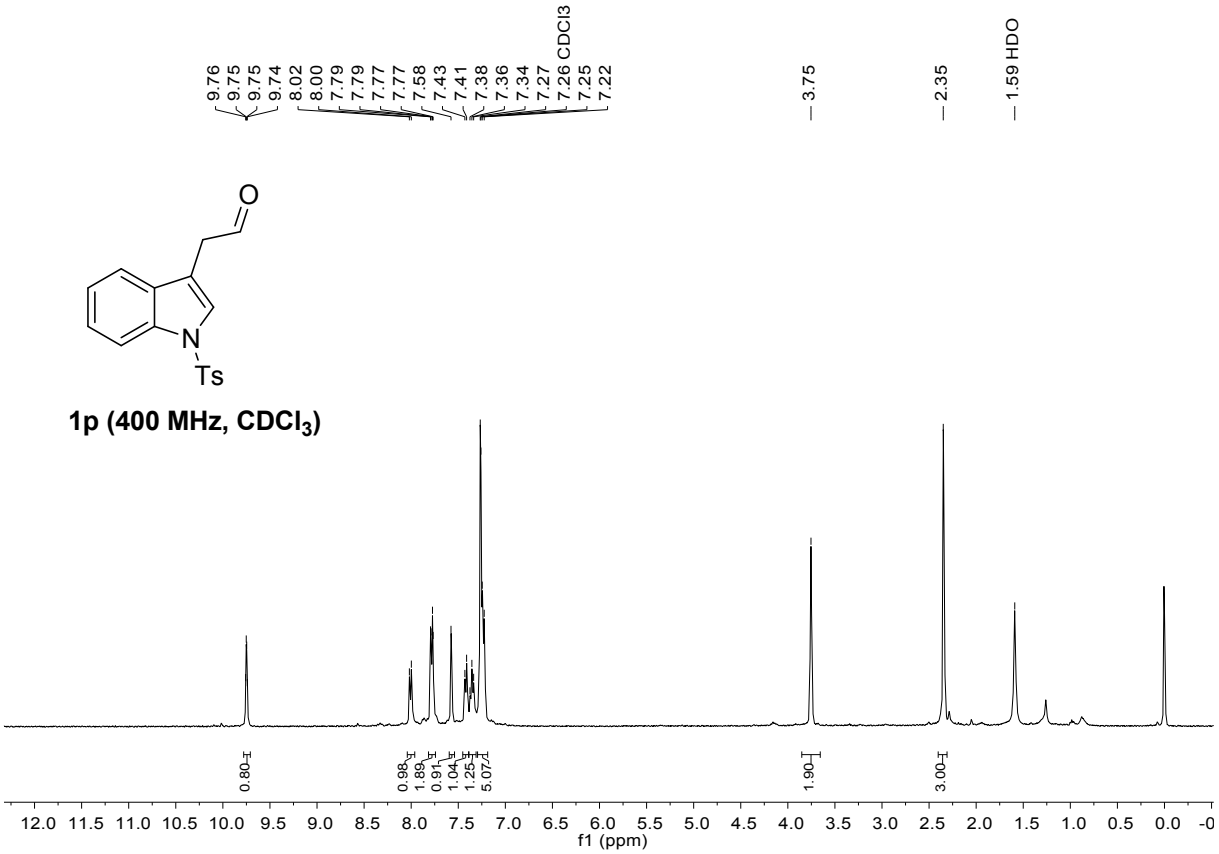
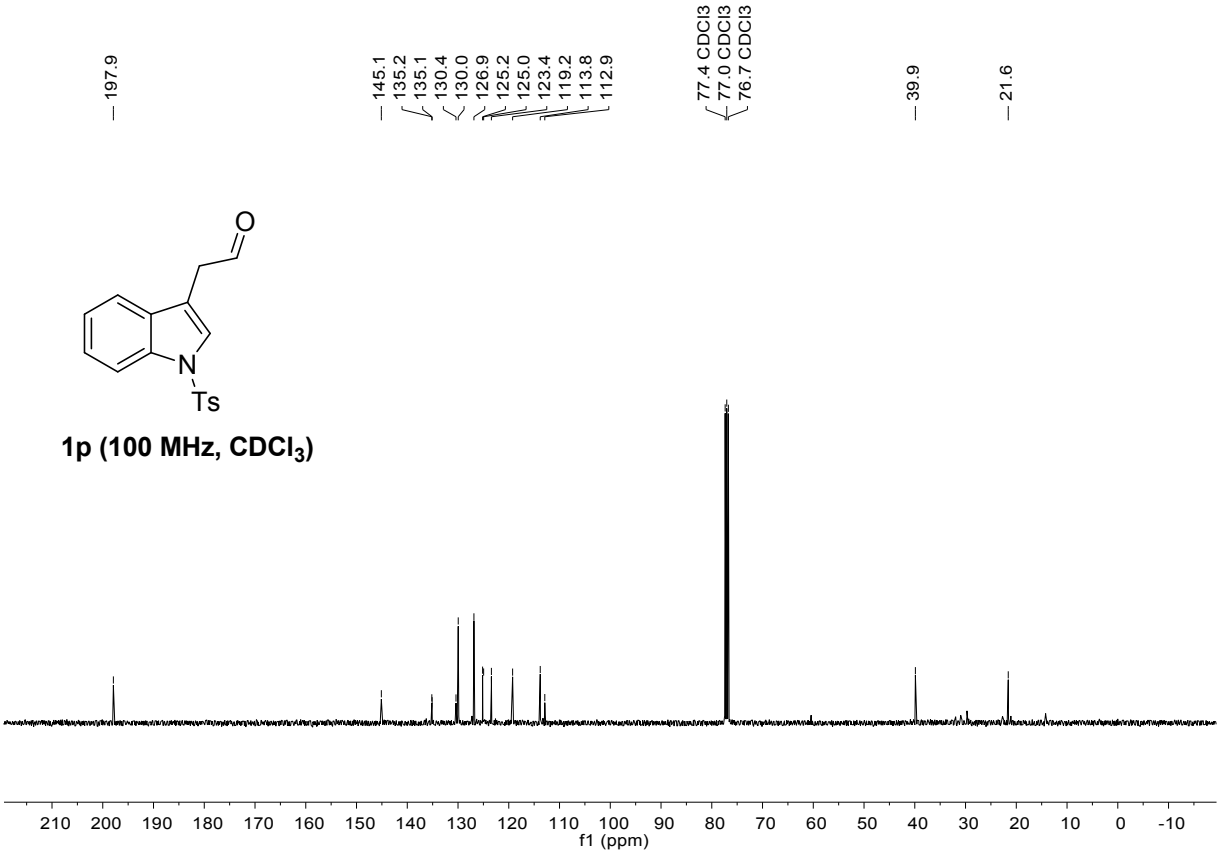
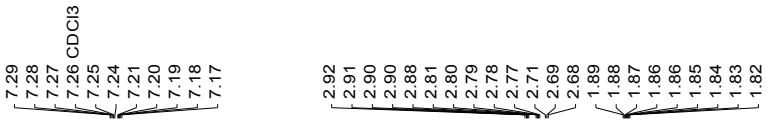
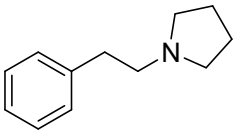


1p (400 MHz, $CDCl_3$)

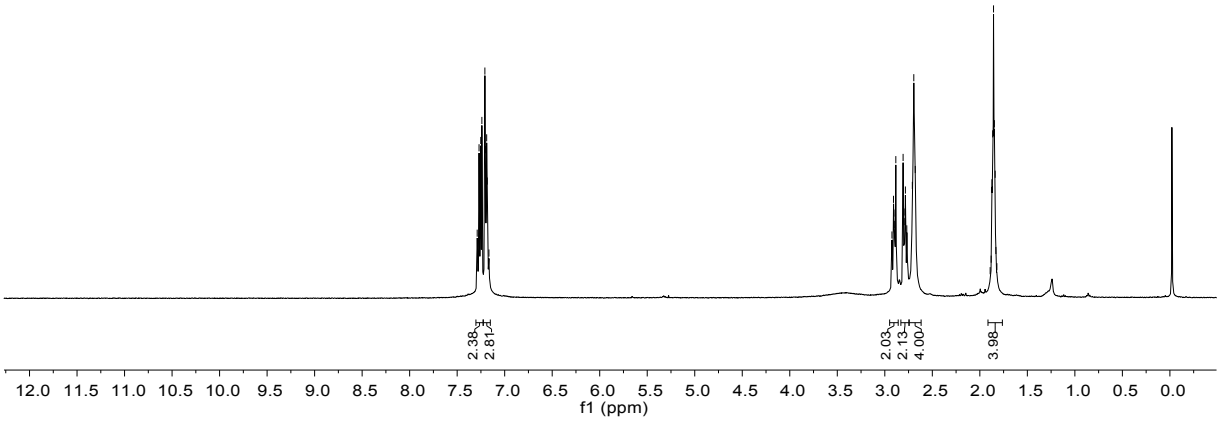


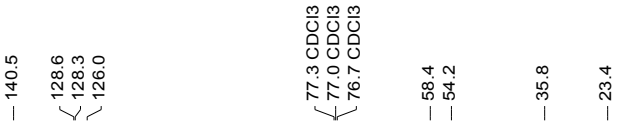
1p (100 MHz, $CDCl_3$)

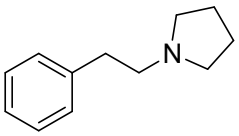




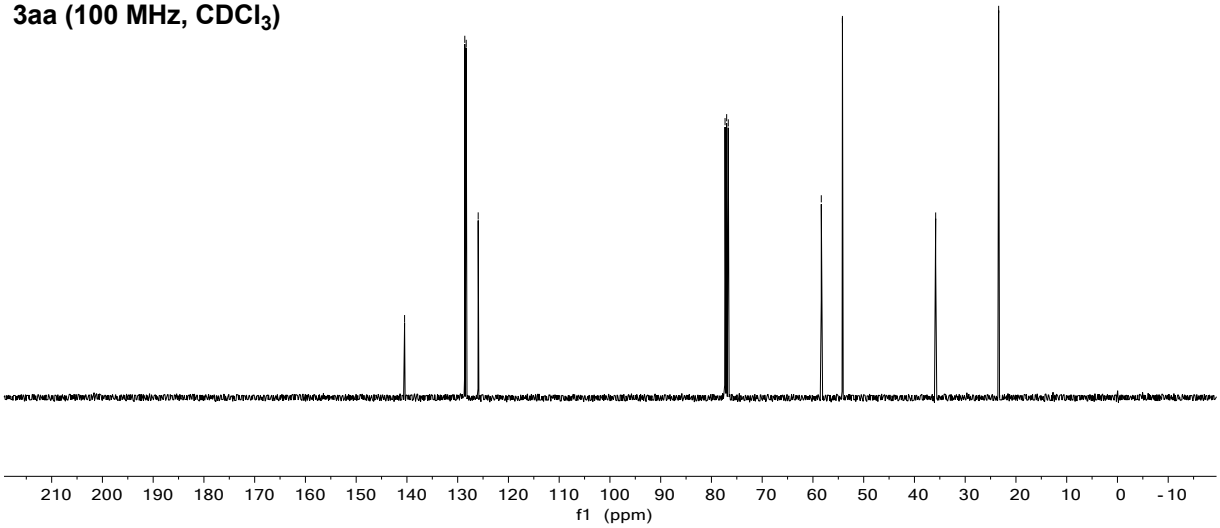
3aa (400 MHz, $CDCl_3$)

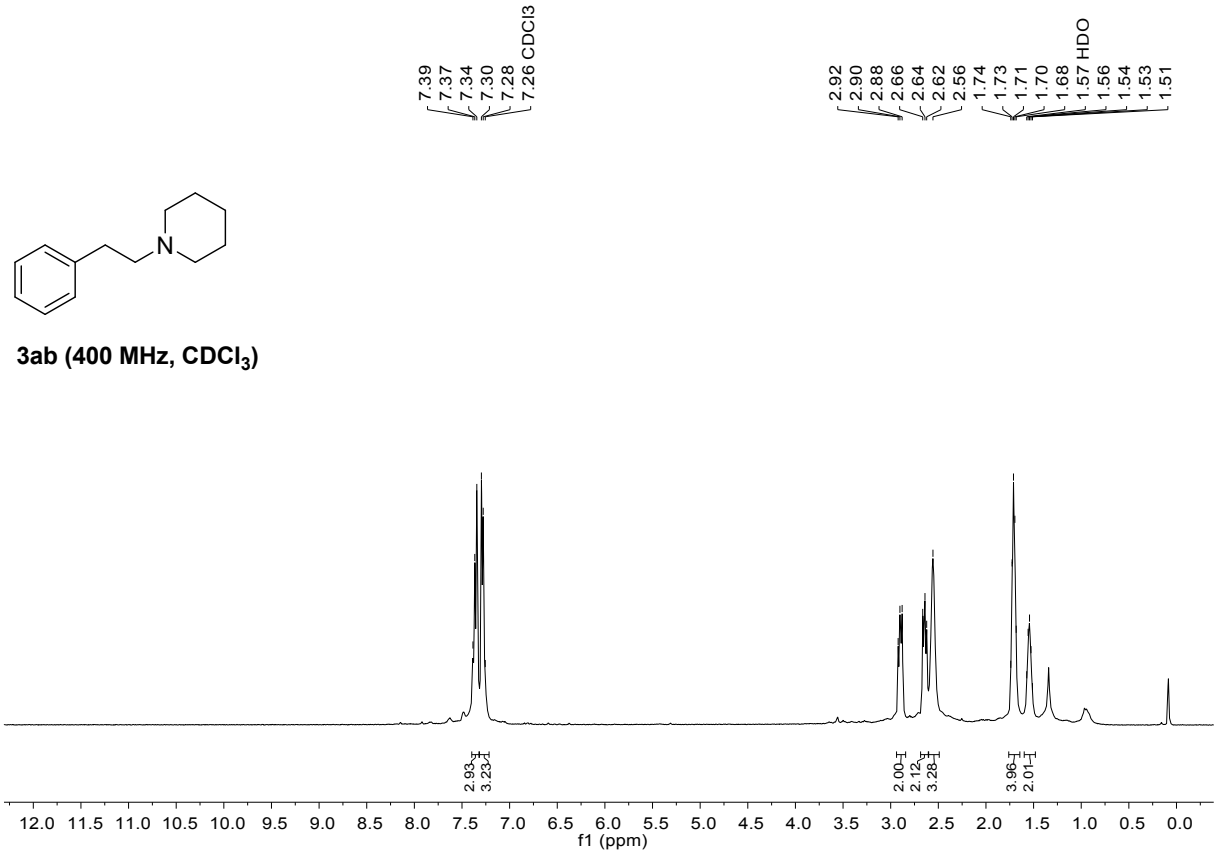




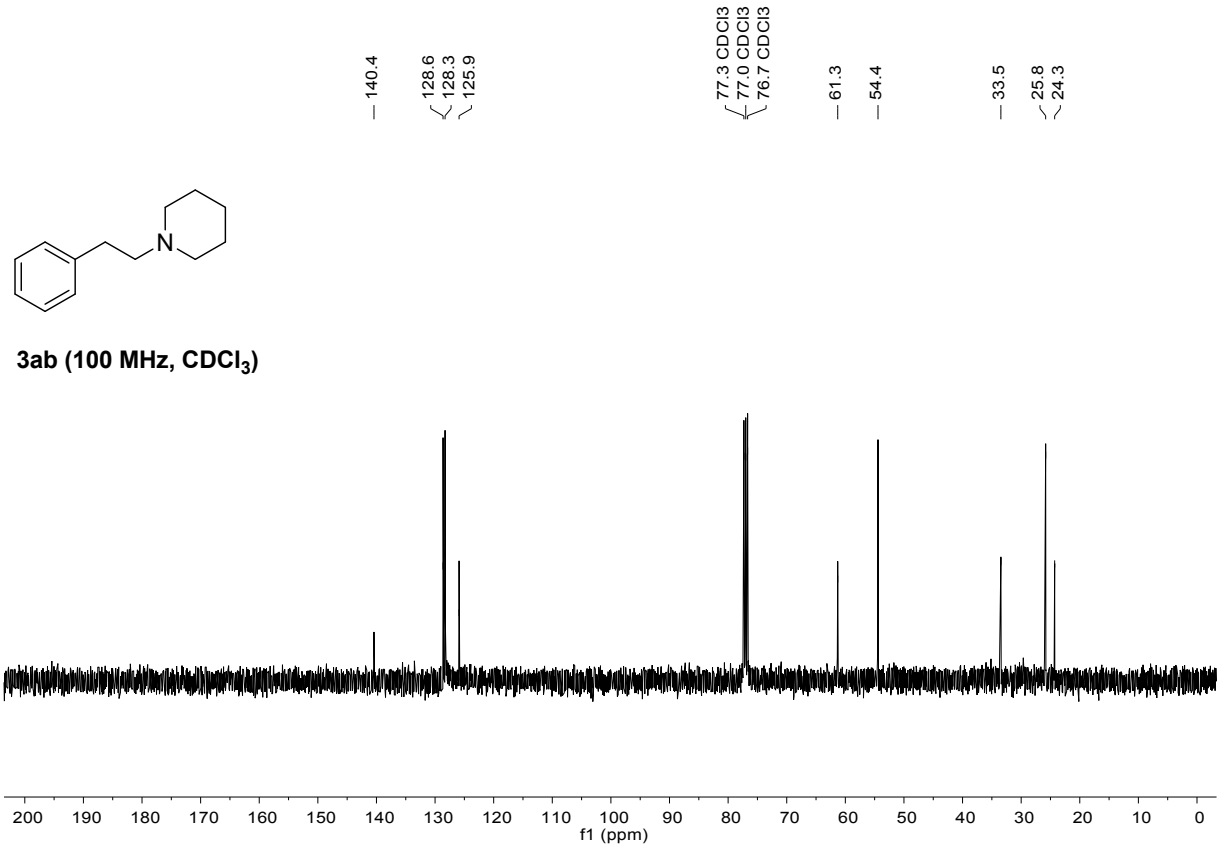


3aa (100 MHz, $CDCl_3$)

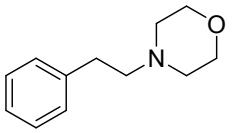




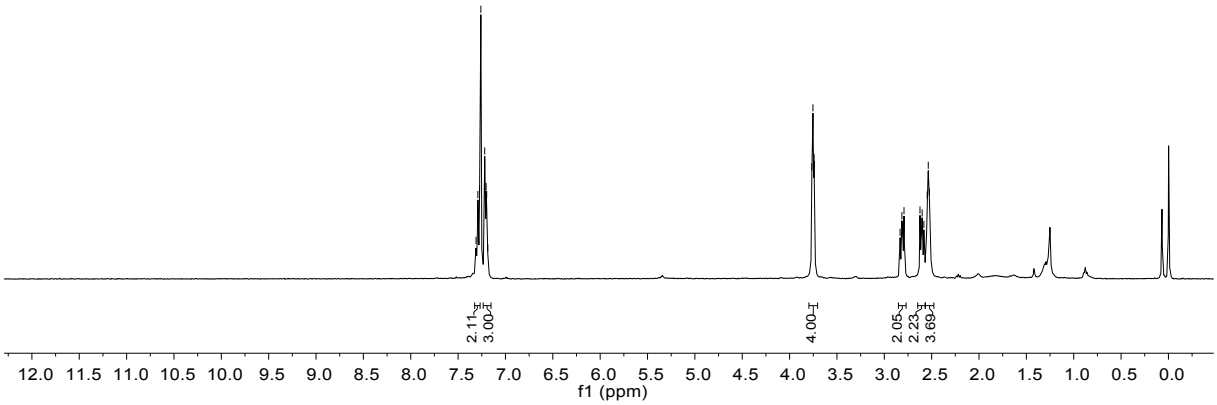
3ab (400 MHz, $CDCl_3$)

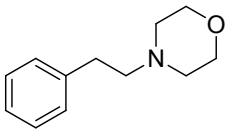


3ab (100 MHz, $CDCl_3$)

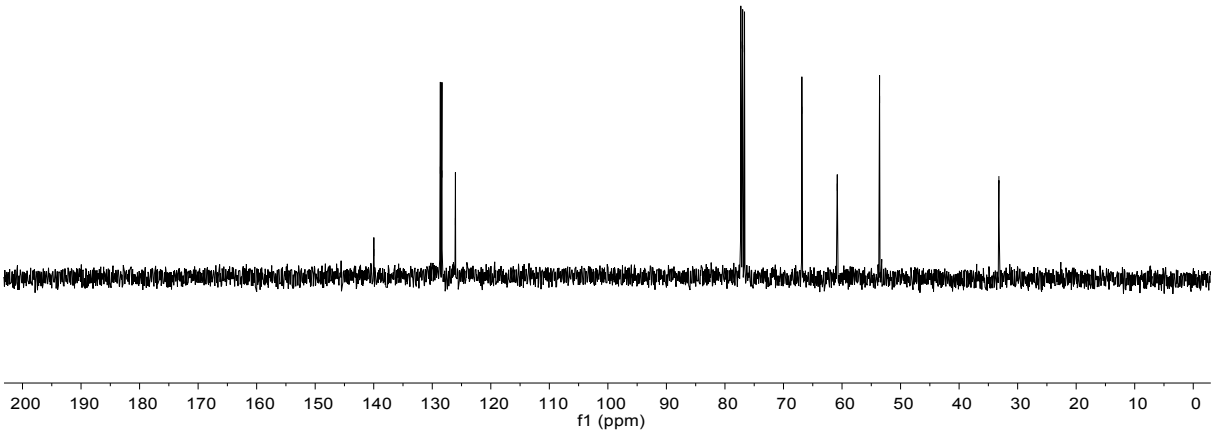


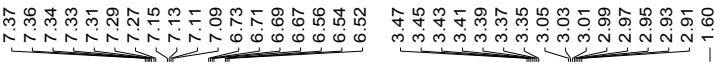
3ac (400 MHz, $CDCl_3$)

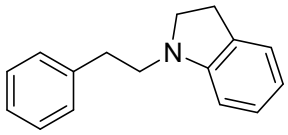




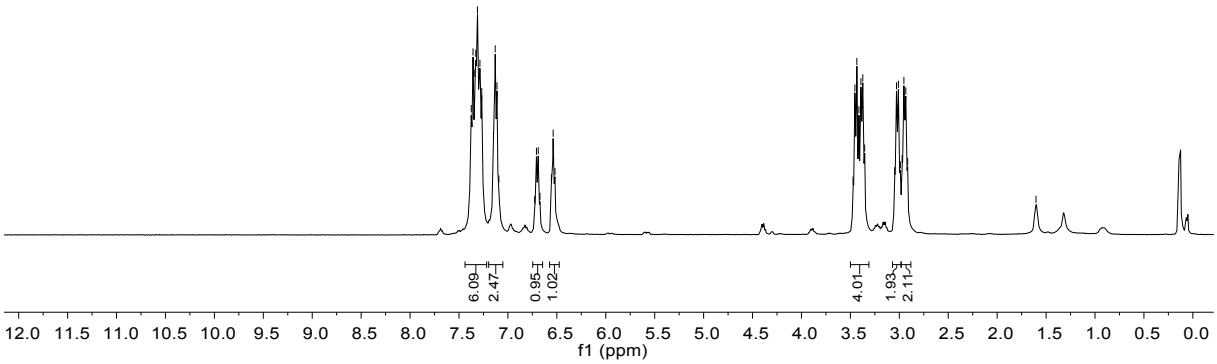
3ac (100 MHz, $CDCl_3$)

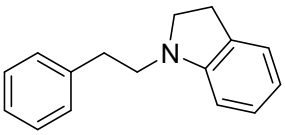




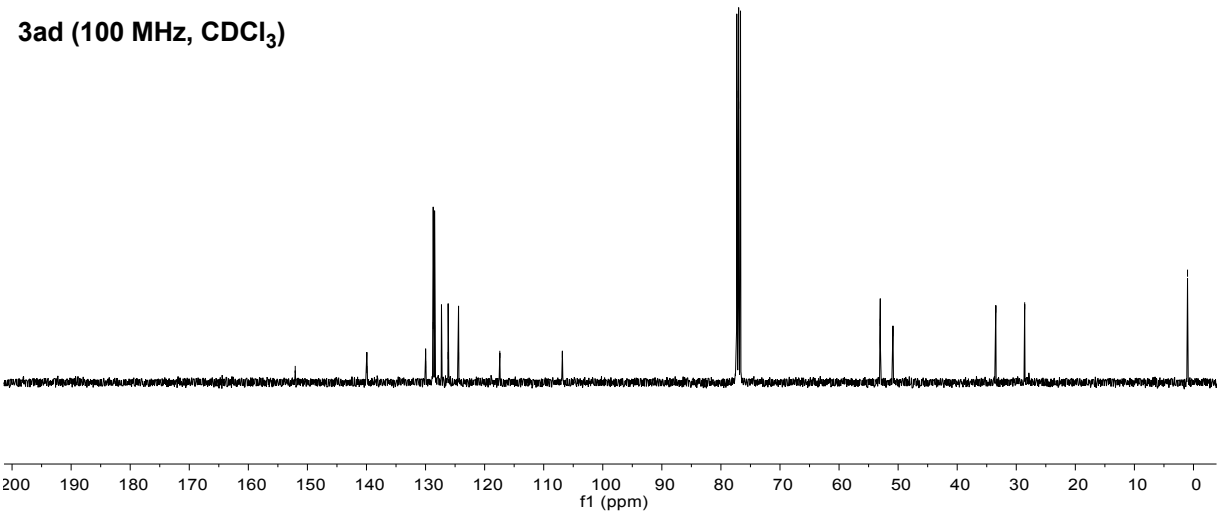


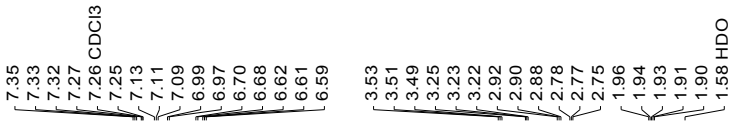
3ad (400 MHz, $CDCl_3$)

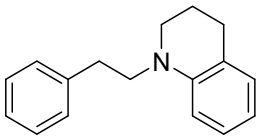




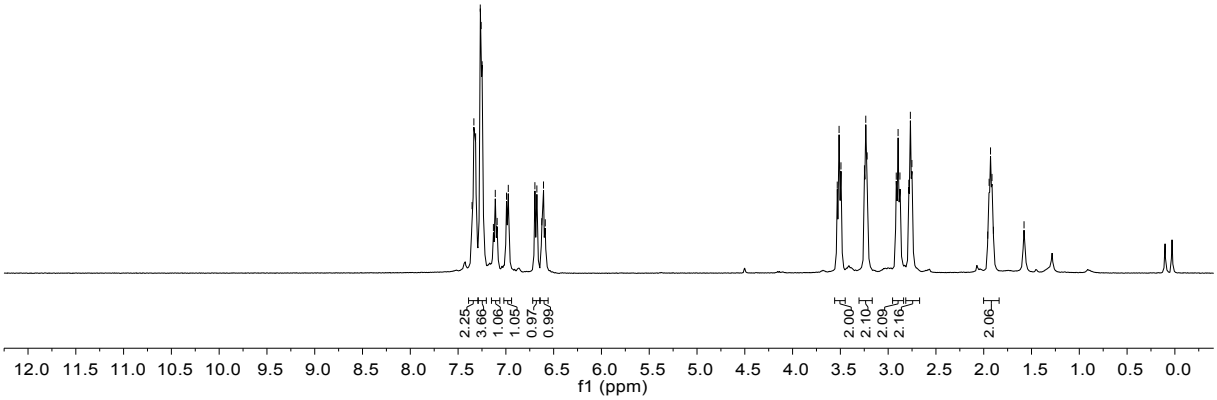
3ad (100 MHz, $CDCl_3$)

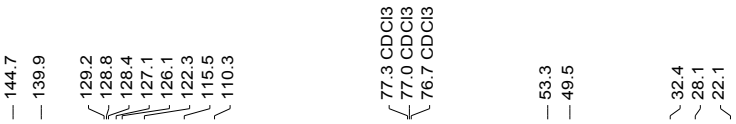


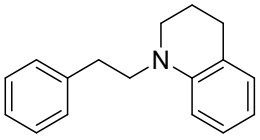




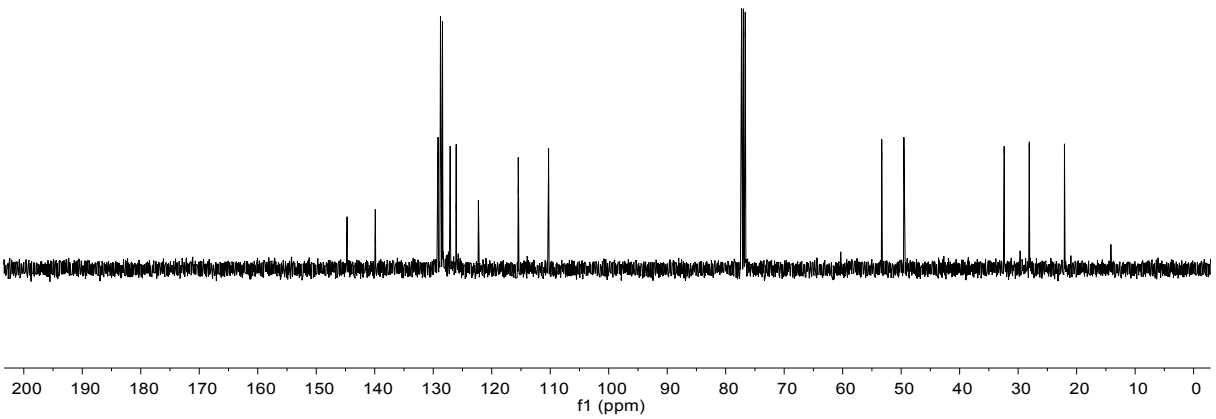
3ae (400 MHz, $CDCl_3$)

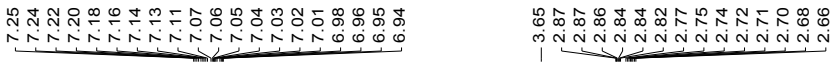


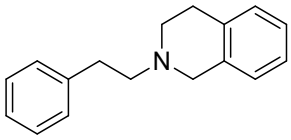




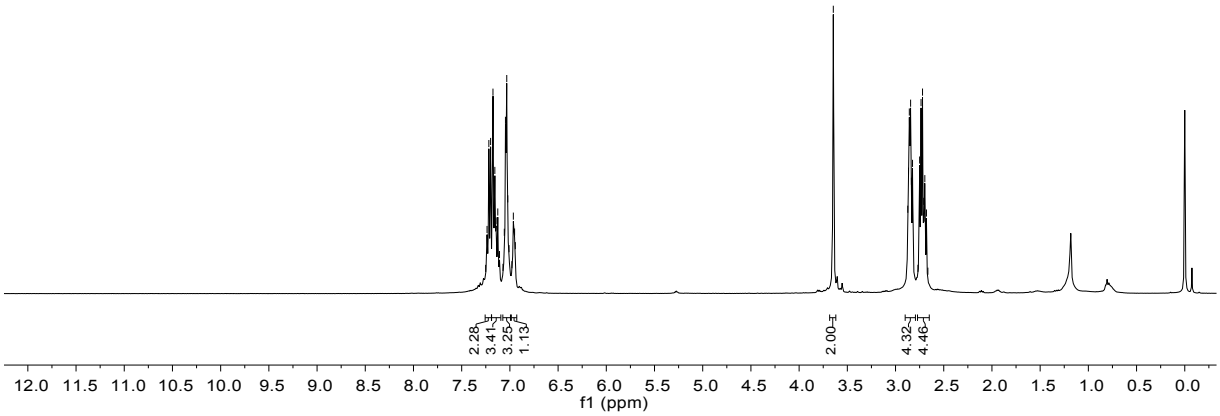
3ae (100 MHz, $CDCl_3$)

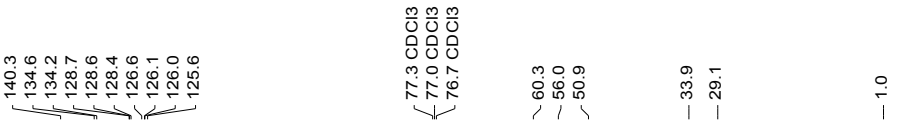


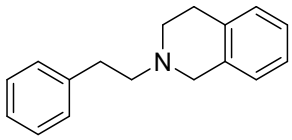




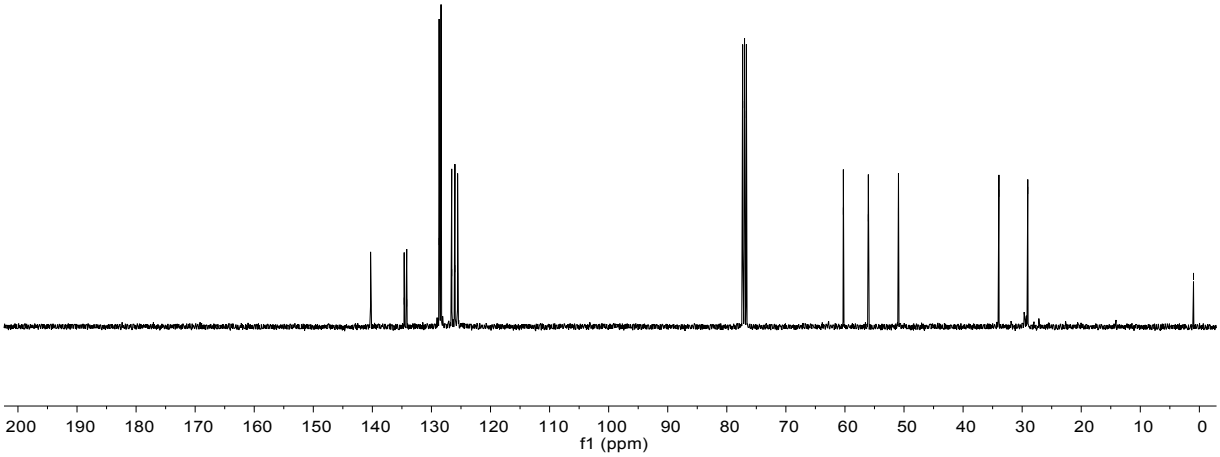
3af (400 MHz, $CDCl_3$)

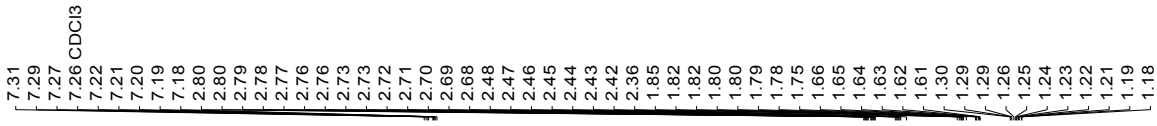


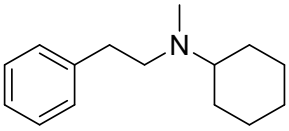




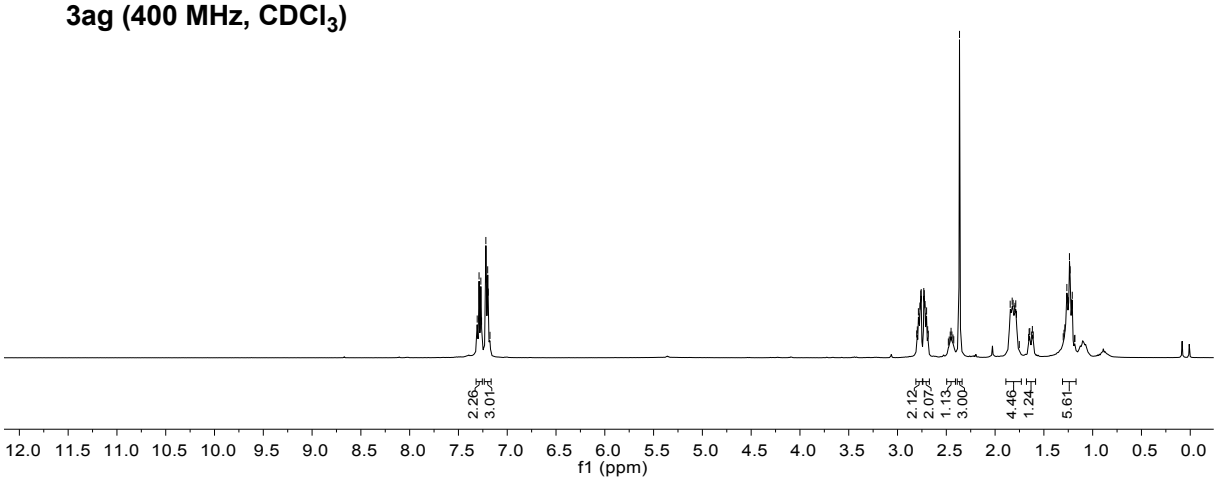
3af (100 MHz, $CDCl_3$)

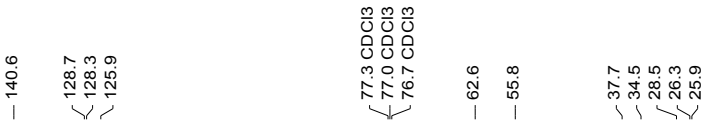


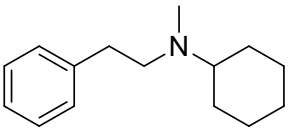




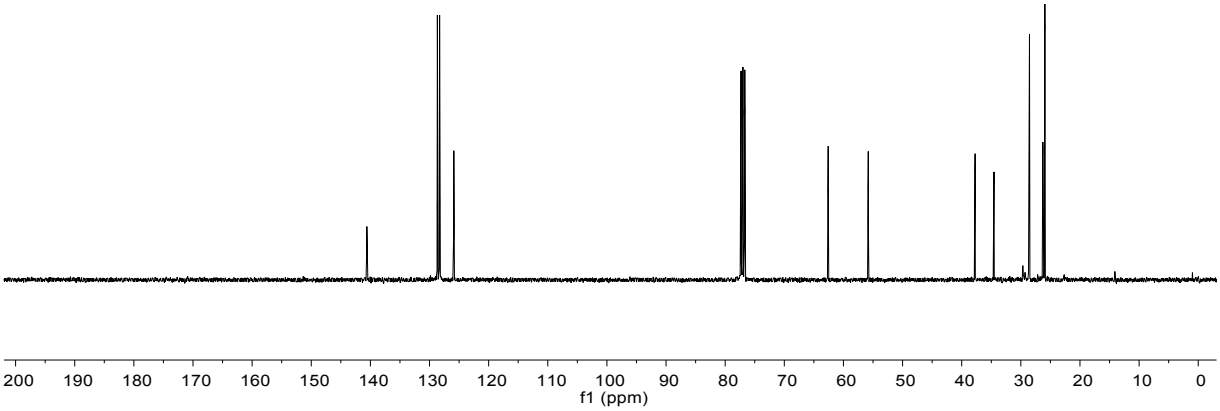
3ag (400 MHz, $CDCl_3$)

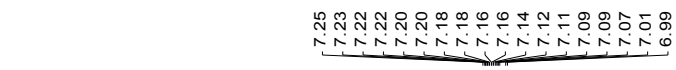


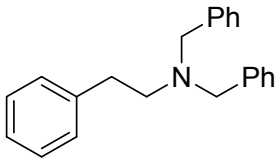




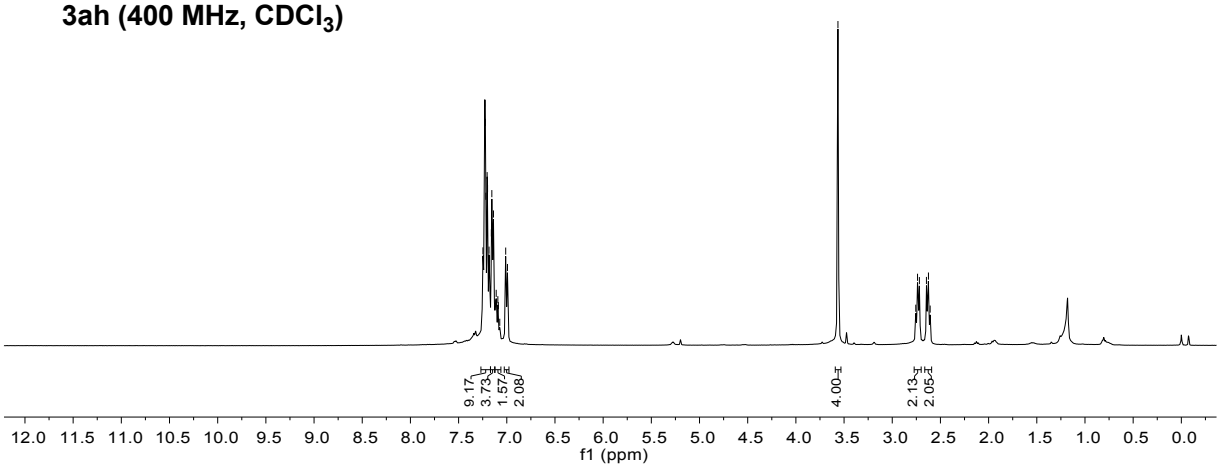
3ag (100 MHz, $CDCl_3$)

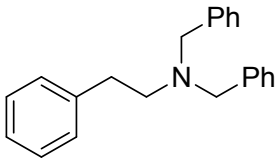




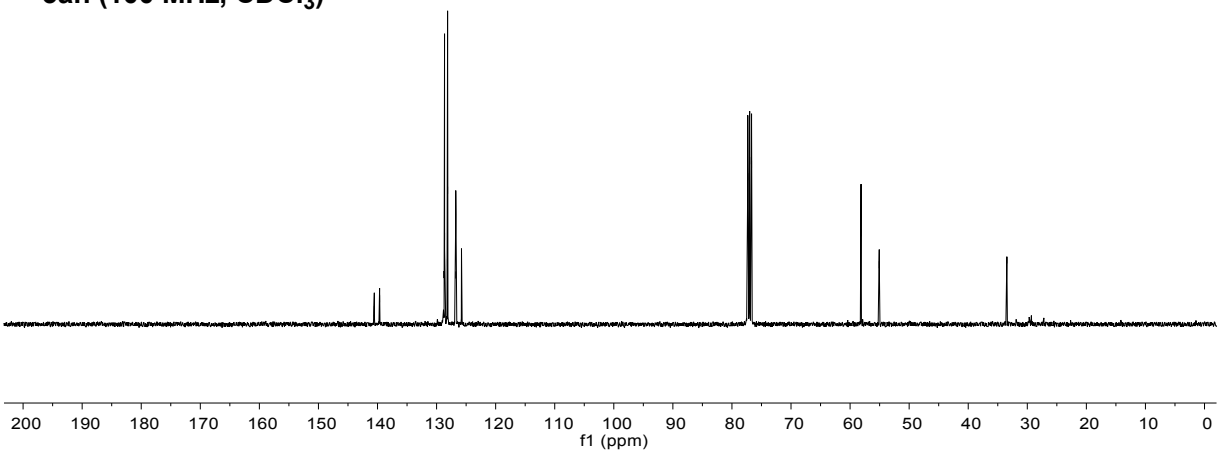


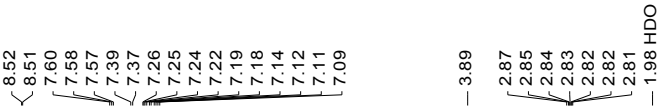
3ah (400 MHz, $CDCl_3$)

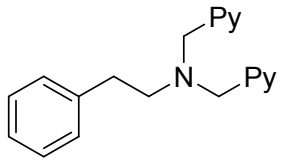




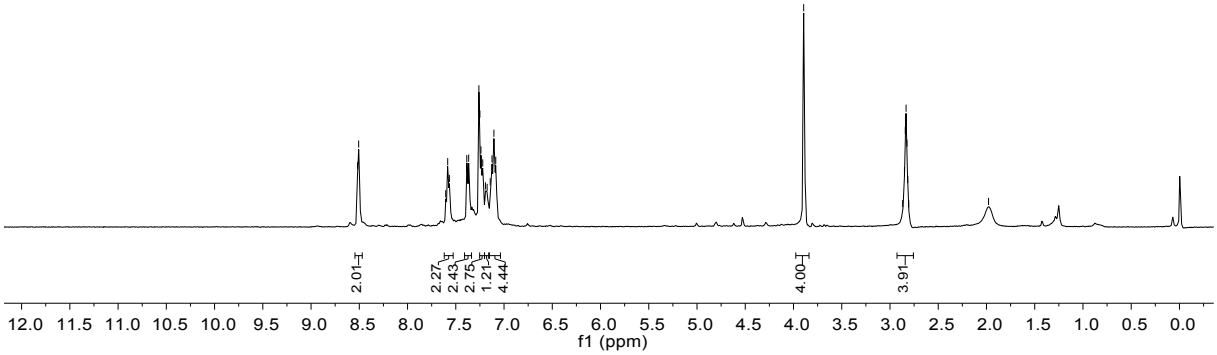
3ah (100 MHz, $CDCl_3$)

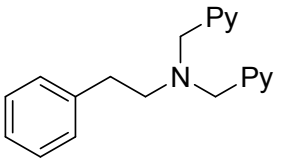




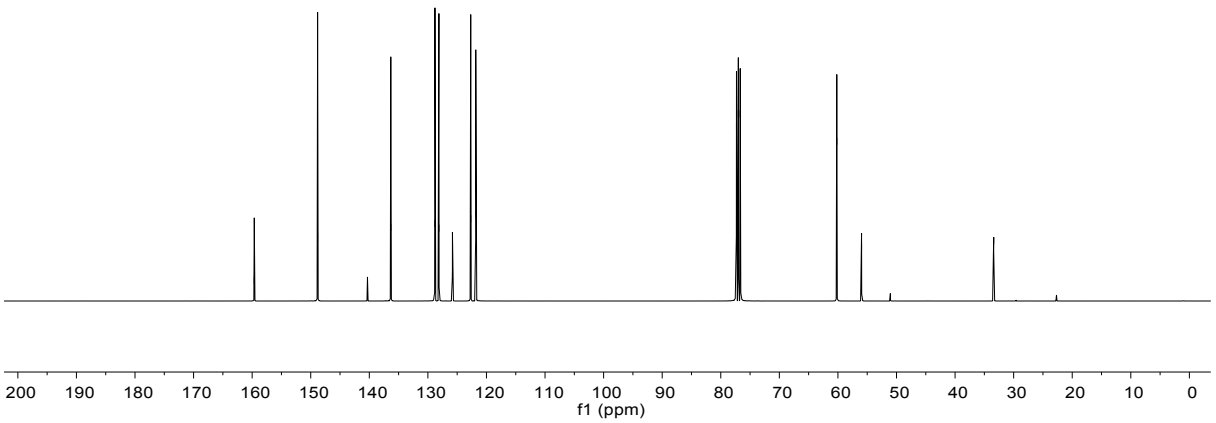


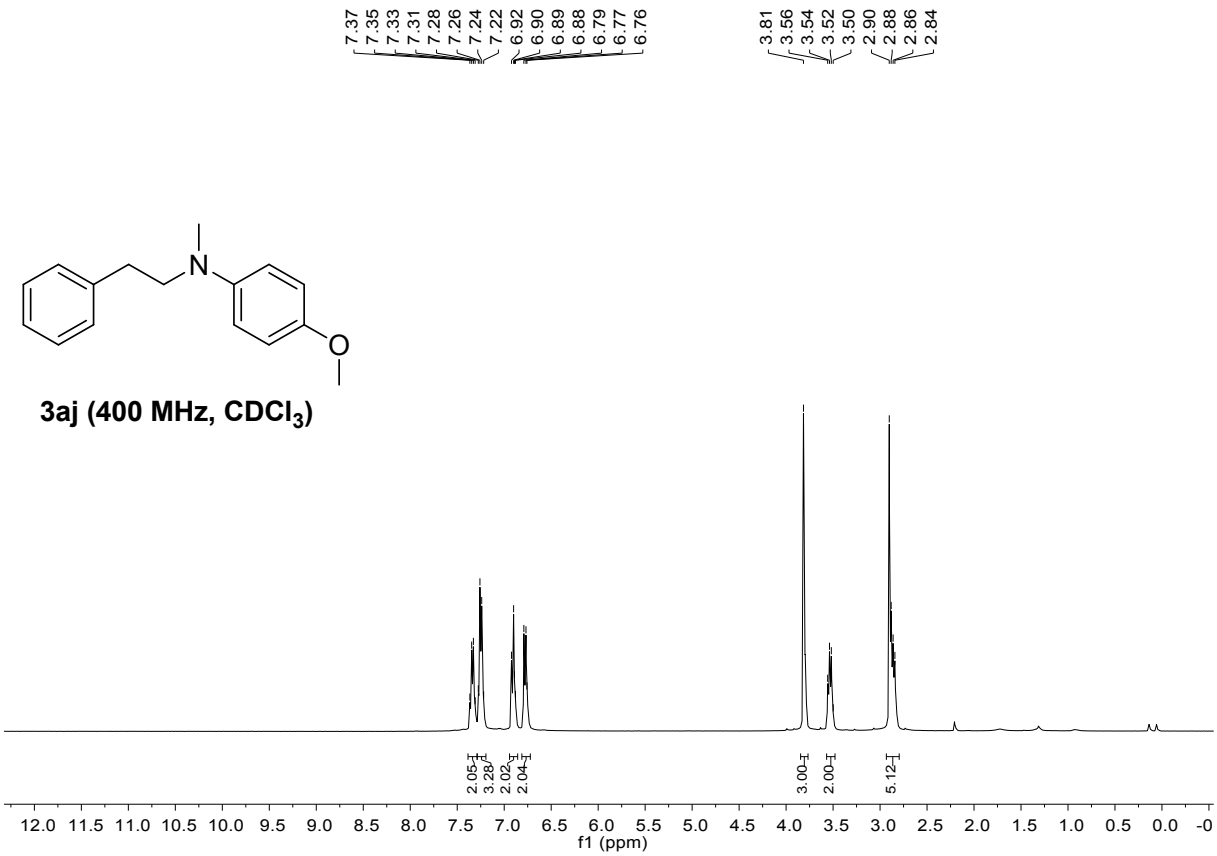
3ai (400 MHz, $CDCl_3$)



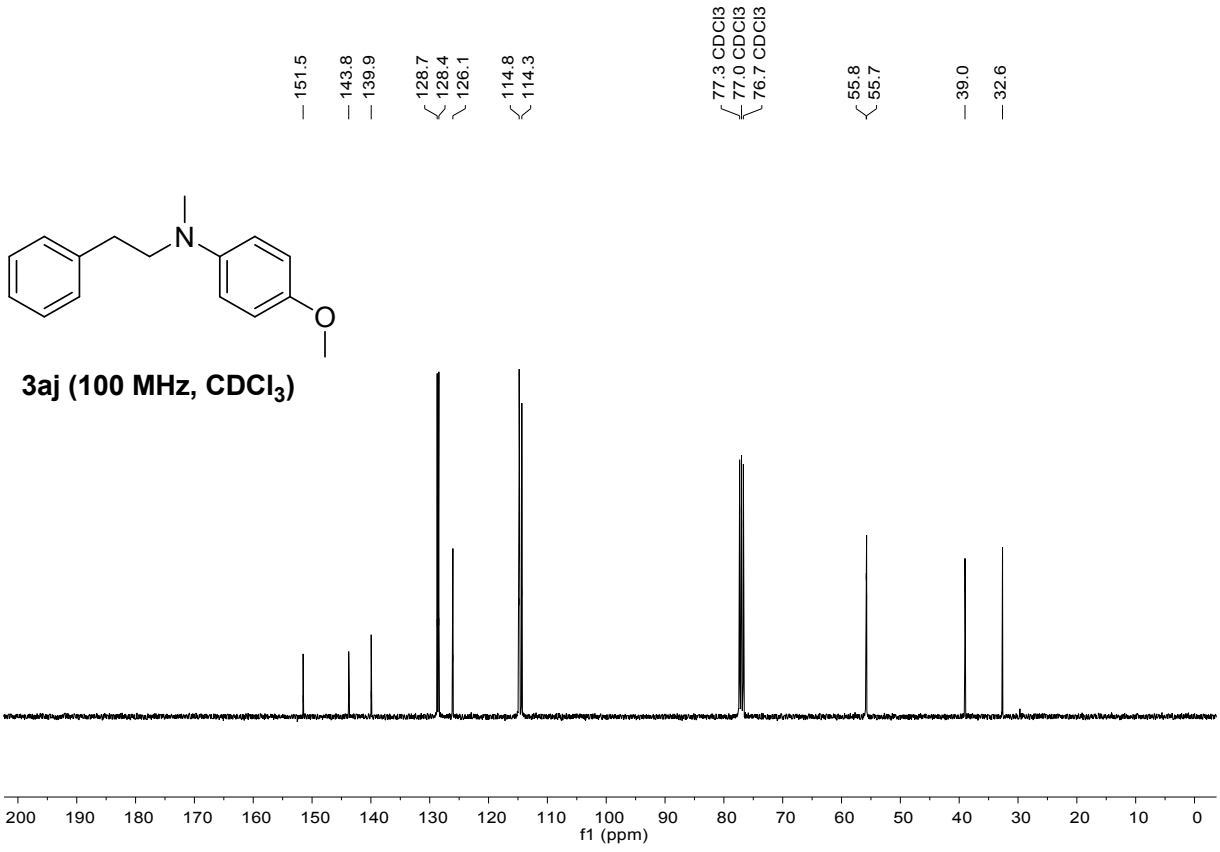


3ai (100 MHz, $CDCl_3$)

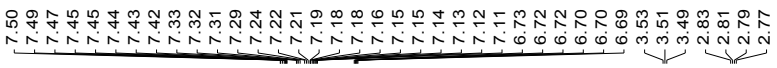


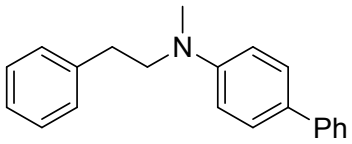


3aj (400 MHz, $CDCl_3$)

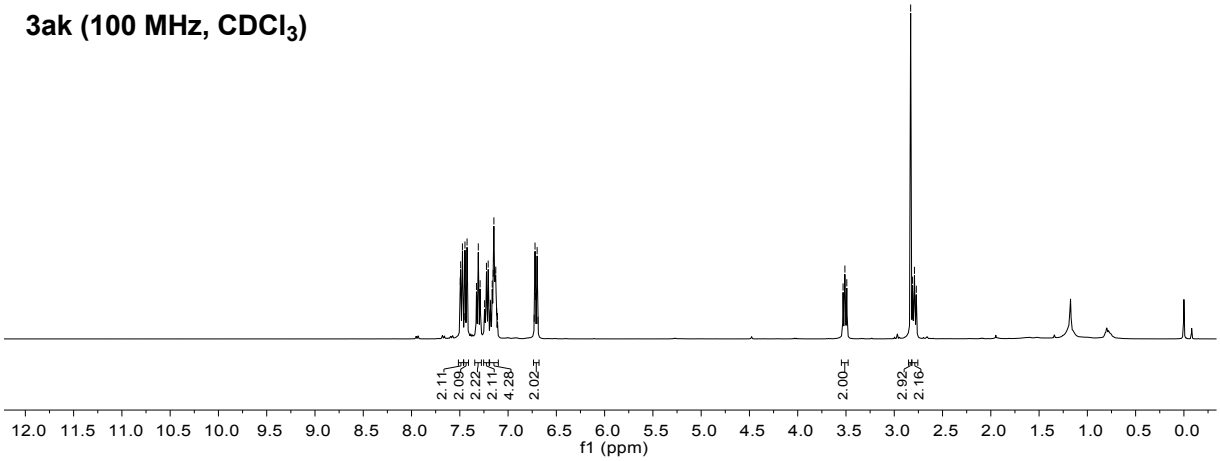


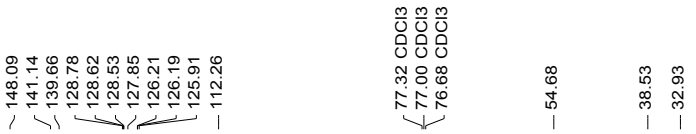
3aj (100 MHz, $CDCl_3$)

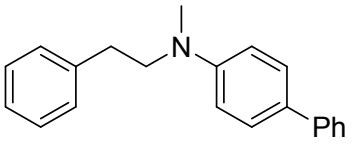




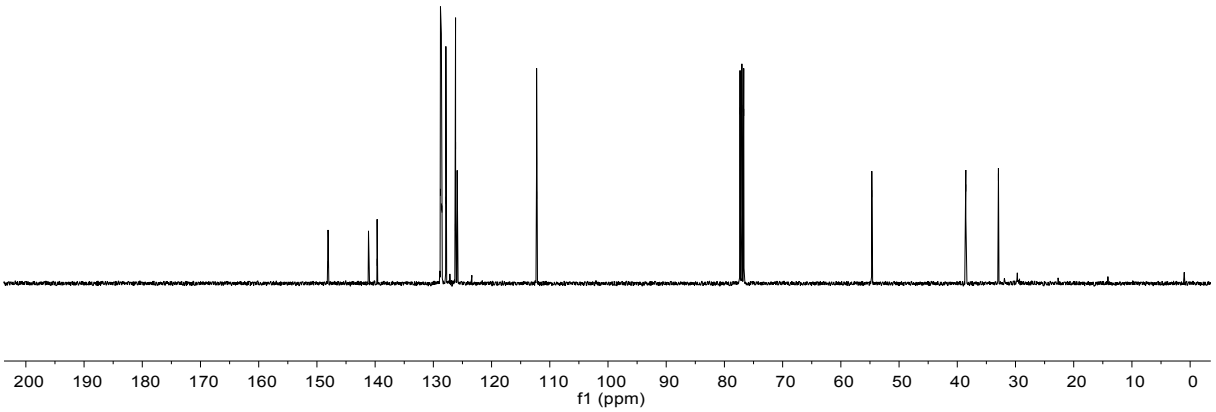
3ak (100 MHz, $CDCl_3$)

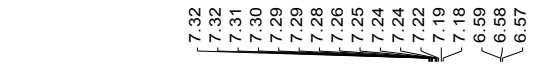


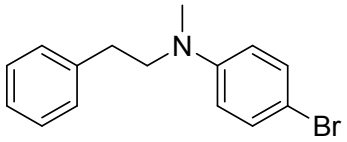




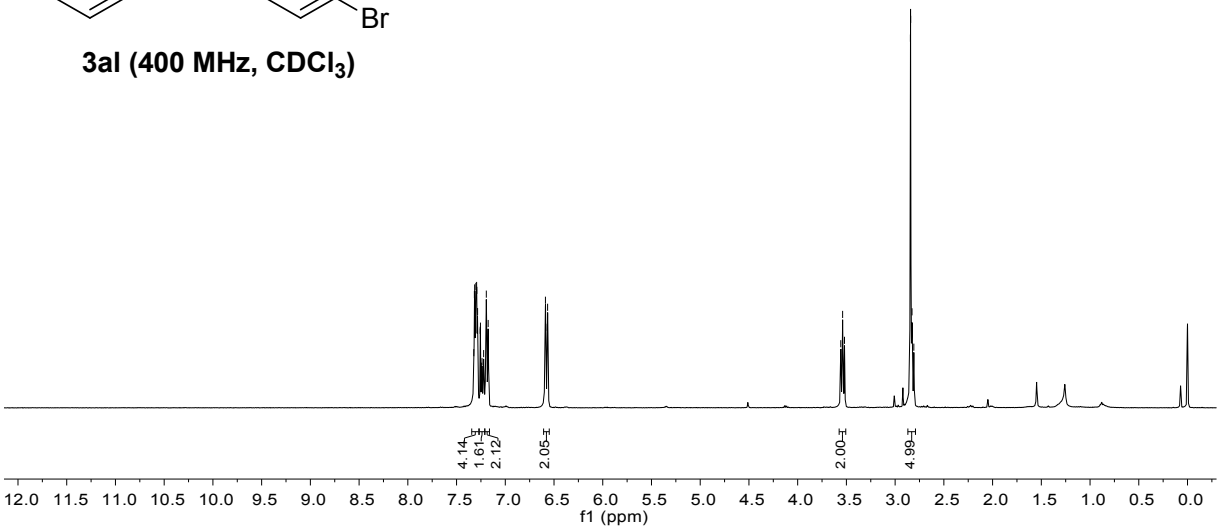
3ak (400 MHz, $CDCl_3$)

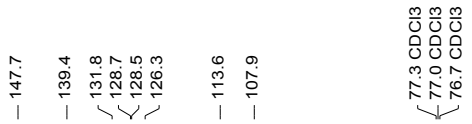


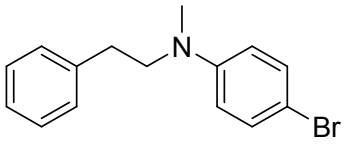




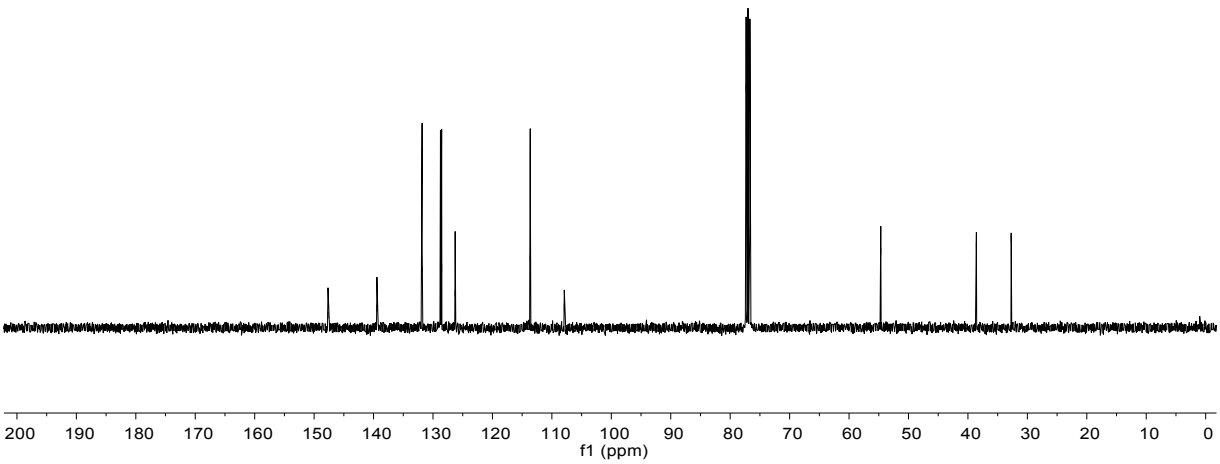
3al (400 MHz, $CDCl_3$)

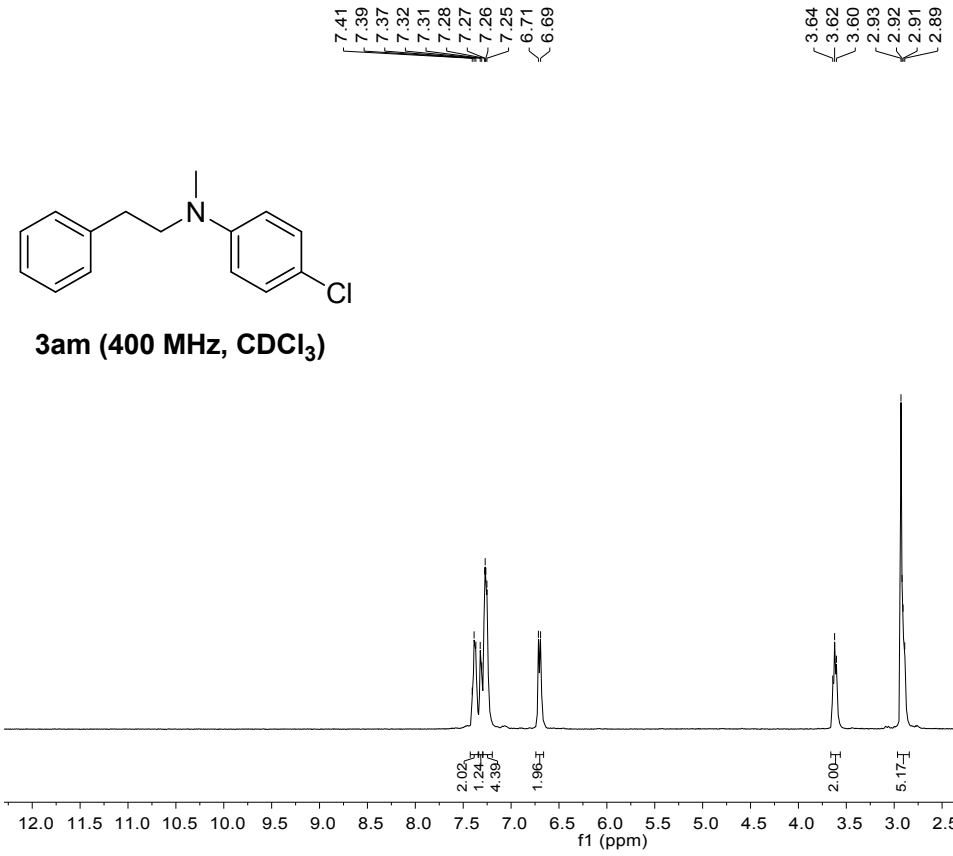




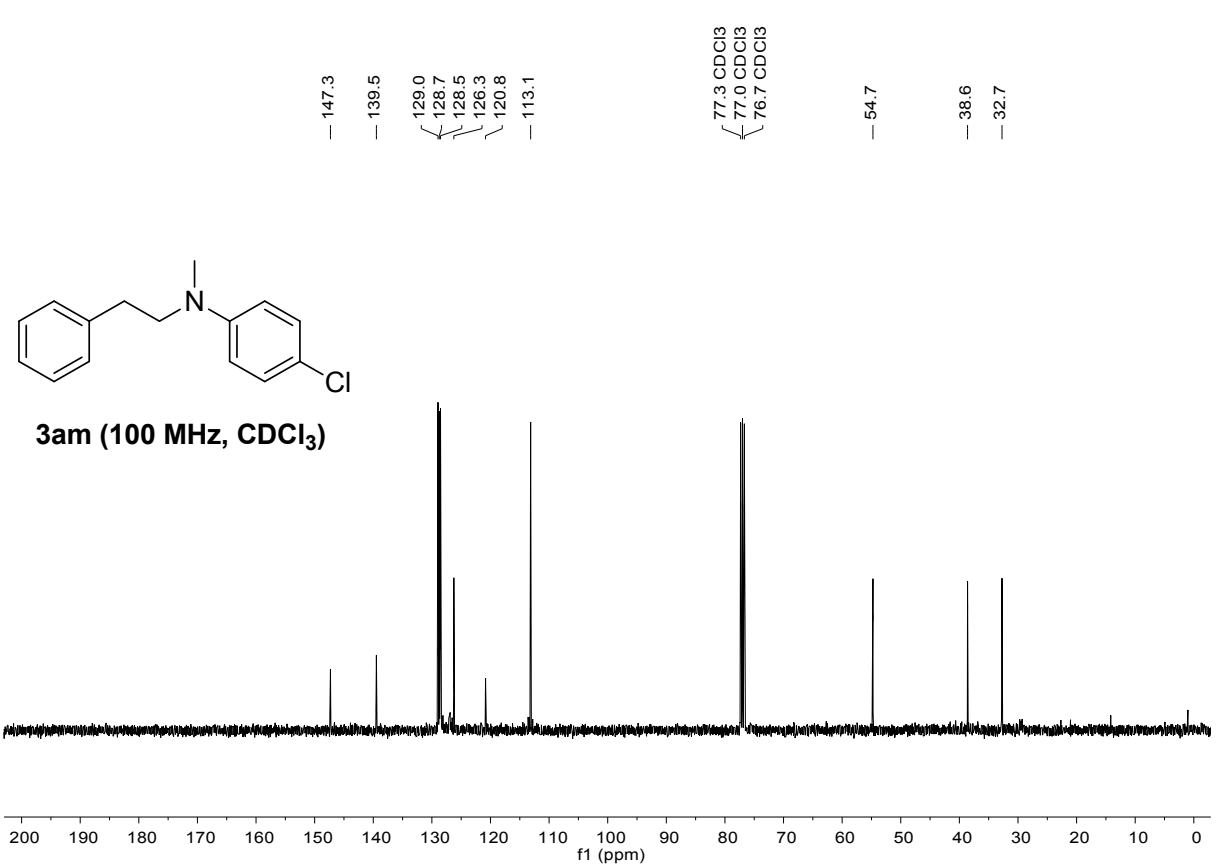


3al (100 MHz, $CDCl_3$)

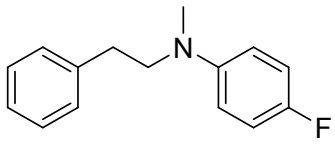




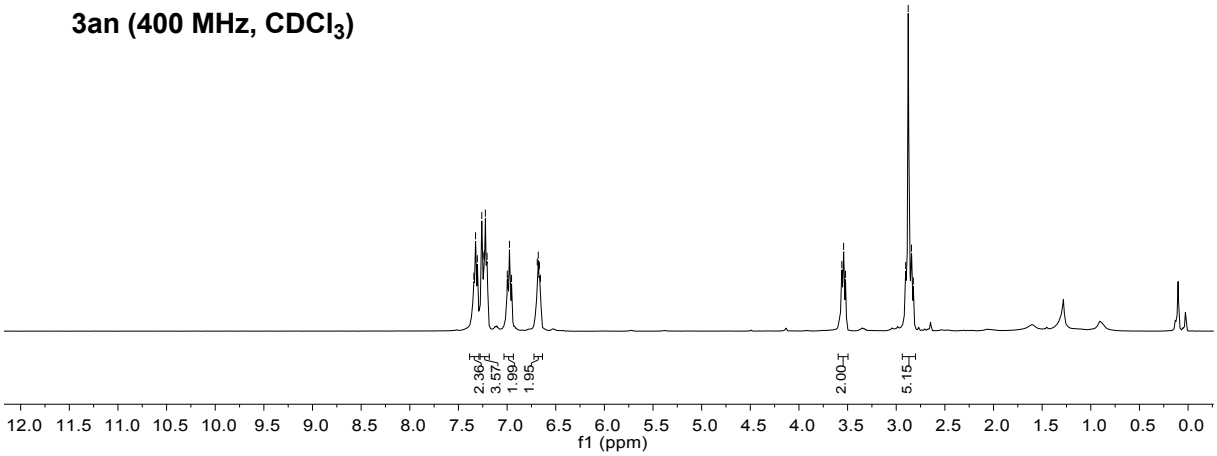
3am (400 MHz, $CDCl_3$)

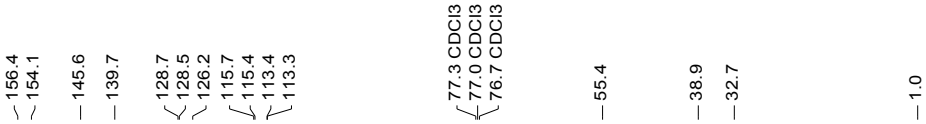


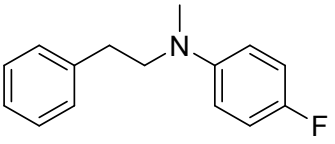
3am (100 MHz, $CDCl_3$)



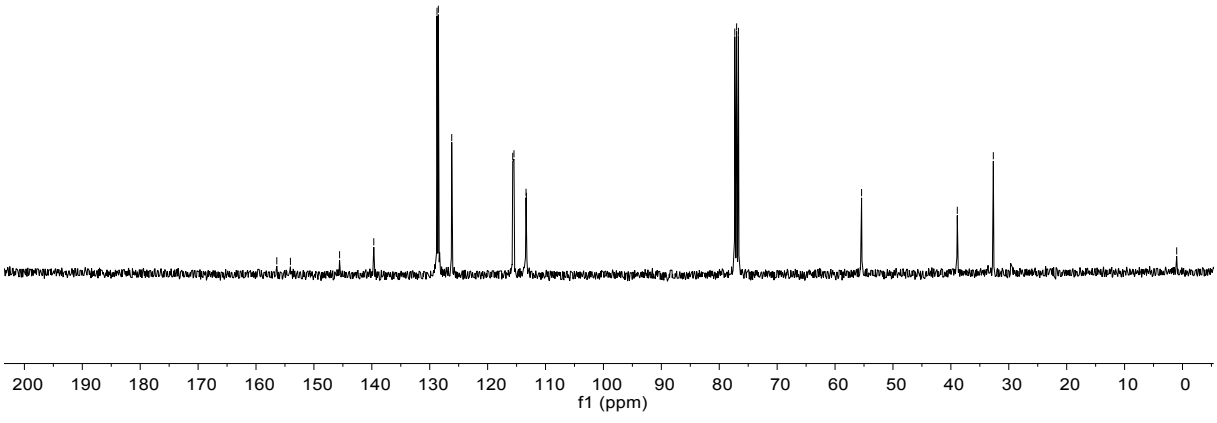
3an (400 MHz, $CDCl_3$)

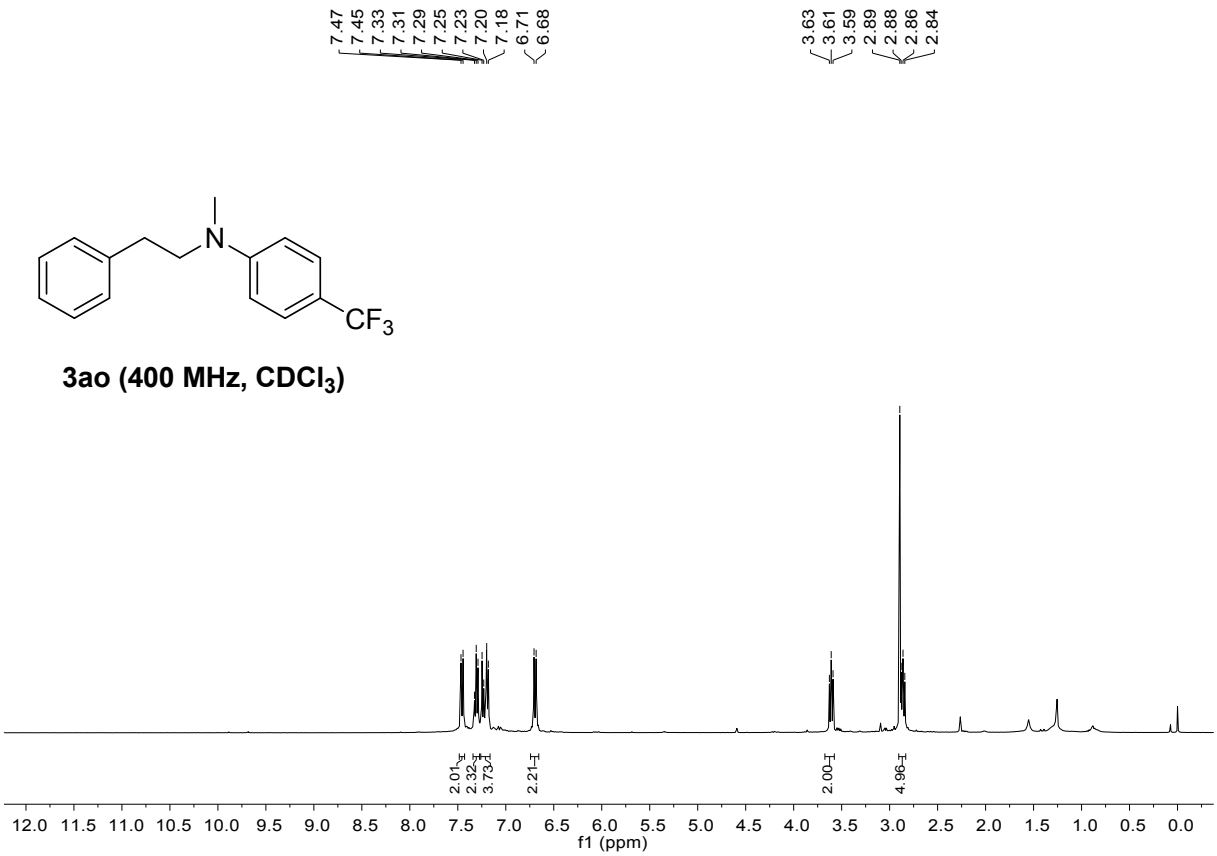




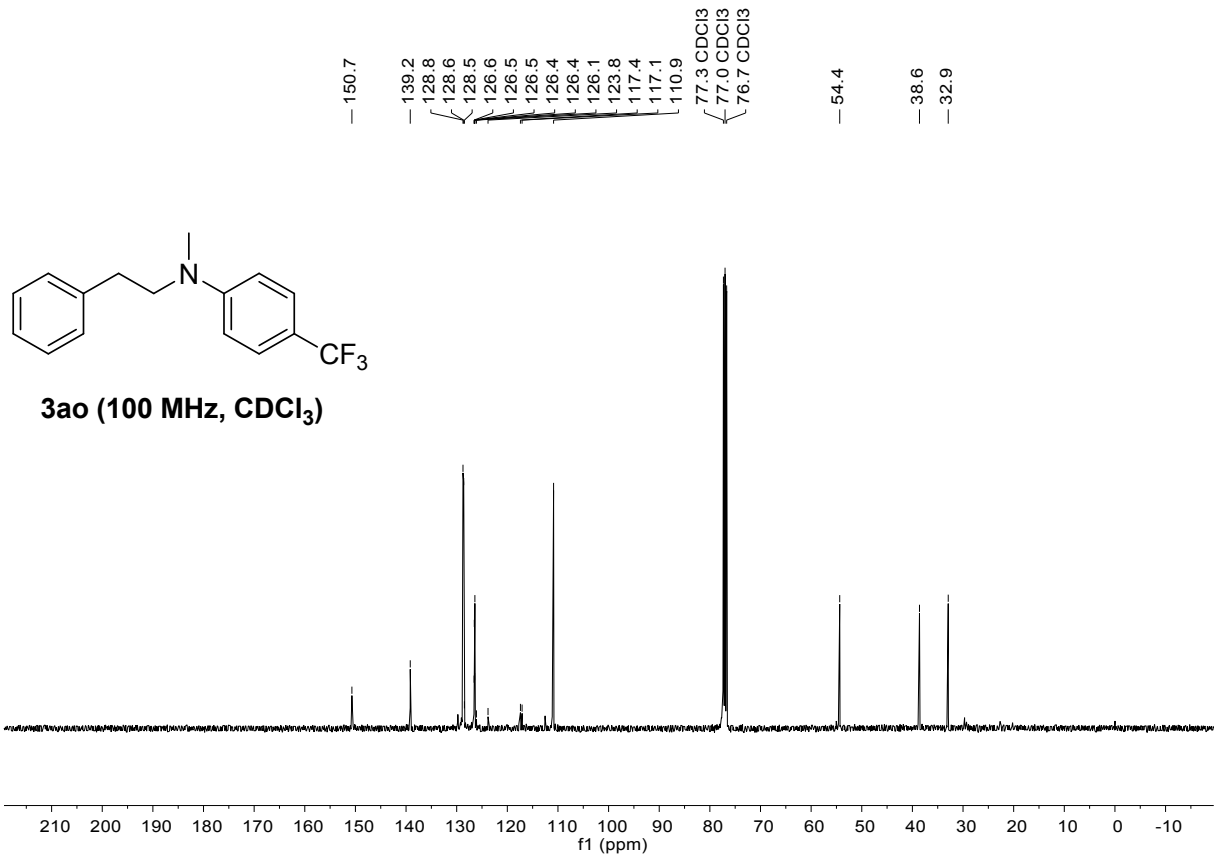


3an (100 MHz, $CDCl_3$)

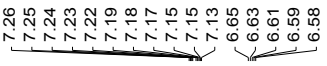


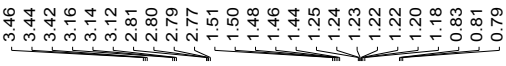


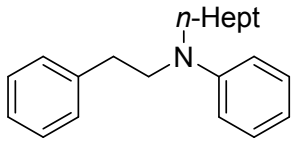
3ao (400 MHz, $CDCl_3$)



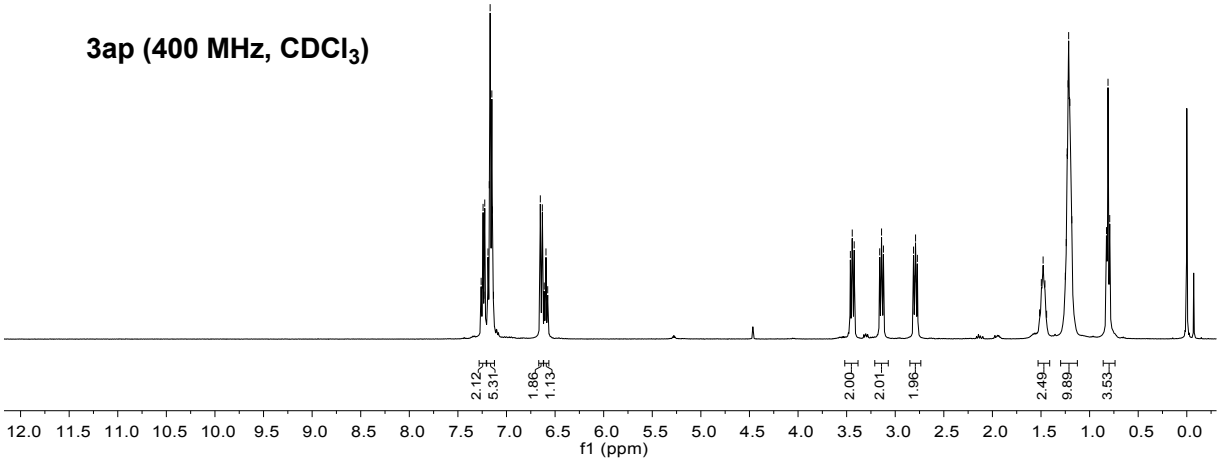
3ao (100 MHz, $CDCl_3$)

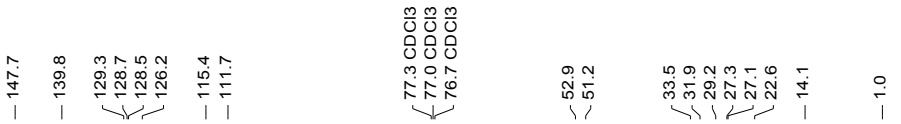


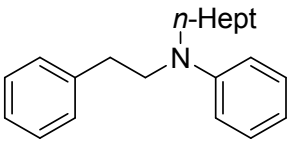




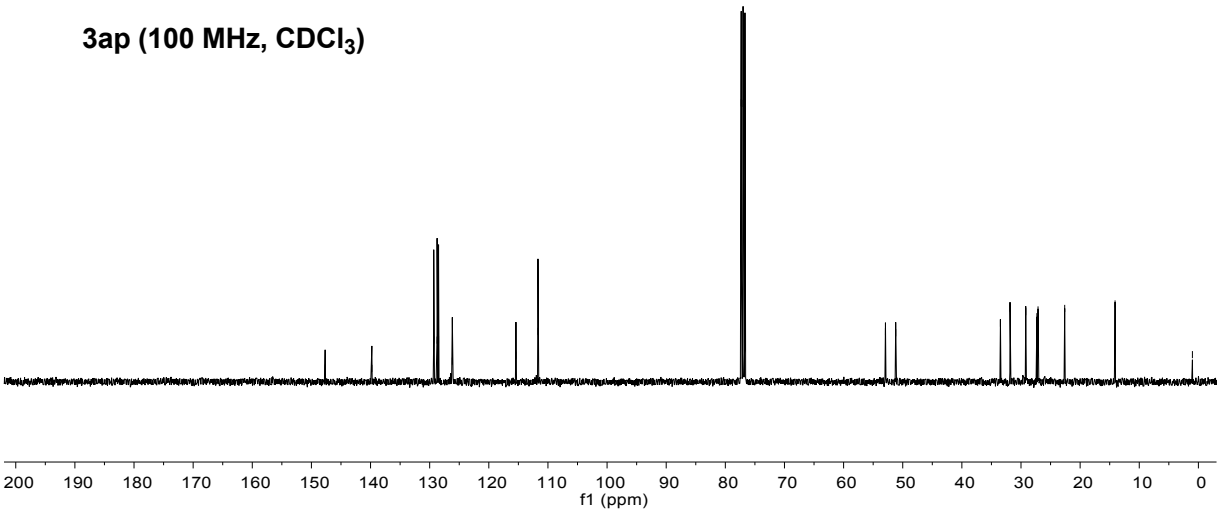
3ap (400 MHz, $CDCl_3$)

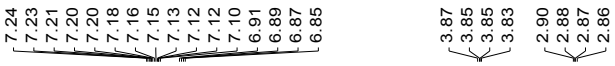


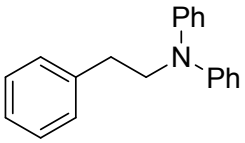




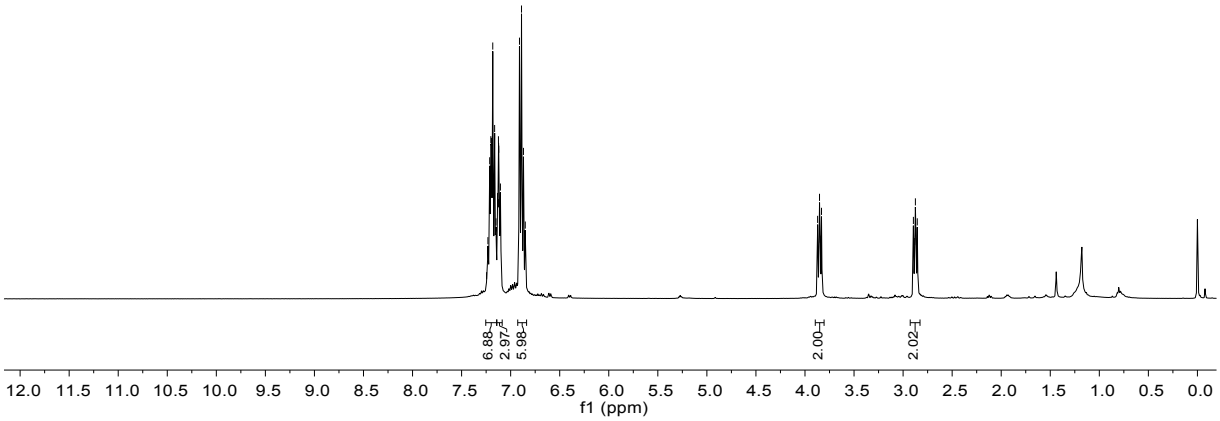
3ap (100 MHz, $CDCl_3$)

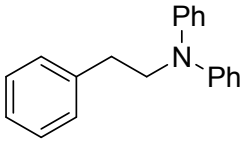




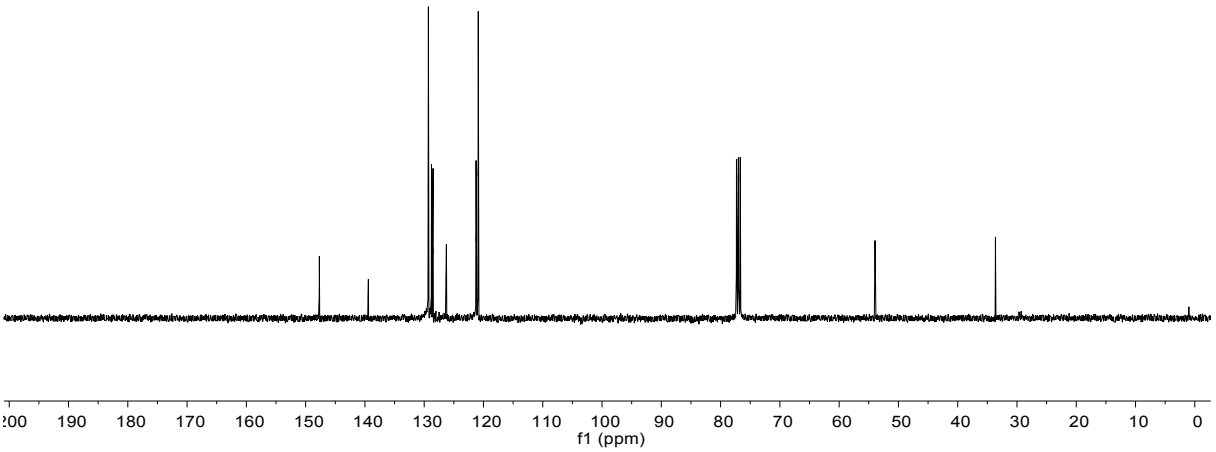


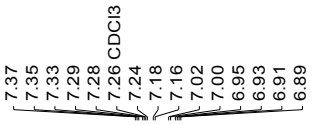
3aq (400 MHz, $CDCl_3$)

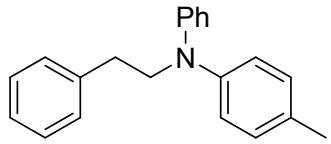




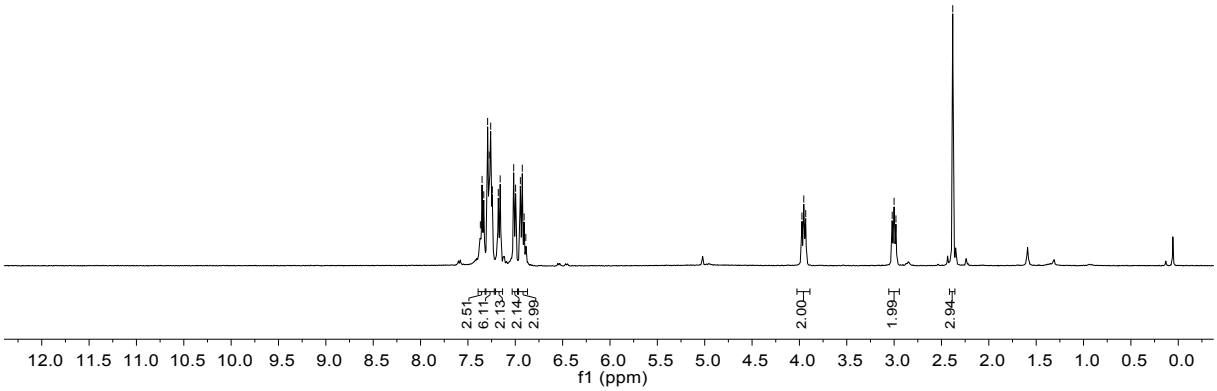
3aq (100 MHz, $CDCl_3$)

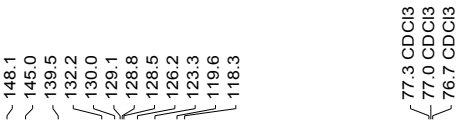


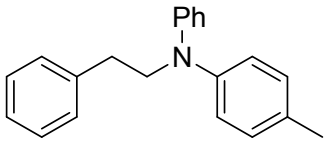




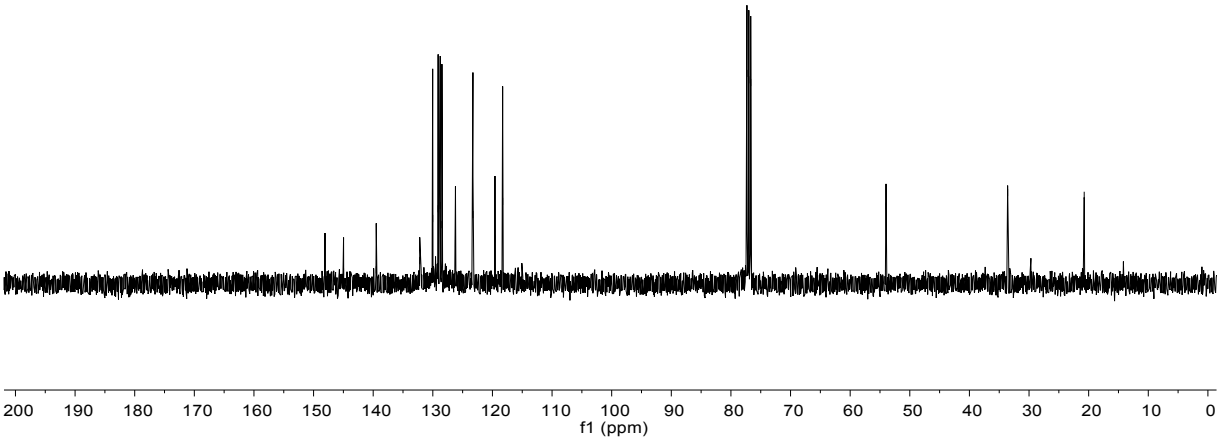
3ar (400 MHz, $CDCl_3$)

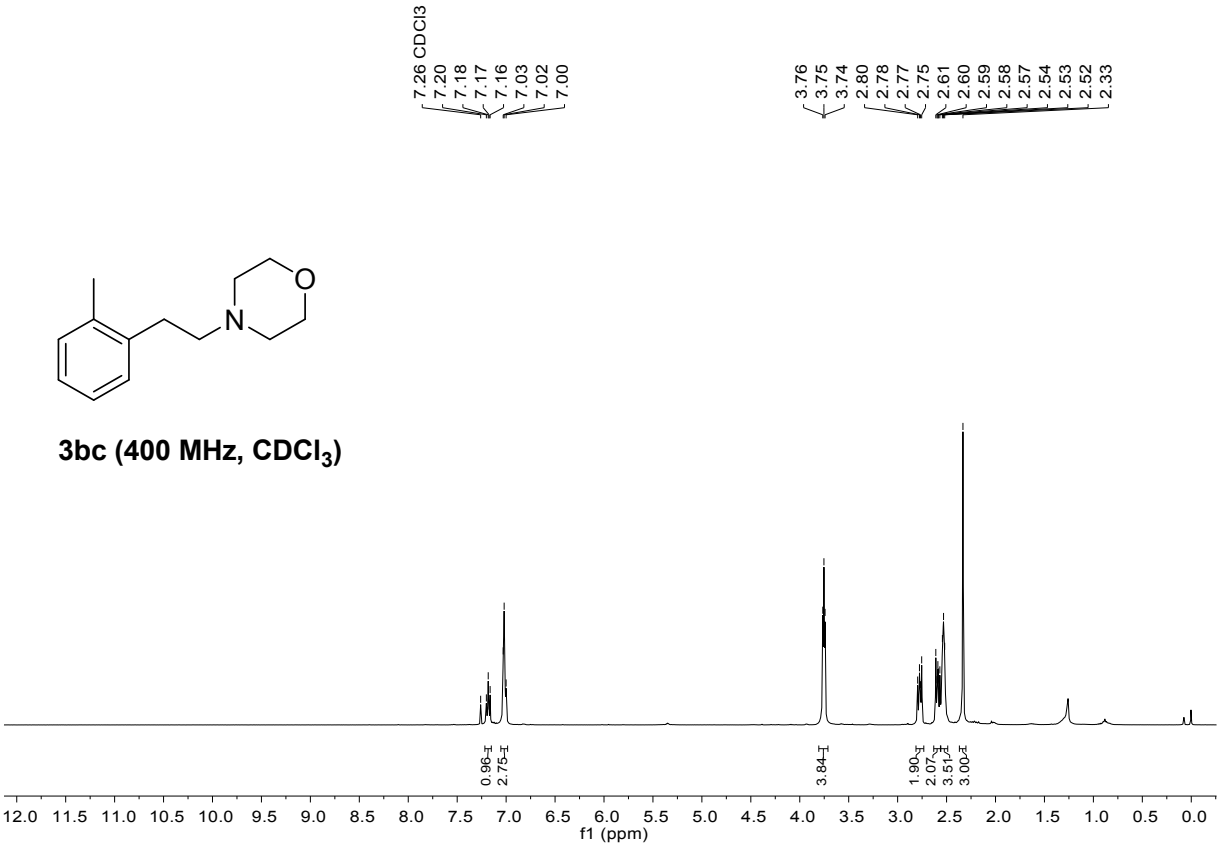




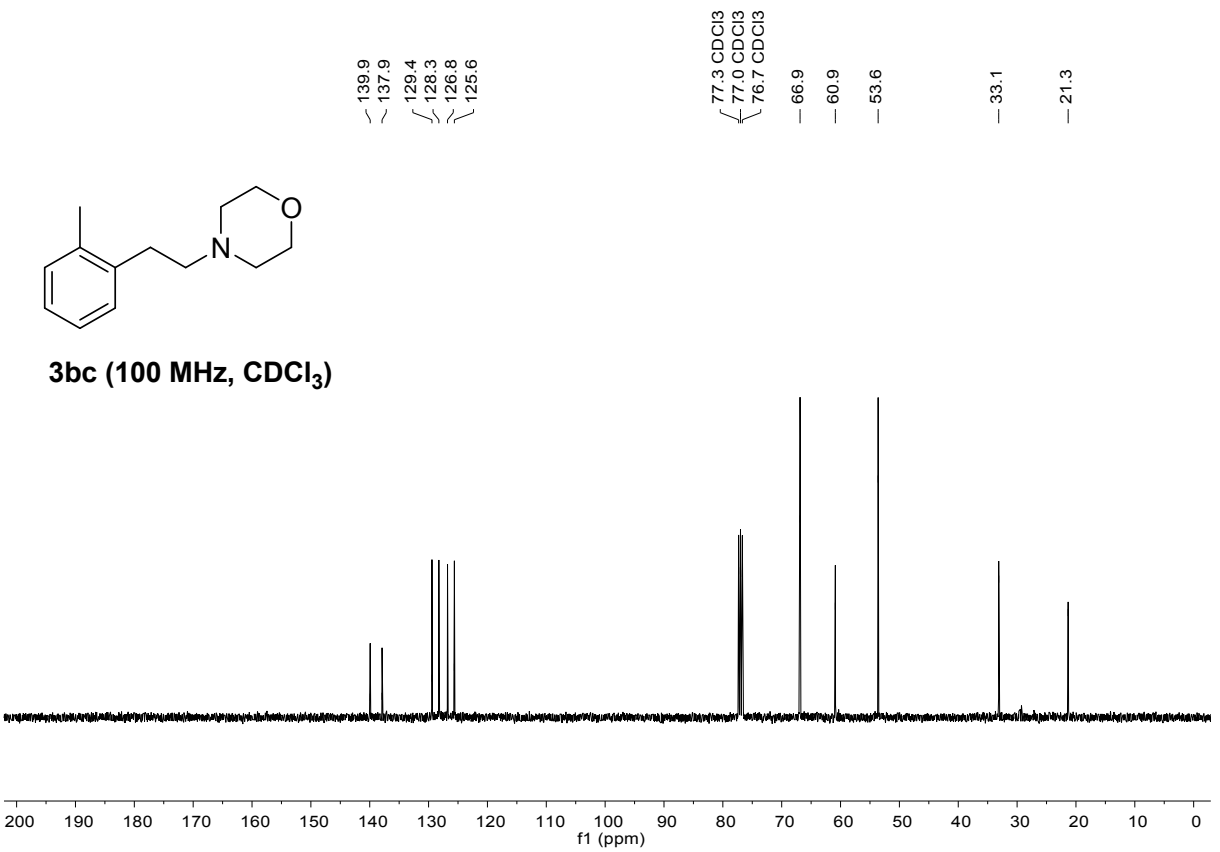


3ar (100 MHz, $CDCl_3$)

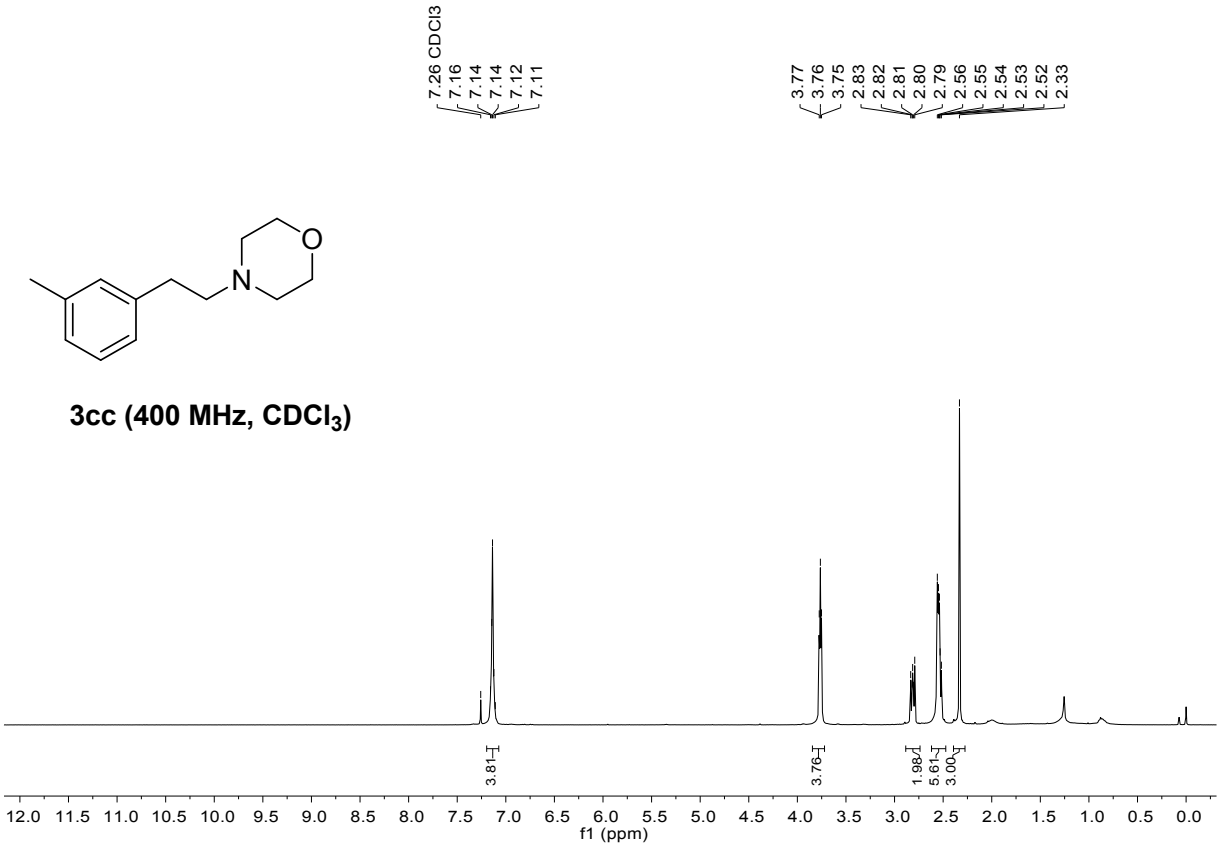




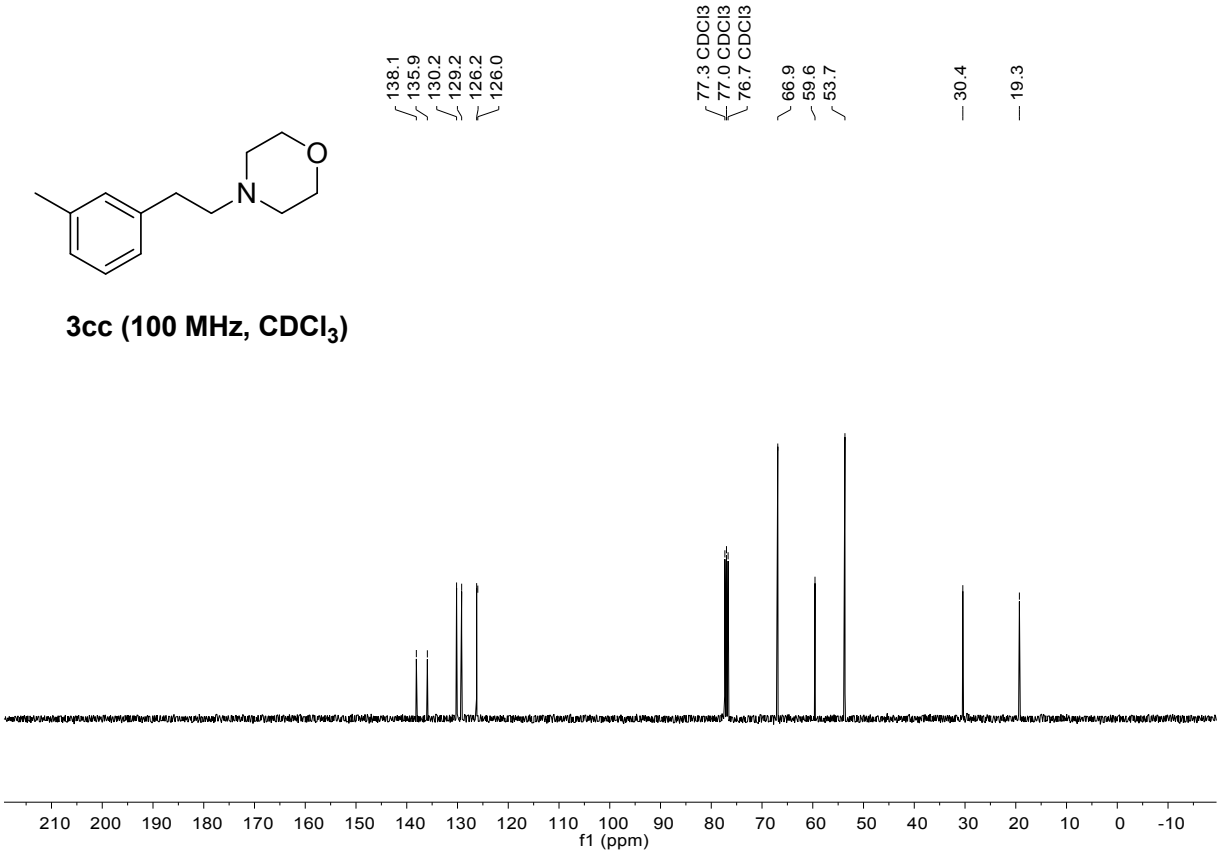
3bc (400 MHz, $CDCl_3$)



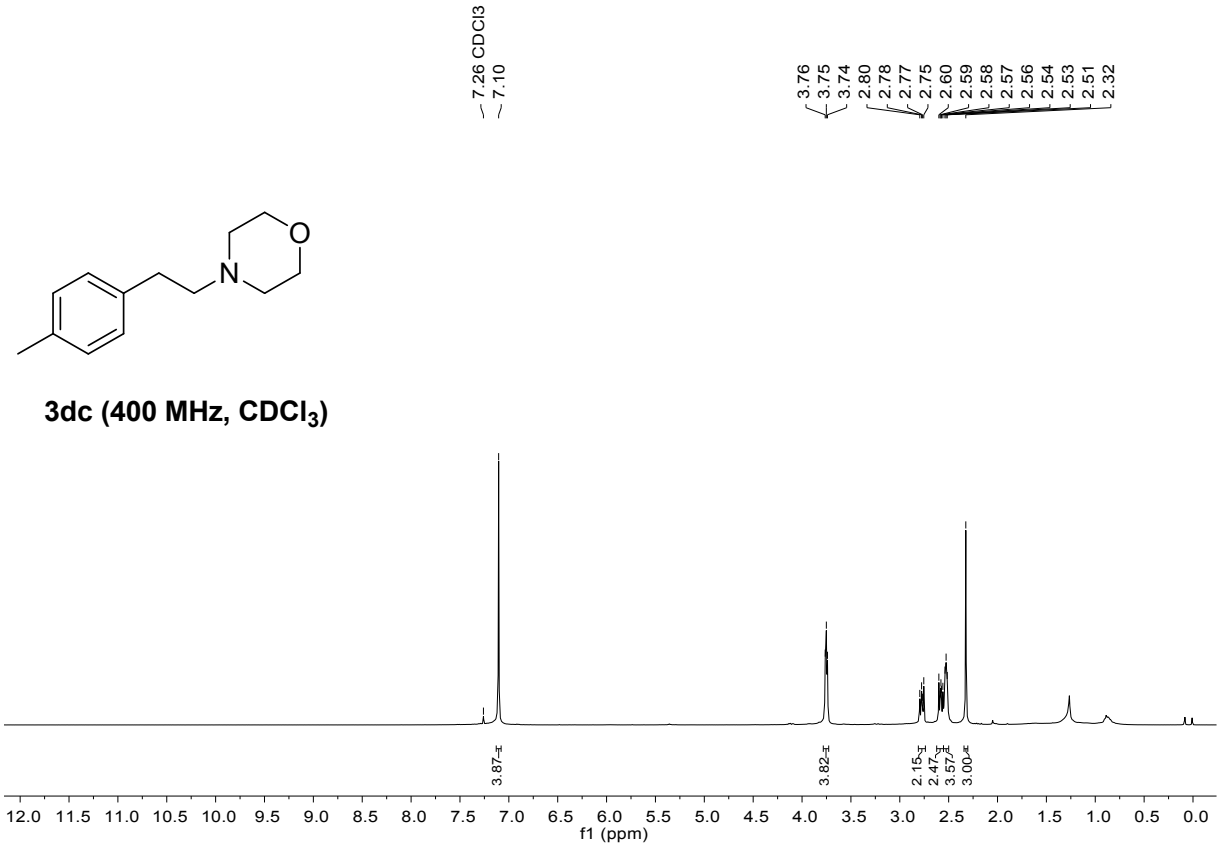
3bc (100 MHz, $CDCl_3$)

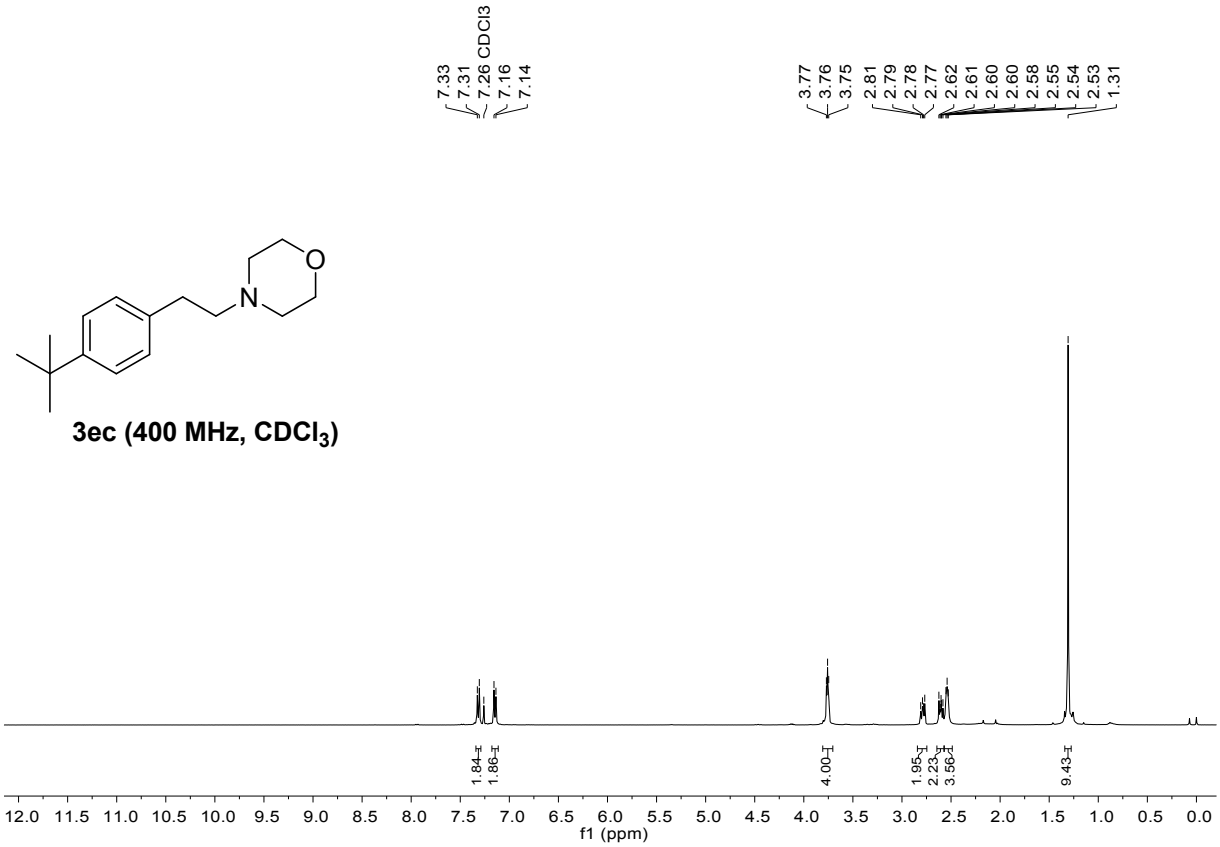


3cc (400 MHz, $CDCl_3$)

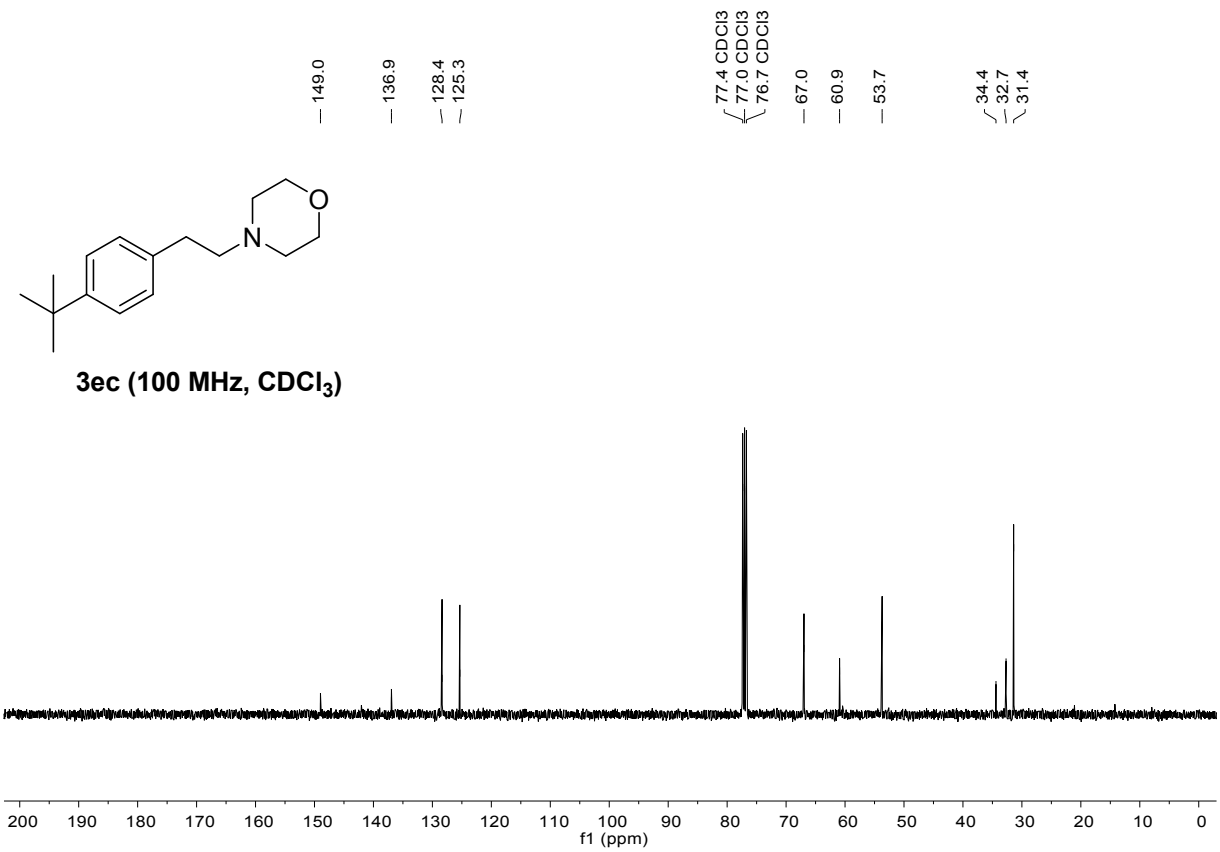


3cc (100 MHz, $CDCl_3$)

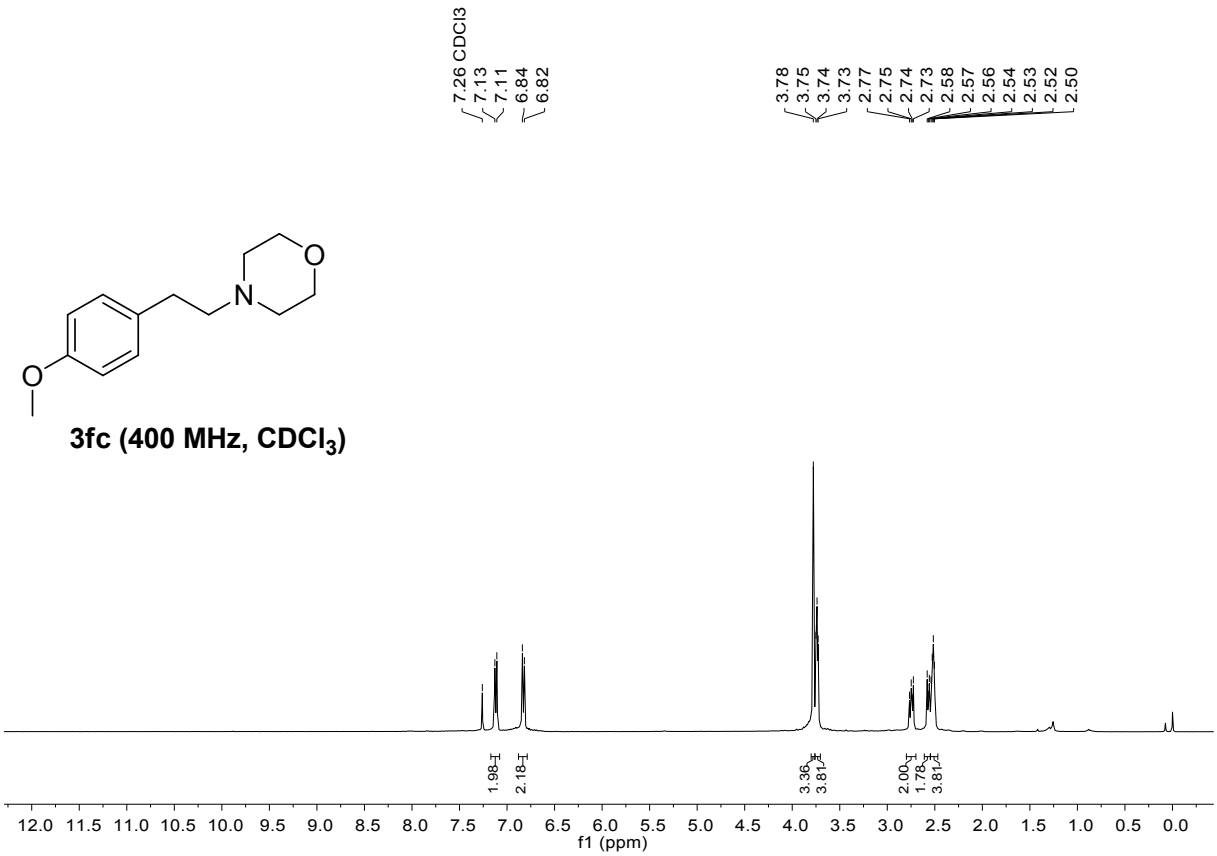

7.26 CDCl3
7.10
3.76
3.75
3.74
2.80
2.78
2.77
2.75
2.60
2.59
2.58
2.57
2.56
2.54
2.53
2.51
2.32
O
N
3dc (400 MHz, CDCl3)
3.87
3.82
2.15
2.47
3.57
3.00
12.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)
136.9
135.5
129.0
128.5
77.3 CDCl3
77.0 CDCl3
76.7 CDCl3
66.9
61.0
53.7
32.8
21.0
O
N
3dc (100 MHz, CDCl3)
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)



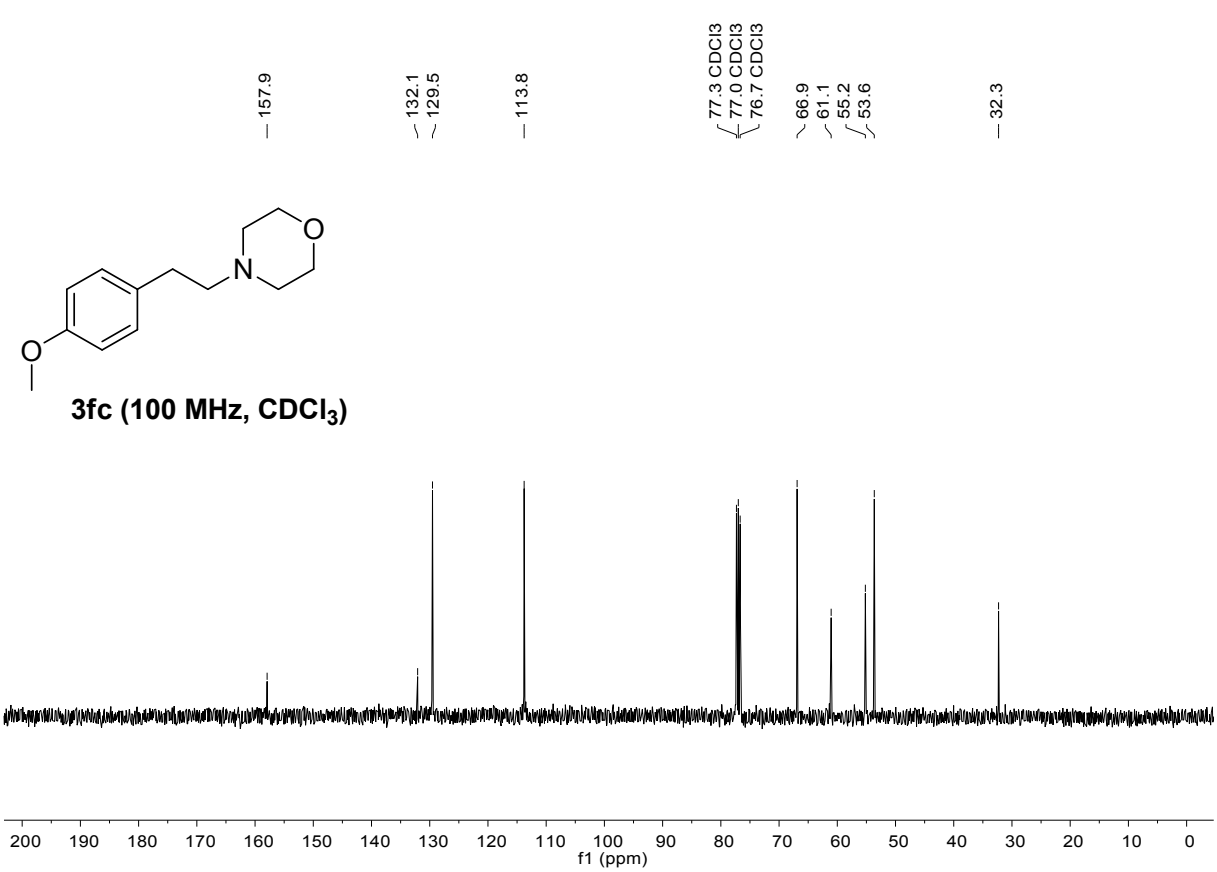
3ec (400 MHz, $CDCl_3$)



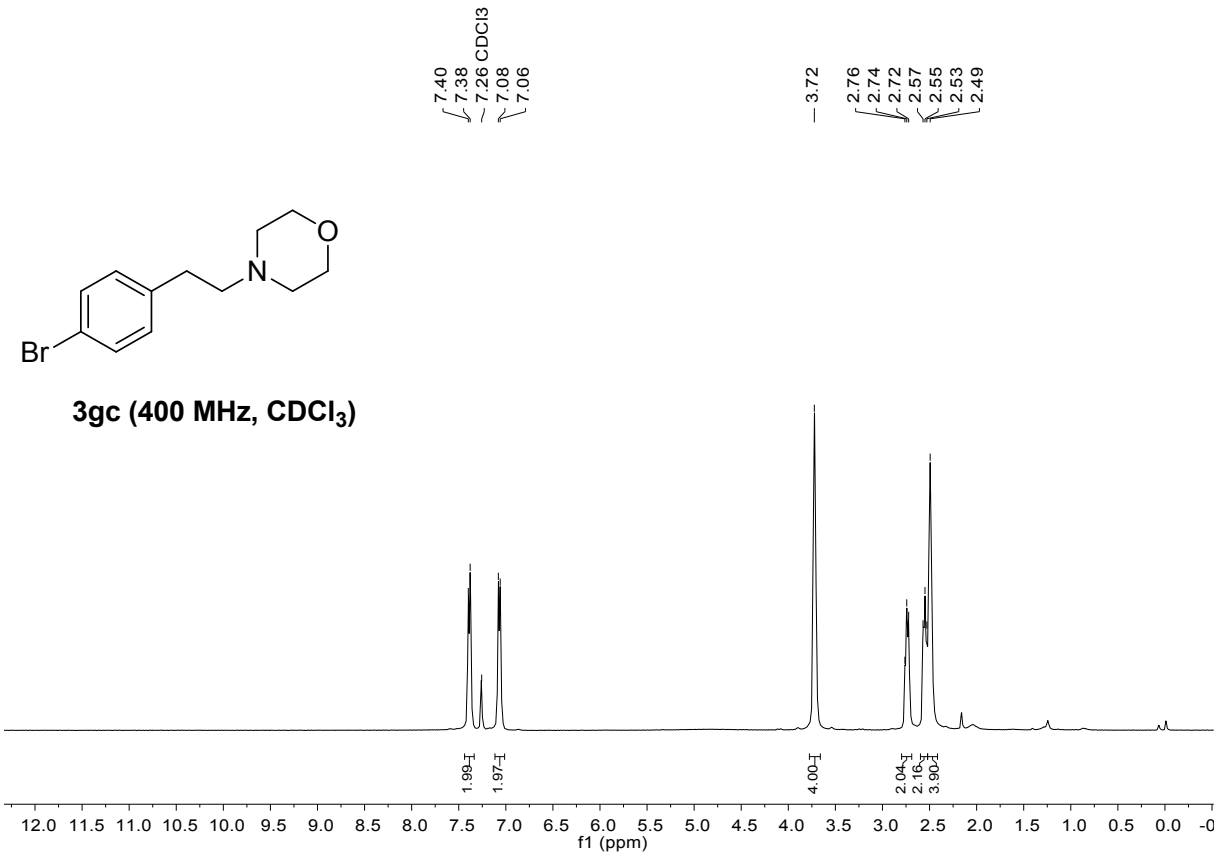
3ec (100 MHz, $CDCl_3$)



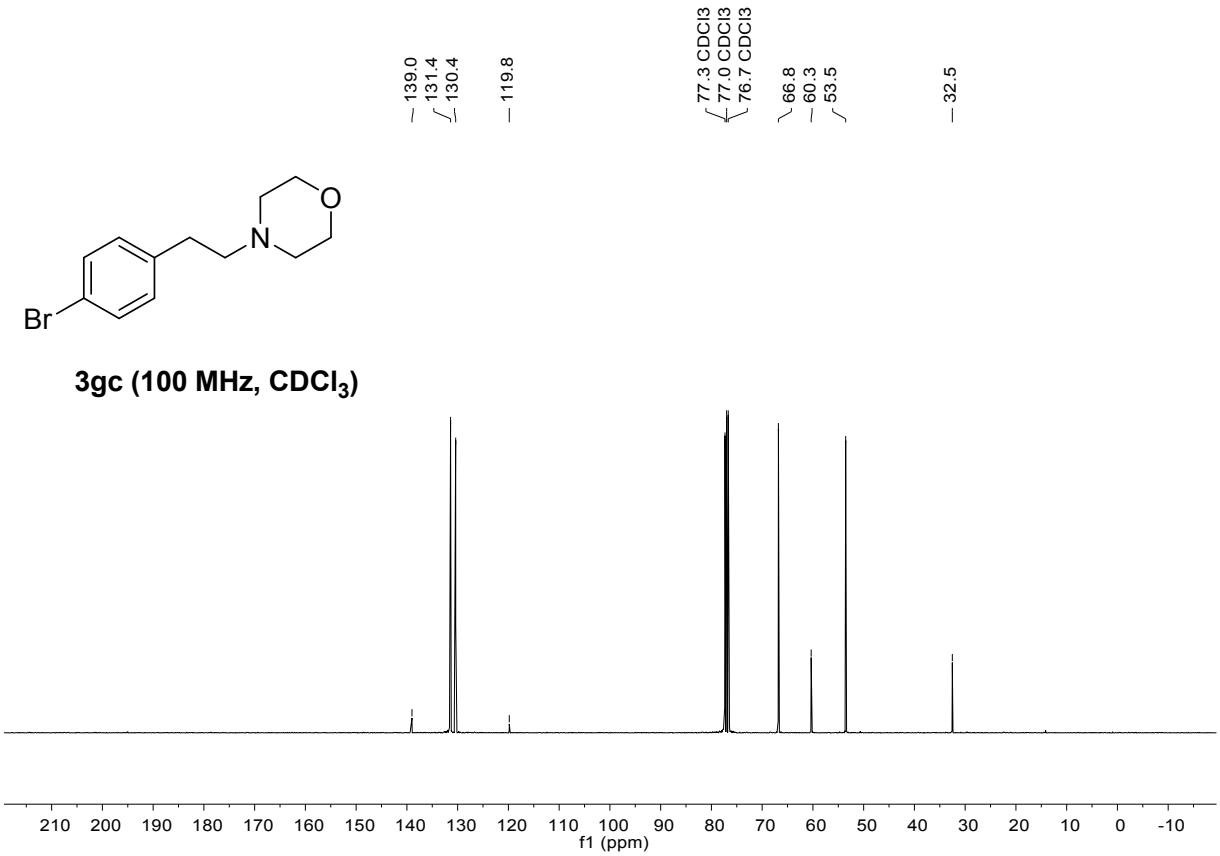
3fc (400 MHz, $CDCl_3$)



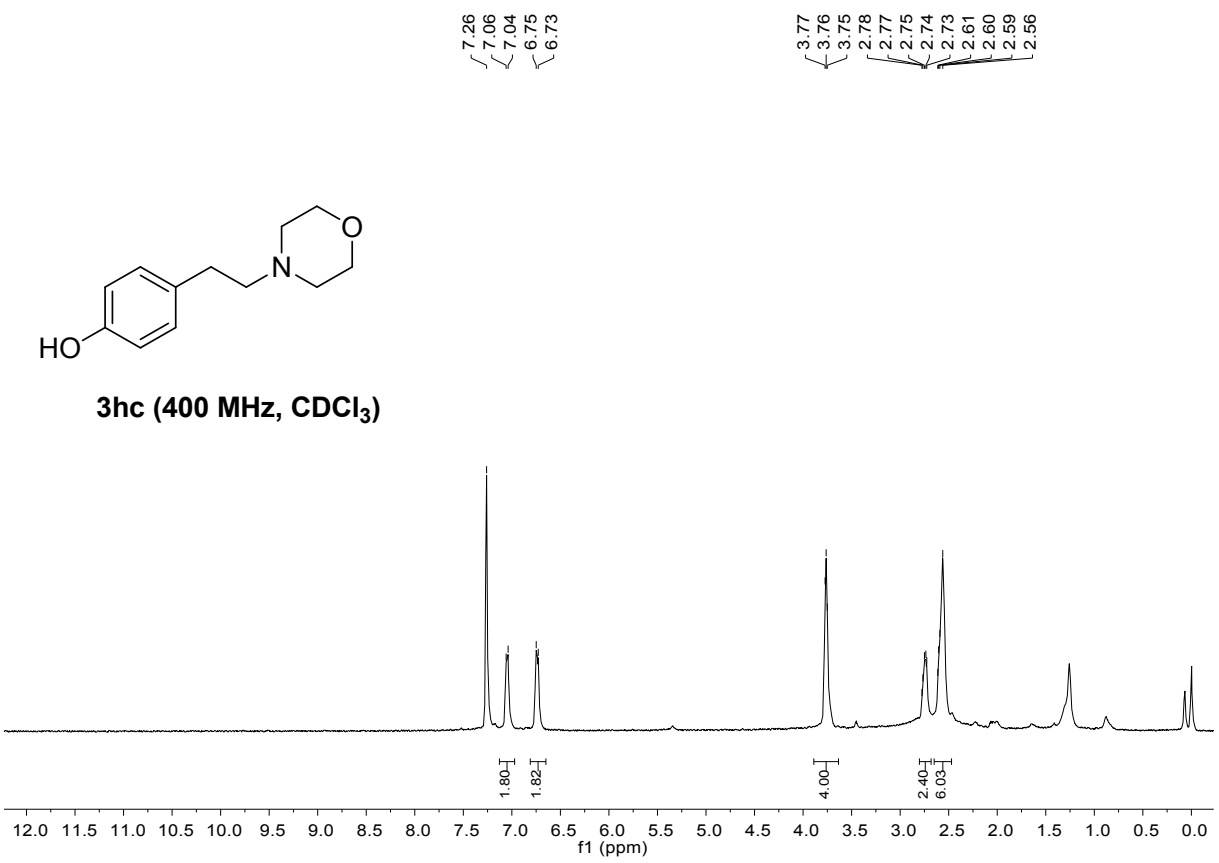
3fc (100 MHz, $CDCl_3$)



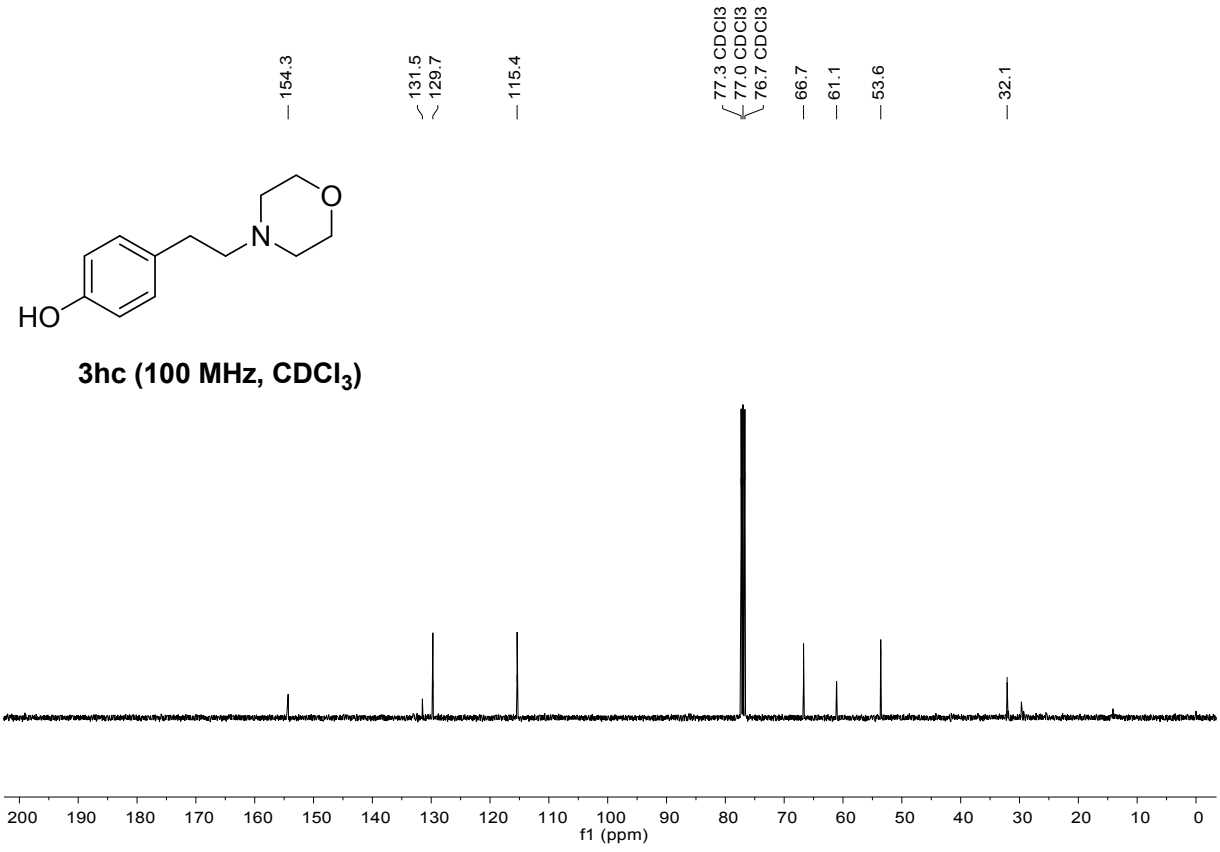
3gc (400 MHz, $CDCl_3$)



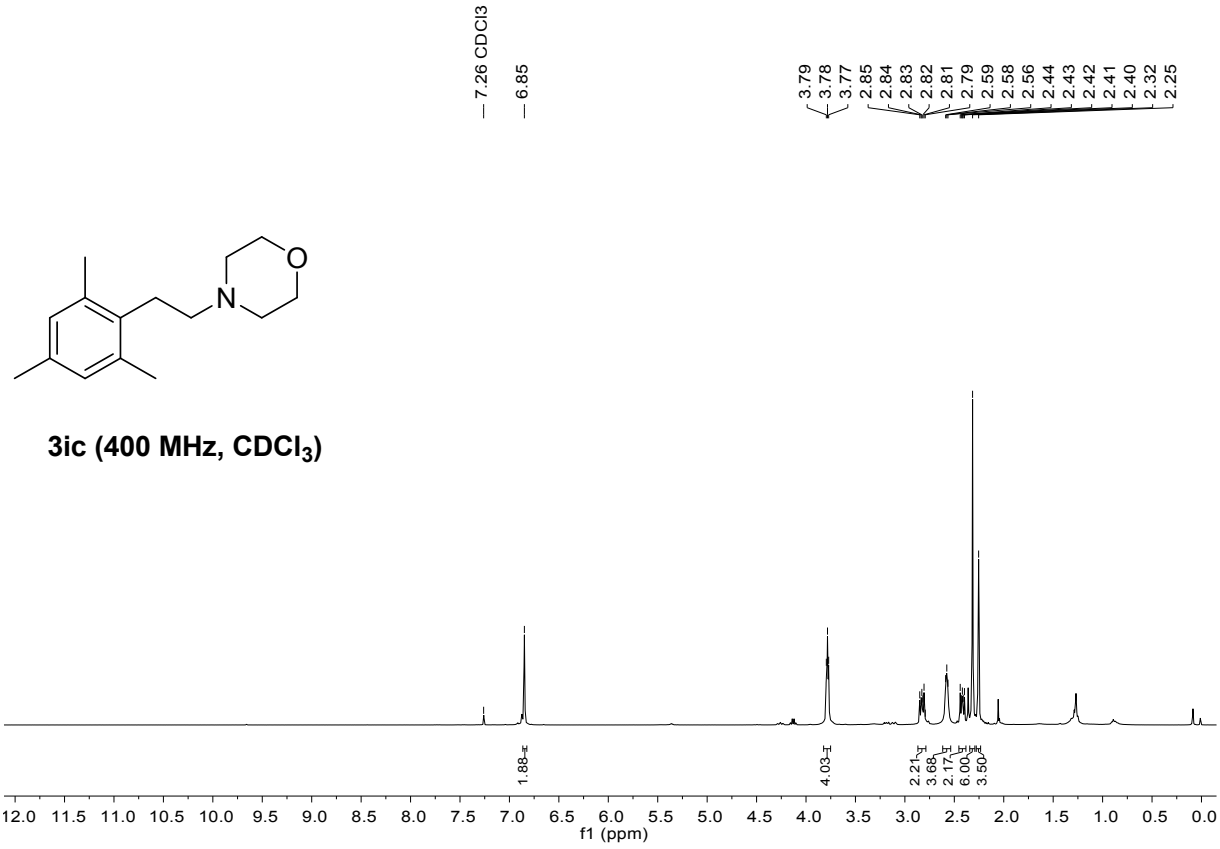
3gc (100 MHz, $CDCl_3$)



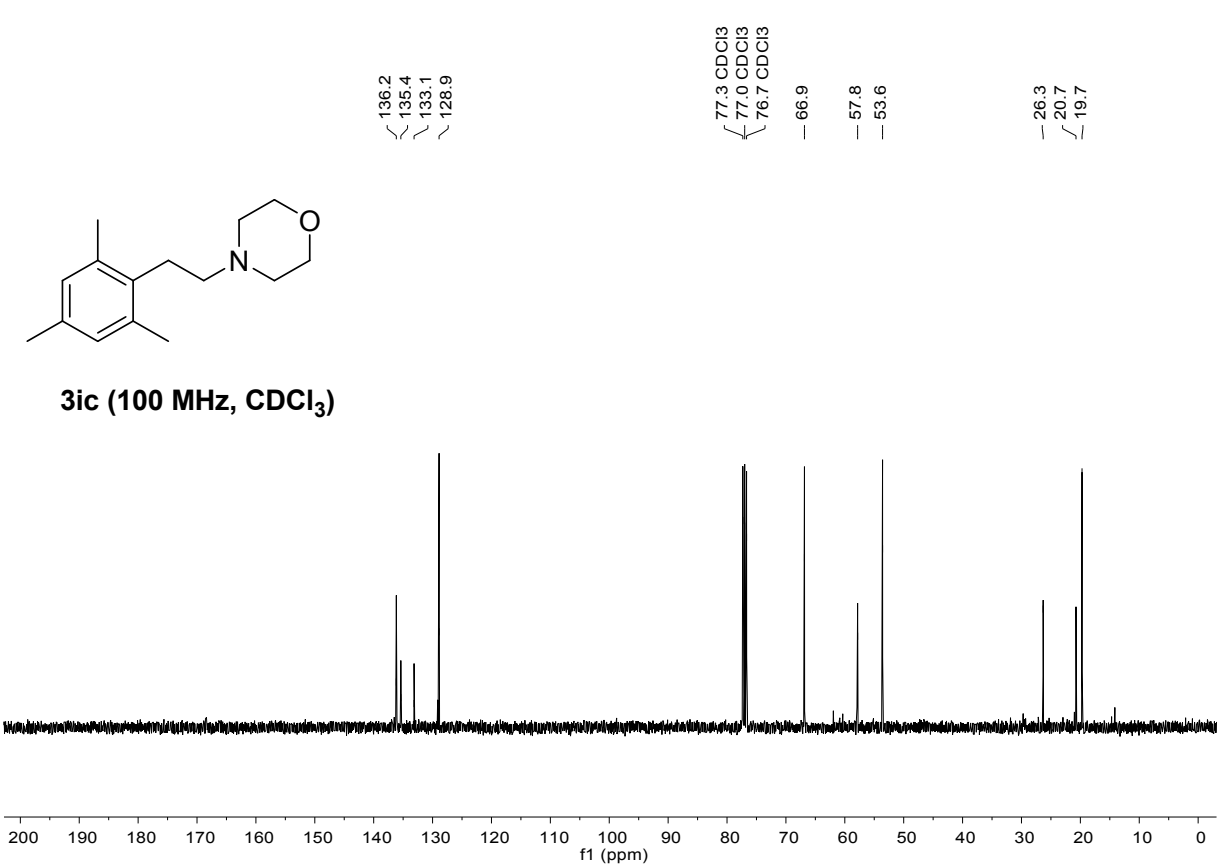
3hc (400 MHz, $CDCl_3$)



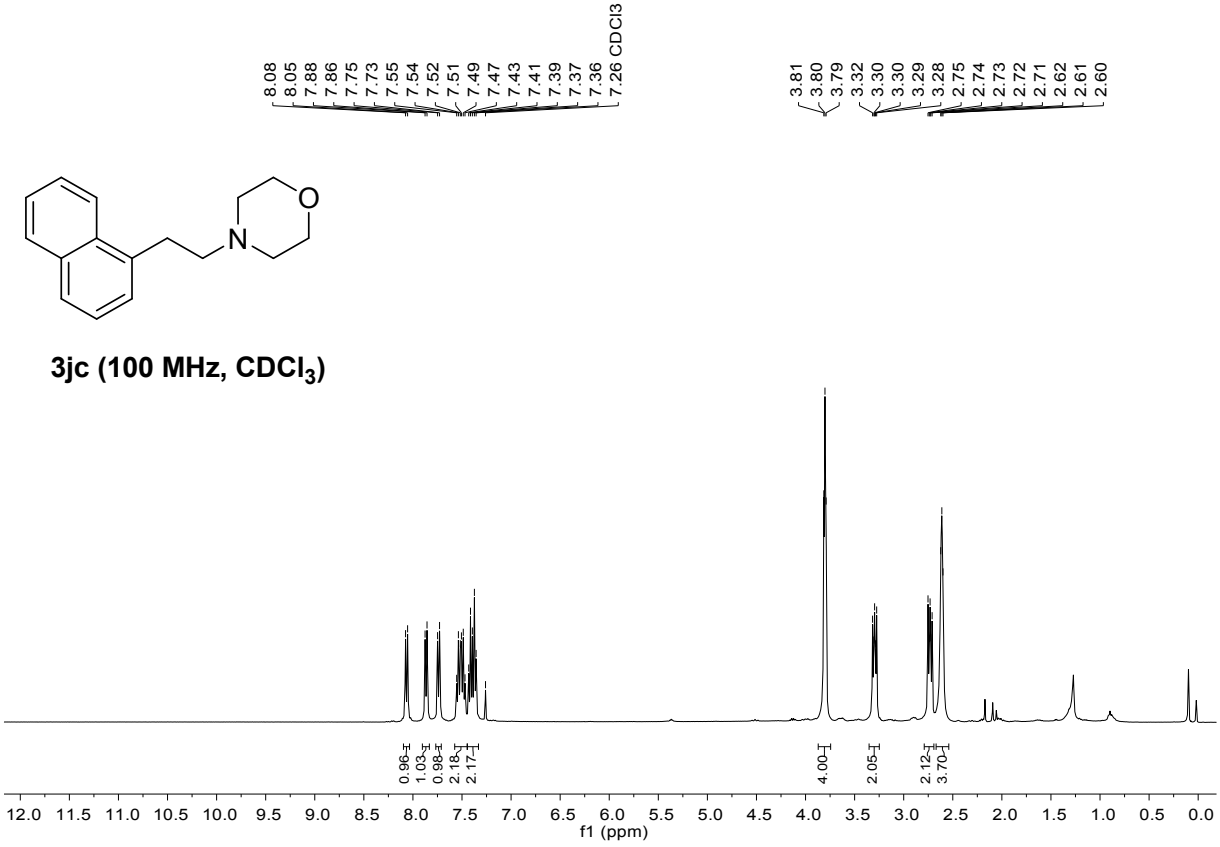
3hc (100 MHz, $CDCl_3$)



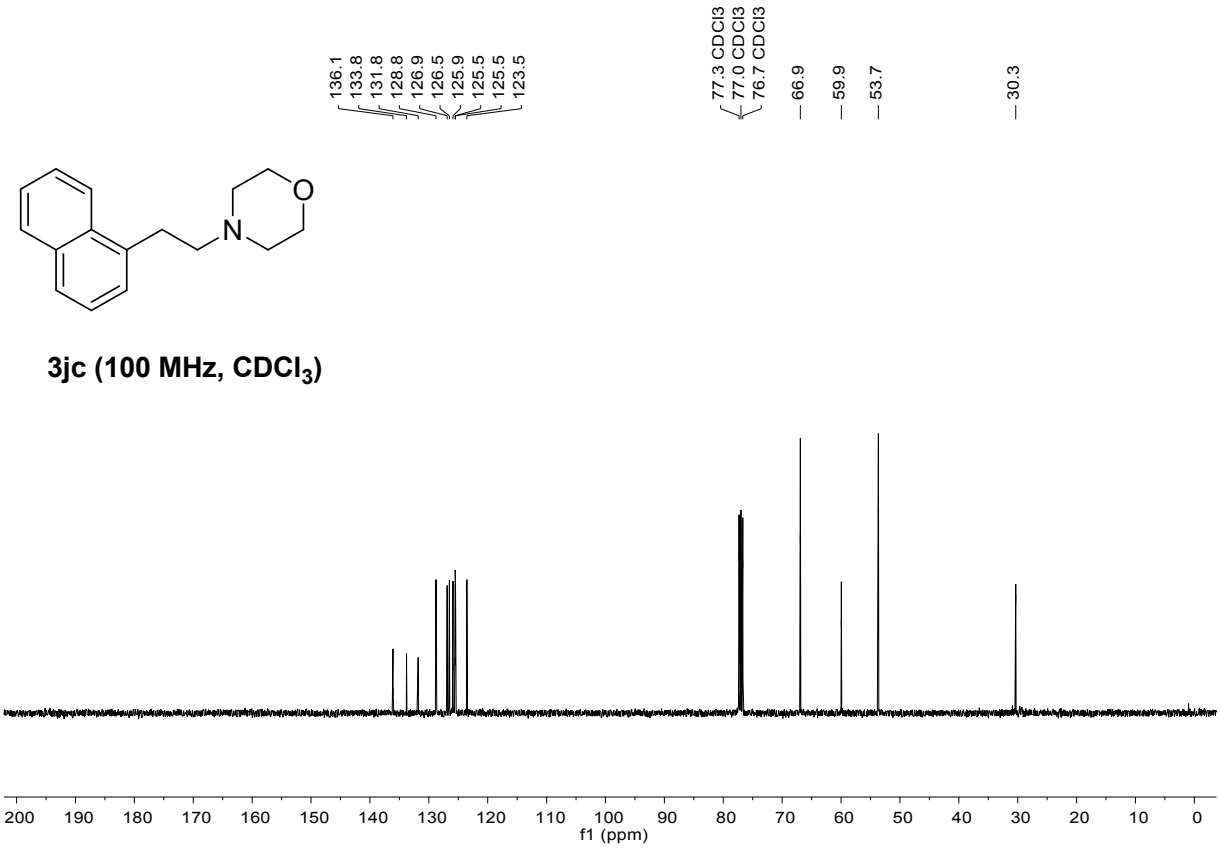
3ic (400 MHz, $CDCl_3$)



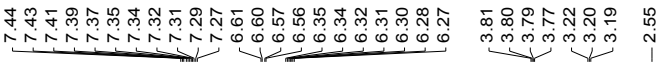
3ic (100 MHz, $CDCl_3$)

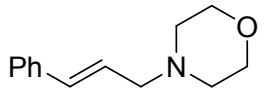


3jc (100 MHz, $CDCl_3$)

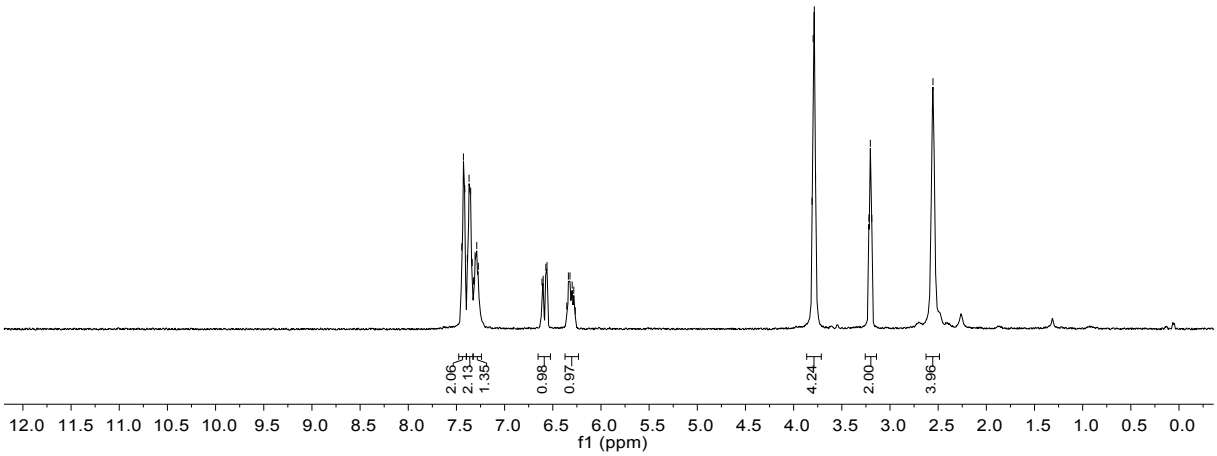


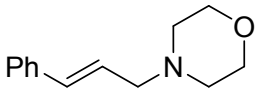
3jc (100 MHz, $CDCl_3$)



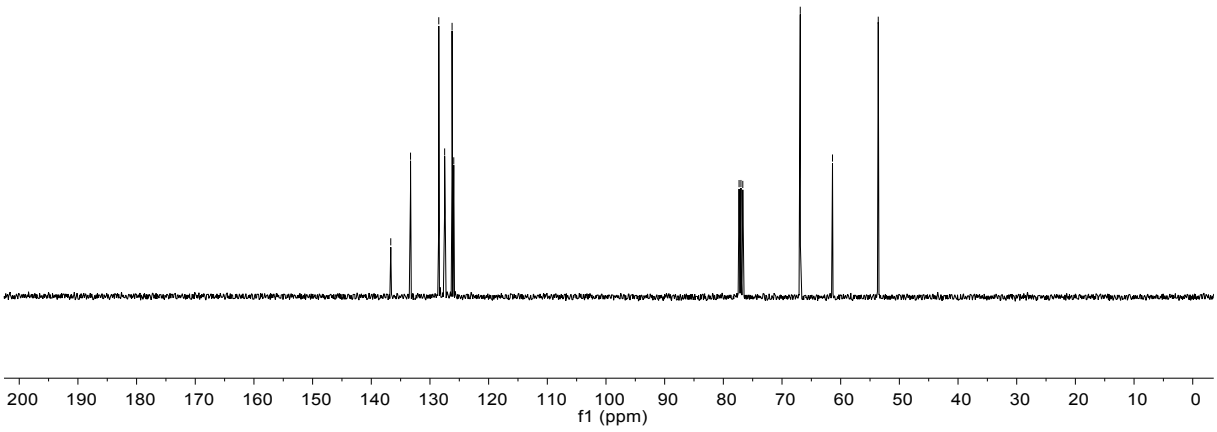


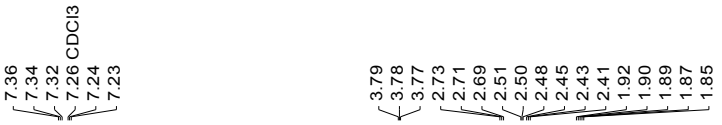
3kc (400 MHz, $CDCl_3$)

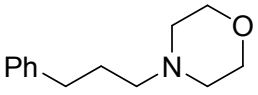




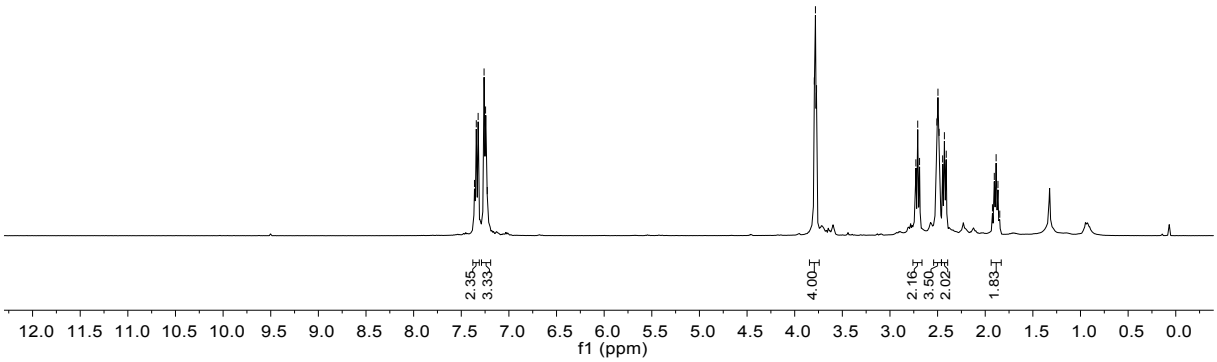
3kc (100 MHz, $CDCl_3$)

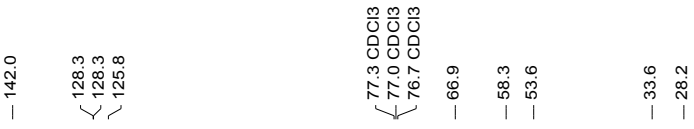


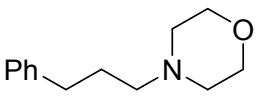




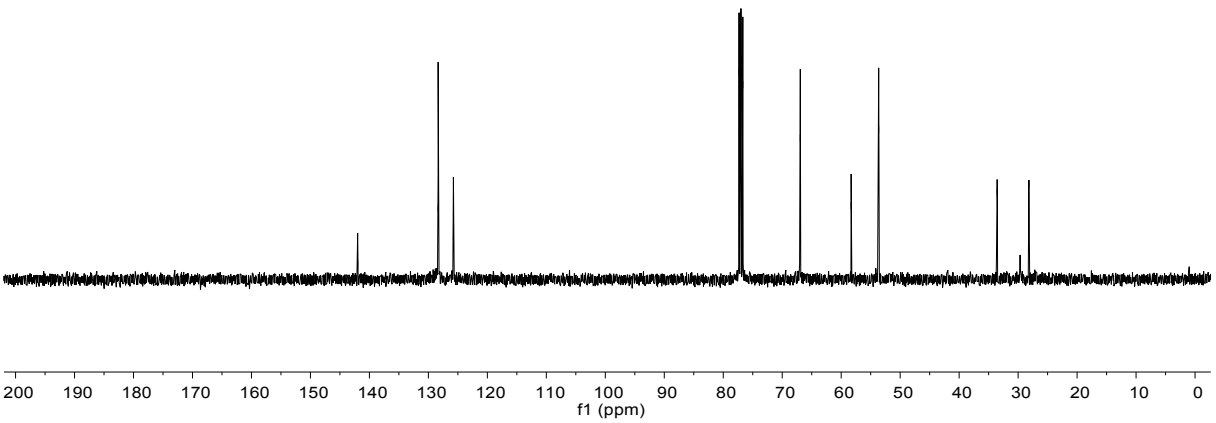
3lc (400 MHz, $CDCl_3$)

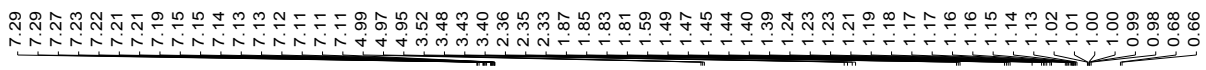


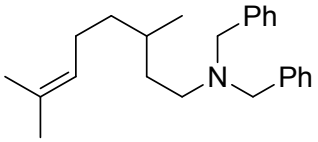




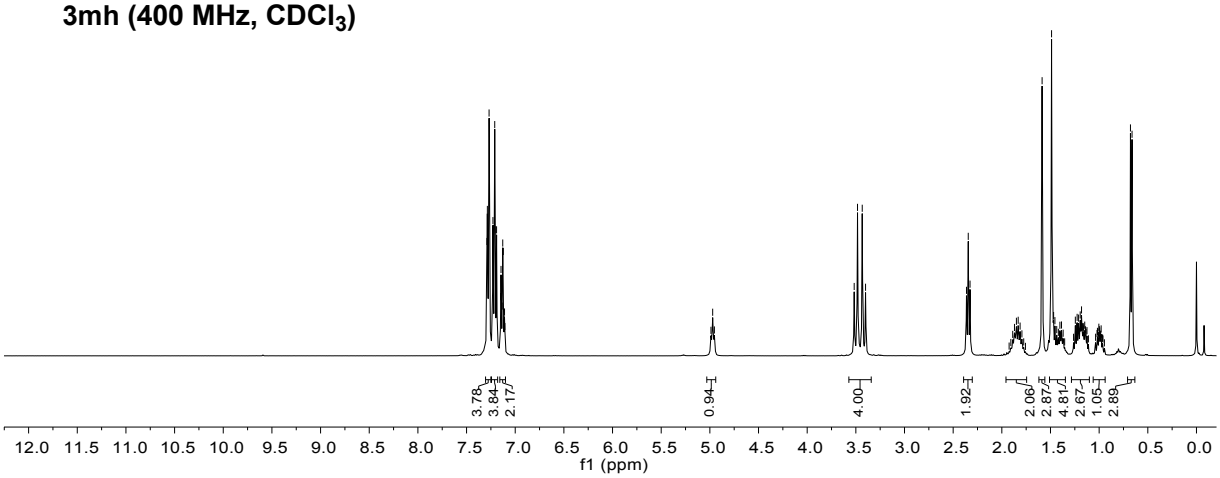
3lc (100 MHz, $CDCl_3$)

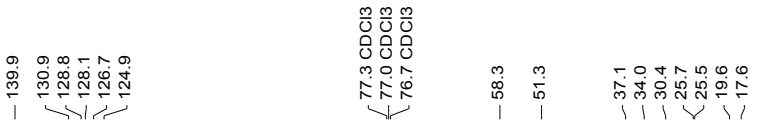


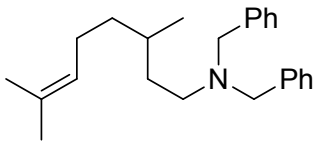




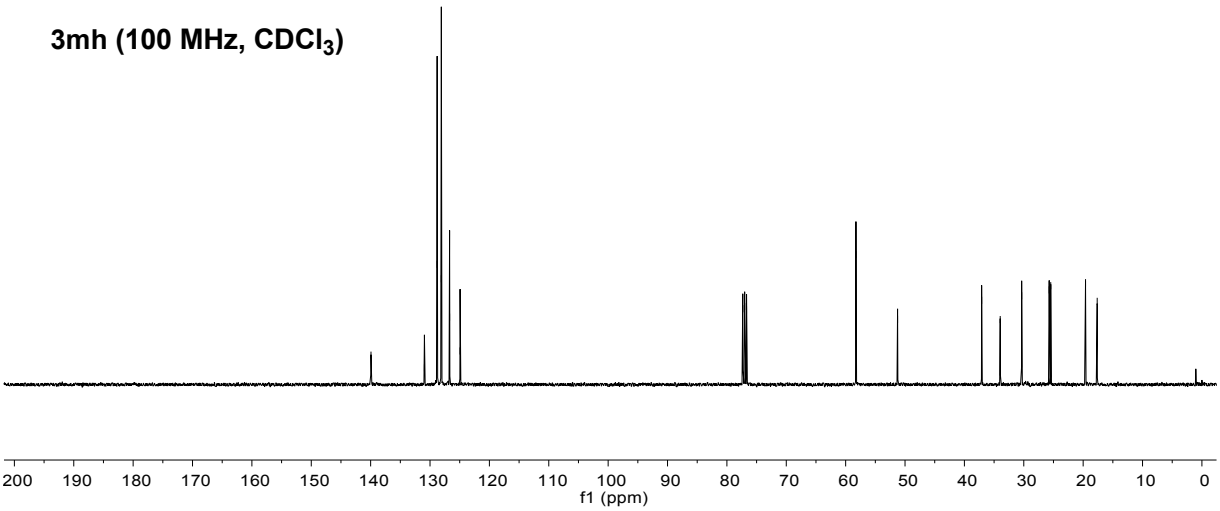
3mh (400 MHz, $CDCl_3$)

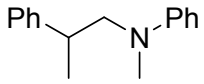




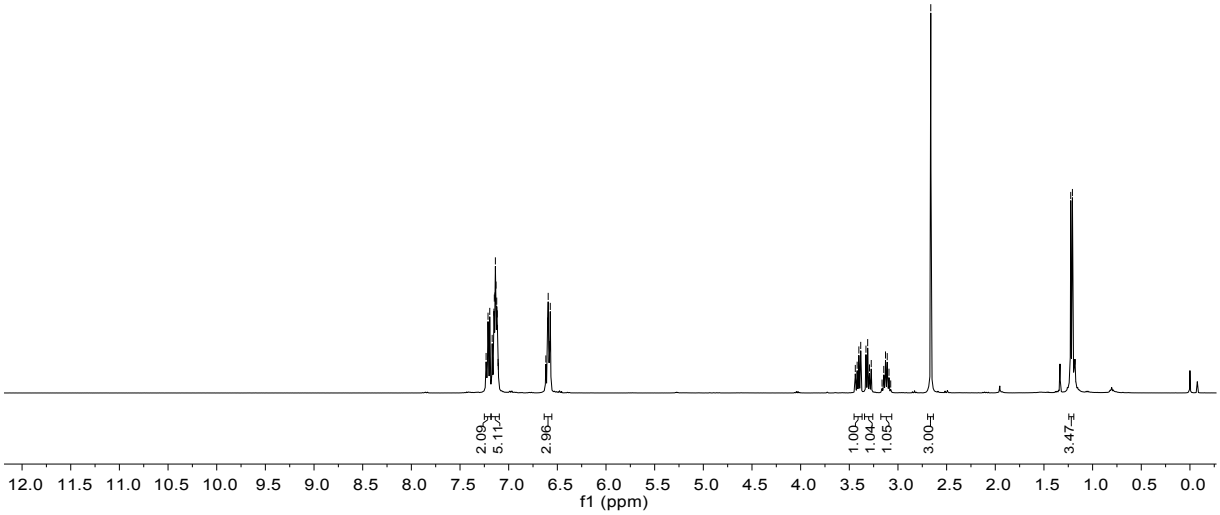


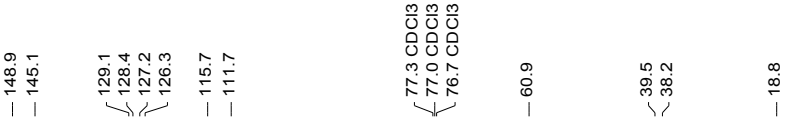
3mh (100 MHz, $CDCl_3$)

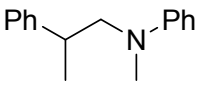




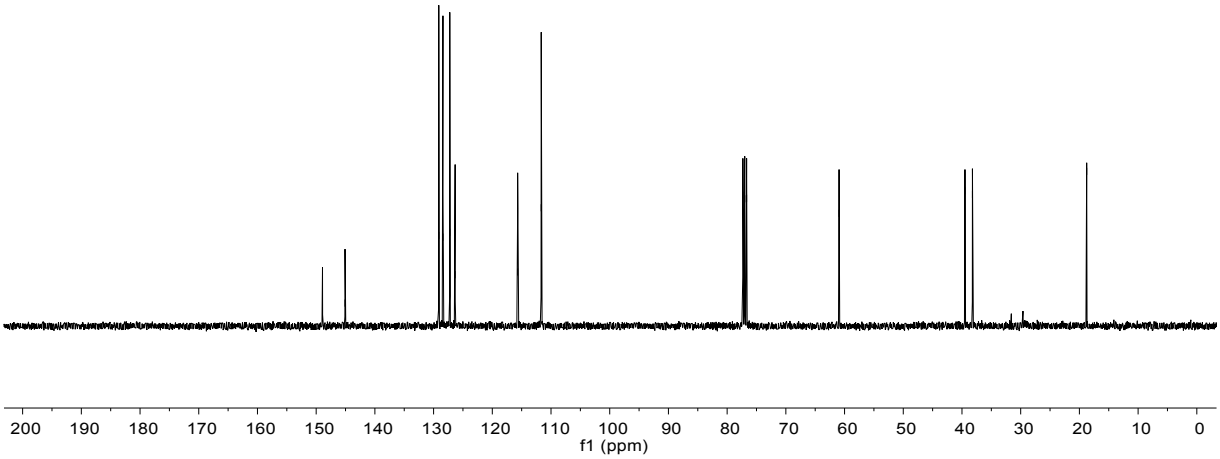
3ns (400 MHz, $CDCl_3$)

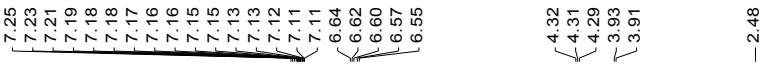


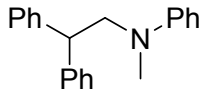




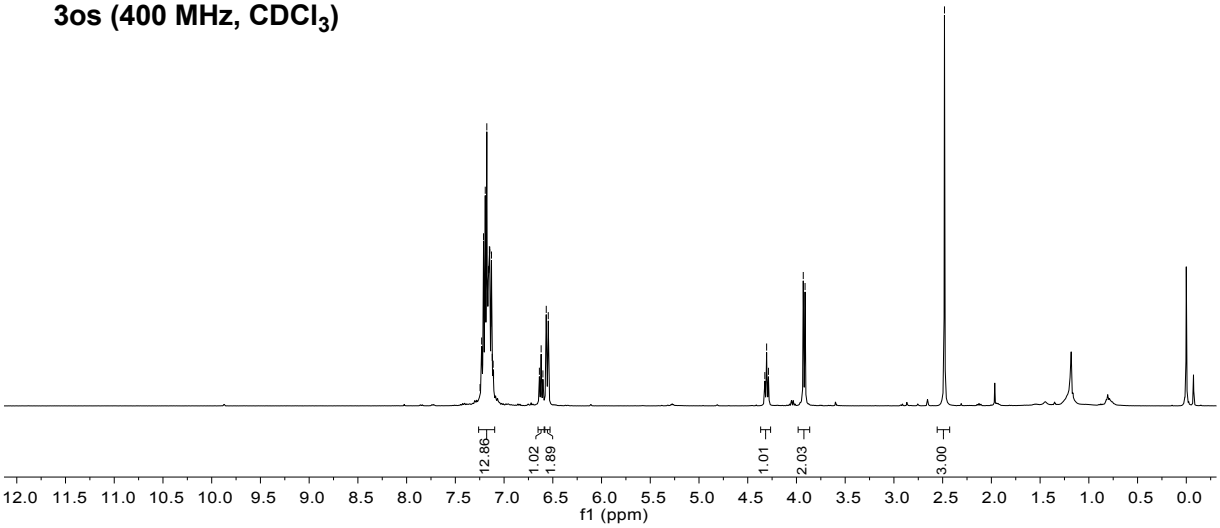
3ns (100 MHz, $CDCl_3$)

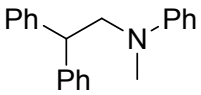




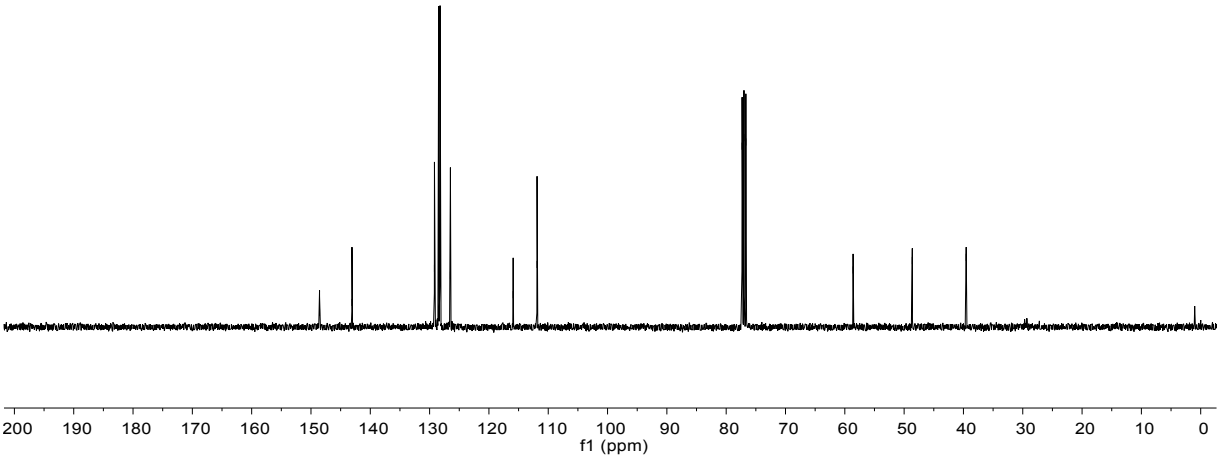


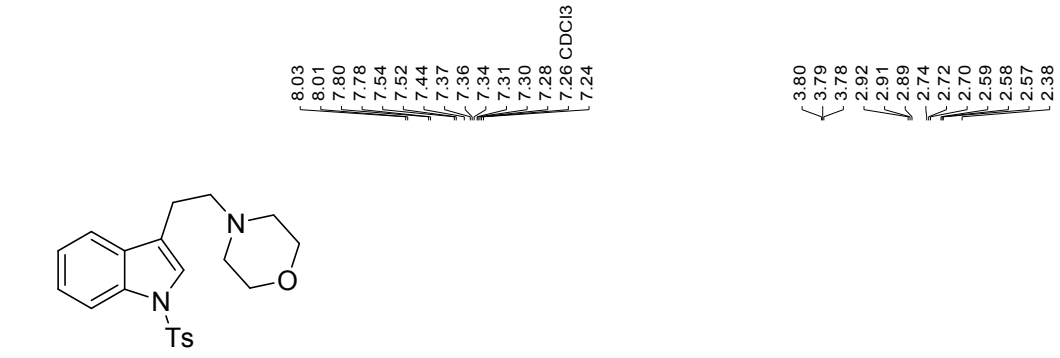
3os (400 MHz, $CDCl_3$)



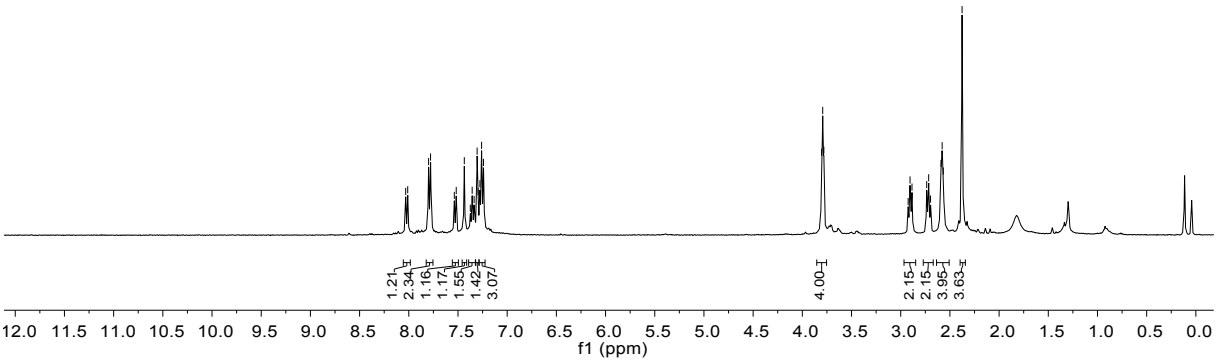


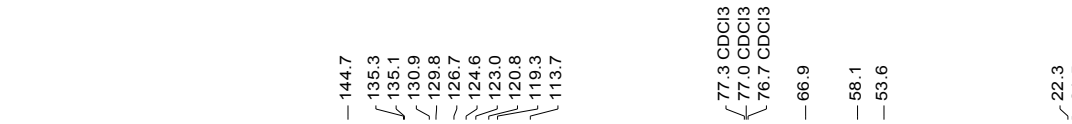
3os (100 MHz, $CDCl_3$)

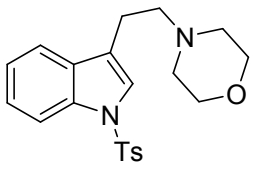




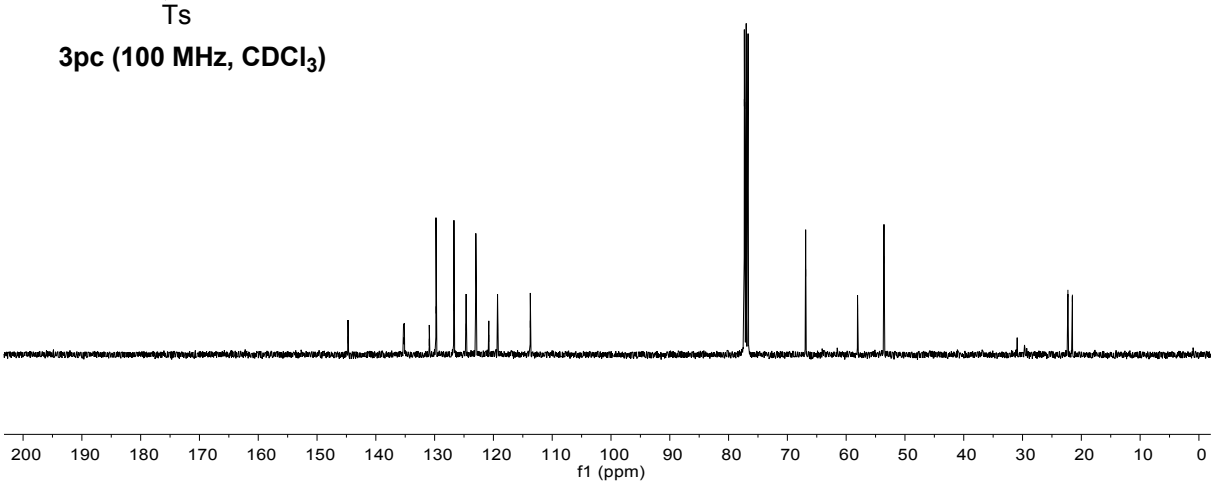
3pc (400 MHz, $CDCl_3$)

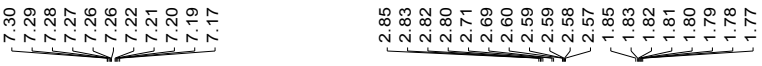


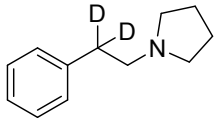




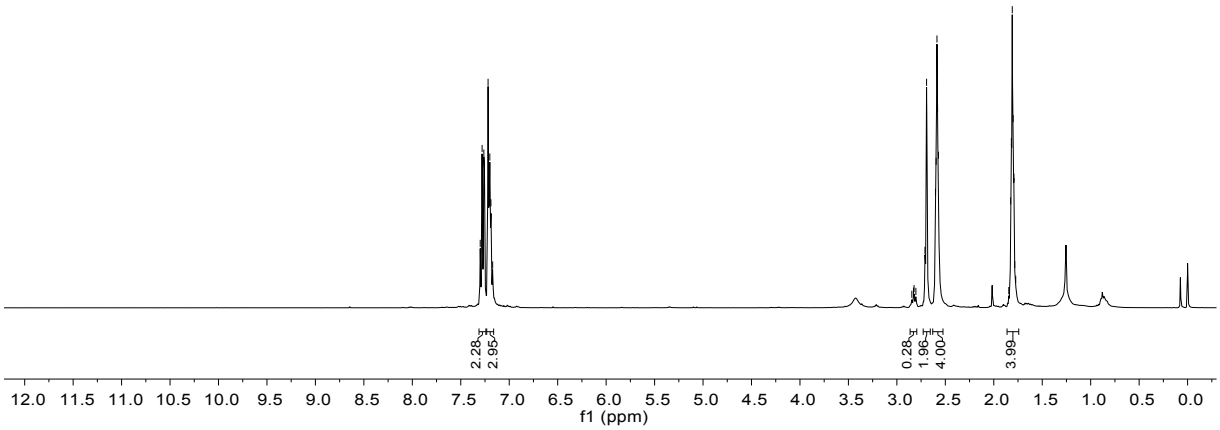
3pc (100 MHz, $CDCl_3$)

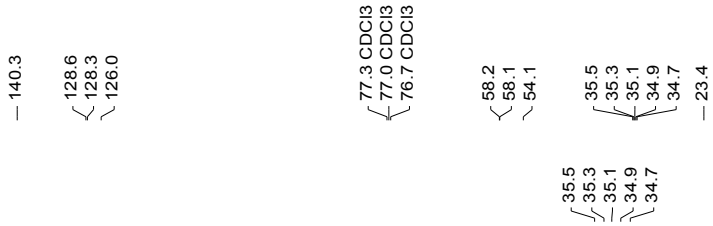


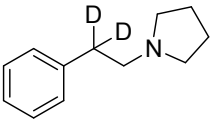




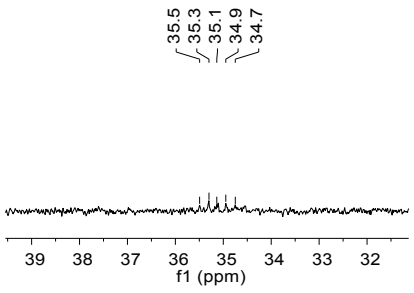
3aa-*d* (400 MHz, $CDCl_3$)

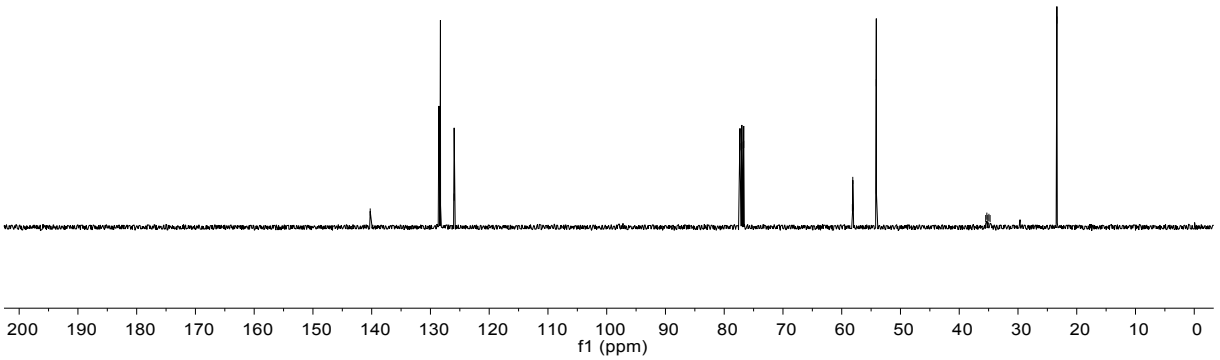


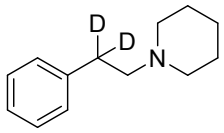




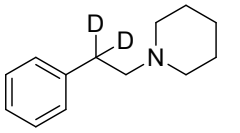
3aa-*d* (100 MHz, $CDCl_3$)



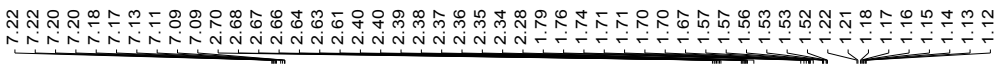


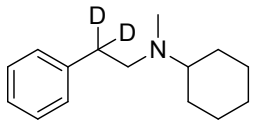


3ab-*d* (400 MHz, $CDCl_3$)

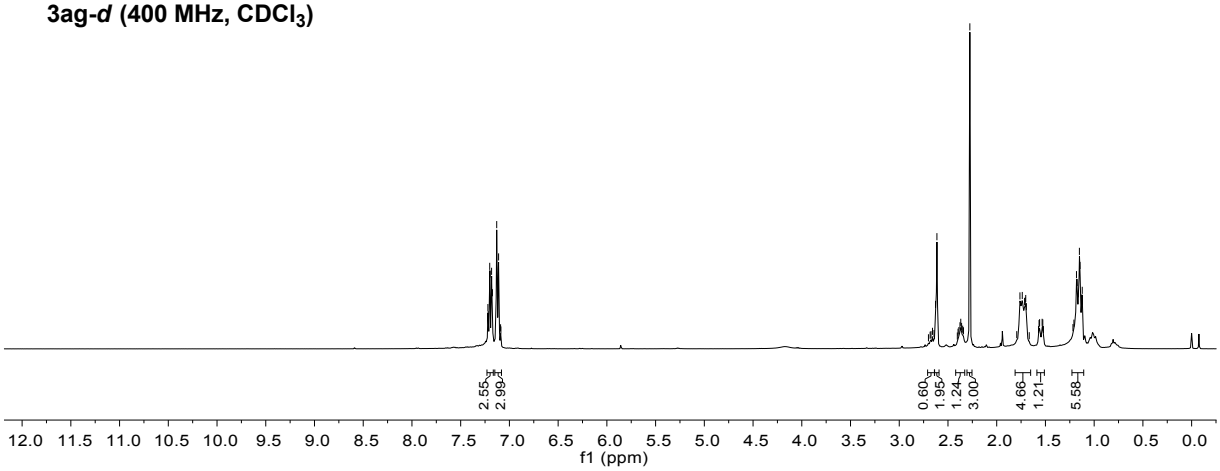


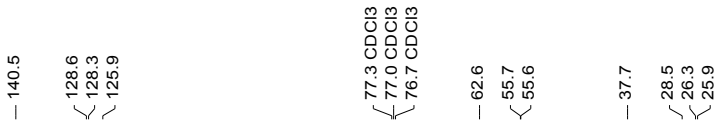
3ab-*d* (100 MHz, $CDCl_3$)

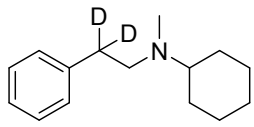




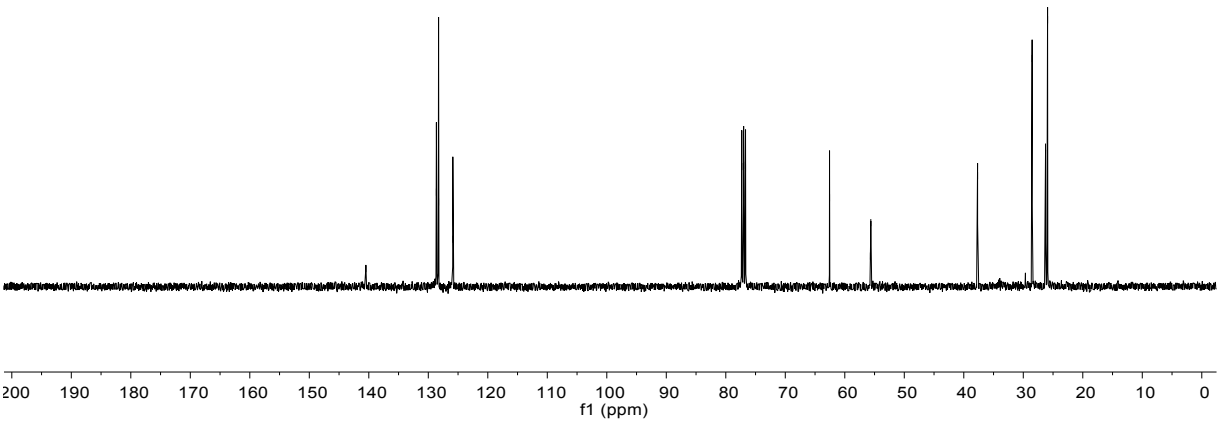
3ag-*d* (400 MHz, $CDCl_3$)

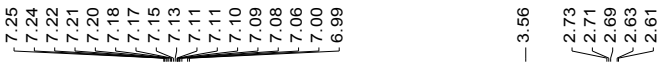


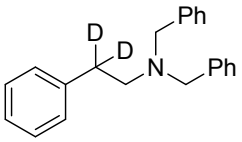




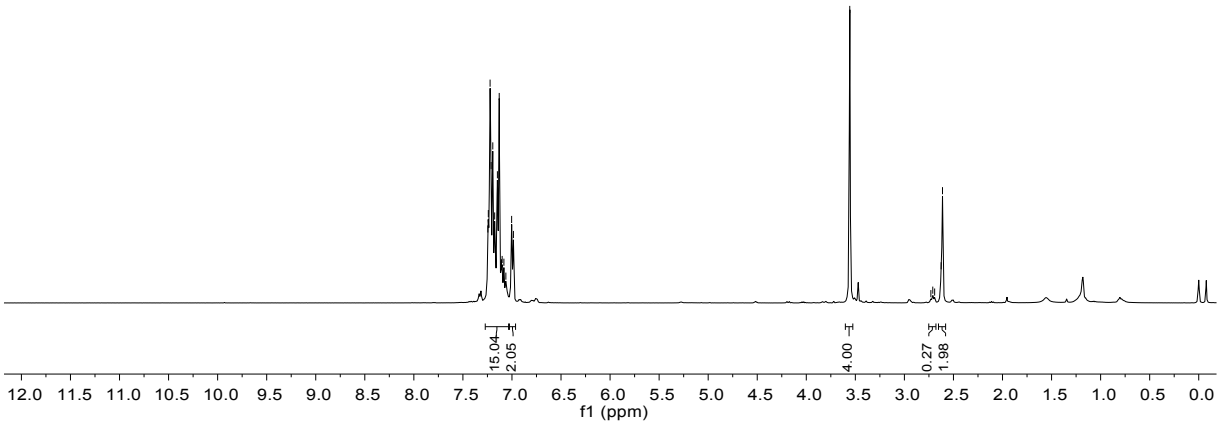
3ag-*d* (100 MHz, $CDCl_3$)

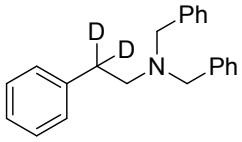




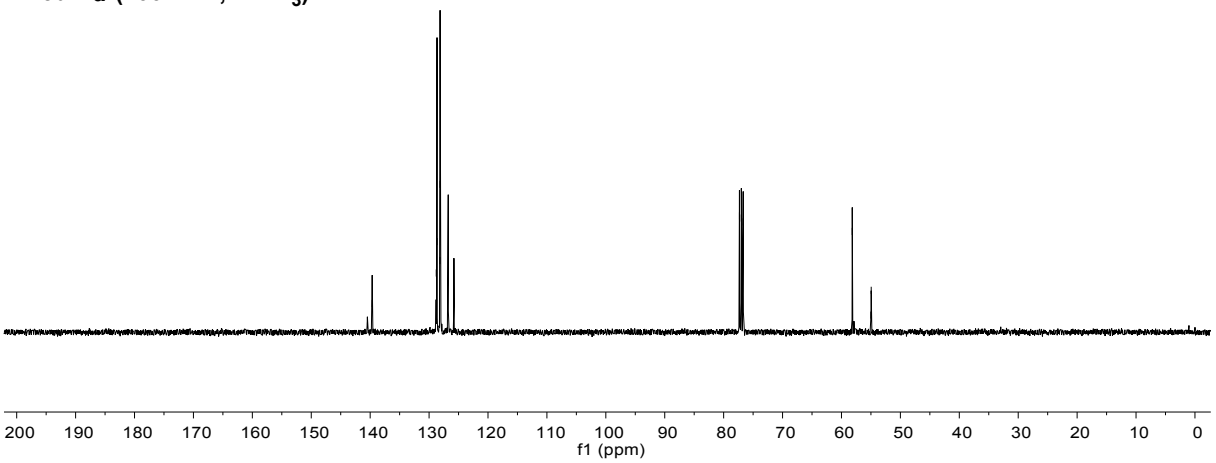


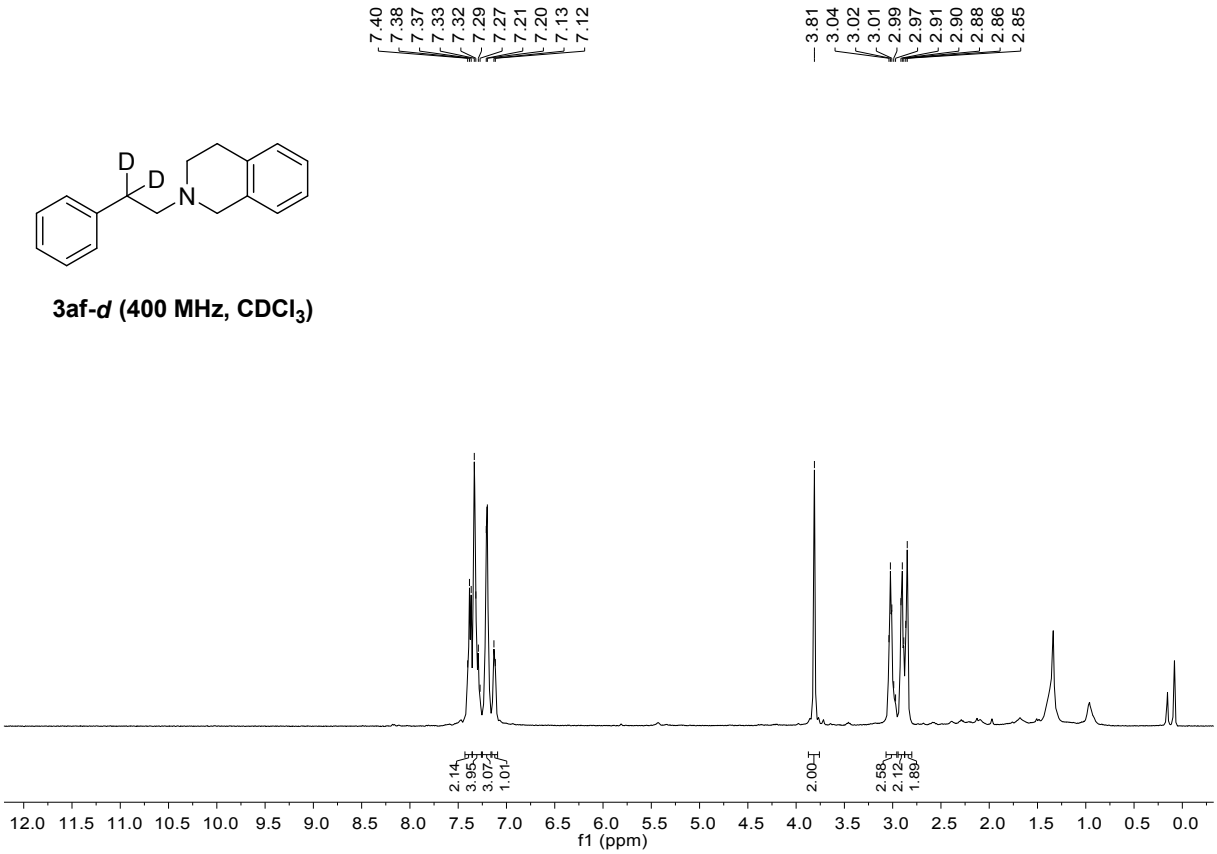
3dh-*d* (400 MHz, $CDCl_3$)



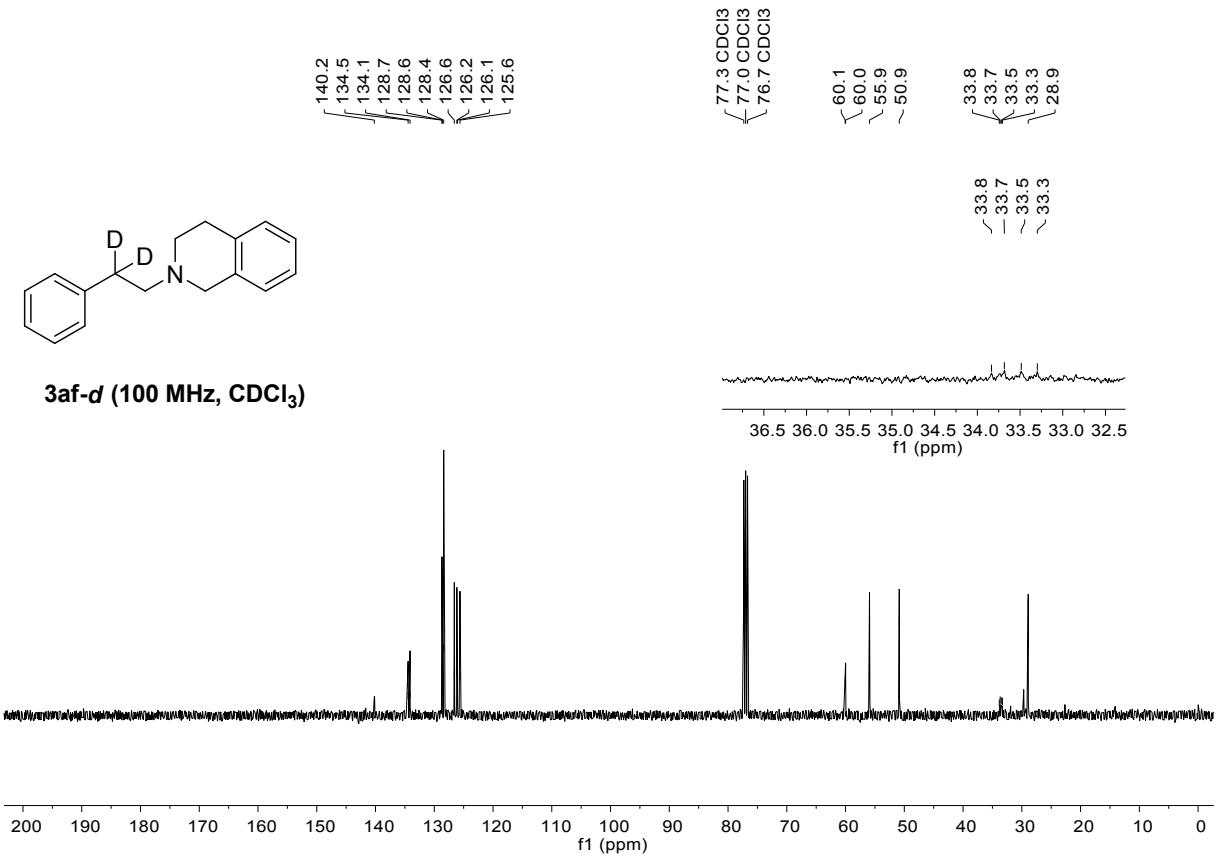


3dh-*d* (100 MHz, $CDCl_3$)

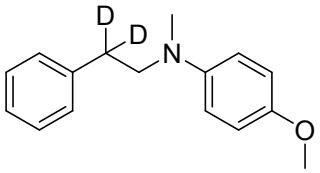




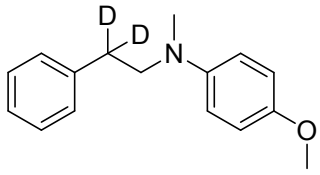
3af-*d* (400 MHz, $CDCl_3$)



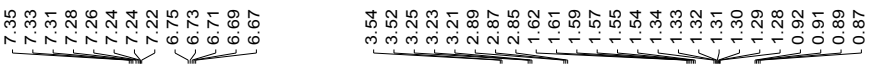
3af-*d* (100 MHz, $CDCl_3$)

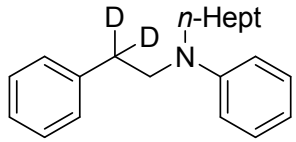


3aj-*d* (400 MHz, $CDCl_3$)

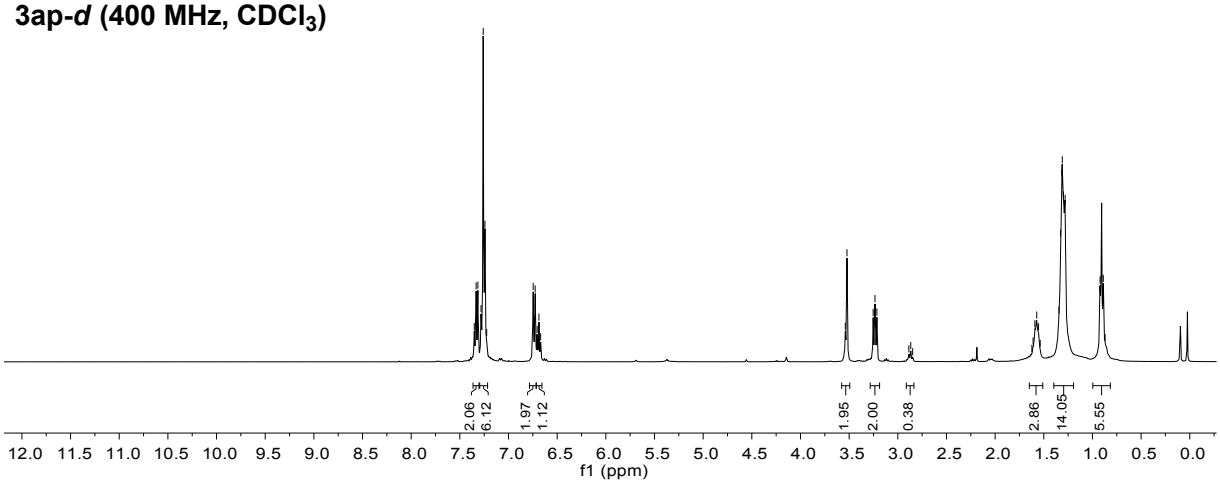


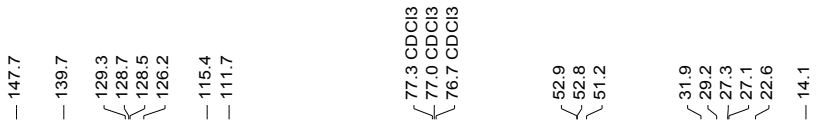
3aj-*d* (100 MHz, $CDCl_3$)

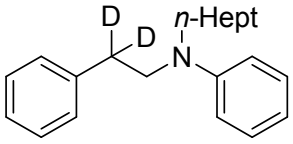




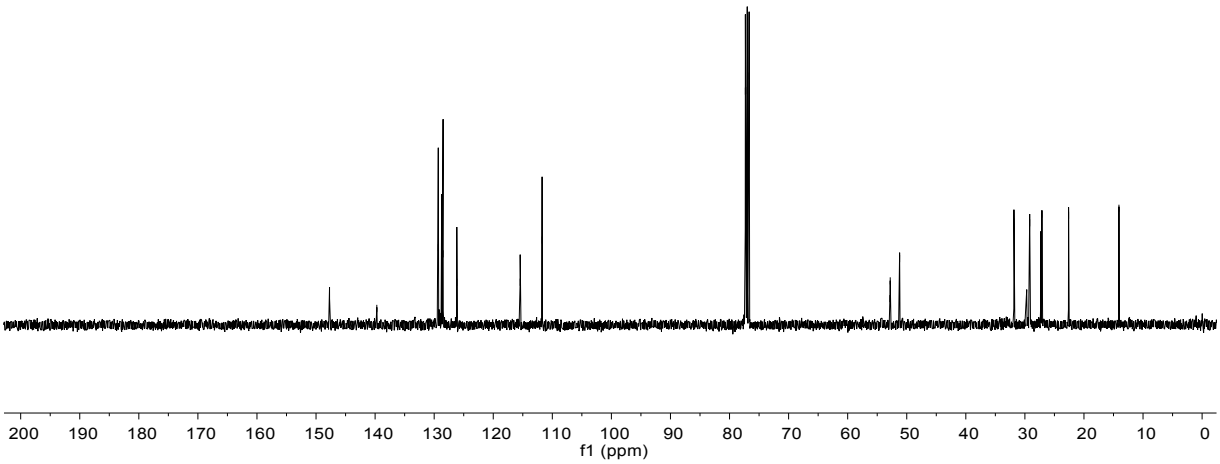
3ap-*d* (400 MHz, $CDCl_3$)

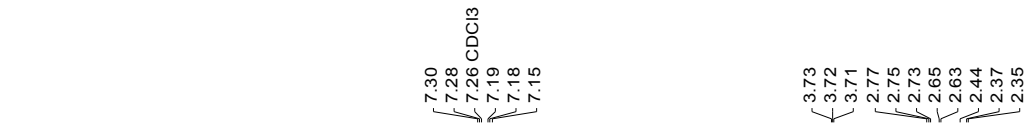


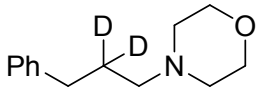




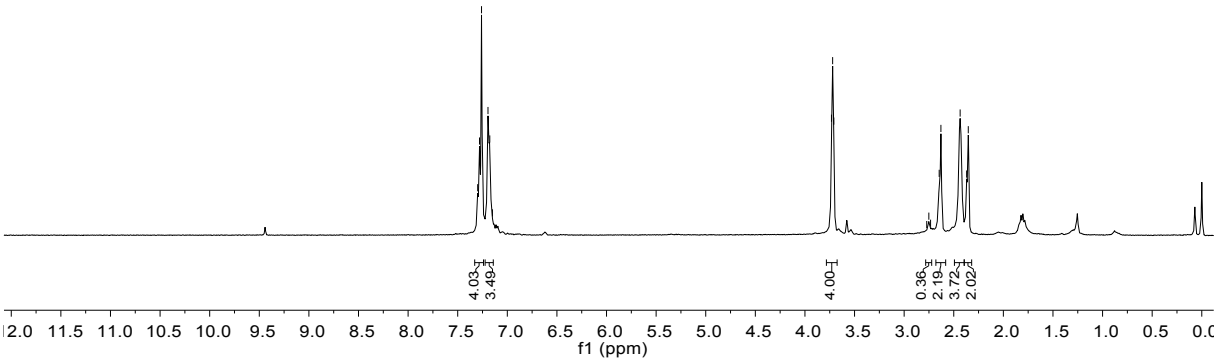
3ap-*d* (100 MHz, $CDCl_3$)

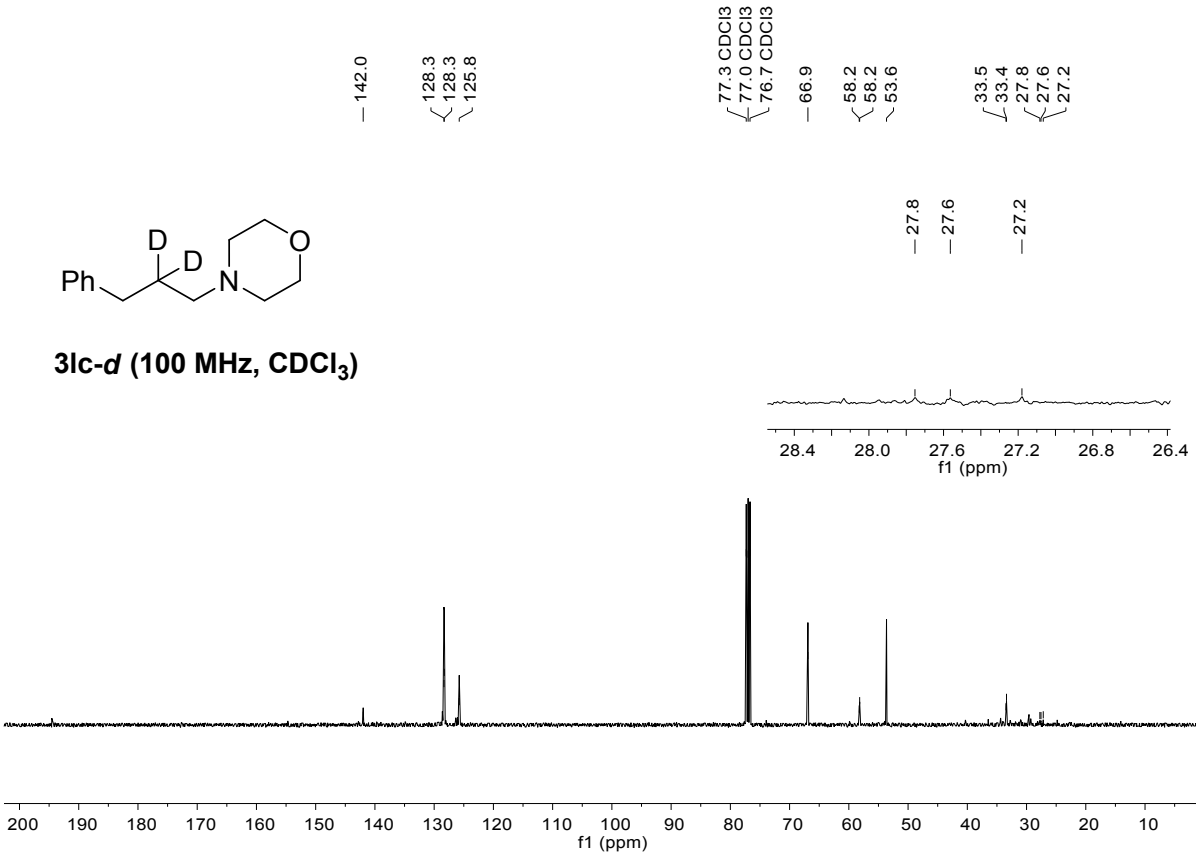




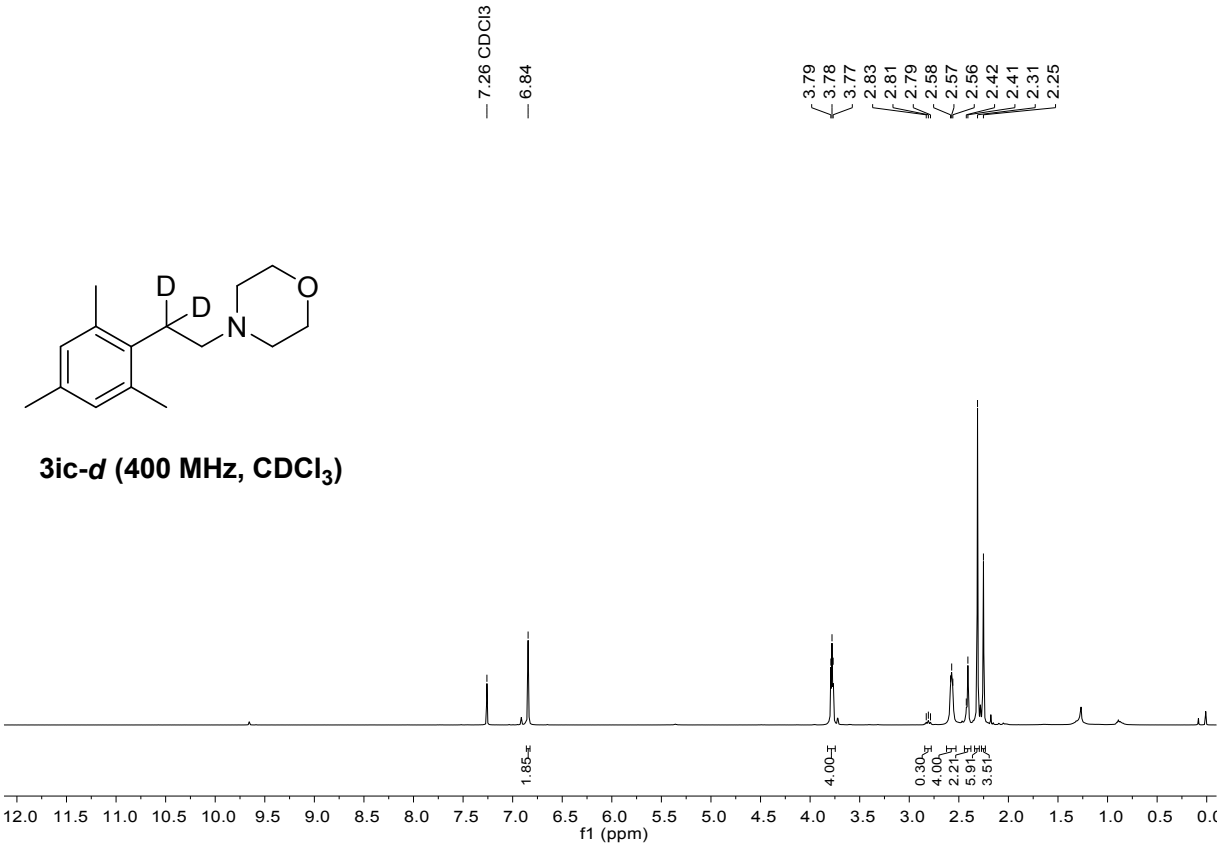


3lc-*d* (400 MHz, $CDCl_3$)

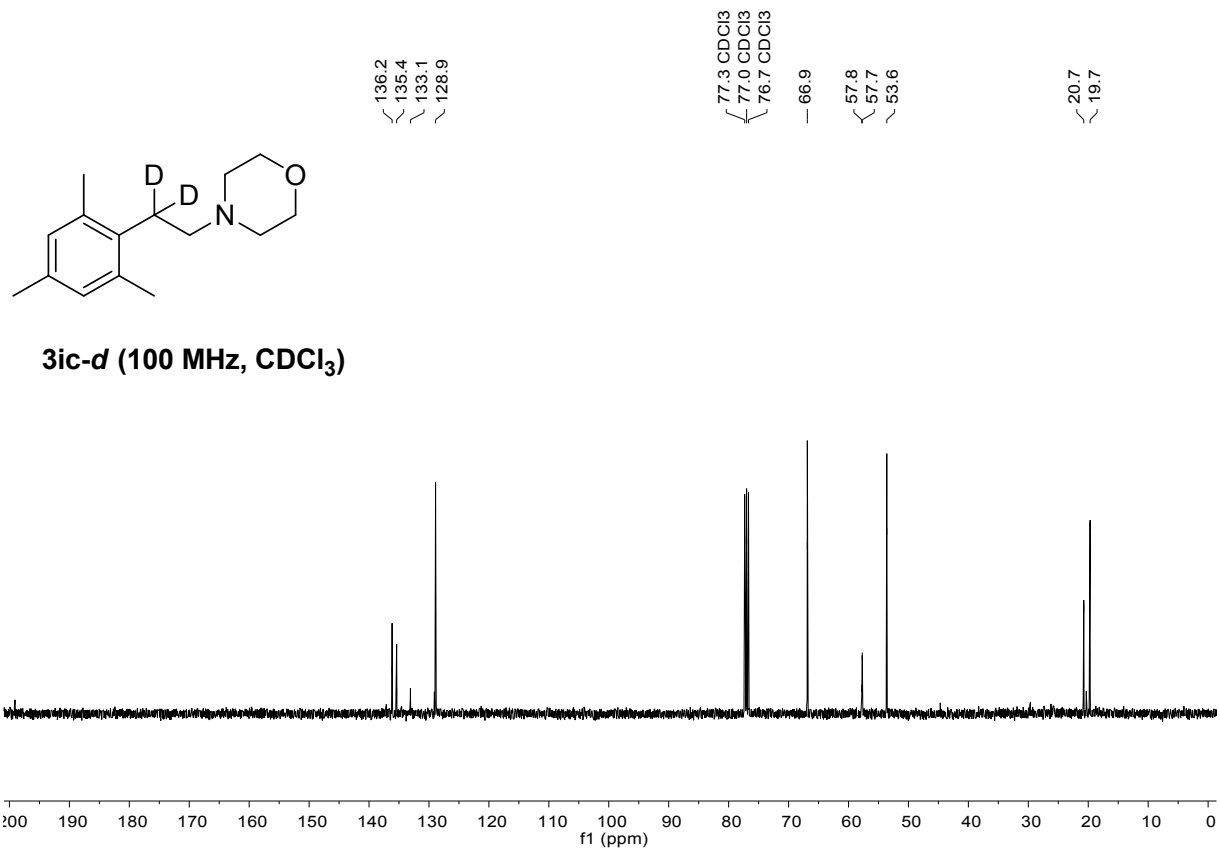




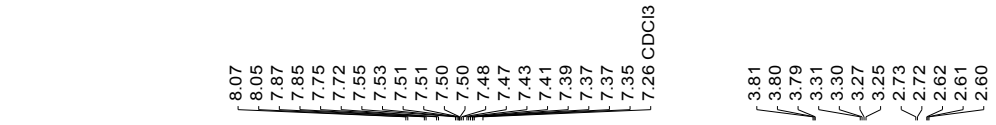
3lc-*d* (100 MHz, $CDCl_3$)

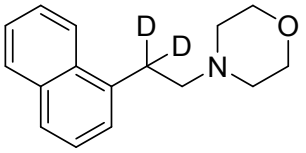


3ic-*d* (400 MHz, $CDCl_3$)

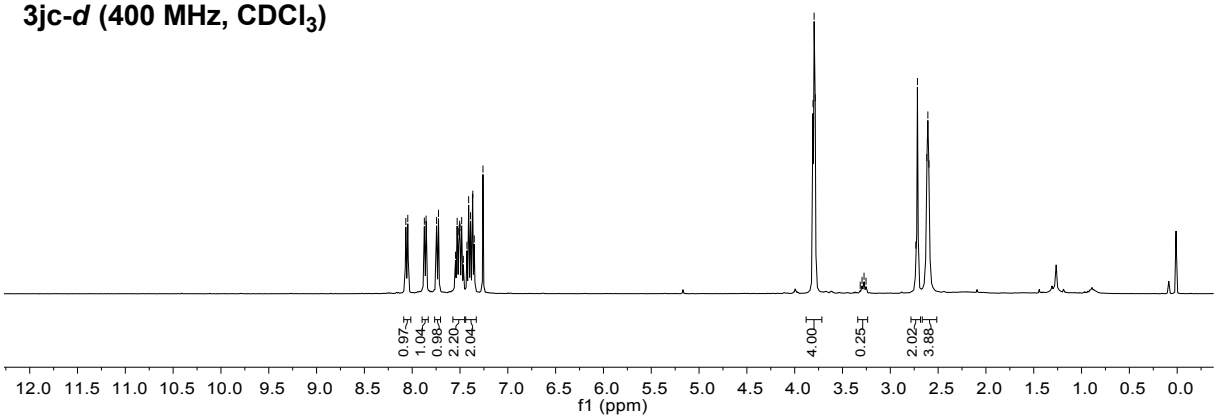


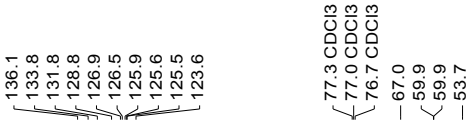
3ic-*d* (100 MHz, $CDCl_3$)

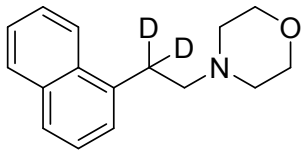




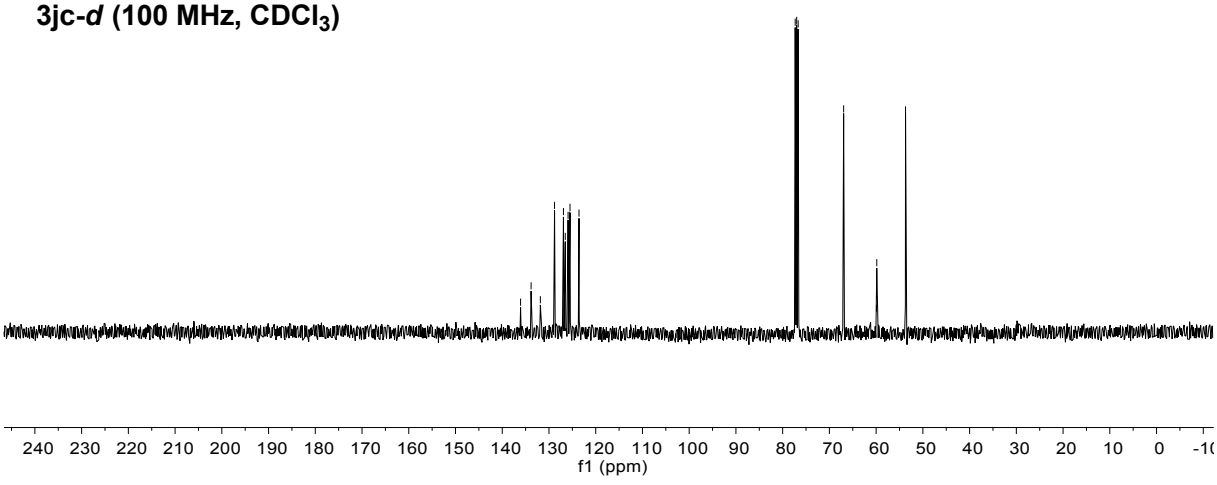
3jc-*d* (400 MHz, $CDCl_3$)

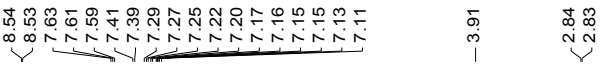


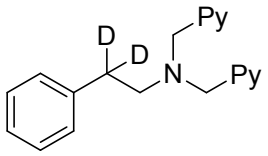




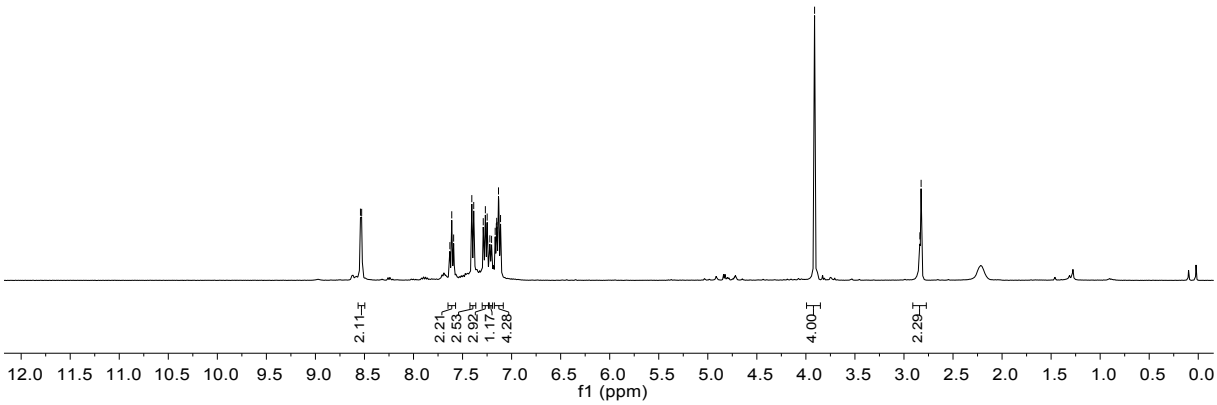
3jc-*d* (100 MHz, $CDCl_3$)

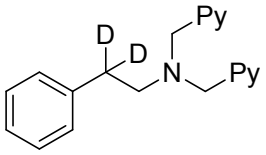




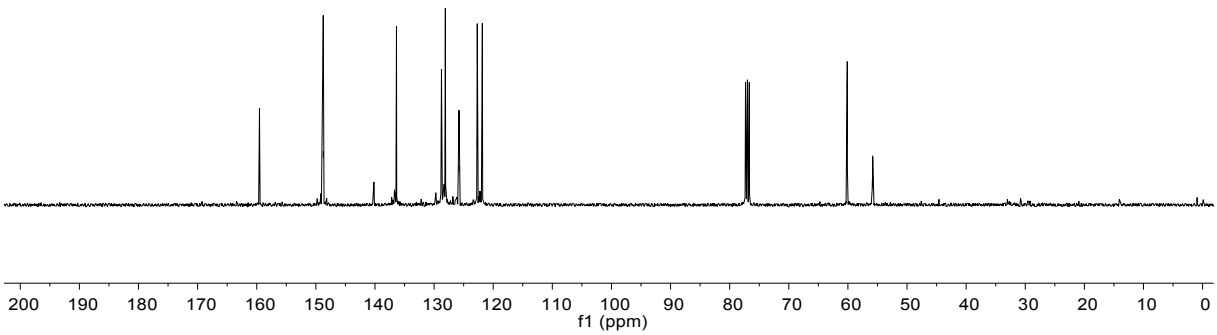


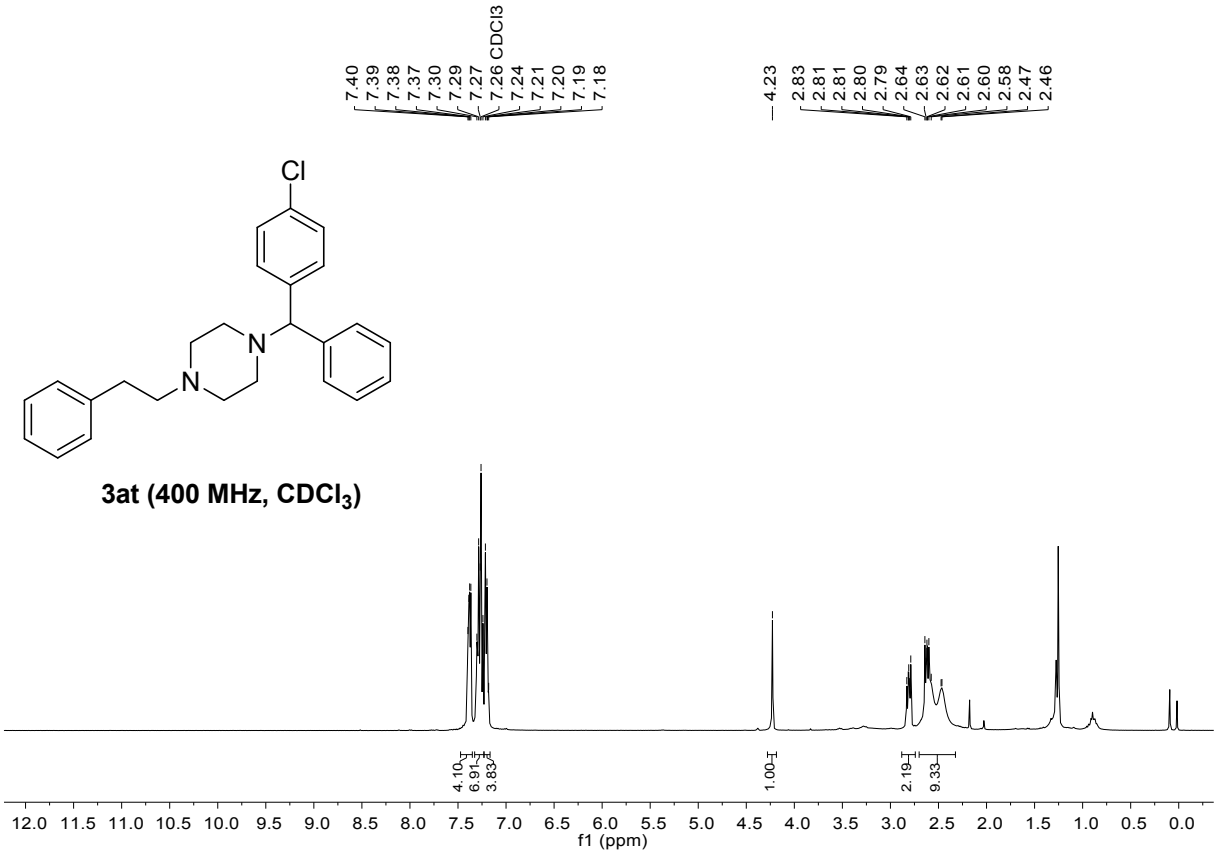
3ai-*d* (400 MHz, $CDCl_3$)



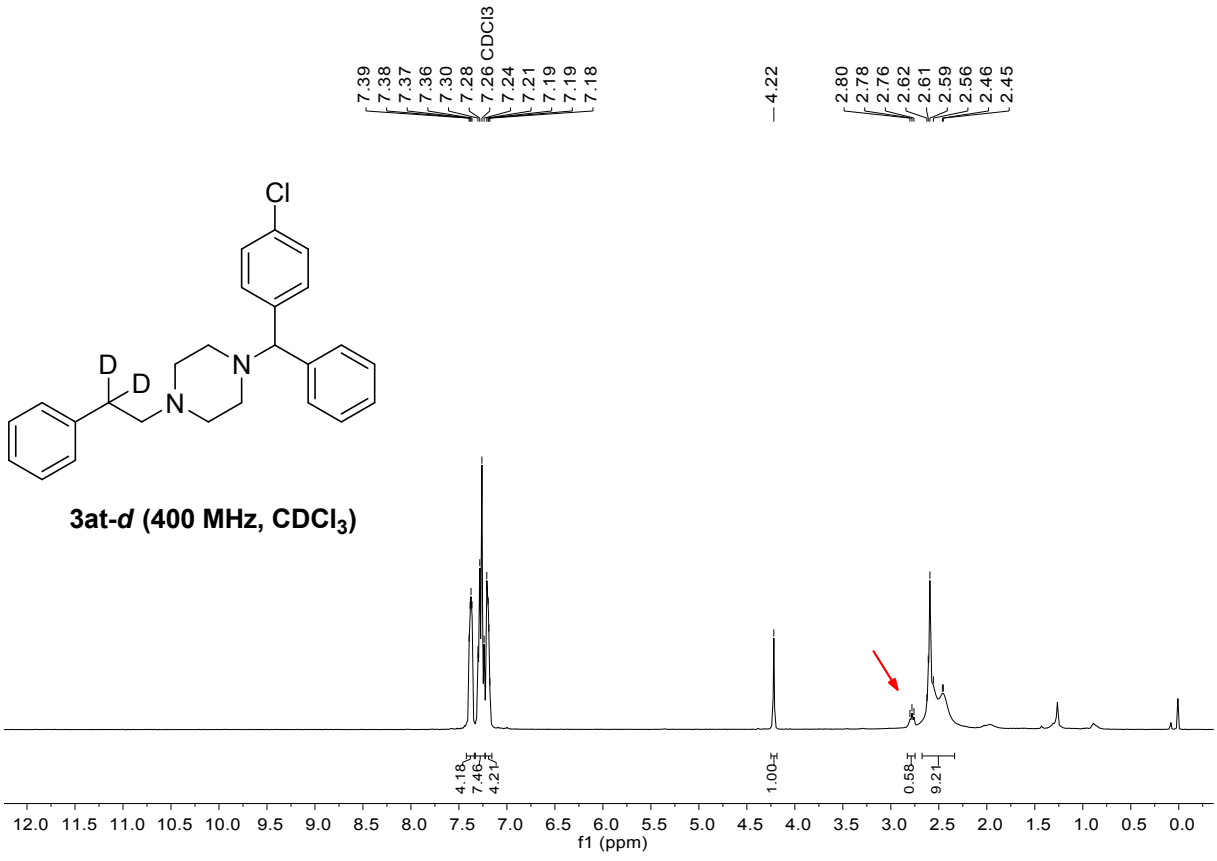


3ai-*d* (100 MHz, $CDCl_3$)

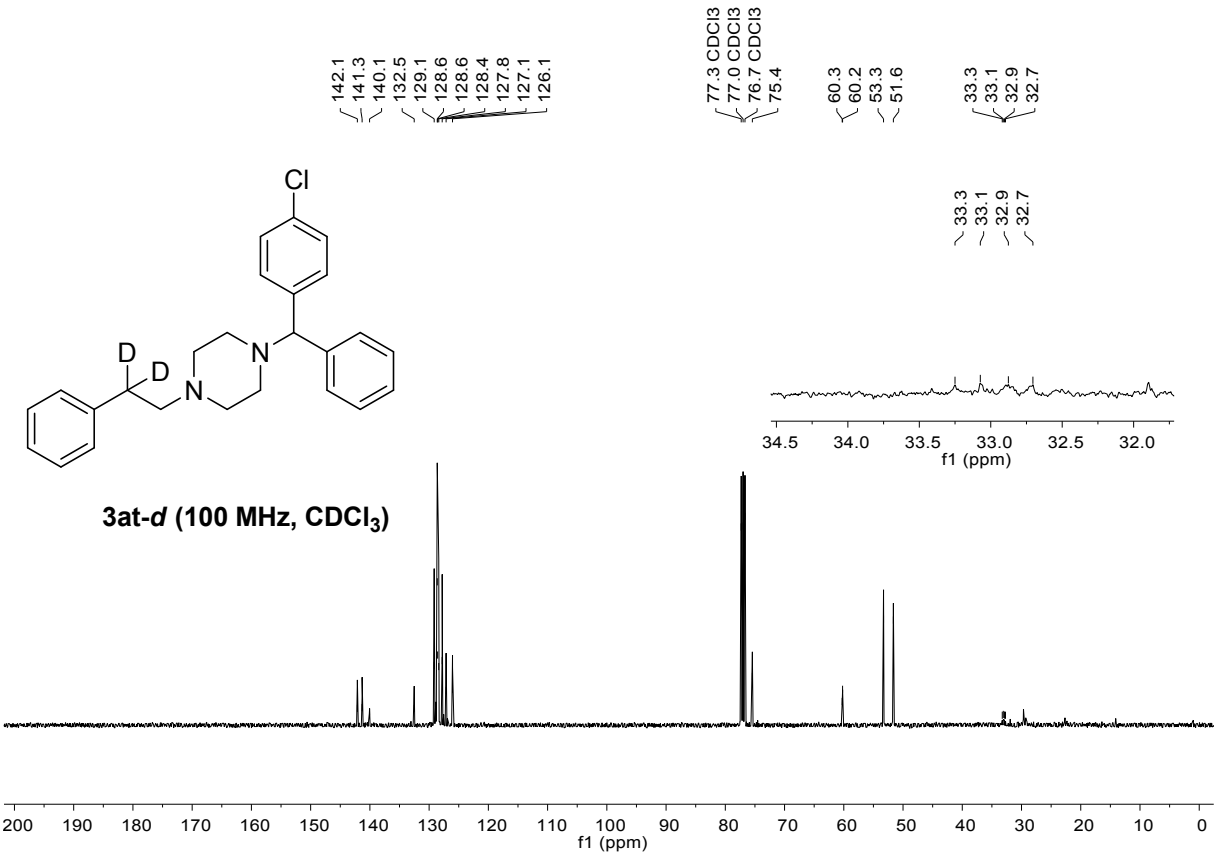




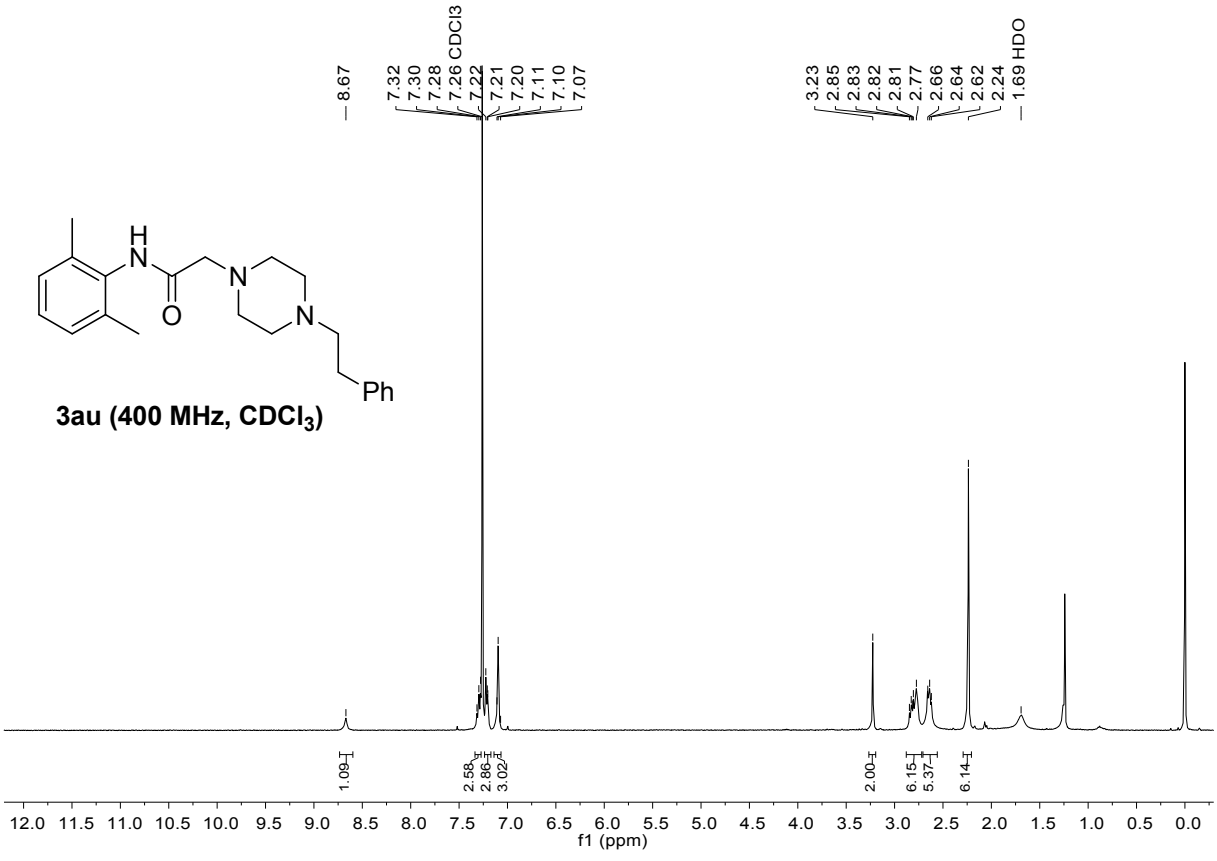
3at (400 MHz, $CDCl_3$)



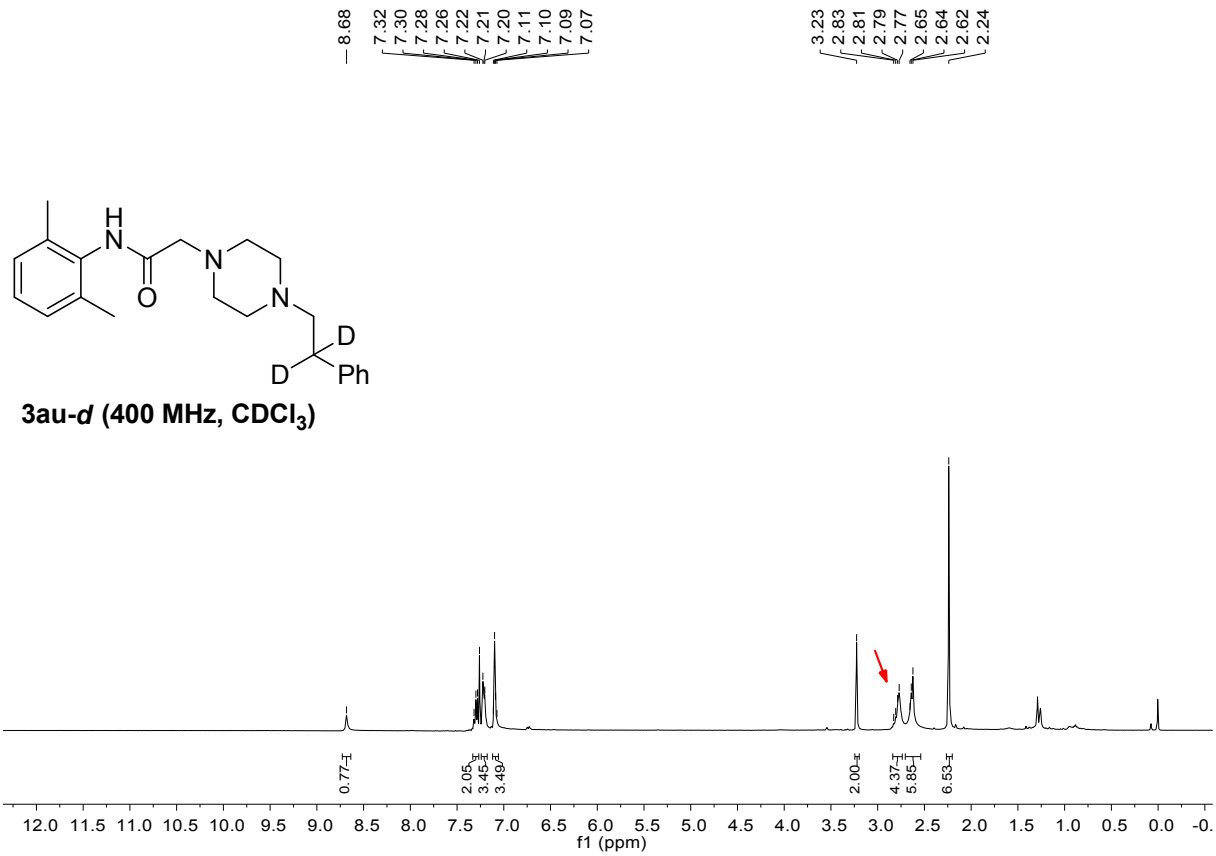
3at-*d* (400 MHz, $CDCl_3$)



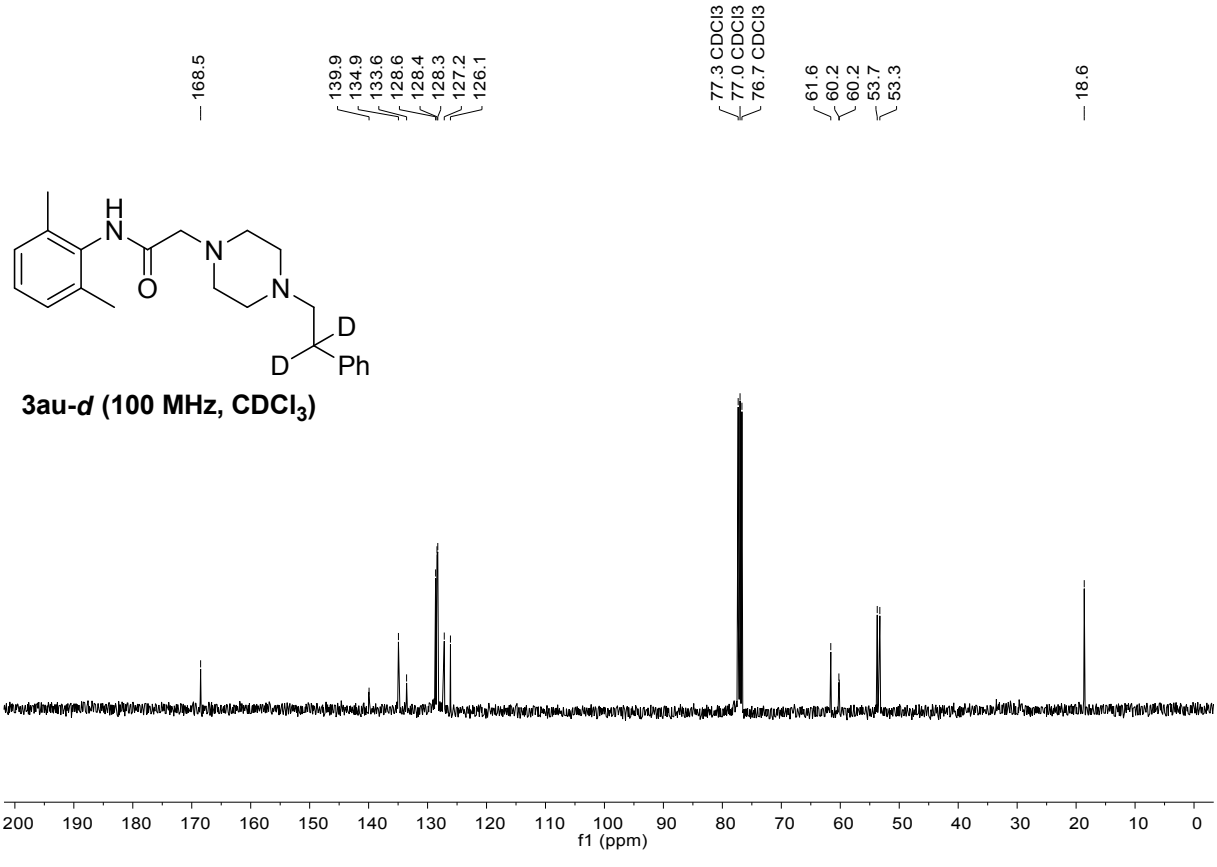
3at-*d* (100 MHz, $CDCl_3$)



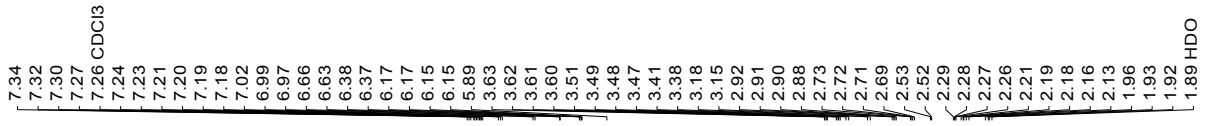
3au (400 MHz, $CDCl_3$)

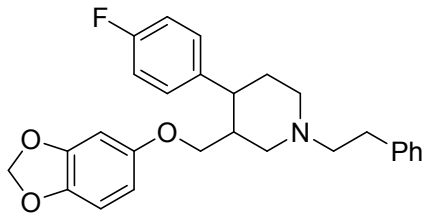


3au-*d* (400 MHz, $CDCl_3$)

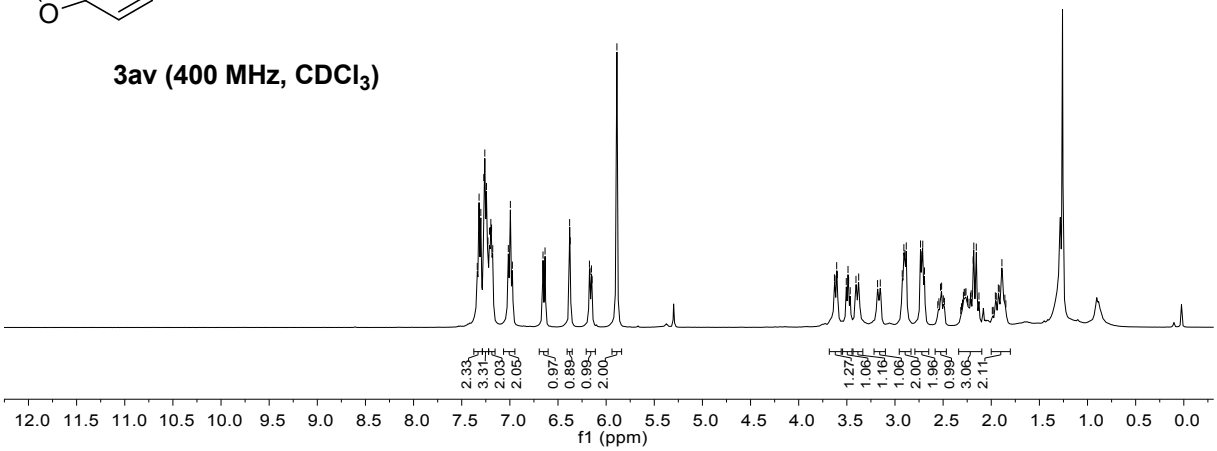


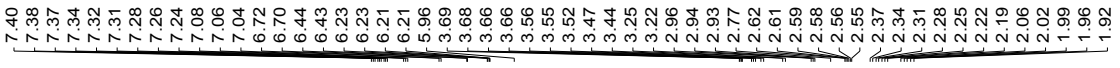
3au-*d* (100 MHz, $CDCl_3$)

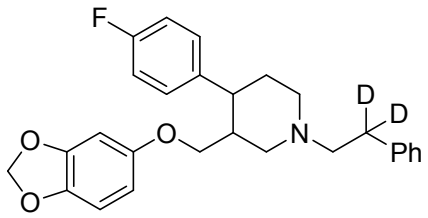




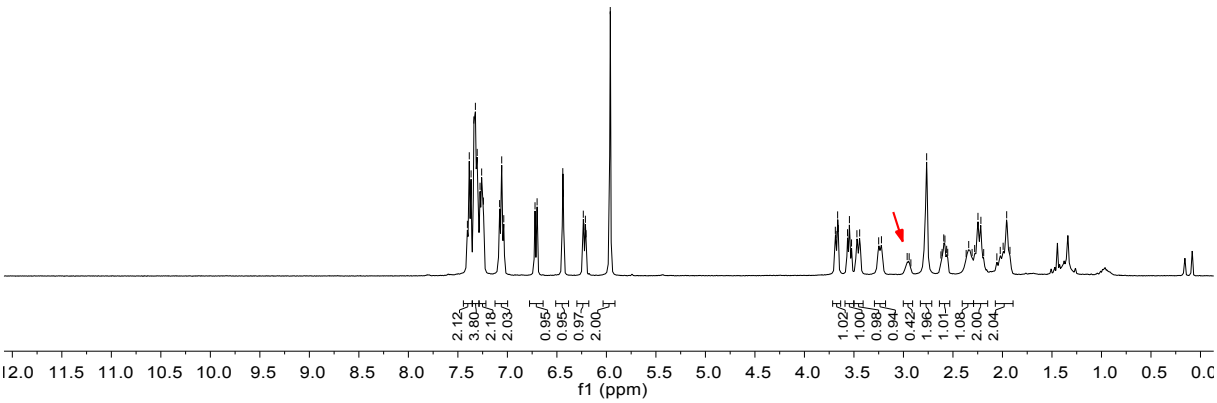
3av (400 MHz, $CDCl_3$)

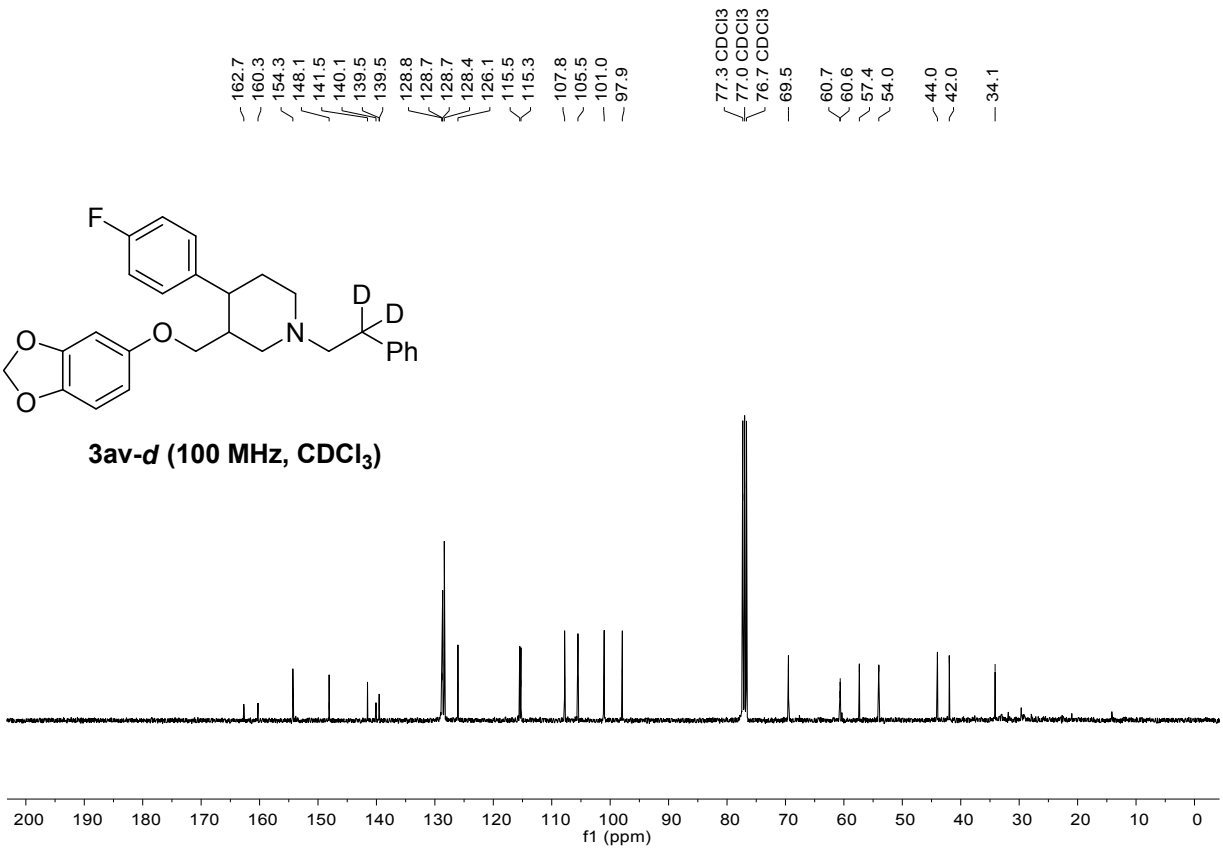






3av-*d* (400 MHz, $CDCl_3$)





3av-*d* (100 MHz, $CDCl_3$)