Electronic Supplementary Material (ESI) for Chemical Communications

Supporting Information

[4+3] Cycloaddition of *in-situ* generated azoalkenes with C,N-cyclic azomethine imines: Efficient synthesis of tetrazepine derivatives

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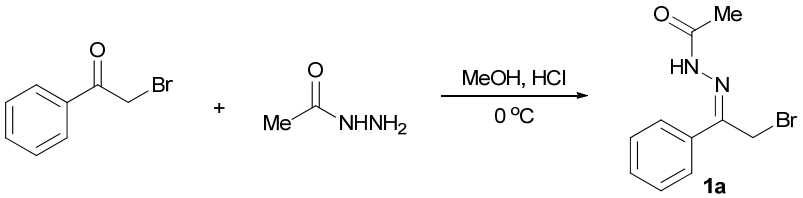
Contents

1. General Information

Unless otherwise noted, materials were purchased from commercial suppliers and used without further purification. All the solvents were treated according to general methods. Flash column chromatography was performed using 200-300 mesh silica gel. ^{1}H NMR spectra were recorded on 400/600 MHz spectrophotometers. Chemical shifts (δ) are reported in ppm from the solvent resonance as the internal standard ($CDCl_3$: 7.26 ppm). Data are reported as follows: chemical shift, multiplicity (s = singlet, d = doublet, t = triplet, dd = doublet of doublets, m = multiplet), coupling constants (Hz) and integration. ^{13}C NMR spectra were recorded on Varian Mercury 400 (100 MHz) with complete proton decoupling spectrophotometers ($CDCl_3$: 77.0 ppm, DMSO-d^6: 39.5 ppm). Mass spectra were measured on a MS spectrometer.

2. Preparation and Spectral Data of Substrates

2.1 General procedure for preparation of N-acetyl hydrazones 1.[1,2]



To a stirred solution of 2-bromo-1-phenylethanone (20 mmol, 1.0 eq.) in Methanol (20 mL), the acethydrazide (30 mmol, 1.5 eq.) and HCl (conc., 0.5 mL) was added at 0 °C. The mixture was stirred at the same temperature for 3 h and filtered then washed with Et_2O (15 mL). The crude product was then recrystallized from MeOH. Compound **1a** was obtained as a white solid (3.20 g, 63% yield).

The other N-acetyl hydrazones were prepared according to the above procedure.

The C,N-cyclic azomethines were also prepared according to Maruoka's procedure.[3]

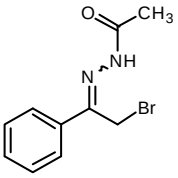
References: [1] J. M. Hatcher and D. M. Coltart, *J. Am. Chem. Soc.*, 2010, **132**, 4546.

[2] J. R. Chen, W. R. Dong, M. Candy, F. F. Pan, M. Jorres and C. Bolm, *J. Am. Chem. Soc.*, 2012, **134**, 6924.

[3] T. Hashimoto, Y. Maeda, M. Omote, H. Nakatsu and K. Maruoka, *J. Am. Chem. Soc.*, 2010, **132**, 4076.

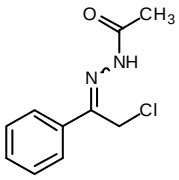
2.2 Spectral Data of Substrates

Hydrazone 1a



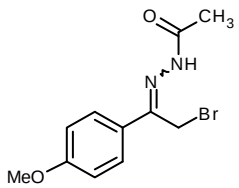
^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, major+minor) 9.66 (1 H, s, major), 8.39 (1 H, s, minor), 7.75 (2 H, d, *J* = 4.5 Hz, major), 7.49 – 7.46 (3 H, m, minor), 7.40 (3 H, s, major), 7.27 – 7.24 (2 H, m, minor), 4.33 (2 H, s, major), 4.27 (2 H, s, minor), 2.40 (3 H, s, major), 2.31 (3 H, s, minor). ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) δ (ppm) 174.42, 172.64, 147.32, 143.49, 135.18, 130.28, 130.00, 129.34, 128.35, 127.29, 125.66, 34.73, 20.48, 20.18, 18.94. M.P.: 130 – 132 °C. IR (in KBr): 3060, 3023, 1669, 1552, 1364, 1264, 1074, 776, 692, 535 cm^{-1}.

Hydrazone 1b



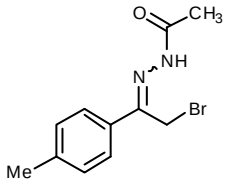
^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, major+minor) 9.75 (1 H, s, major), 8.40 (1 H, s, minor), 7.74 (2 H, s, major), 7.49 (3 H, s, minor), 7.40 (3 H, s, major), 7.26 (2 H, d, *J* = 6.6 Hz, minor), 4.51 (2 H, s, major), 4.38 (2 H, s, minor), 2.41 (3 H, s, major), 2.31 (3 H, s, minor). ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) δ (ppm) 174.18, 172.53, 147.07, 143.25, 135.20, 129.89, 129.28, 129.13, 128.23, 127.12, 125.59, 47.20, 32.94, 20.35, 20.03. M.P.: 111 – 113 °C. IR (in KBr): 2969, 1681, 1589, 1342, 1022, 777, 687, 613 cm^{-1}.

Hydrazone 1c



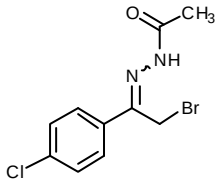
^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, only one isomer) 9.36 (1 H, s), 7.72 (2 H, d, *J* = 7.9 Hz), 6.94 (2 H, d, *J* = 7.9 Hz), 4.29 (2 H, s), 3.85 (3 H, s), 2.41 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) δ (ppm) 174.41, 160.64, 143.61, 128.90, 127.69, 127.25, 114.90, 113.86, 55.18, 20.53, 18.78. M.P.: 128 – 129 °C. IR (in KBr): 3095, 2971, 1682, 1606, 1341, 1251, 837, 800, 618 cm^{-1}.

Hydrazone 1d



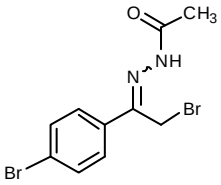
^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, only one isomer) 9.58 (1 H, s), 7.66 (2 H, d, *J* = 7.2 Hz), 7.23 (2 H, d, *J* = 7.3 Hz), 4.32 (2 H, s), 2.42 (3 H, s), 2.39 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) δ (ppm) 174.30, 172.54, 147.51, 143.58, 140.19, 139.46, 132.36, 130.03, 129.06, 127.19, 127.15, 126.16, 125.58, 34.86, 21.07, 20.99, 20.47, 20.16, 18.90. M.P.: 142 – 143 °C. IR (in KBr): 3186, 3097, 1669, 1609, 1384, 1337, 1020, 861, 612, 536 cm^{-1}.

Hydrazone 1e



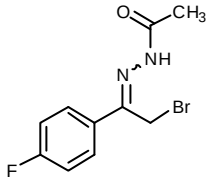
^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, major+minor) 9.67 (1 H, s, major), 8.39 (1 H, s, minor), 7.71 (2 H, d, *J* = 8.2 Hz, major), 7.50 (2 H, d, *J* = 8.1 Hz, minor), 7.40 (2 H, d, *J* = 8.2 Hz, major), 7.25 (2 H, d, *J* = 7.9 Hz, minor), 4.32 (2 H, s, major), 4.27 (2 H, s, minor), 2.41 (3 H, s, major), 2.33 (3 H, s, minor). ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) δ (ppm) 173.55, 141.35, 134.38, 133.43, 129.13, 128.61, 127.98, 127.98, 127.20, 126.59, 21.23, 20.18, 19.75, 18.77. M.P.: 157 – 159 °C. IR (in KBr): 3187, 3086, 1669, 1598, 1485, 1378, 1336, 1020, 605, 529 cm^{-1}.

Hydrazone 1f



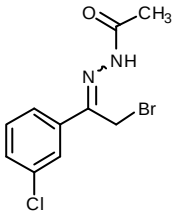
^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, major+minor) 9.60 (1 H, s, major), 8.37 (1 H, s, minor), 7.64 (4 H, d, *J* = 6.9 Hz, major+minor), 7.55 (2 H, d, *J* = 7.8 Hz, major), 7.17 (2 H, s, minor), 4.30 (2 H, s, major), 4.27 (2 H, s, minor), 2.41 (3 H, s, major), 2.32 (3 H, s, minor). ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) δ (ppm) 173.38, 141.23, 133.85, 130.84, 126.79, 122.65, 20.13, 18.73. M.P.: 157 – 159 °C. IR (in KBr): 3187, 3086, 1669, 1598, 1485, 1378, 1336, 1020, 605, 529 cm^{-1}.

Hydrazone 1g



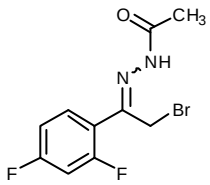
^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, major+minor) 9.38 (1 H, s, major), 8.34 (1 H, s, minor), 7.77 – 7.75 (2 H, m, major), 7.31 – 7.29 (2 H, m, minor), 7.22 (2 H, t, *J* = 7.9 Hz, minor), 7.12 (2 H, t, *J* = 7.9 Hz, major), 4.29 (2 H, s, major), 4.28 (2 H, s, minor), 2.41 (3 H, s, major), 2.33 (3 H, s, minor). ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) δ (ppm) 174.34, 172.76, 164.52, 162.03, 146.26, 142.53, 131.46, 131.43, 129.72, 129.64, 127.72, 127.64, 116.80, 116.59, 115.52, 115.30, 34.83, 20.51, 20.23, 18.89. M.P.: 141 – 142 °C. IR (in KBr): 3105, 1684, 1604, 1513, 1337, 1235, 1151, 1123, 838, 613, 534 cm^{-1}.

Hydrazone 1h



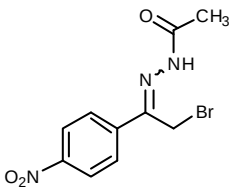
^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, major+minor) 9.55 (1 H, s, major), 8.36 (1 H, s, minor), 7.76 (1 H, s, major), 7.63 (1 H, d, *J* = 7.3 Hz, major), 7.47 (2 H, d, *J* = 6.7 Hz, minor), 7.38 (2 H, m, major), 7.29 (1 H, s, minor), 7.19 (1 H, d, *J* = 6.1 Hz, minor), 4.30 (2 H, s, major), 4.27 (2 H, s, minor), 2.43 (3 H, s, major), 2.33 (3 H, s, minor). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 175.16, 173.22, 145.84, 142.68, 137.10, 135.63, 134.63, 132.25, 130.89, 130.36, 129.78, 129.47, 127.65, 125.94, 125.81, 124.00, 34.52, 20.65, 20.38, 18.67. M.P.: 138 – 140 °C. IR (in KBr): 3189, 3093, 1683, 1583, 1513, 1336, 1267, 1157, 787, 678, 632 cm^{-1}.

Hydrazone 1i



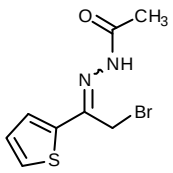
^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, major+minor) 9.49 (1 H, s, major), 8.36 (1 H, s, minor), 7.61 (1 H, q, *J* = 8.0, 12.0 Hz, major), 7.33 – 7.28 (1 H, m, minor), 7.05 (1 H, t, *J* = 7.8 Hz, minor), 7.00 (1 H, d, *J* = 8.9 Hz, minor), 6.95 (1 H, t, *J* = 8.0 Hz, major), 6.89 (1 H, t, *J* = 9.7 Hz, major), 4.30 (4 H, s, major+minor), 2.35 (3 H, s, major), 2.34 (3 H, s, minor). ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) δ (ppm) 172.84, 166.49, 163.27, 163.15, 160.77, 160.71, 160.65, 160.60, 158.20, 158.08, 143.20, 139.09, 131.18, 130.46, 120.05, 110.73, 110.55, 103.52, 103.26, 103.00, 102.74, 21.63, 21.56, 20.72, 19.60. M.P.: 115 – 118 °C. IR (in KBr): 2989, 1689, 1665, 1556, 1501, 1266, 1145, 1094, 868, 821, 614 cm^{-1}.

Hydrazone 1j



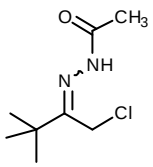
^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, only one isomer) 9.06 (1 H, s), 8.29 (2 H, d, *J* = 8.6 Hz), 7.93 (2 H, d, *J* = 8.5 Hz), 4.27 (2 H, s), 2.45 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$/TFA) δ (ppm) 178.41, 177.84, 150.52, 148.59, 146.29, 140.59, 136.74, 130.17, 129.12, 127.26, 124.87, 124.04, 20.13, 17.38. M.P.: 186 – 188 °C. IR (in KBr): 3109, 2976,1726, 1690, 1566, 1517, 1377, 1186, 1016, 911, 855, 699, 605 cm^{-1}.

Hydrazone 1k



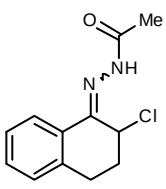
^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, only one isomer) 9.03 (1 H, s), 8.27 (1 H, d, *J* = 8.6 Hz), 7.91 (1 H, d, *J* = 8.5 Hz), 7.24 (2 H, s), 4.26 (1 H, s), 2.43 (1 H, s), 1.54 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) δ (ppm) 173.87, 140.53, 139.78, 127.50, 127.01, 125.76, 20.15, 18.79. M.P.: 144 – 146 °C. IR (in KBr): 3174, 2961,1728, 1674, 1445, 1378, 1338, 1156, 1105, 706, 637, 605, 518 cm^{-1}.

Hydrazone 1l



^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, only one isomer) 9.15 (1 H, s), 4.08 (2 H, s), 2.27 (3 H, s), 1.18 (9 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 174.42, 153.21, 38.36, 32.14, 27.39, 20.22. M.P.: 83 – 86 °C. IR (in KBr): 3229, 2969, 1674, 1463, 1391, 1295, 1138, 1074, 926, 863, 767, 727, 627 cm^{-1}.

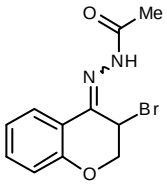
Hydrazone 1m



^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, only one isomer) 9.55 (1 H, s), 8.10 (1 H, d, *J* = 7.7 Hz), 7.30 (1 H, t, *J* = 7.3 Hz), 7.26 (1 H, d, *J* = 7.3 Hz), 7.18 (1 H, d, *J* = 7.3 Hz), 5.20 (1 H, s), 3.27 (1 H, t, *J* = 13.2 Hz), 2.75 (1 H, d, *J* = 16.5 Hz), 2.44 (3 H, s), 2.39 (1 H, d, *J* = 14.5 Hz), 2.27 (1 H, t, *J* = 13.4 Hz). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 174.39, 142.87,

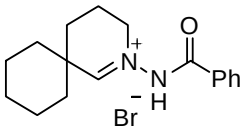
137.98, 129.66, 129.46, 128.56, 126.72, 124.77, 48.24, 30.83, 23.95, 20.60. M.P.: 163 – 165 °C. IR (in KBr): 3177, 3079,1714, 1666, 1383, 1138, 958, 778, 725, 620, 569 cm^{-1}.

Hydrazone 1n



^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm, major+minor) 10.04 (1 H, s, minor), 9.83 (1 H, s, major), 8.06 (1 H, s, minor), 7.99 (1 H, d, *J* = 7.8 Hz, major), 7.40 (1 H, d, *J* = 8.2 Hz, minor), 7.33 (1 H, t, *J* = 7.7 Hz, major), 7.04 (1 H, t, *J* = 7.5 Hz, major), 7.01 (1 H, d, *J* = 8.2 Hz, major), 6.90 (1 H, d, *J* = 8.0 Hz, minor), 5.25 (1 H, s, minor), 5.20 (1 H, s, major), 4.57 (1 H, d, *J* = 12.9 Hz, major), 4.41 (1 H, d, *J* = 13.0 Hz, major), 2.45 (3 H, s, minor), 2.44 (3 H, s, major). ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) δ (ppm) 173.74, 155.06, 137.94, 130.67, 124.38, 121.69, 117.62, 117.15, 69.68, 35.55, 20.25. M.P.: 173 – 175 °C. IR (in KBr): 3183, 2930, 1667, 1613, 1397, 1138, 1140, 1059, 946, 774, 596 cm^{-1}.

C,N-cyclic azomethine imine salt 2g

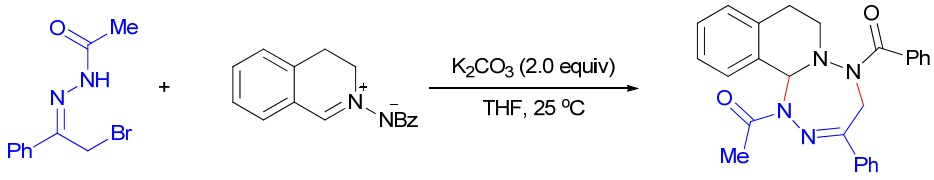


Prepared according to the known literature procedure.[3]

^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 8.65 (1 H, s), 8.19 (2 H, d, *J* = 7.5), 7.61 (1 H, t, *J* = 7.4), 7.51 (2 H, t, *J* = 7.7), 4.26 (2 H, m), 2.16 (2 H, m), 1.90 (4 H, t, *J* = 9.6), 1.79 – 1.73 (4 H, m), 1.66 (1 H, s), 1.56 (2 H, d, *J* = 10.7), 1.49 (1 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 184.26, 164.15, 133.26, 128.41, 128.35, 128.17, 55.78, 40.78, 33.34, 26.05, 24.24, 19.59, 18.02.

3. General Procedure and Spectral Data of Products

3.1 General Procedure

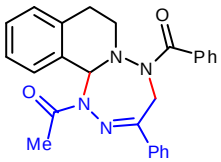


1a **2a** **3aa**

1a (0.3 mmol, 76.5 mg), K_2CO_3 (0.6 mmol, 82.9 mg) and **2a** (0.33 mmol, 82.6 mg) were dissolved in THF (3 mL). Then, the mixture was stirred at room temperature until the reaction was completed monitored by TLC analysis. The crude product was purified by flash chromatography on silica gel (petroleum ether/ethyl acetate 5:1~3:1) to give the desired product **3aa** as a white solid.

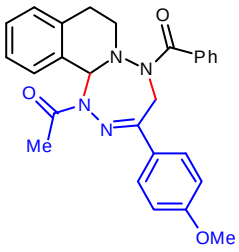
3.2 Spectral Data of Products.

Product 3aa



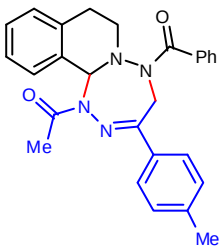
Yield of **3aa** : 85% and 86% respectively. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.61 (2 H, d, *J* = 7.6 Hz), 7.56 (2 H, d, *J* = 7.1 Hz), 7.38 (4 H, m), 7.34 (2 H, t, *J* = 7.5 Hz), 7.17 – 7.16 (2 H, m), 7.12 – 7.10 (2 H, m), 6.98 (1 H, d, *J* = 4.9 Hz), 5.65 (1 H, d, *J* = 18.0 Hz), 3.95 (1 H, d, *J* = 18.0 Hz), 3.39 (2 H, m), 3.08 – 3.02 (1 H, m), 2.74 (1 H, m), 2.46 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.66, 173.25, 155.76, 135.66, 135.11, 134.86, 132.75, 130.20, 129.83, 129.03, 128.37, 127.80, 127.64, 126.99, 126.82, 125.87, 70.62, 49.26, 48.21, 24.78, 22.06. M.P.: 192 – 195 °C. IR (in KBr): 2974, 2912, 1686, 1613, 1657, 1380, 1278, 1032, 1016, 755, 701 cm^{-1}. HRMS (EI): m/z [M + H^+] calcd for $C_{26}H_{24}N_4O_2$: 425.1978; found: 425.1973.

Product 3ca



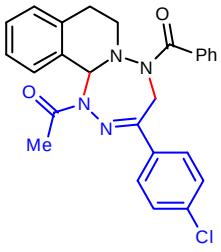
Yield of **3ca** : 92%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.65 (2 H, d, *J* = 6.7 Hz), 7.56 (2 H, d, *J* = 7.3 Hz), 7.39 (3 H, m), 7.17 – 7.16 (2 H, m), 7.10 (1 H, d, *J* = 3.2 Hz), 7.02 (2 H, d, *J* = 4.4 Hz), 6.87 (2 H, d, *J* = 8.3 Hz), 5.66 (1 H, d, *J* = 18.0 Hz), 3.99 (1 H, d, *J* = 18.0 Hz), 3.80 (3 H, s), 3.39 (2 H, s), 3.04 – 2.99 (1 H, m), 2.75 (1 H, d, *J* = 16.8 Hz), 2.43 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.46, 172.88, 161.36, 156.59, 135.08, 134.79, 132.66, 129.76, 128.82, 128.54, 127.73, 127.69, 127.58, 127.52, 126.63, 125.93, 113.70, 70.76, 55.20, 48.89, 47.46, 25.13, 22.11. M.P.: 154 – 156 °C. IR (in KBr): 2964, 2925, 1679, 1653, 1607, 1377, 1250, 1173, 1034, 790, 701 cm^{-1}. HRMS (EI): m/z [M + H^+] calcd for $C_{27}H_{26}N_4O_3$: 455.2083; found: 455.2079.

Product 3da



Yield of **3da** : 84%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.53 (2 H, d, *J* = 7.5 Hz), 7.50 (2 H, d, *J* = 7.5 Hz), 7.38 – 7.33 (3 H, m), 7.13 – 7.12 (4 H, m), 7.07 – 7.05 (2 H, m), 6.97 – 6.95 (1 H, m), 5.63 (1 H, d, *J* = 18.0 Hz), 3.92 (1 H, d, *J* = 18.0 Hz), 3.35 (2 H, d, *J* = 5.4 Hz), 3.03 – 2.97 (1 H, m), 2.71 (1 H, d, *J* = 17.0 Hz), 2.42 (3 H, s), 2.31 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.54, 173.12, 156.40, 140.62, 135.18, 134.88, 132.78, 130.79, 129.81, 129.09, 128.94, 127.76, 127.64, 126.96, 126.77, 125.97, 70.68, 49.15, 47.98, 25.01, 22.08, 21.22. M.P.: 164 – 167 °C. IR (in KBr): 2912, 1676, 1655, 1383, 1356, 1305, 905, 772, 747 cm^{-1}. HRMS (EI): m/z [M + H^+] calcd for $C_{27}H_{26}N_4O_2$: 439.2134; found: 439.2127.

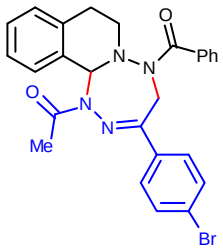
Product 3ea



Yield of **3ea** : 91%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.57 (2 H, s), 7.55 (2 H, s), 7.44 – 7.36 (3 H, m), 7.32 (2 H, d, *J* = 8.2 Hz), 7.17 (2 H, m), 7.13 (2 H, s), 6.94 (1 H,

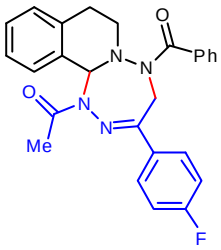
d, J = 7.0 Hz), 5.60 (1 H, d, J = 18.0 Hz), 3.92 (1 H, d, J = 18.0 Hz), 3.41 (2 H, m), 3.07 (1 H, m), 2.76 (1 H, d, J = 16.7 Hz), 2.46 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.85, 173.31, 153.94, 136.31, 134.92, 134.83, 134.02, 132.58, 129.95, 129.18, 128.59, 128.23, 127.89, 127.69, 127.61, 126.84, 125.70, 70.68, 49.31, 48.04, 24.53, 22.07. M.P.: 171 – 173 ^{o}C. IR (in KBr): 2916, 1678, 1656, 1378, 1281, 1103, 736, 702, 618 cm^{-1}. HRMS (EI): m/z [M + H^{+}] calcd for $C_{26}H_{23}ClN_4O_2$: 459.1588; found: 459.1582.

Product 3fa



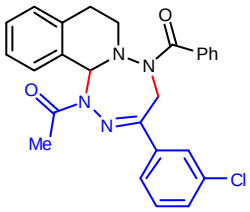
Yield of **3fa** : 76%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.54 (2 H, d, J = 7.1 Hz), 7.46 (4 H, s), 7.39 – 7.35 (3 H, m), 7.15 (2 H, m), 7.11 (2 H, d, J = 6.8 Hz), 6.92 (1 H, d, J = 7.5 Hz), 5.58 (1 H, d, J = 18.0 Hz), 3.90 (1 H, d, J = 18.0 Hz), 3.43 – 3.37 (2 H, m), 3.09 – 3.03 (1 H, m), 2.75 (1 H, d, J = 17.2 Hz), 2.45 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.80, 173.24, 153.90, 134.89, 134.79, 134.46, 132.54, 131.50, 129.90, 129.15, 128.41, 127.85, 127.64, 127.57, 126.80, 125.63, 124.63, 70.60, 49.27, 47.98, 24.45, 22.02. M.P.: 196 – 198 ^{o}C. IR (in KBr): 2915, 1680, 1657, 1377, 1304, 1023, 752, 727, 636 cm^{-1}. HRMS (EI): m/z [M + H^{+}] calcd for $C_{26}H_{23}BrN_4O_2$: 503.1083; found: 503.1075.

Product 3ga



Yield of **3ga** : 85%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.63 (2 H, s), 7.56 (2 H, d, J = 7.3 Hz), 7.41 – 7.37 (3 H, m), 7.18 – 7.16 (2 H, m), 7.13 – 7.10 (2 H, m), 7.03 (2 H, t, J = 8.6 Hz), 6.96 (1 H, d, J = 6.6 Hz), 5.61 (1 H, d, J = 18.0 Hz), 3.94 (1 H, d, J = 18.0 Hz), 3.41 (2 H, s), 3.08 – 3.04 (1 H, m), 2.77 (1 H, d, J = 17.1 Hz), 2.46 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.86, 173.19, 165.23, 162.73, 154.57, 135.02, 134.90, 132.74, 131.82, 129.94, 129.07, 128.99, 127.88, 127.68, 126.82, 125.80, 115.56, 115.34, 70.79, 49.27, 48.11, 24.78, 22.06. M.P.: 187 – 189 ^{o}C. IR (in KBr): 2980, 1684, 1662, 1381, 1278, 1020, 755, 648 cm^{-1}. HRMS (EI): m/z [M + H^{+}] calcd for $C_{26}H_{23}FN_4O_2$: 443.1883; found: 443.1878.

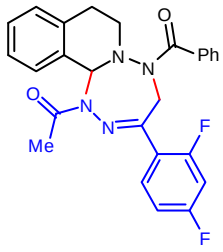
Product 3ha



Yield of **3ha** : 85%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.60 (1 H, s), 7.57 (2 H, d, J = 7.4 Hz), 7.51 (1 H, d, J = 7.5 Hz), 7.42 (3 H, m), 7.37 (1 H, d, J = 7.9 Hz), 7.30 (1 H, d, J = 7.9 Hz), 7.23 – 7.16 (3 H, m), 7.15 (1 H, d, J = 7.1 Hz), 6.94 (1 H, d, J = 7.1 Hz), 5.60 (1 H, d, J = 18.0 Hz), 3.94 (1 H, d, J = 18.0 Hz), 3.48 – 3.41 (2 H, m), 3.13 – 3.08 (1 H, m), 2.78 (1 H, d, J = 17.0 Hz), 2.51 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.86, 173.29, 153.22, 137.44, 134.97, 134.83, 134.41, 132.62, 130.09, 129.93, 129.62, 129.19, 128.70, 127.92, 127.67, 127.63, 126.91, 125.63, 124.99, 70.58, 49.41, 48.20, 24.49, 22.02. M.P.: 182 – 184 ^{o}C. IR

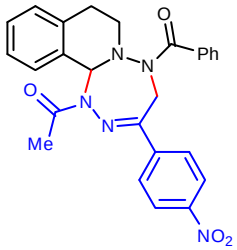
(in KBr): 2959, 2918, 1685, 1658, 1359, 1306, 1033, 753, 704 cm^{-1}. HRMS (EI): m/z [M + H^+] calcd for $C_{26}H_{23}ClN_4O_2$: 459.1588; found: 459.1582.

Product 3ia



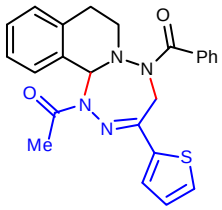
Yield of **3ia** : 78%. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) 7.58 – 7.56 (1 H, m), 7.54 (2 H, d, *J* = 7.3 Hz), 7.38 (3 H, m), 7.23 – 7.20 (2 H, m), 7.17 – 7.14 (2 H, m), 6.88 – 6.85 (2 H, m), 6.73 (1 H, t, *J* = 9.8 Hz), 5.26 (1 H, d, *J* = 18.0 Hz), 4.03 (1 H, d, *J* = 18.0 Hz), 3.45 (1 H, m), 3.39 (1 H, m), 3.12 (1 H, m), 2.76 (1 H, d, *J* = 17.1 Hz), 2.47 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 174.14, 173.22, 164.94, 162.32, 161.86, 159.34, 150.93, 135.16, 134.92, 132.64, 130.96, 129.98, 129.16, 127.94, 127.72, 127.01, 125.81, 121.15, 121.05, 111.90, 111.69, 104.53, 104.27, 104.02, 70.47, 50.28, 49.58, 24.55, 21.95. M.P.: 152 – 154 °C. IR (in KBr): 2977, 1689, 1658, 1384, 1354, 1302, 1018, 849, 734 cm^{-1}. HRMS (EI): m/z [M + H^+] calcd for $C_{26}H_{22}F_2N_4O_2$: 461.1789; found: 461.1783.

Product 3ja



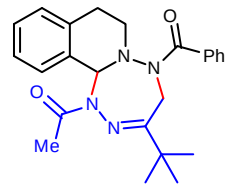
Yield of **3ja** : 92%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 8.21 (2 H, d, *J* = 8.6 Hz), 7.78 (2 H, d, *J* = 8.6 Hz), 7.58 (2 H, d, *J* = 7.3 Hz), 7.46 (1 H, t, *J* = 7.3 Hz), 7.42 (2 H, t, *J* = 7.3 Hz), 7.23 – 7.19 (4 H, m), 6.91 (1 H, d, *J* = 7.4 Hz), 5.61 (1 H, d, *J* = 18.0 Hz), 3.98 (1 H, d, *J* = 18.0 Hz), 3.58 – 3.40 (2 H, m), 3.16 (1 H, m), 2.82 (1 H, d, *J* = 17.9 Hz), 2.53 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 174.03, 173.31, 151.49, 148.37, 141.65, 134.77, 134.71, 132.41, 130.00, 129.36, 127.98, 127.67, 127.65, 127.53, 126.91, 125.30, 123.47, 70.58, 49.50, 48.24, 24.12, 21.92. M.P.: 178 – 181 °C. IR (in KBr): 2924, 1690, 1635, 1491, 1382, 1308, 1264, 907, 873, 760 cm^{-1}. HRMS (EI): m/z [M + H^+] calcd for $C_{26}H_{23}N_5O_4$: 470.1828; found: 470.1822.

Product 3ka



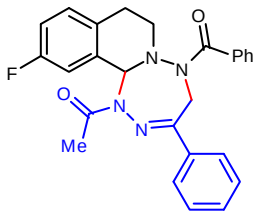
Yield of **3ka** : 81%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.55 (2 H, d, *J* = 7.2 Hz), 7.41 – 7.35 (5 H, m), 7.15 (2 H, m), 7.11 (1 H, d, *J* = 7.1 Hz), 7.02 – 6.99 (2 H, m), 6.97 (1 H, d, *J* = 7.4 Hz), 5.70 (1 H, d, *J* = 18.0 Hz), 4.00 (1 H, d, *J* = 18.0 Hz), 3.39 (2 H, d, *J* = 5.9 Hz), 3.06 – 3.02 (1 H, m), 2.77 (1 H, m), 2.45 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.53, 172.96, 151.16, 139.59, 134.97, 134.88, 132.51, 129.93, 129.32, 129.14, 128.88, 127.96, 127.86, 127.67, 127.54, 126.87, 125.96, 71.09, 49.11, 46.67, 25.16, 22.06. M.P.: 178 – 180 °C. IR (in KBr): 2968, 2919, 1674, 1652, 1379, 1355, 1277, 1174, 1006, 984, 778, 737, 716 cm^{-1}. HRMS (EI): m/z [M + H^+] calcd for $C_{24}H_{22}N_4O_2S$: 431.1542; found: 431.1534.

Product 3la



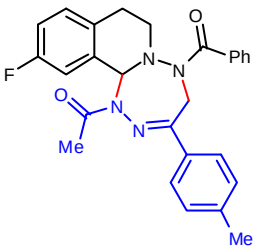
Yield of **3la** : 83%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.53 (2 H, d, *J* = 6.6 Hz), 7.39 – 7.35 (3 H, m), 7.20 (1 H, t, *J* = 7.2 Hz), 7.16 (1 H, t, *J* = 7.3 Hz), 7.12 (1 H, d, *J* = 7.4 Hz), 7.09 (1 H, s), 6.89 (1 H, d, *J* = 7.7 Hz), 5.27 (1 H, d, *J* = 18.0 Hz), 3.56 (1 H, d, *J* = 18.0 Hz), 3.37 – 3.35 (1 H, m), 3.28 (1 H, m), 3.06 – 3.00 (1 H, m), 2.71 (1 H, d, *J* = 17.2 Hz), 2.40 (3 H, s), 1.00 (9 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.49, 173.36, 164.42, 135.31, 134.94, 132.81, 129.63, 128.88, 127.60, 127.55, 126.45, 126.26, 69.83, 49.11, 46.02, 38.41, 27.66, 24.42, 21.90. M.P.: 179 – 181 °C. IR (in KBr): 2973, 1681, 1660, 1363, 1331, 1262, 1095, 984, 752, 704, 697 cm^{-1}. HRMS (EI): m/z [M + H$^+$] calcd for $C_{24}H_{28}N_4O_2$: 450.2291; found: 450.2285.

Product 3bb



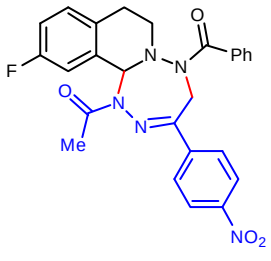
Yield of **3bb** : 80%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.66 (2 H, d, *J* = 6.9 Hz), 7.56 (2 H, d, *J* = 7.1 Hz), 7.41 (2 H, d, *J* = 6.4 Hz), 7.38 (4 H, s), 7.08 (2 H, s), 6.89 (1 H, t, *J* = 7.4 Hz), 6.72 (1 H, d, *J* = 8.7 Hz), 5.69 (1 H, d, *J* = 18.0 Hz), 3.95 (1 H, d, *J* = 18.0 Hz), 3.40 (2 H, s), 3.02 – 2.99 (1 H, m), 2.72 (1 H, d, *J* = 16.7 Hz), 2.47 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.67, 173.14, 162.67, 160.21, 156.19, 135.44, 134.99, 130.65, 130.58, 130.58, 130.43, 129.94, 128.49, 127.69, 127.62, 127.04, 115.28, 115.07, 112.77, 112.55, 70.45, 49.23, 48.10, 24.18, 22.01. M.P.: 189 – 190 °C. IR (in KBr): 2975, 2916, 1682, 1654, 1495, 1311, 1021, 927, 764, 698 cm^{-1}. HRMS (EI): m/z [M + H$^+$] calcd for $C_{26}H_{23}FN_4O_2$: 443.1883; found: 443.1871.

Product 3db



Yield of **3db** : 77%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.58 – 7.56 (4 H, m), 7.43 – 7.37 (3 H, m), 7.19 (1 H, s), 7.18 (1 H, s), 7.09 – 7.04 (1 H, m), 7.04 (1 H, s), 6.89 (1 H, t, *J* = 7.7 Hz), 6.73 (1 H, d, *J* = 8.9 Hz), 5.69 (1 H, d, *J* = 17.8 Hz), 3.95 (1 H, d, *J* = 18.0 Hz), 3.39 (2 H, s), 3.03 – 2.97 (1 H, m), 2.72 (1 H, d, *J* = 18.0 Hz), 2.46 (3 H, s), 2.36 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.60, 173.10, 162.63, 160.18, 156.78, 140.94, 134.99, 134.80, 132.48, 130.59, 130.51, 129.94, 129.21, 127.71, 127.61, 127.03, 115.27, 115.06, 112.87, 112.65, 70.45, 49.13, 47.85, 24.32, 22.11, 21.29. M.P.: 185 – 188 °C . IR (in KBr): 2917, 1677, 1653, 1494, 1380, 1289, 1255, 1019, 815, 705, 628 cm^{-1}. HRMS (EI): m/z [M + H$^+$] calcd for $C_{27}H_{25}FN_4O_2$: 457.2040; found: 457.2028.

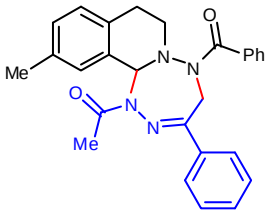
Product 3jb



Yield of **3jb** : 81%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 8.22 (2 H, d, *J* = 8.2 Hz), 7.80 (2 H, d, *J* = 8.0 Hz), 7.54 (2 H, d, *J* = 7.4 Hz), 7.46 – 7.43 (1 H, m), 7.40 (2 H,

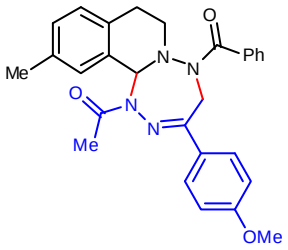
t, J = 7.3 Hz), 7.16 – 7.14 (2 H, m), 6.94 (1 H, t, J = 8.1 Hz), 6.63 (1 H, d, J = 8.8 Hz), 5.61 (1 H, d, J = 18.0 Hz), 3.95 (1 H, d, J = 18.0 Hz), 3.46 (2 H, s), 3.09 – 3.07 (1 H, m), 2.78 (1 H, d, J = 16.8 Hz), 2.51 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 174.09, 173.23, 162.66, 160.20, 151.80, 148.57, 141.46, 134.56, 131.05, 130.43, 130.17, 127.75, 127.58, 123.61, 115.52, 115.31, 112.27, 112.05, 70.42, 49.54, 48.14, 23.54, 21.92. M.P.: 210 – 212 °C. IR (in KBr): 2921, 1692, 1631, 1520, 1367, 1346, 1261, 869, 695, 634 cm^{-1}. HRMS (EI): m/z [M + H$^+$] calcd for $C_{26}H_{22}FN_5O_4$: 488.1734; found: 488.1729.

Product 3bc



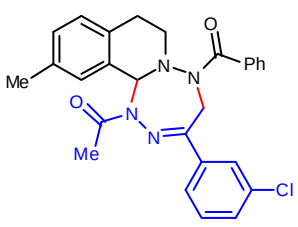
Yield of **3bc** : 85%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.64 (2 H, d, J = 7.2 Hz), 7.58 (2 H, d, J = 7.0 Hz), 7.43 – 7.40 (3 H, m), 7.39 – 7.36 (3 H, m), 7.12 (1 H, s), 7.01 – 6.98 (2 H, m), 6.79 (1 H, s), 5.68 (1 H, d, J = 18.0 Hz), 3.98 (1 H, d, J = 18.0 Hz), 3.40 – 3.38 (2 H, m), 3.05 – 2.99 (1 H, m), 2.71 (1 H, d, J = 16.9 Hz), 2.49 (3 H, s), 2.28 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.67, 173.23, 155.80, 136.37, 135.67, 135.11, 132.39, 131.62, 130.16, 129.78, 129.01, 128.88, 128.60, 128.33, 127.59, 127.02, 126.22, 70.52, 49.36, 48.25, 24.27, 22.08, 21.05. M.P.: 201 – 203 °C. IR (in KBr): 2932, 1686, 1655, 1382, 1367, 1279, 1022, 904, 768, 691 cm^{-1}. HRMS (EI): m/z [M + H$^+$] calcd for $C_{27}H_{26}N_4O_2$: 439.2134; found: 439.2124.

Product 3cc



Yield of **3cc** : 82%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.63 (2 H, s), 7.54 (2 H, s), 7.35 (3 H, d, J = 5.9 Hz), 6.95 (3 H, s), 6.86 (2 H, s), 6.79 (1 H, s), 5.63 (1 H, d, J = 18.0 Hz), 3.98 (1 H, d, J = 18.0 Hz), 3.80 (3 H, d, J = 4.6 Hz), 3.34 (2 H, d, J = 21.8 Hz), 2.94 (1 H, s), 2.68 (1 H, d, J = 12.9 Hz), 2.41 (3 H, s), 2.25 (3 H, d, J = 4.5 Hz). ^{13}C NMR (100 MHz, $CDCl_3$) 173.41, 172.80, 161.38, 156.76, 136.29, 136.19, 135.12, 132.46, 132.41, 131.62, 129.71, 128.64, 128.58, 127.81, 127.54, 126.36, 113.70, 70.66, 55.18, 49.02, 47.53, 24.76, 22.10, 21.01. M.P.: 166 – 168 °C. IR (in KBr): 2918, 1679, 1651, 1610, 1514, 1380, 1254, 1173, 1023, 740, 695 cm^{-1}. HRMS (EI): m/z [M + H$^+$] calcd for $C_{28}H_{28}N_4O_3$: 469.2240; found: 469.2226.

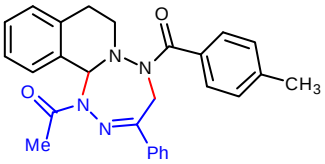
Product 3hc



Yield of **3hc** : 89%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.56 (3 H, d, J = 4.8 Hz), 7.49 (1 H, d, J = 7.1 Hz), 7.41 – 7.35 (4 H, m), 7.28 (1 H, t, J = 7.8 Hz), 7.13 (1 H, s), 7.00 (2 H, s), 6.72 (1 H, s), 5.57 (1 H, d, J = 18.0 Hz), 3.92 (1 H, d, J = 18.0 Hz), 3.39 (2 H, s), 3.05 – 3.01 (1 H, m), 2.71 (1 H, d, J = 16.7 Hz), 2.49 (3 H, s), 2.27 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.91,

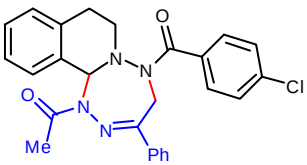
173.32, 153.42, 137.46, 136.54, 134.96, 134.36, 132.29, 131.60, 130.05, 129.90, 129.59, 129.07, 128.74, 127.65, 127.59, 127.11, 126.08, 125.02, 70.53, 49.50, 48.36, 24.00, 22.05, 21.05. M.P.: 187 – 188 °C. IR (in KBr): 2921, 1693, 1654, 1364, 1355, 1167, 984, 709, 687 cm^{-1}. HRMS (EI): m/z [M + H^+] calcd for $C_{27}H_{25}ClN_4O_2$: 473.1744; found: 473.1734.

Product 3ad



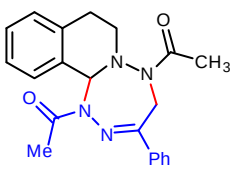
Yield of **3ad** : 88%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.60 (2 H, s), 7.49 (2 H, d, *J* = 7.7 Hz), 7.37 (1 H, d, *J* = 7.0 Hz), 7.34 (2 H, d, *J* = 7.4 Hz), 7.35 – 7.34 (4 H, m), 7.10 (2 H, s), 6.97 (1 H, s), 5.63 (1 H, d, *J* = 18.0 Hz), 3.94 (1 H, d, *J* = 18.0 Hz), 3.47 – 3.41 (2 H, m), 3.06 (1 H, d, *J* = 7.3 Hz), 2.75 (1 H, d, *J* = 17.0 Hz), 2.44 (3 H, s), 2.35 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.60, 173.12, 155.89, 139.98, 135.58, 134.81, 132.67, 131.93, 130.09, 128.94, 128.25, 127.79, 127.69, 126.90, 126.69, 125.76, 70.54, 49.14, 48.24, 24.68, 21.94, 21.28. M.P.: 94 – 96 °C. IR (in KBr): 2921, 1688, 1645, 1382, 1278, 984, 749, 694 cm^{-1}. HRMS (EI): m/z [M + Na^+] calcd for $C_{27}H_{26}N_4O_2$: 461.1953; found: 461.1945.

Product 3ae



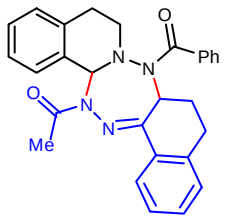
Yield of **3ae** : 81%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.59 (2 H, d, *J* = 7.1 Hz), 7.54 (2 H, d, *J* = 8.0 Hz), 7.38 (1 H, d, *J* = 6.8 Hz), 7.35 (4 H, m), 7.17 (2 H, s), 7.11 (2 H, s), 6.96 (1 H, s), 5.64 (1 H, d, *J* = 18.0 Hz), 3.93 (1 H, d, *J* = 18.0 Hz), 3.45 (1 H, m), 3.36 (1 H, s), 3.05 (1 H, s), 2.76 (1 H, d, *J* = 16.5 Hz), 2.45 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 173.32, 172.43, 155.41, 135.95, 135.48, 134.72, 133.30, 132.47, 130.24, 129.36, 129.07, 128.37, 127.93, 127.89, 126.91, 126.88, 125.82, 70.56, 49.26, 48.26, 24.61, 21.99. M.P.: 85 – 88 °C. IR (in KBr): 2923, 1643, 1552, 1382, 1309, 1089, 839, 750 cm^{-1}. HRMS (EI): m/z [M + Na^+] calcd for $C_{26}H_{23}ClN_4O_2$: 481.1407; found: 481.1400.

Product 3af



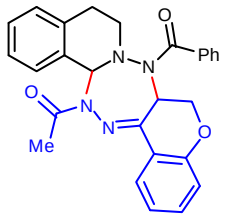
Yield of **3af** : 87%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.54 (2 H, d, *J* = 7.1), 7.37 – 7.35 (1 H, m), 7.31 (2 H, t, *J* = 7.3 Hz), 7.17 (3 H, s), 7.12 (1 H, s), 6.96 (1 H, d, *J* = 4.7 Hz), 5.53 (1 H, d, *J* = 18.0 Hz), 3.74 (1 H, d, *J* = 18.0 Hz), 3.61 (1 H, s), 3.49 (1 H, d, *J* = 13.8 Hz), 3.07 (1 H, s), 2.83 (1 H, d, *J* = 17.2 Hz), 2.41 (3 H, s), 2.23 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 175.51, 173.31, 156.12, 135.56, 134.76, 132.53, 130.09, 129.13, 128.38, 127.79, 126.89, 126.80, 125.88, 70.86, 49.09, 47.27, 24.49, 21.90, 20.85. M.P.: 104 – 106 °C. IR (in KBr): 2923, 1669, 1383, 1263, 1132, 976, 846, 760 cm^{-1}. HRMS (EI): m/z [M + Na^+] calcd for $C_{21}H_{22}N_4O_2$: 385.1640; found: 385.1643.

Product 3ma



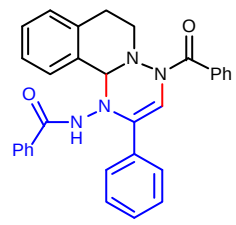
Yield of **3ma** : 64%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 8.07 (1 H, d, *J* = 6.7 Hz), 7.57 (2 H, s), 7.44 (2 H, s), 7.36 (3 H, s), 7.21 – 7.15 (3 H, m), 7.00 (1 H, s), 6.63 (1 H, s), 5.84 (1 H, s), 3.48 (2 H, d, *J* = 7.0 Hz), 3.27 (1 H, s), 2.95 (2 H, s), 2.79 (1 H, s), 2.68 (2 H, d, *J* = 15.6 Hz), 2.11 (3 H, s), 2.00 (1 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 172.09, 171.25, 169.47, 141.36, 135.48, 133.97, 133.03, 132.11, 130.39, 129.81, 128.33, 127.67, 127.36, 127.15, 126.87, 126.78, 126.73, 68.06, 58.32, 47.50, 29.85, 29.75, 28.21, 22.36. M.P.: 197 – 200 °C. IR (in KBr): 2895, 2844, 1657, 1584, 1386, 1323, 1035, 842, 741, 614 cm^{-1}. HRMS (EI): m/z [M + H^+] calcd for $C_{28}H_{26}N_4O_2$: 451.2134; found: 451.2125.

Product 3na



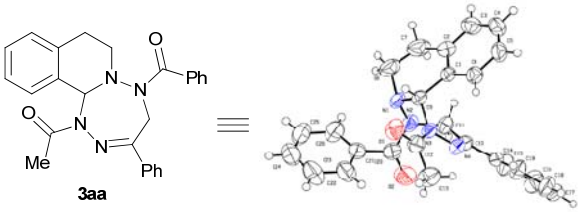
Yield of **3na** : 62%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 8.07 (1 H, s), 7.57 (2 H, s), 7.44 (2 H, s), 7.37 (3 H, s), 7.19 – 7.17 (2 H, m), 7.01 (2 H, s), 6.67 (1 H, s), 6.10 (1 H, s), 4.62 (1 H, s), 4.22 (1 H, s), 3.48 (2 H, d, *J* = 7.6 Hz), 3.25 (1 H, s), 2.77 (2 H, d, *J* = 48.7), 2.12 (3 H, s). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) 172.73, 169.60, 166.88, 158.74, 134.81, 134.35, 133.93, 133.03, 130.28, 129.60, 127.78, 127.24, 127.00, 126.78, 126.59, 126.17, 122.32, 118.69, 117.94, 68.86, 67.75, 56.90, 48.38, 29.89, 22.41. M.P.: 169 – 171 °C. IR (in KBr): 2895, 2844, 1663, 1612, 1595, 1386, 1321, 1037, 771, 741 cm^{-1}. HRMS (EI): m/z [M + H^+] calcd for $C_{27}H_{24}N_4O_3$: 453.1927; found: 453.1922.

Product 5

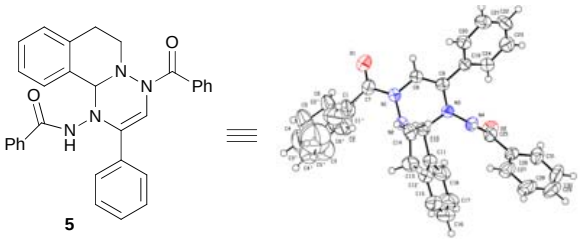


Yield of **5** : 62%. ^{1}H NMR (600 MHz, $CDCl_3$) δ (ppm) 7.65 (2 H, s), 7.48 (2 H, s), 7.30 (5 H, s), 7.21 (4 H, d, *J* = 11.9 Hz), 7.15 (2 H, s), 7.07 – 7.04 (5 H, m), 6.01 (1 H, s), 3.79 (1 H, s), 2.88 (2 H, s), 2.82 (1 H, s). ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) δ (ppm) 166.42, 164.25, 133.14, 132.97, 132.95, 132.46, 132.13, 129.44, 128.47, 128.16, 127.08, 126.74, 126.68, 126.39, 126.29, 126.02, 125.85, 125.60, 123.42, 101.88, 72.31, 44.10, 27.04. M.P.: 186 – 189 °C. IR (in KBr): 3273, 2968, 1644, 1620, 1444, 1314, 1057, 1027, 756, 696 cm^{-1}. HRMS (EI): m/z [M + H^+] calcd for $C_{31}H_{26}N_4O_2$: 487.2134; found: 487.2125

4. X-Ray structure of 3aa and 5

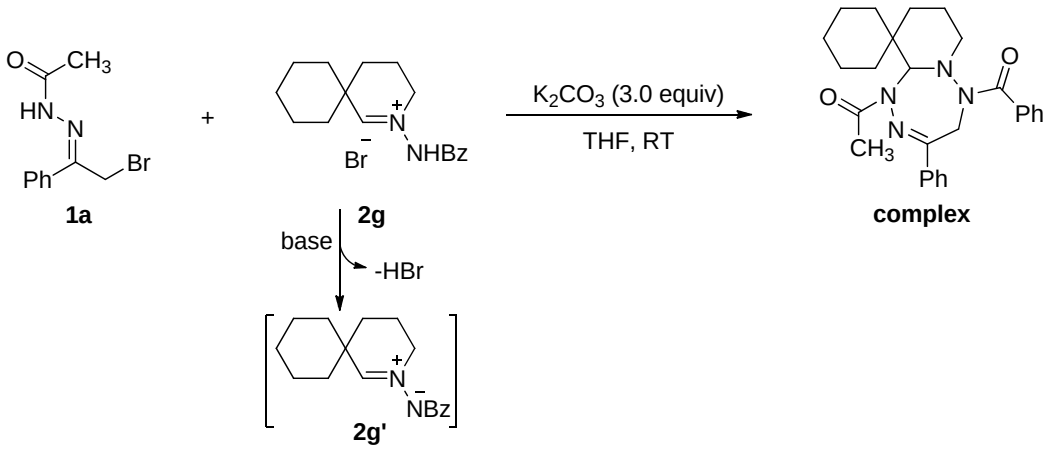


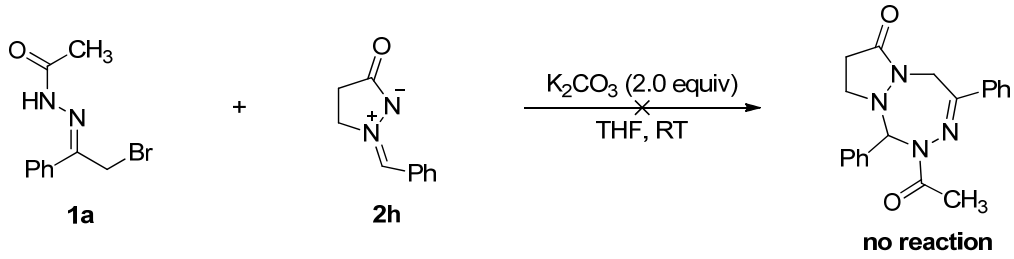
3aa



5

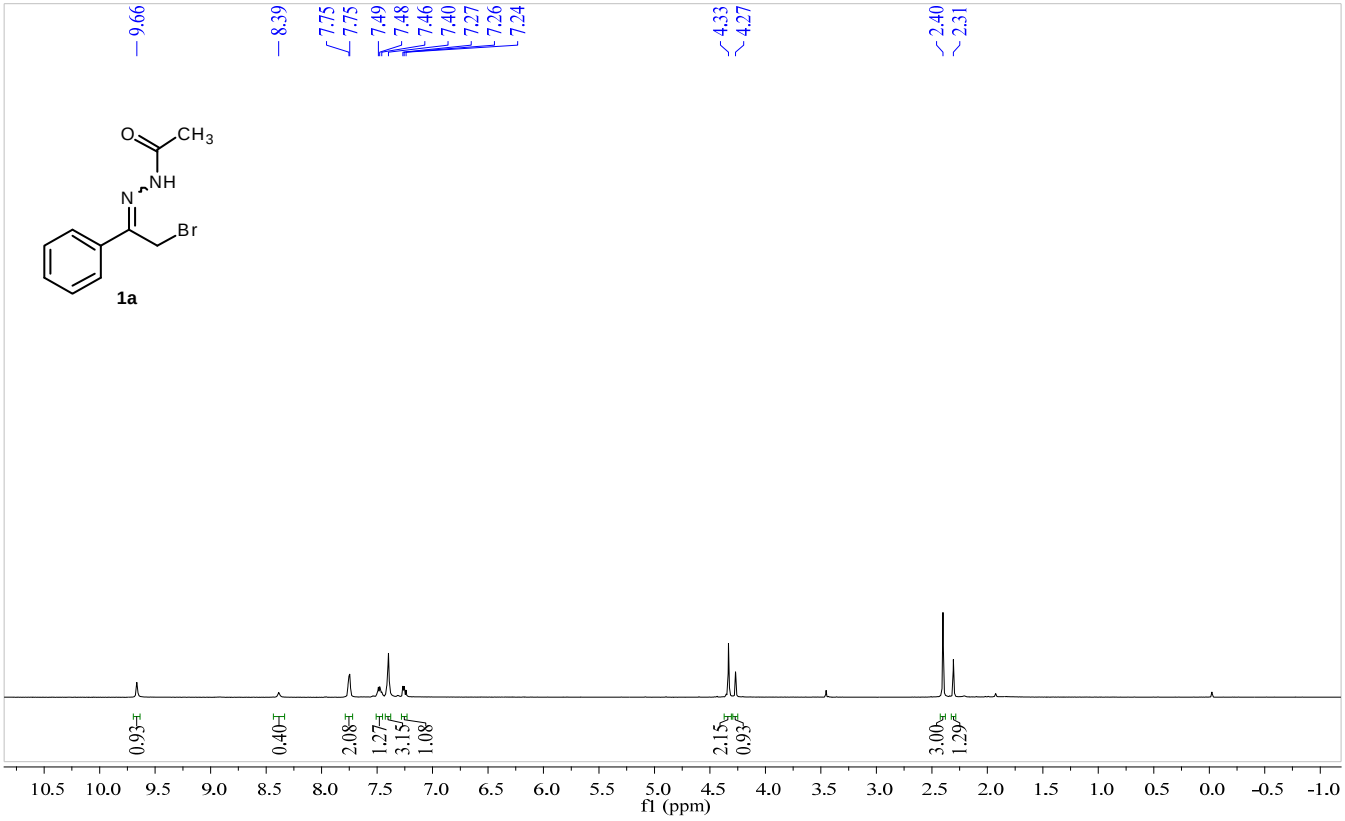
5. Preliminary studies on [4+3] Cycloaddition with in situ generated C,N-cyclic azomethine imine 2g' and N,N'-cyclic azomethine imine 2h.

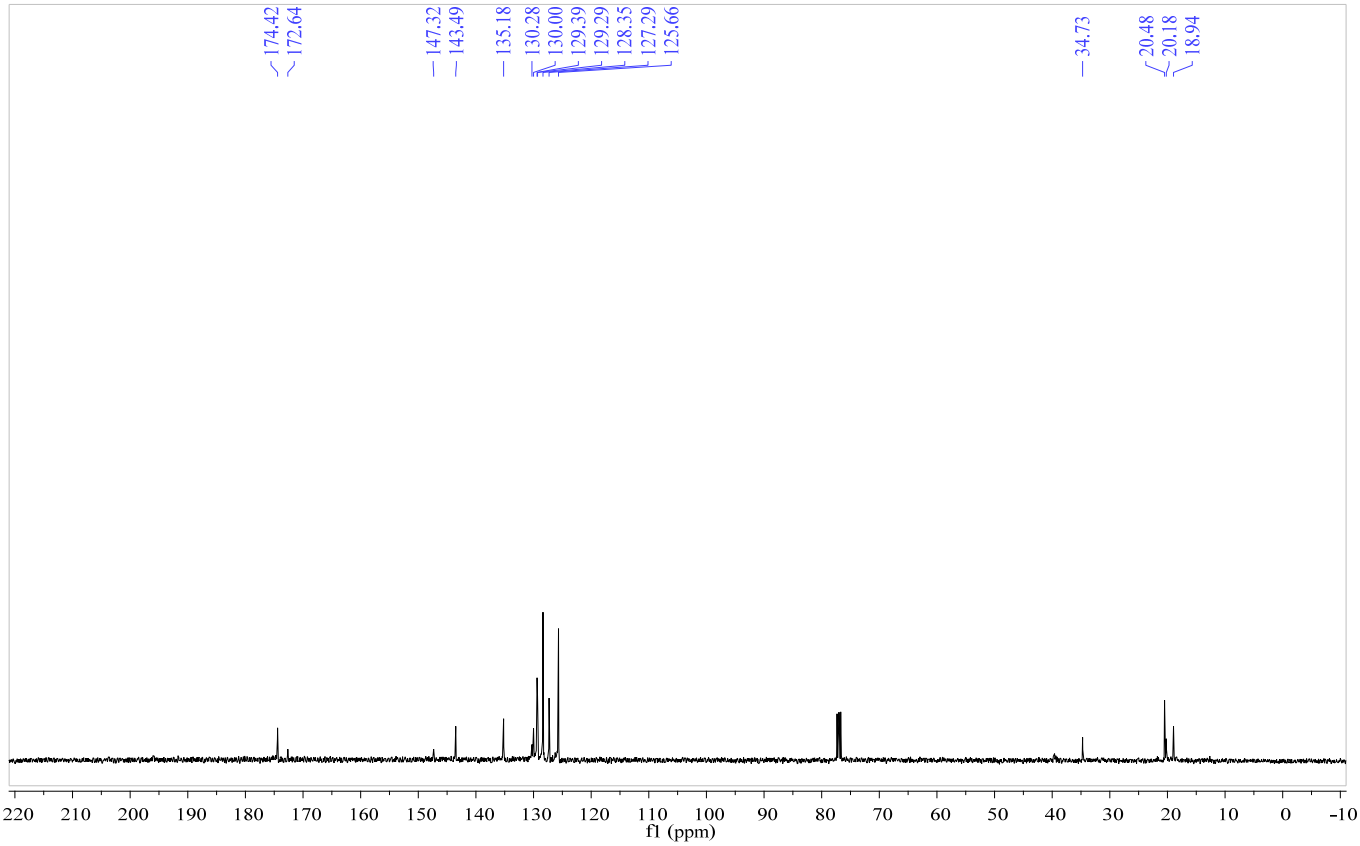




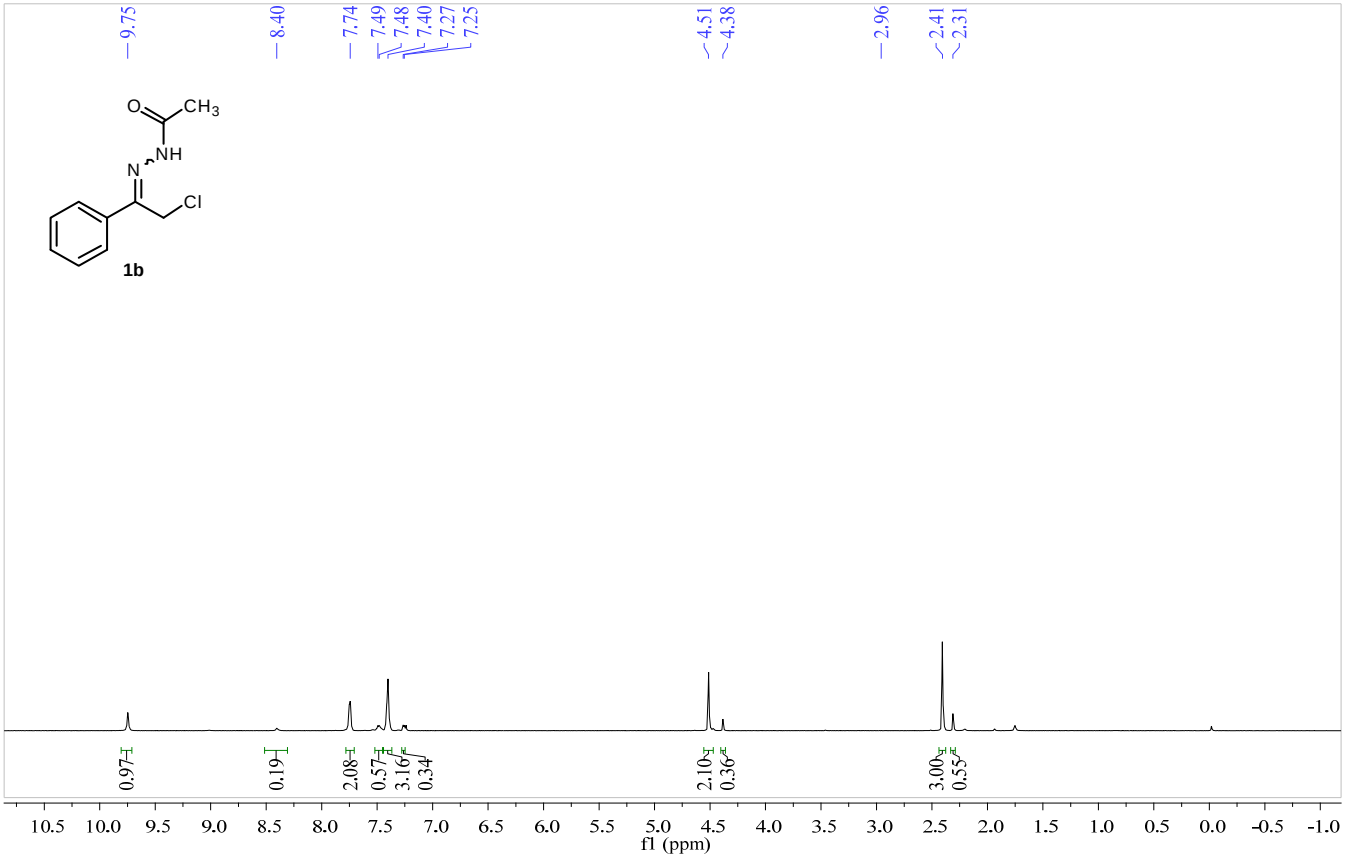
6. NMR and HRMS spectra of Hydrazones, 2g and the Cyclic Products

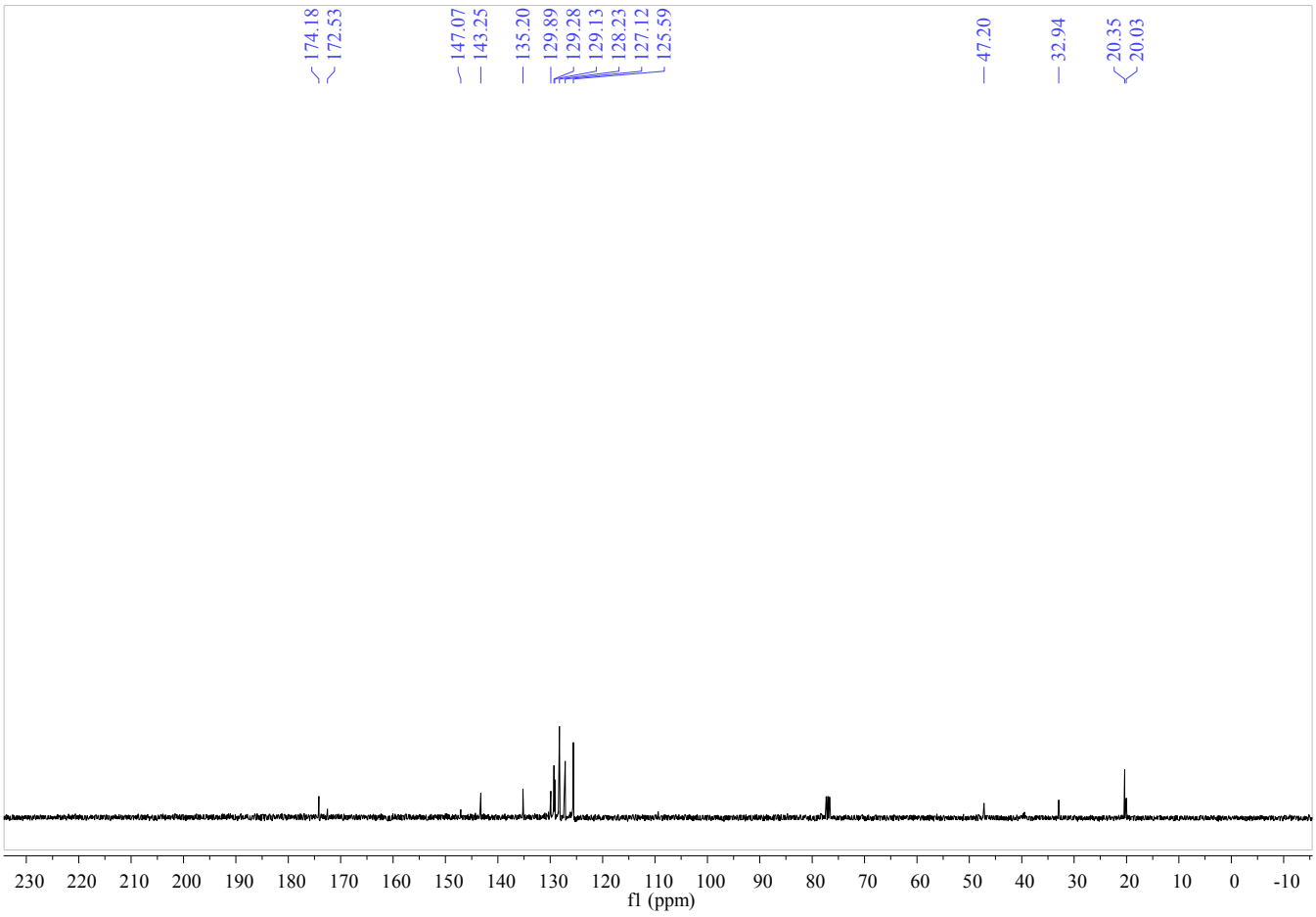
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) spectrum of hydrazone 1a



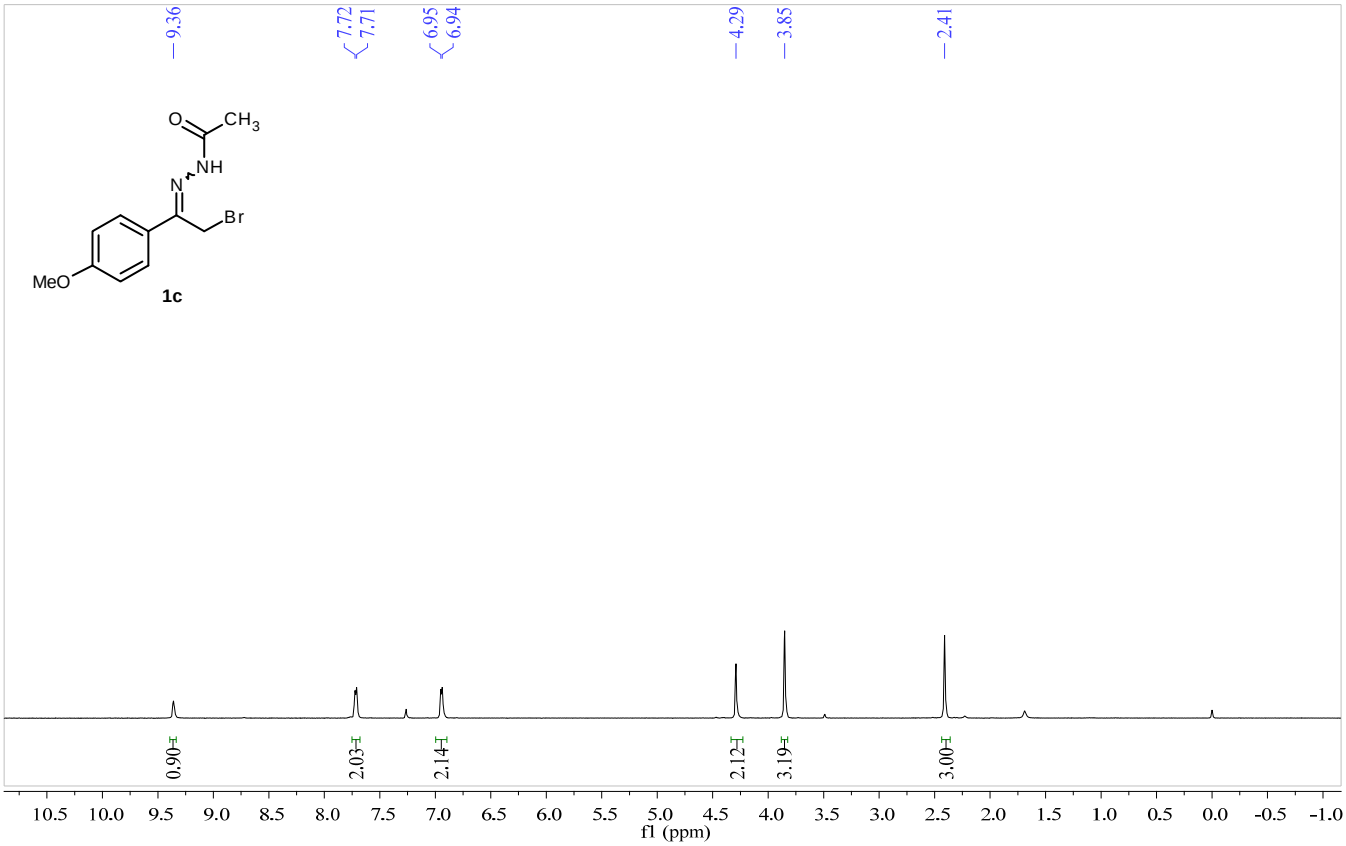


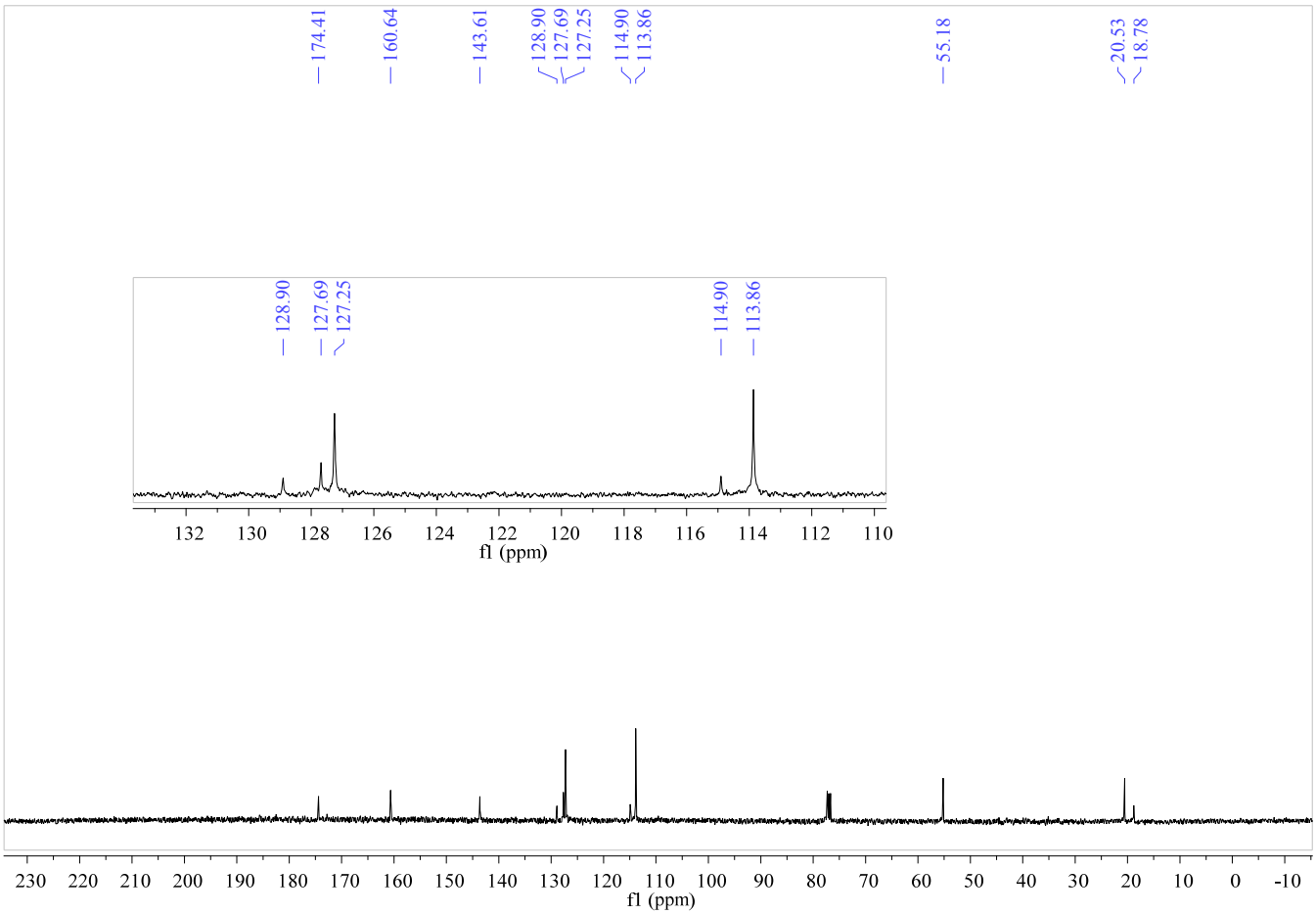
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) spectrum of hydrazone 1b



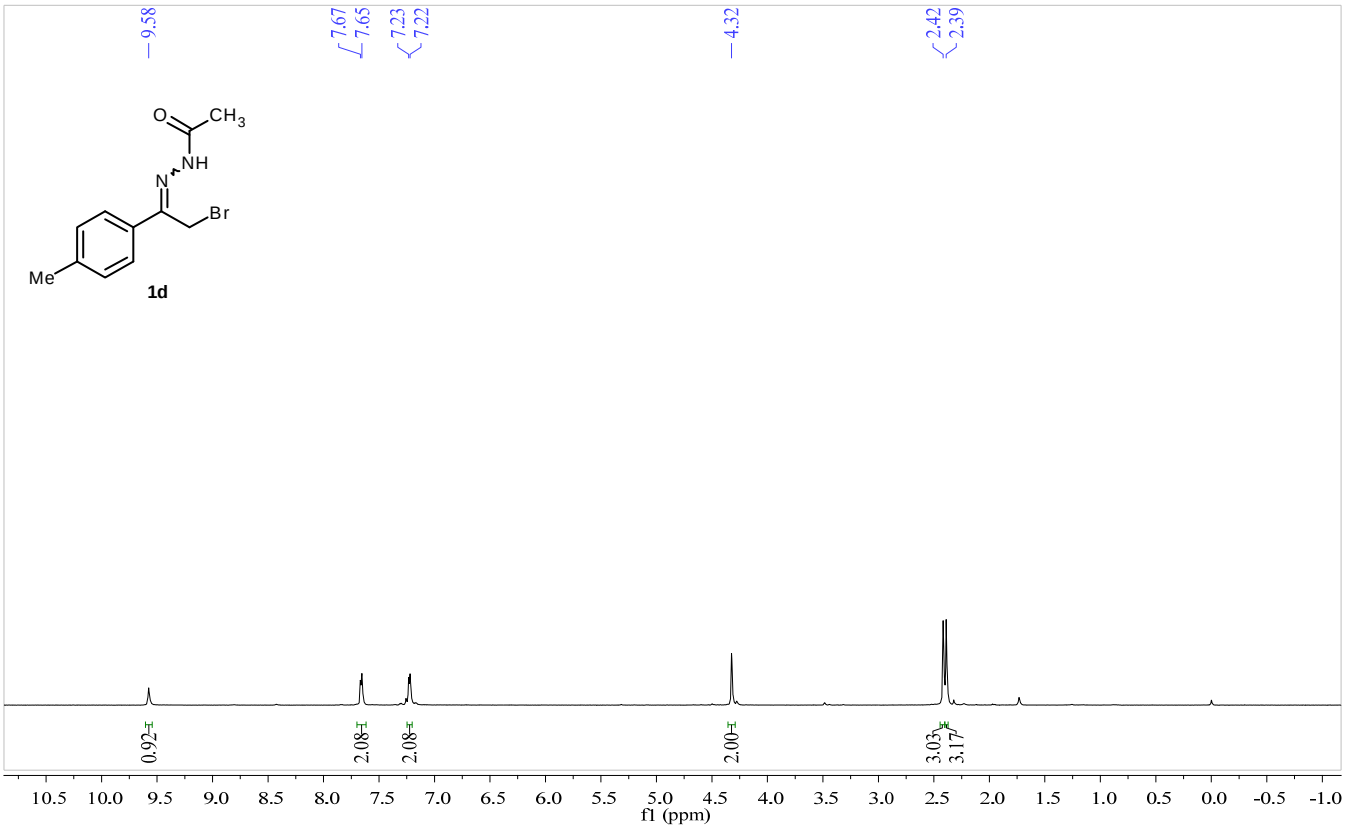


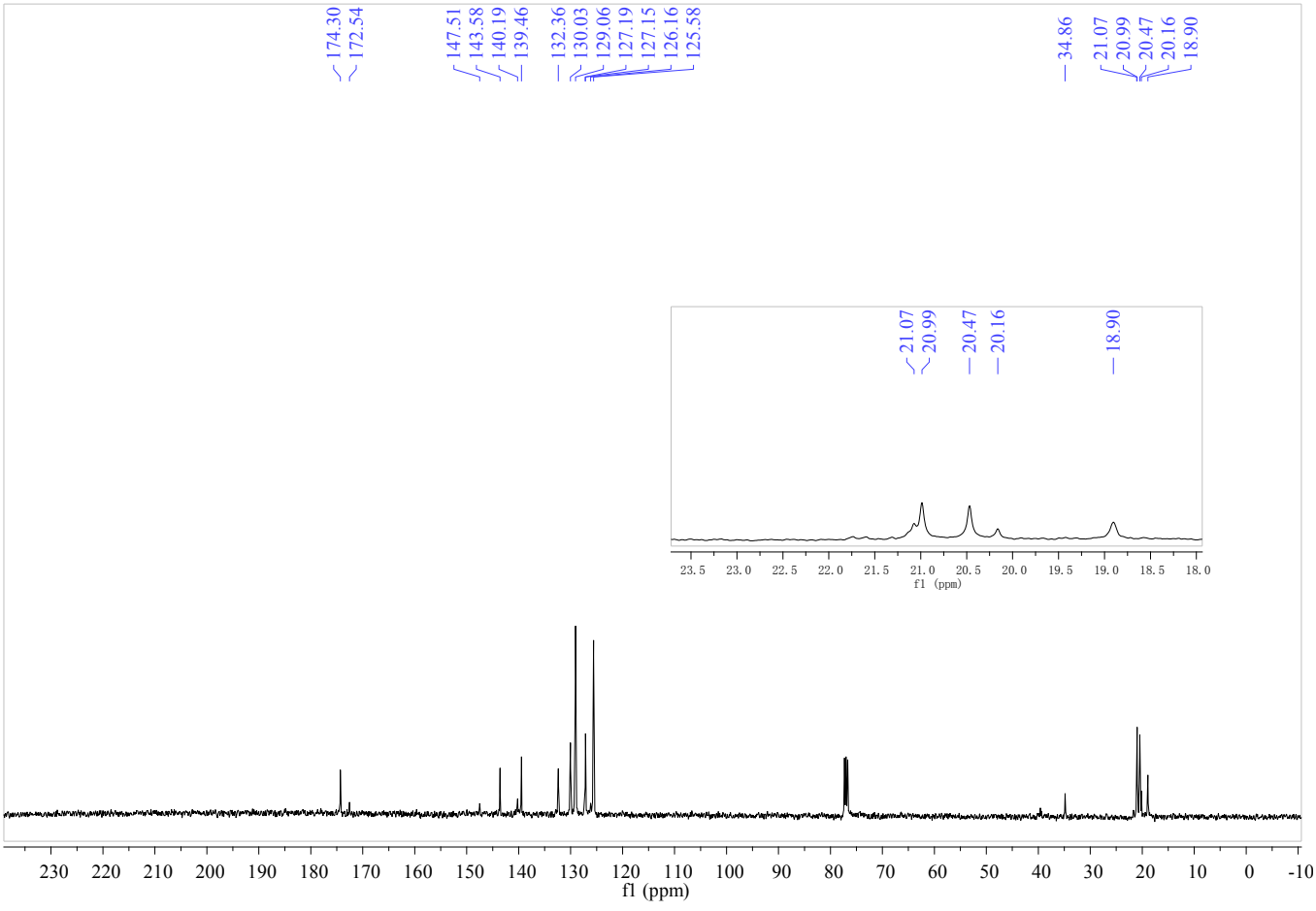
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) spectrum of hydrazone 1c



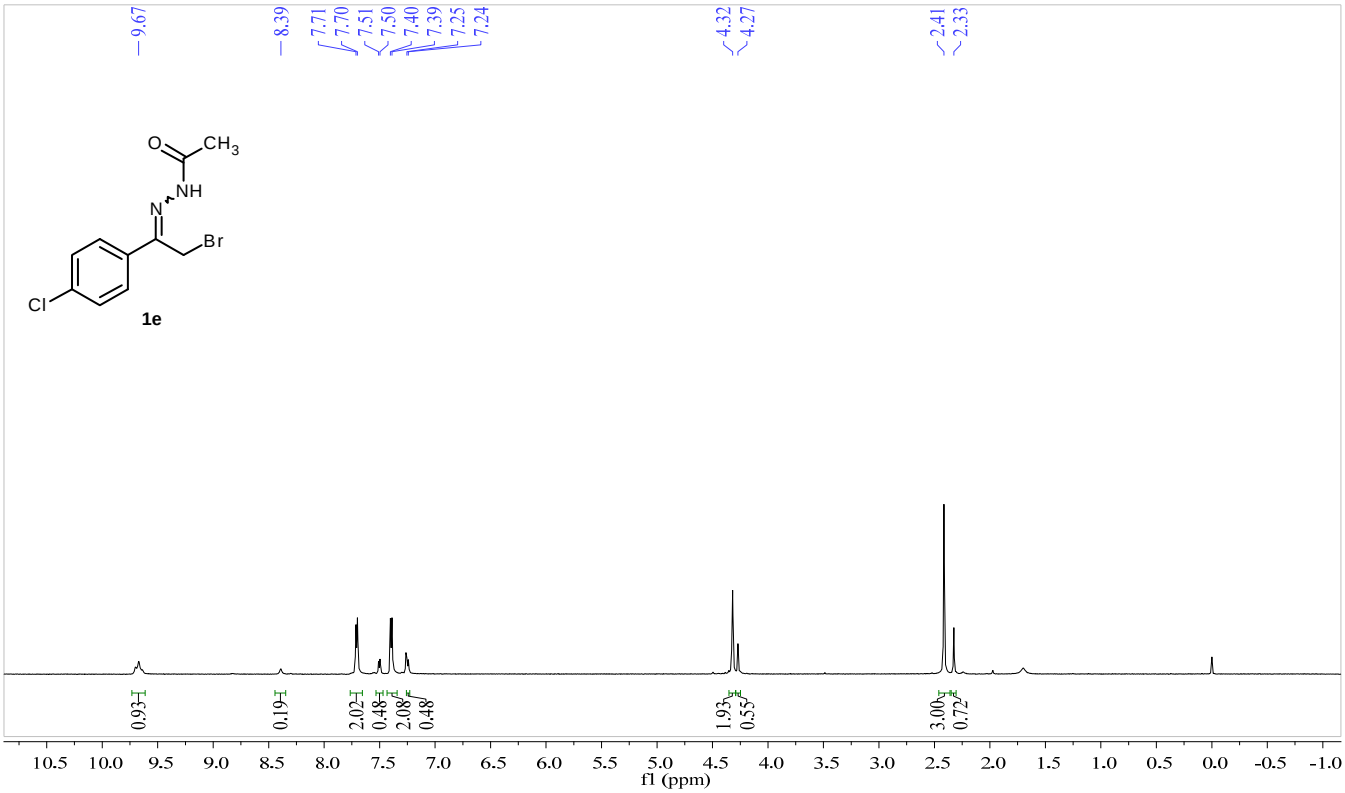


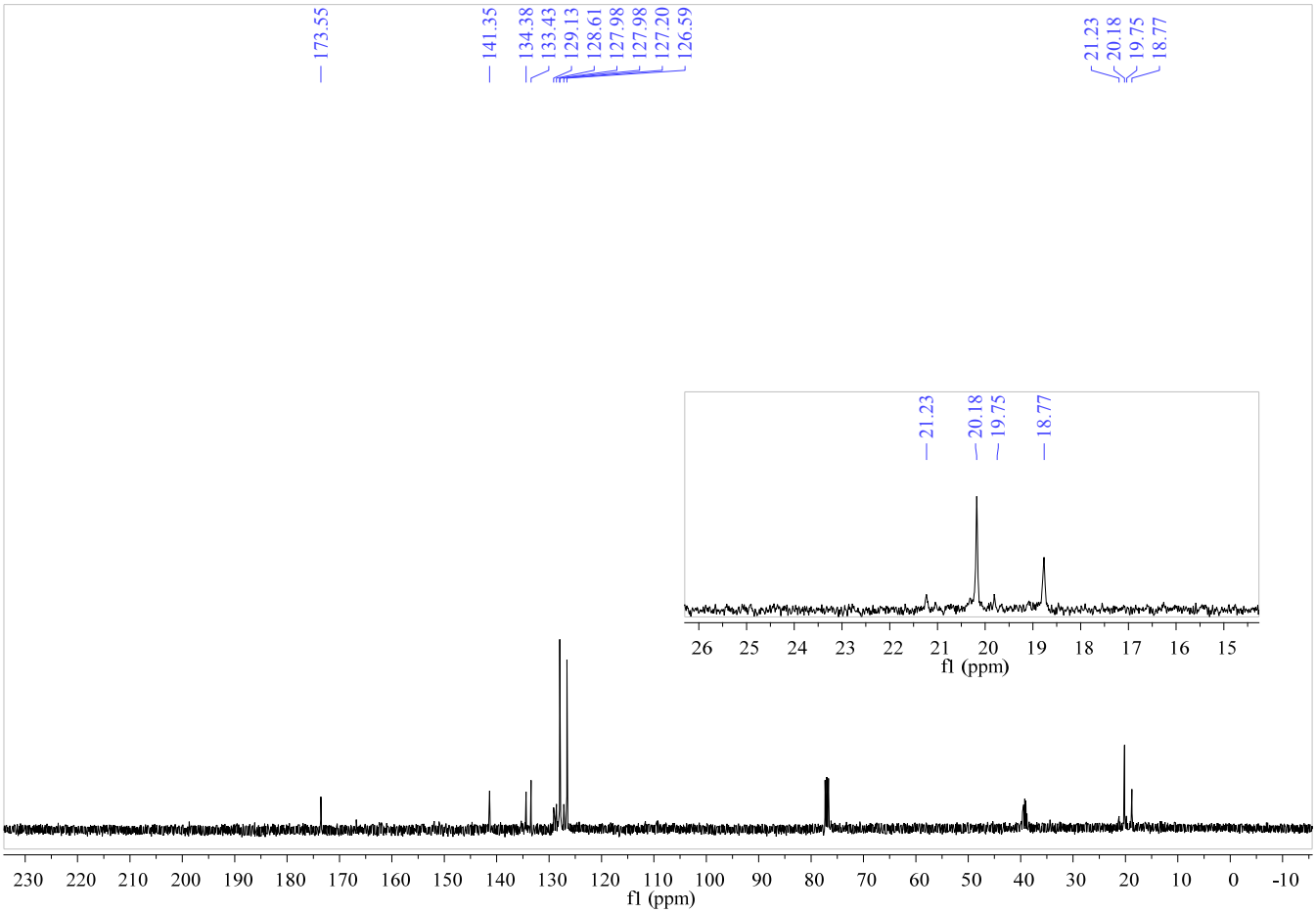
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) spectrum of hydrazone 1d



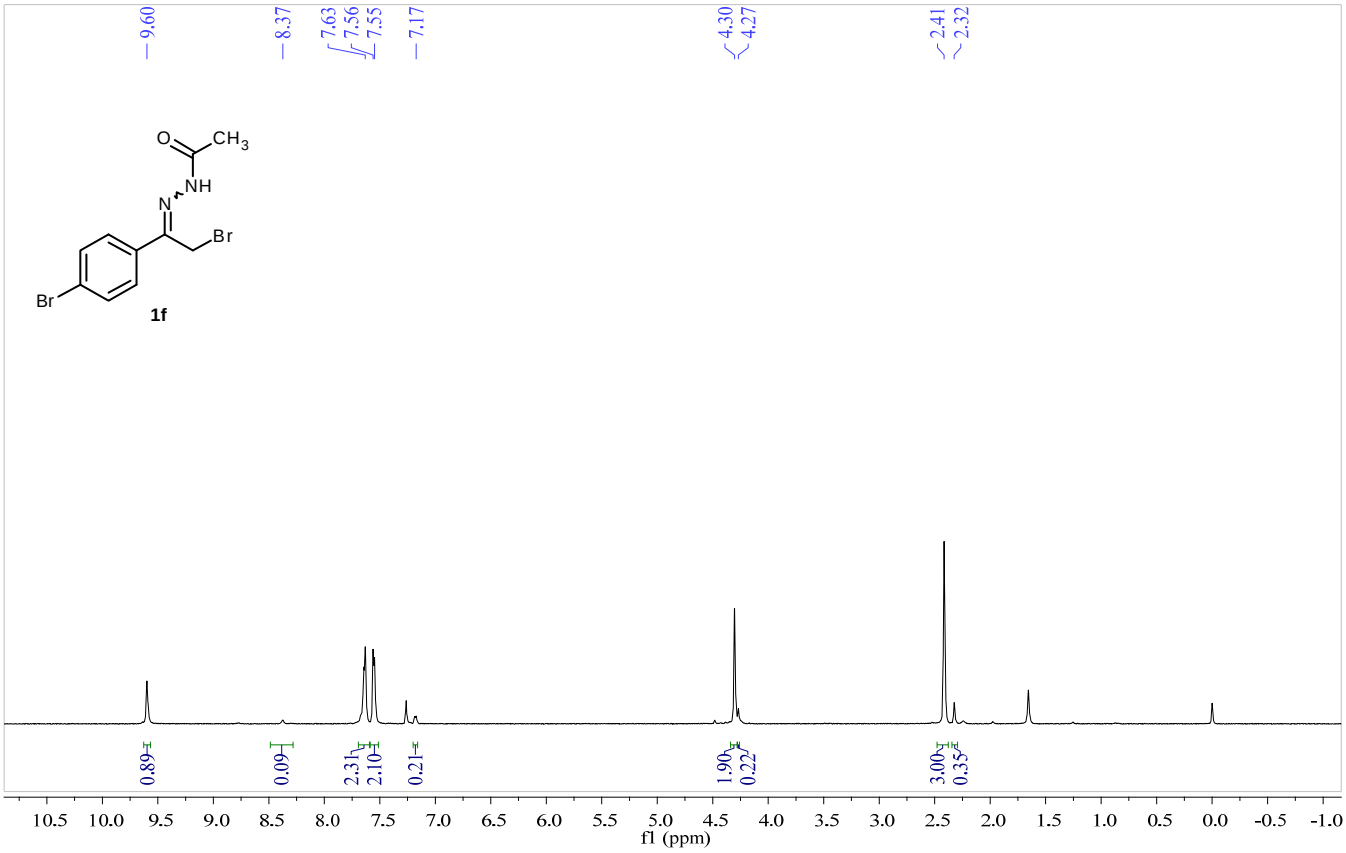


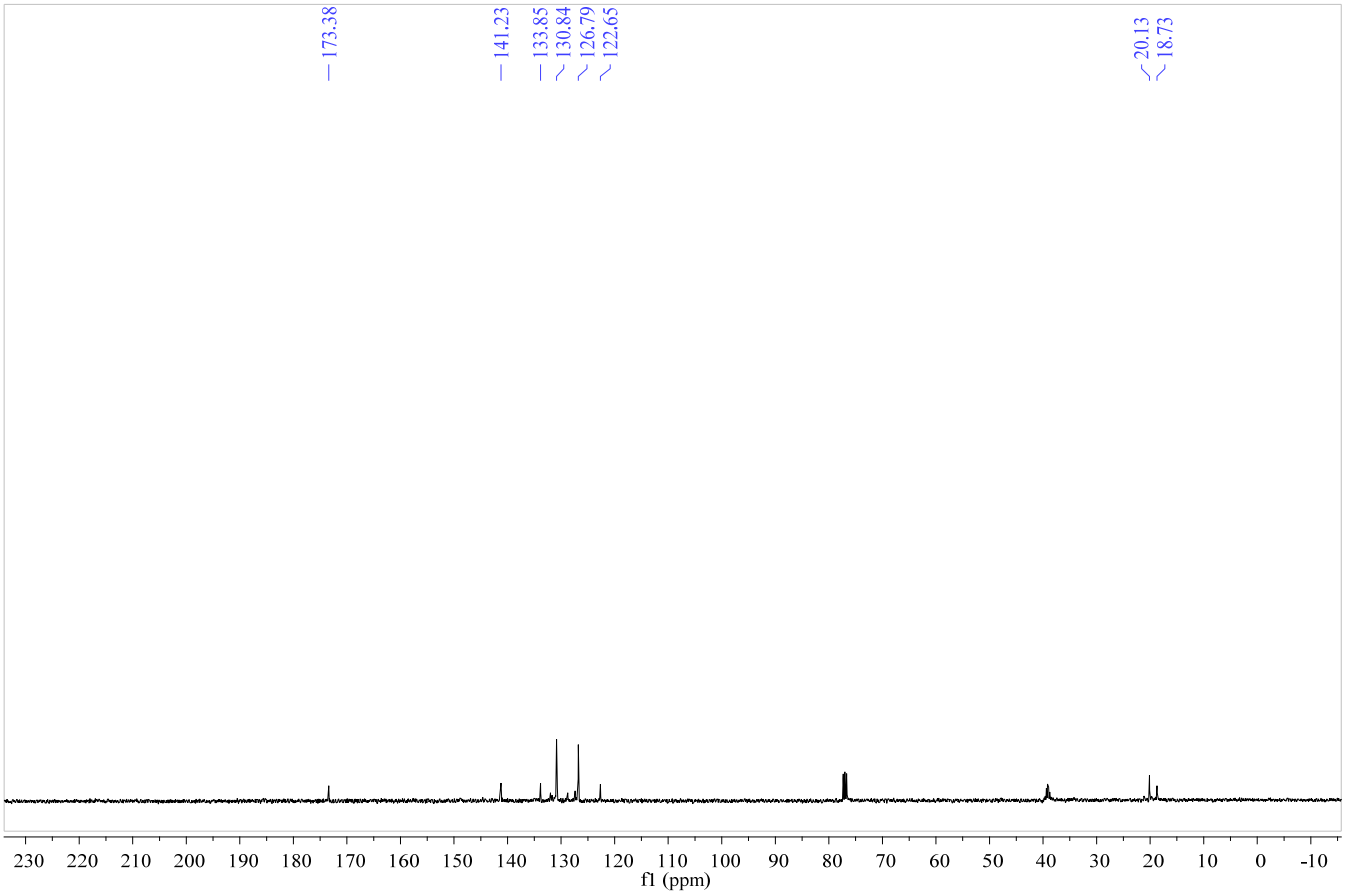
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) spectrum of hydrazone 1e



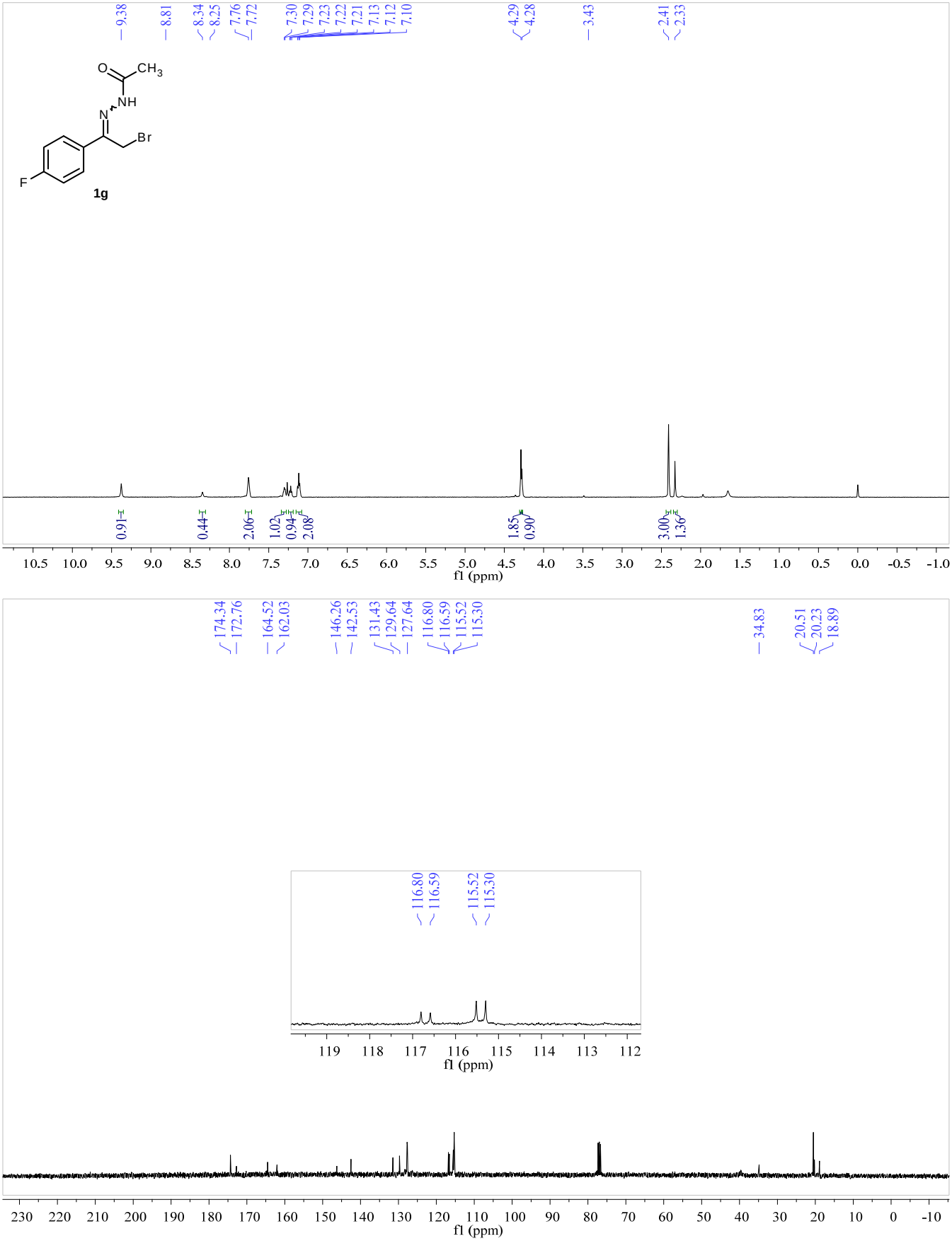


^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) spectrum of hydrazone 1f

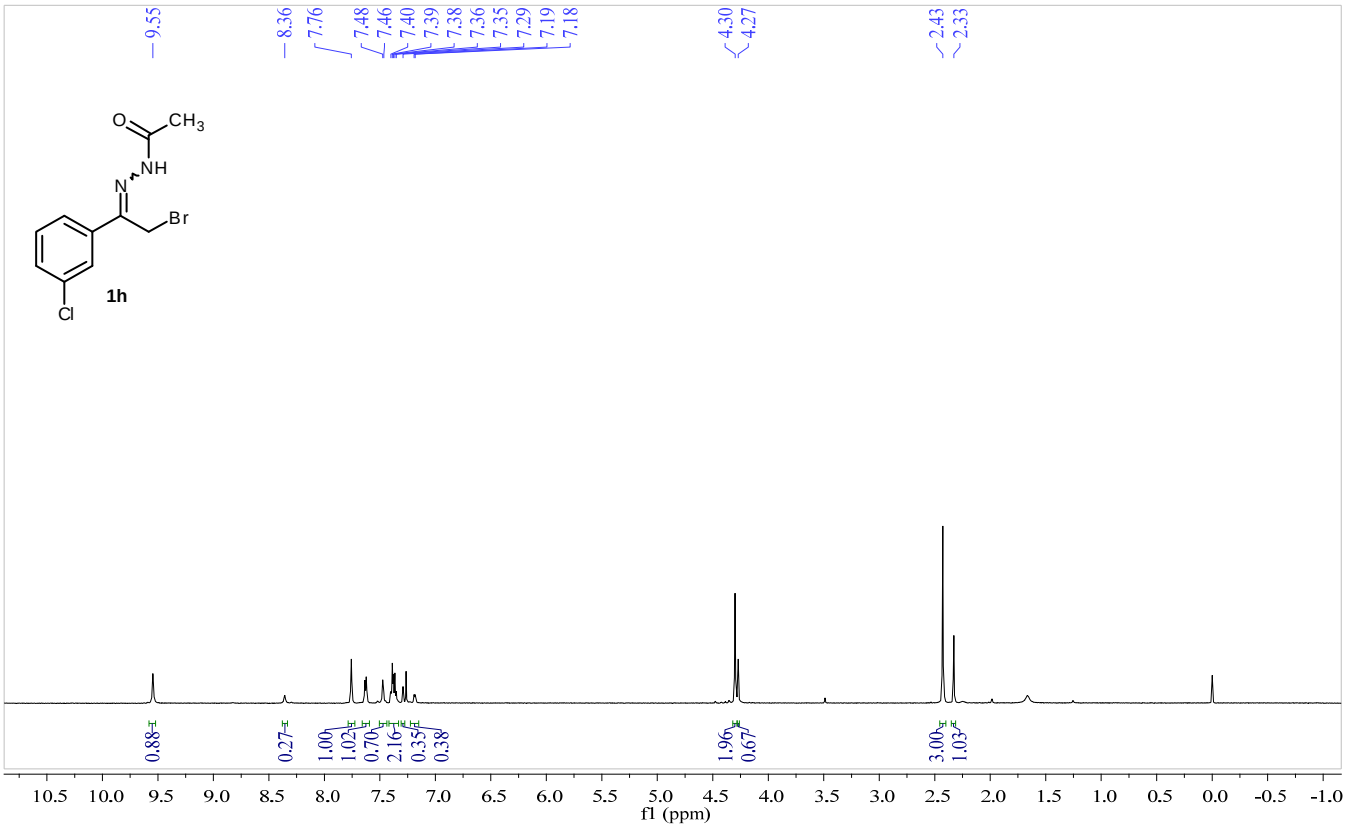


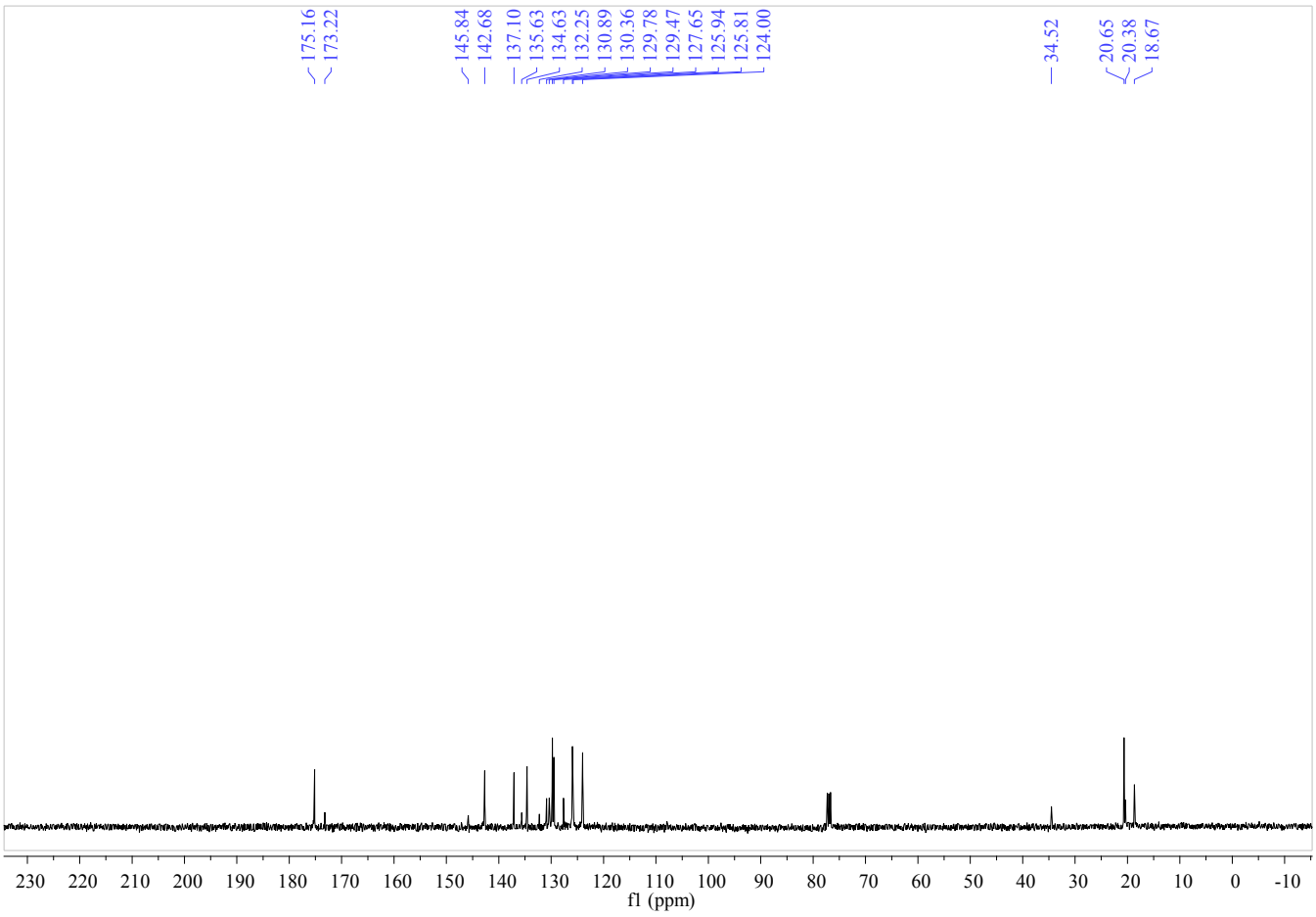


^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) spectrum of hydrazone 1g

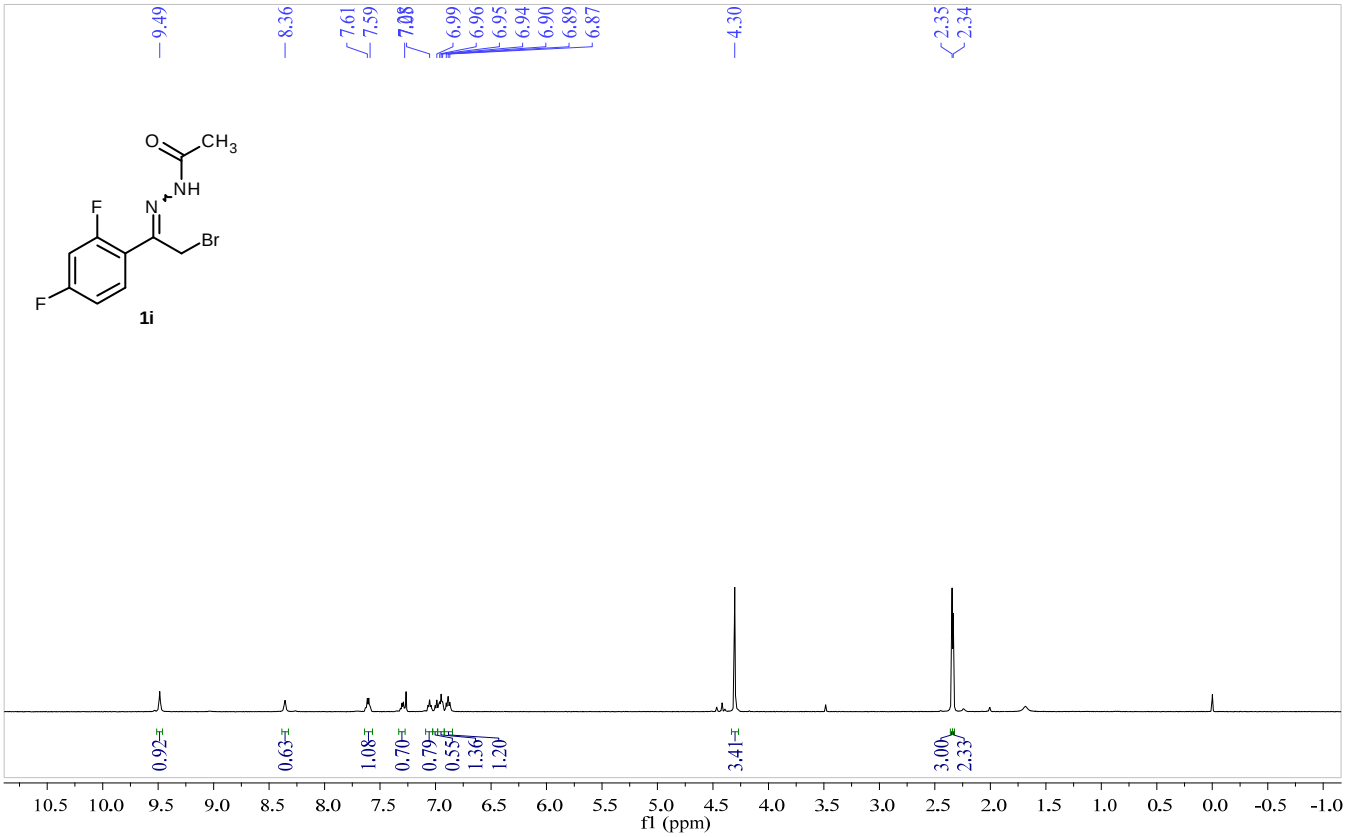


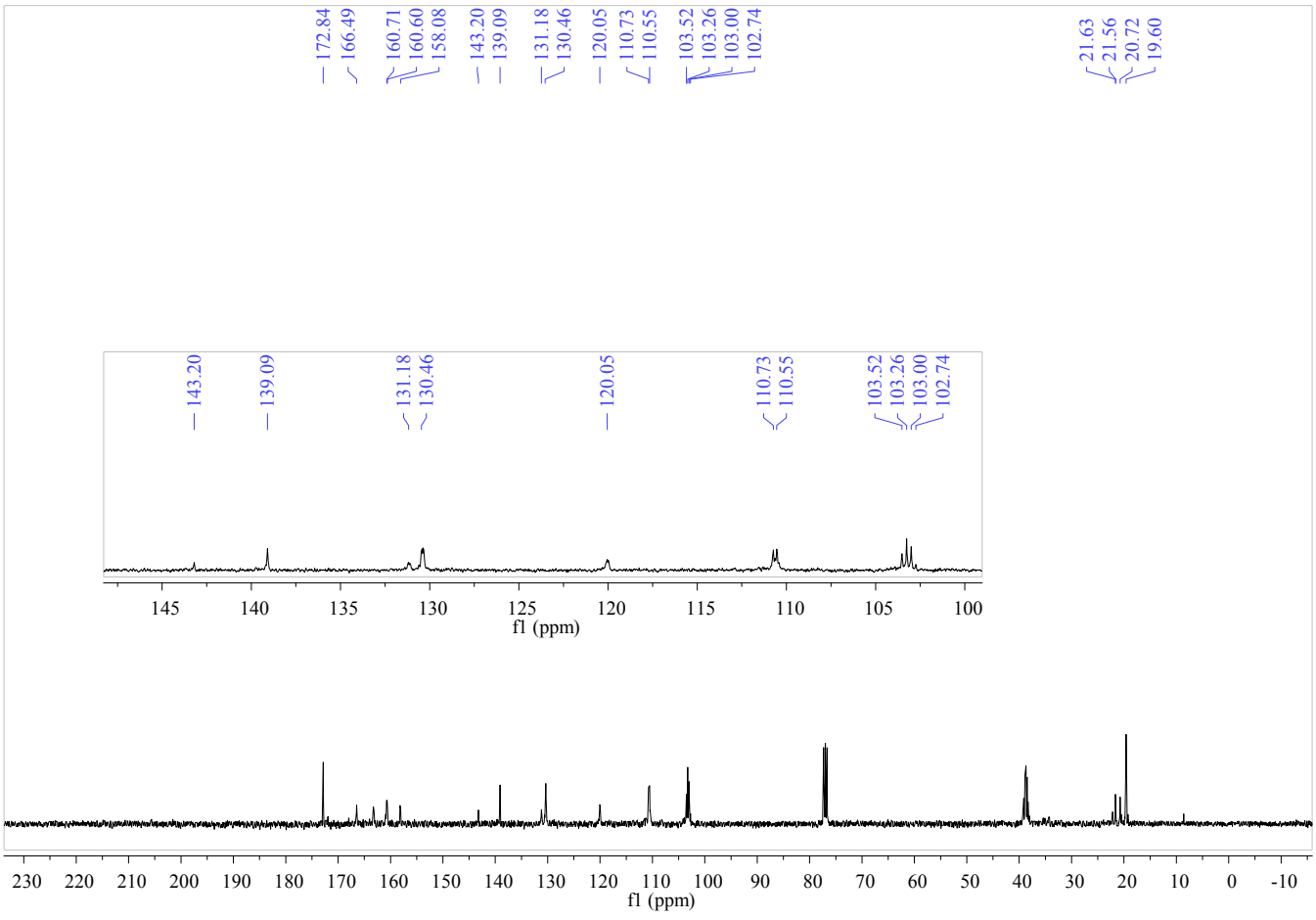
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of hydrazone 1h



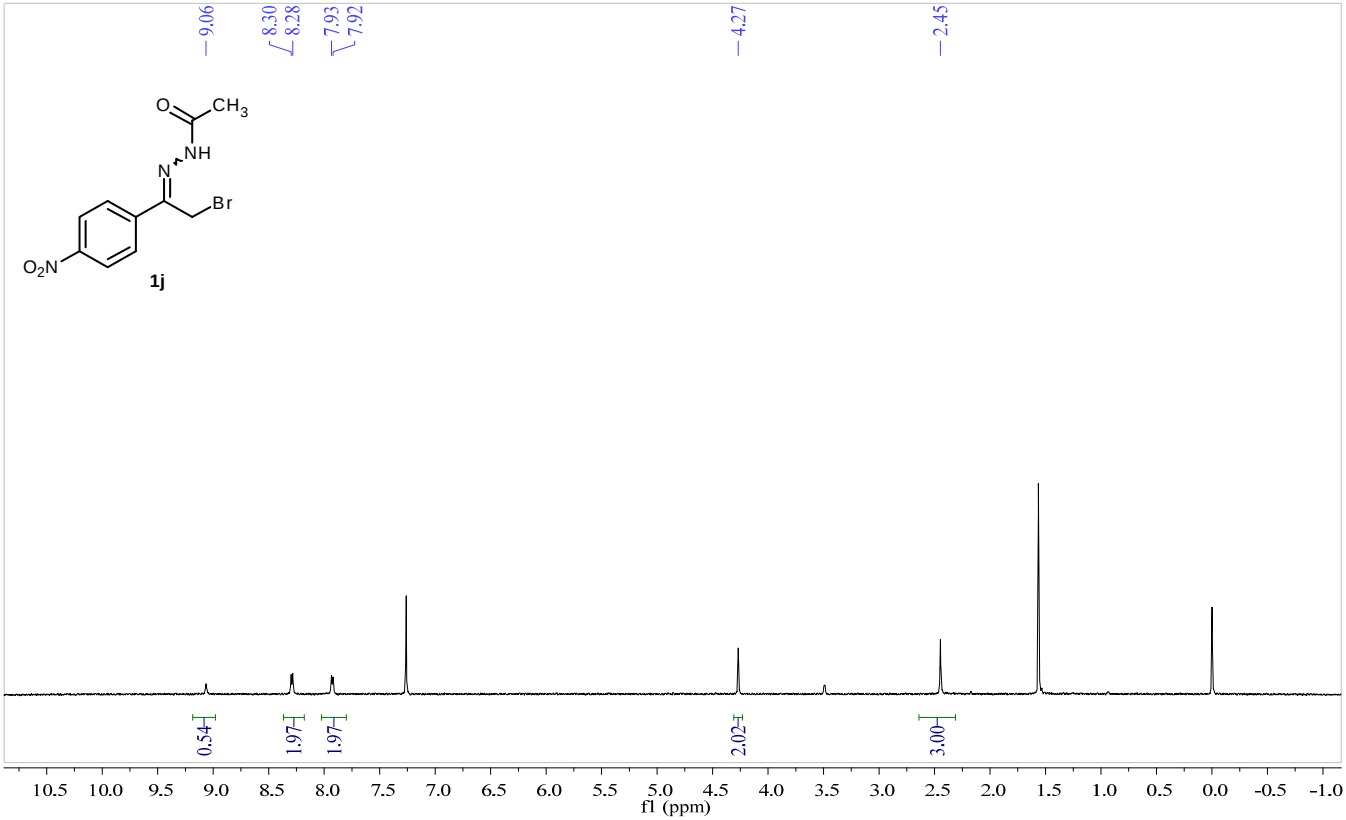


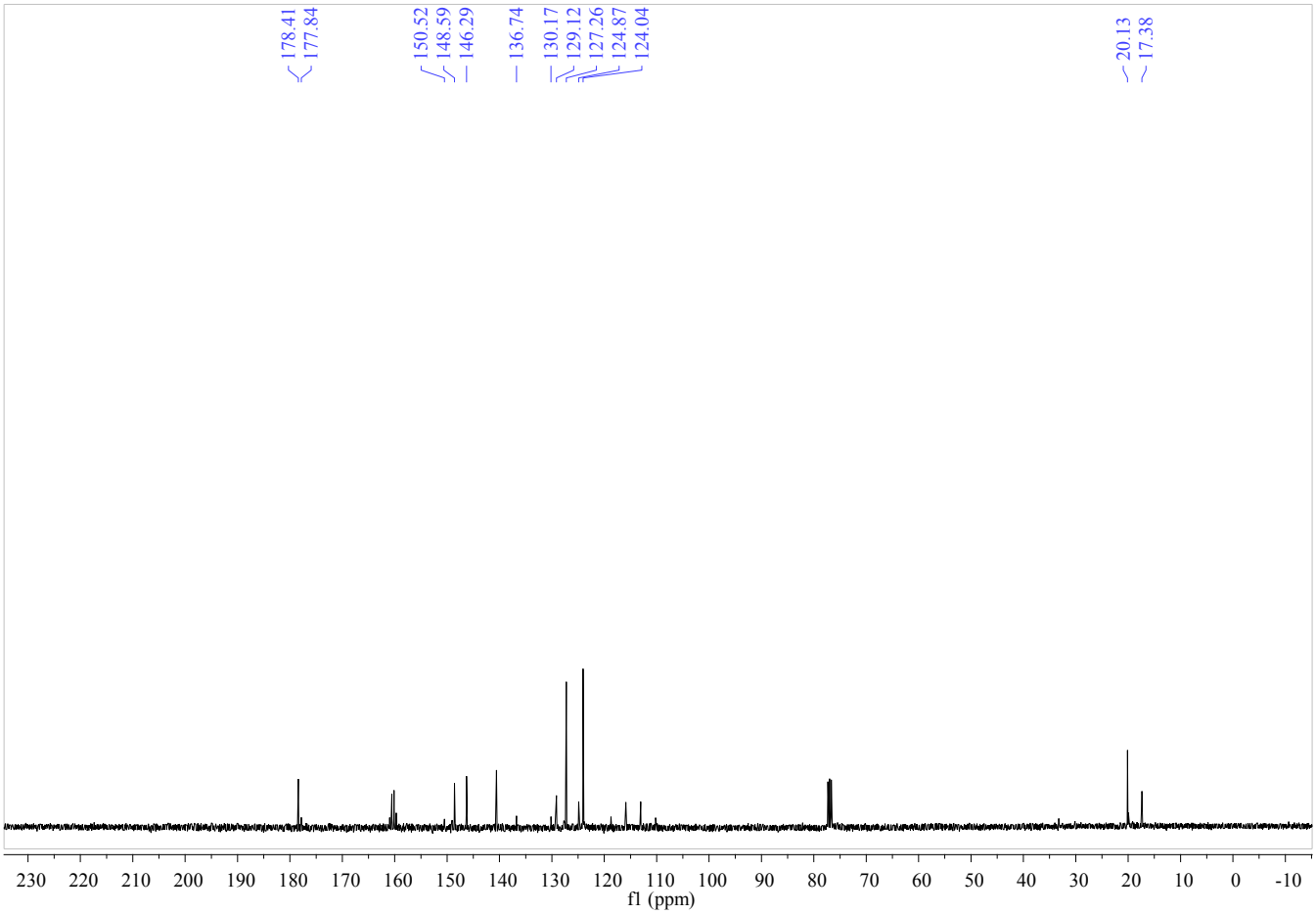
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) spectrum of hydrazone 1i



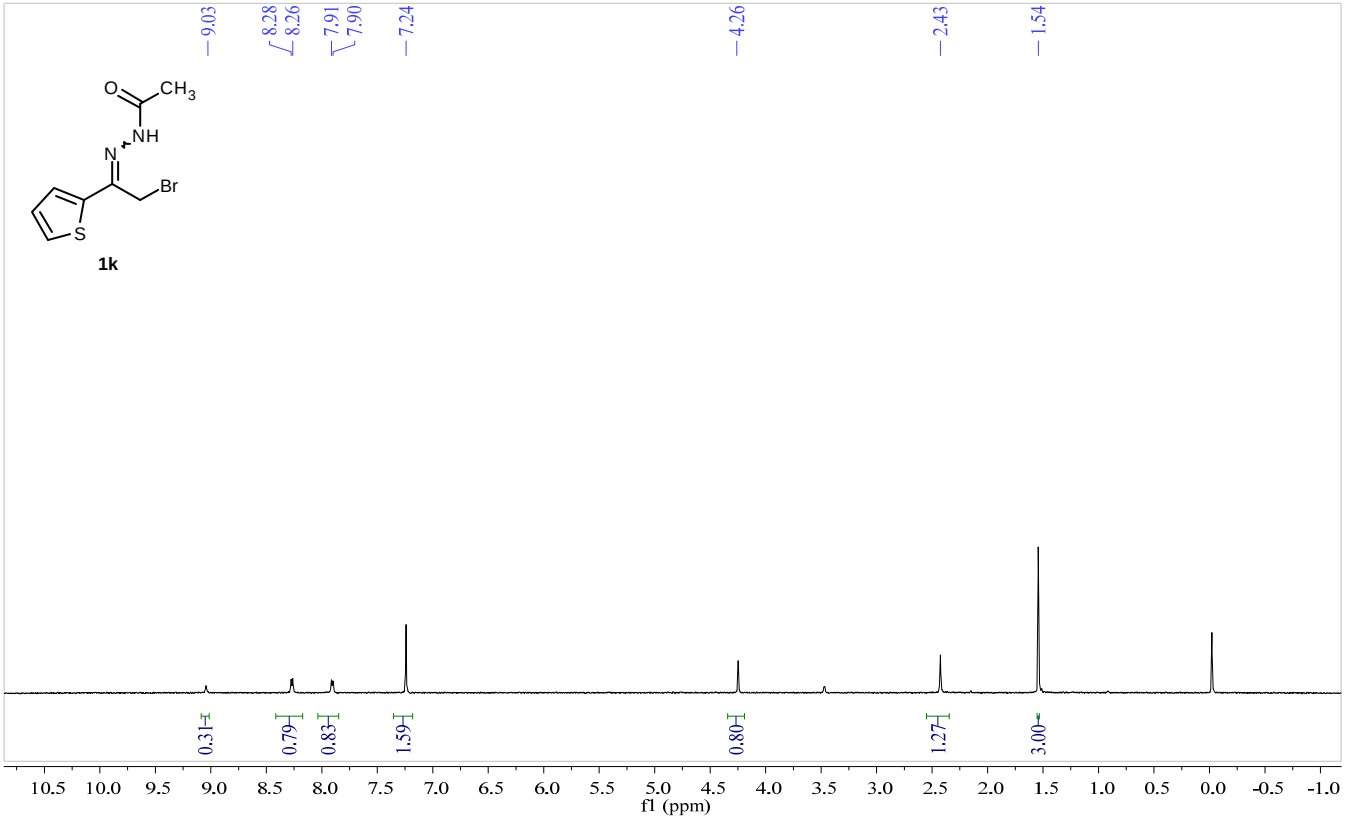


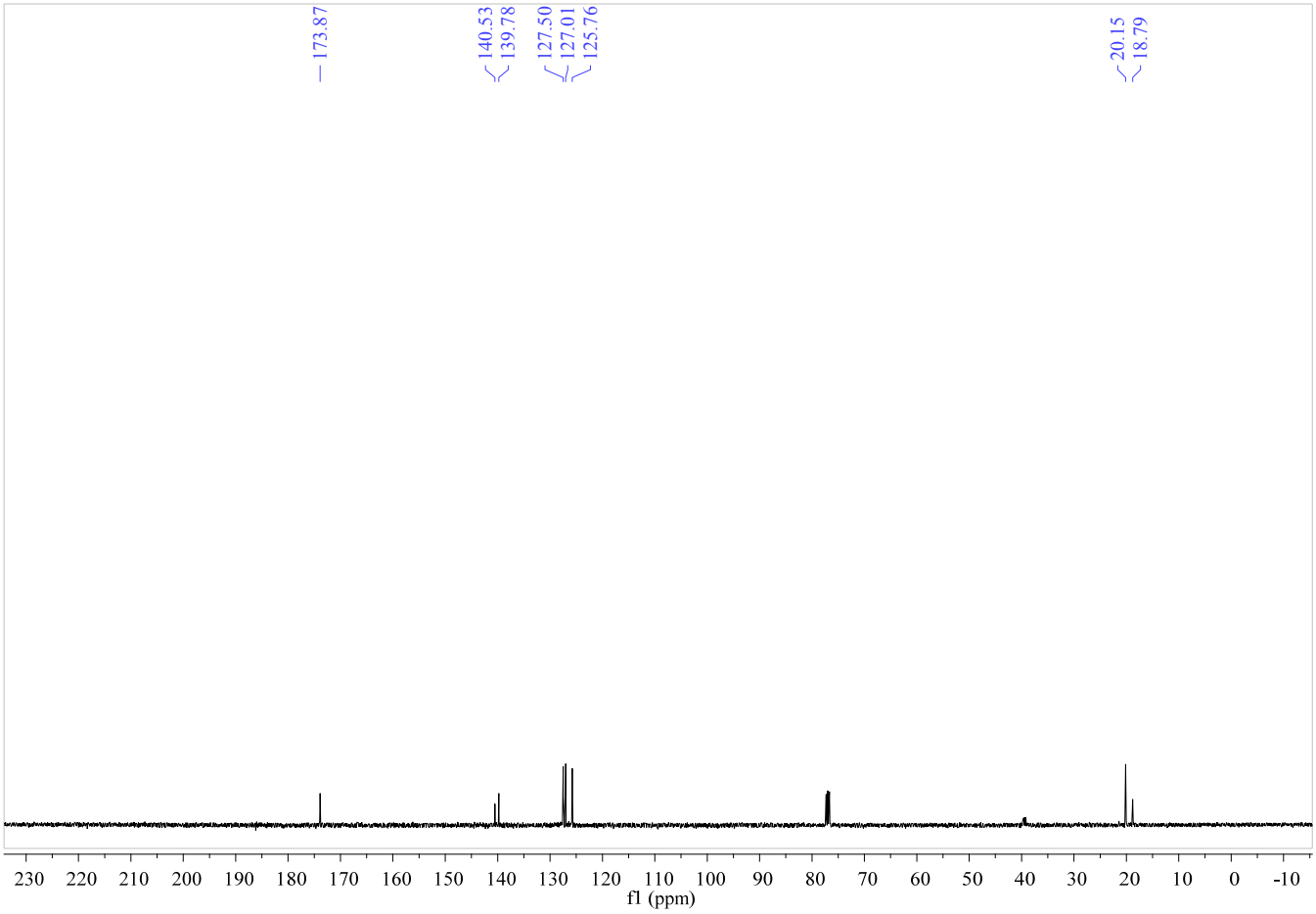
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$/TFA) spectrum of hydrazone 1j



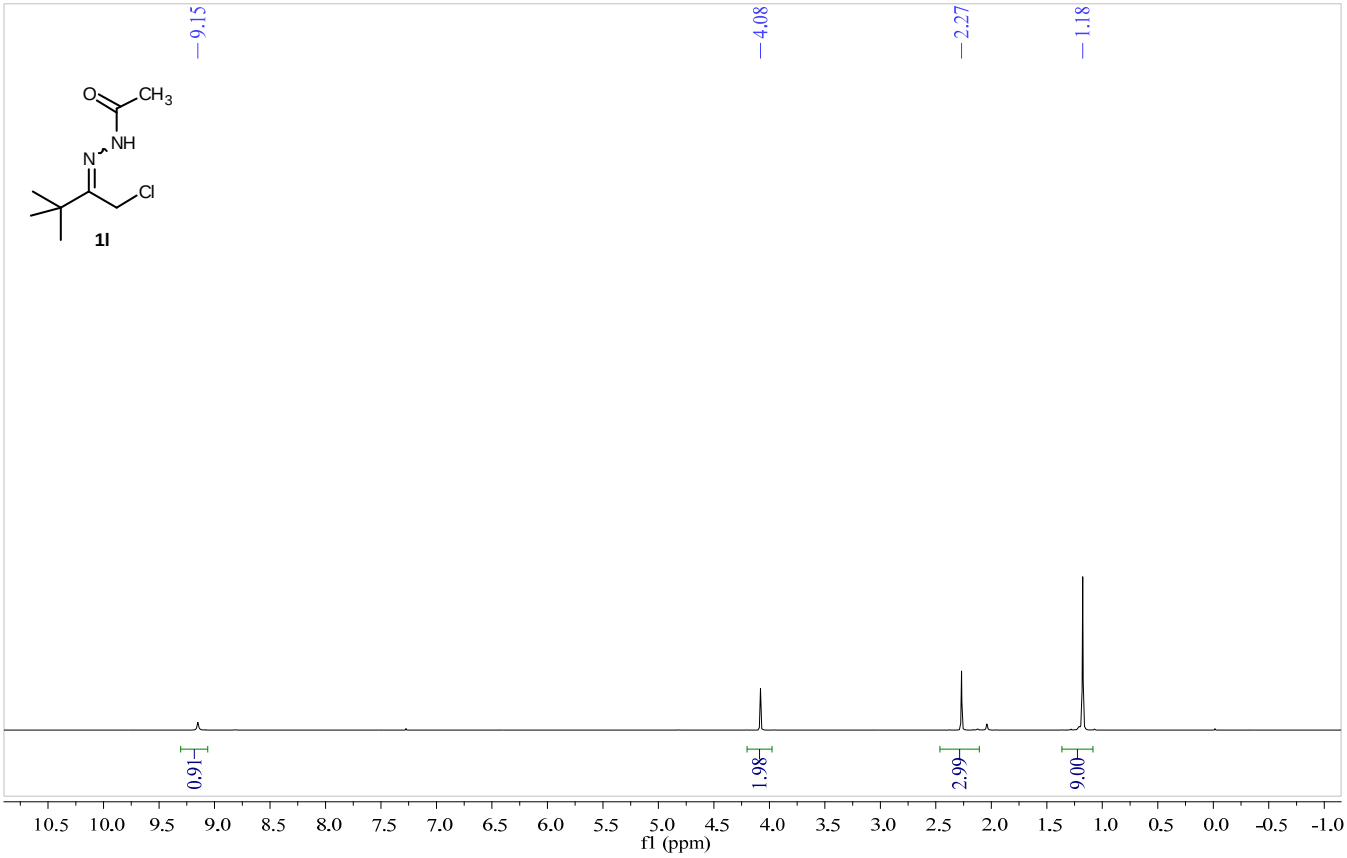


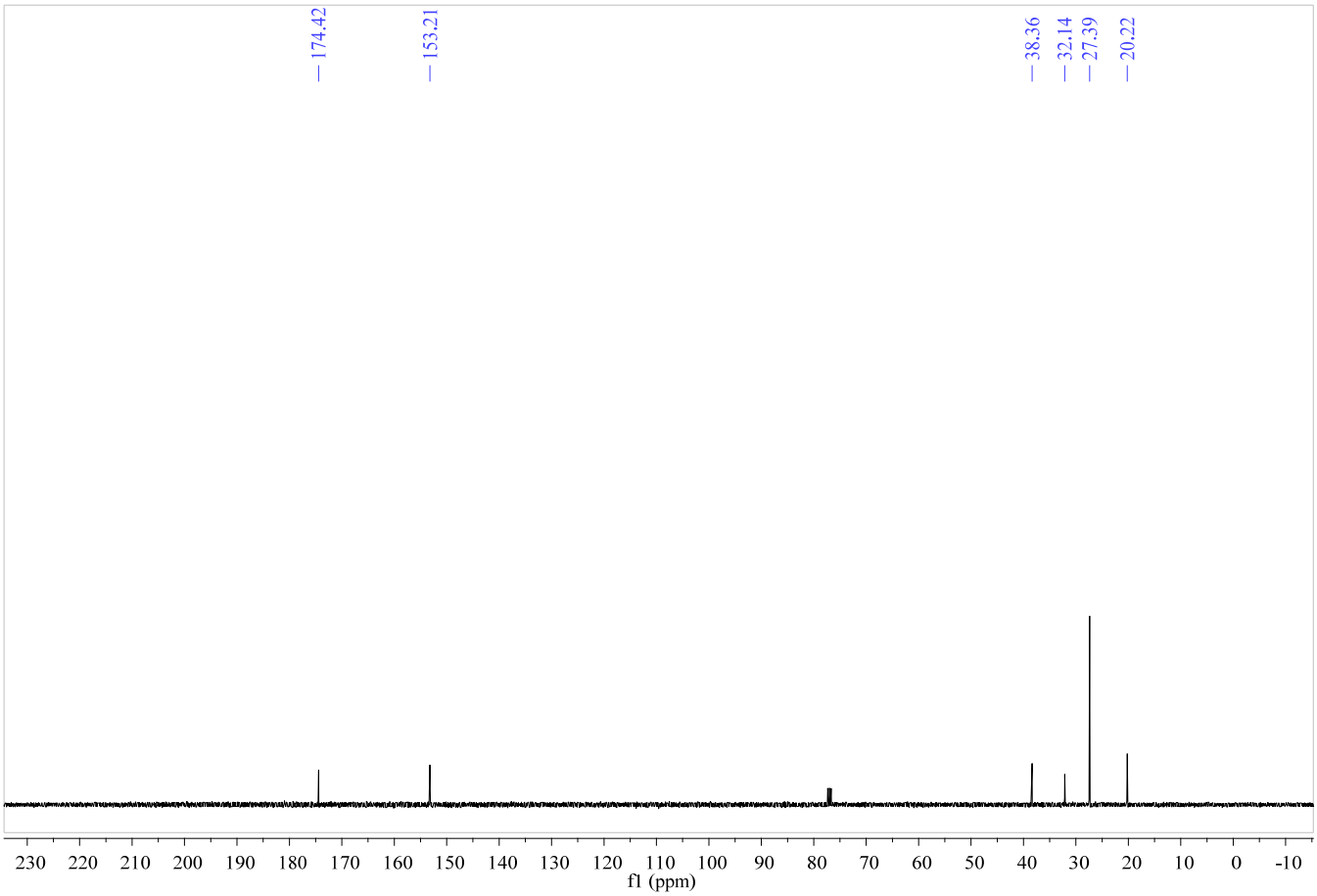
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) spectrum of hydrazone 1k



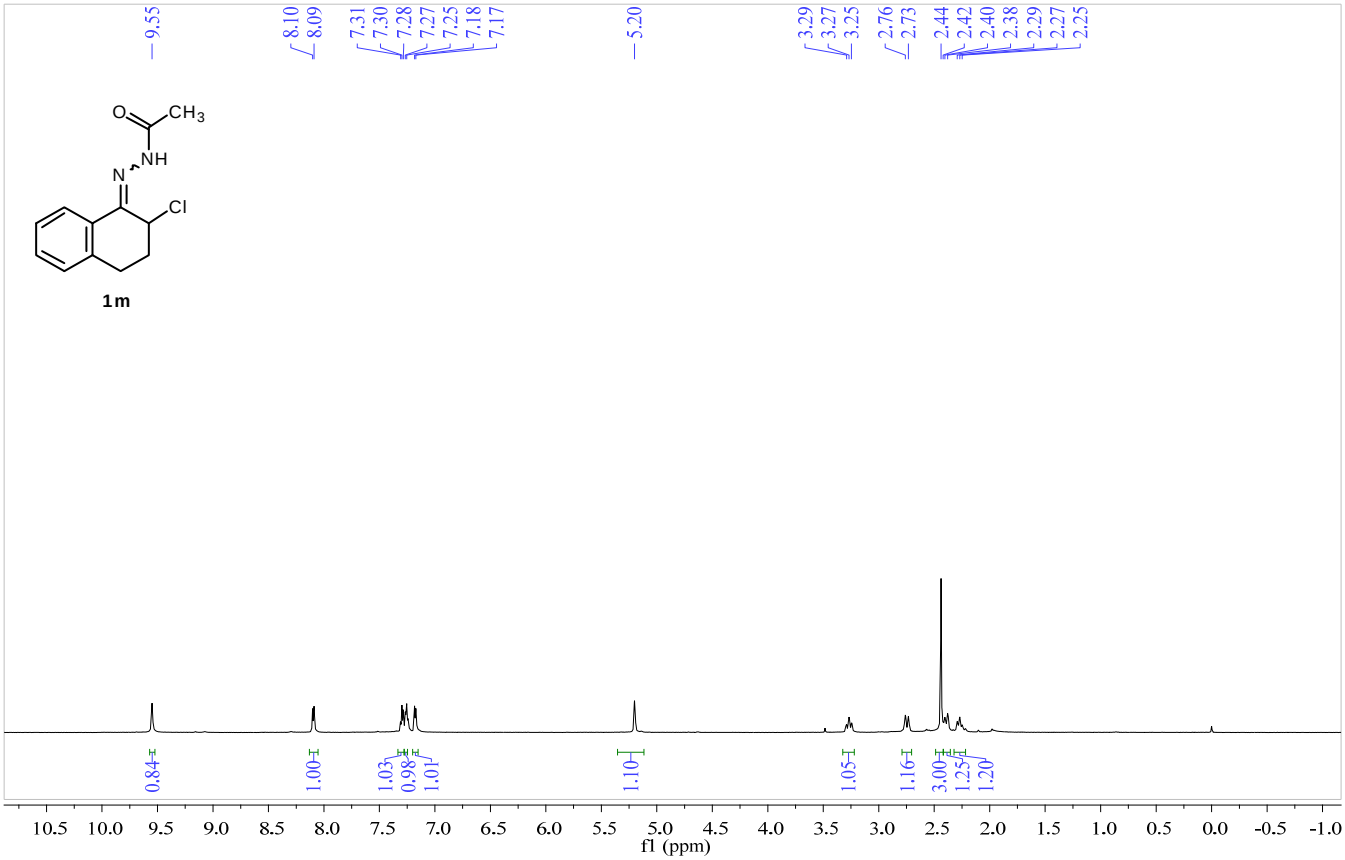


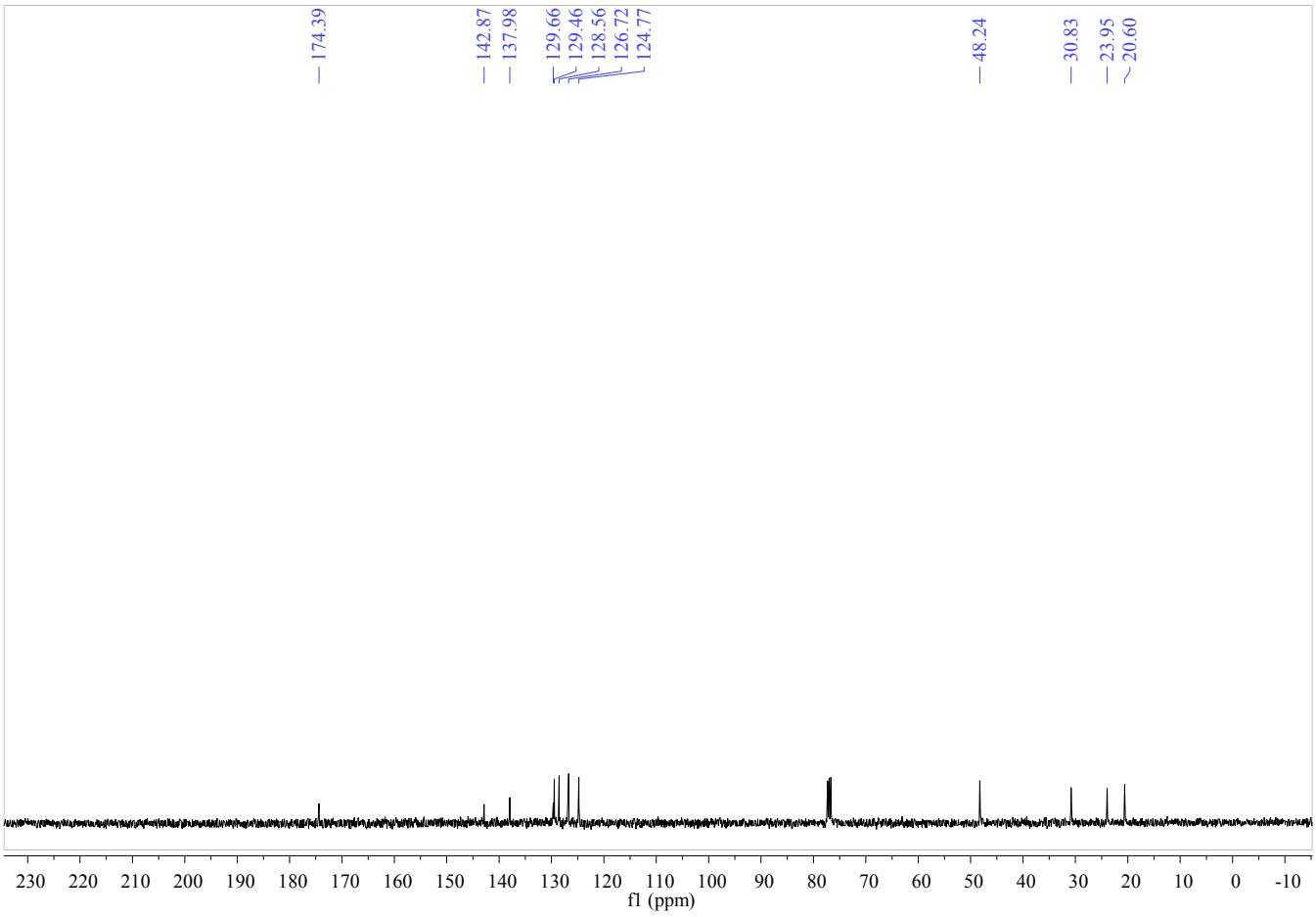
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of hydrazone 1l



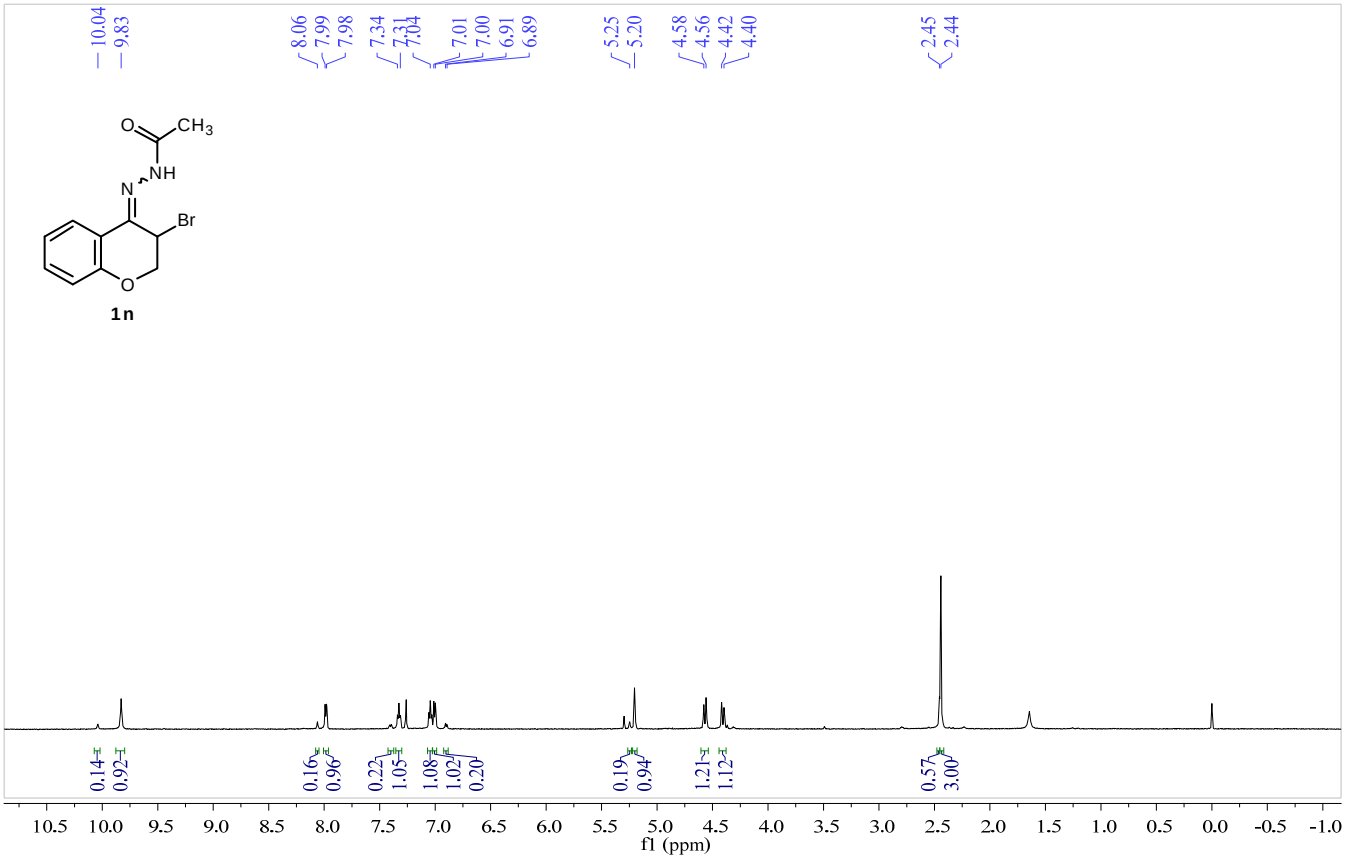


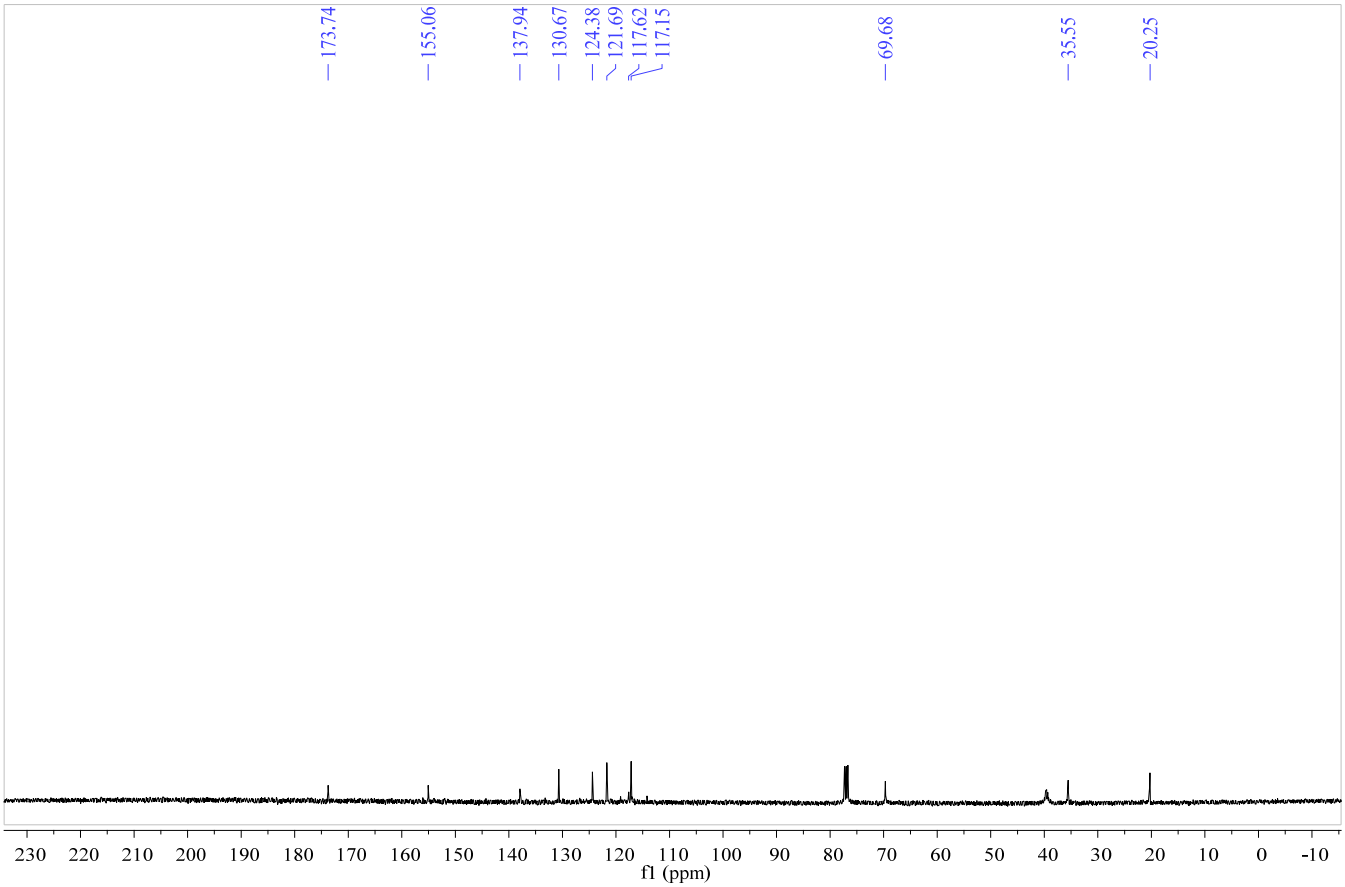
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of hydrazone 1m



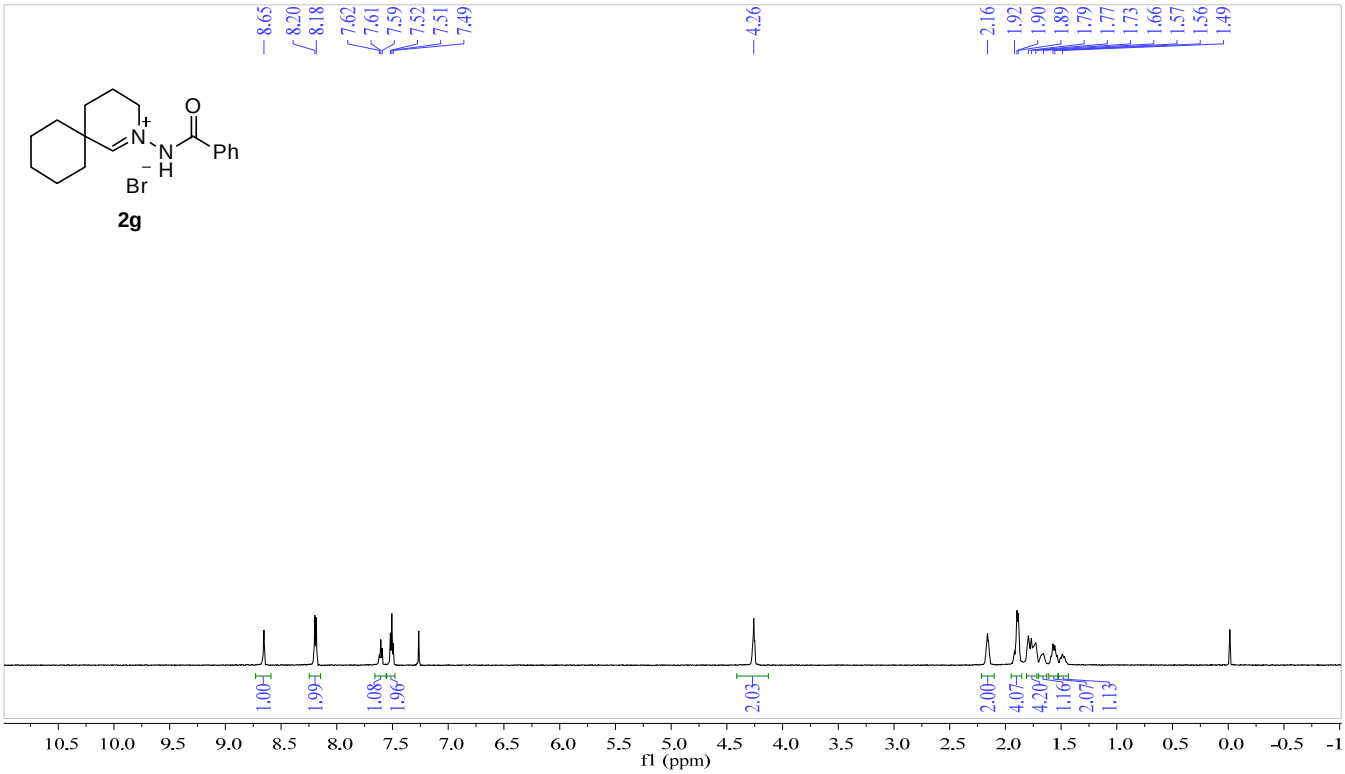


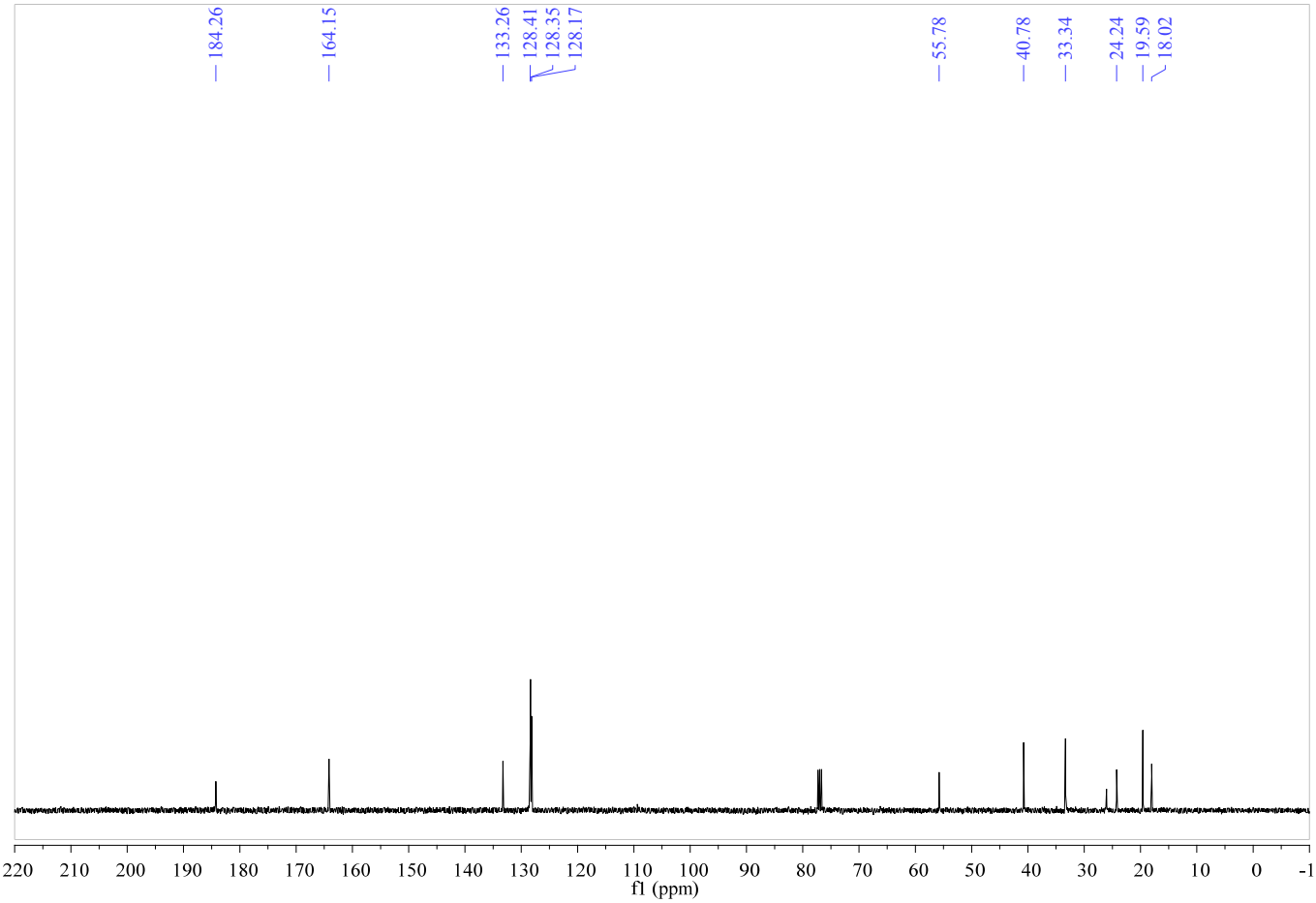
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) spectrum of hydrazone 1n



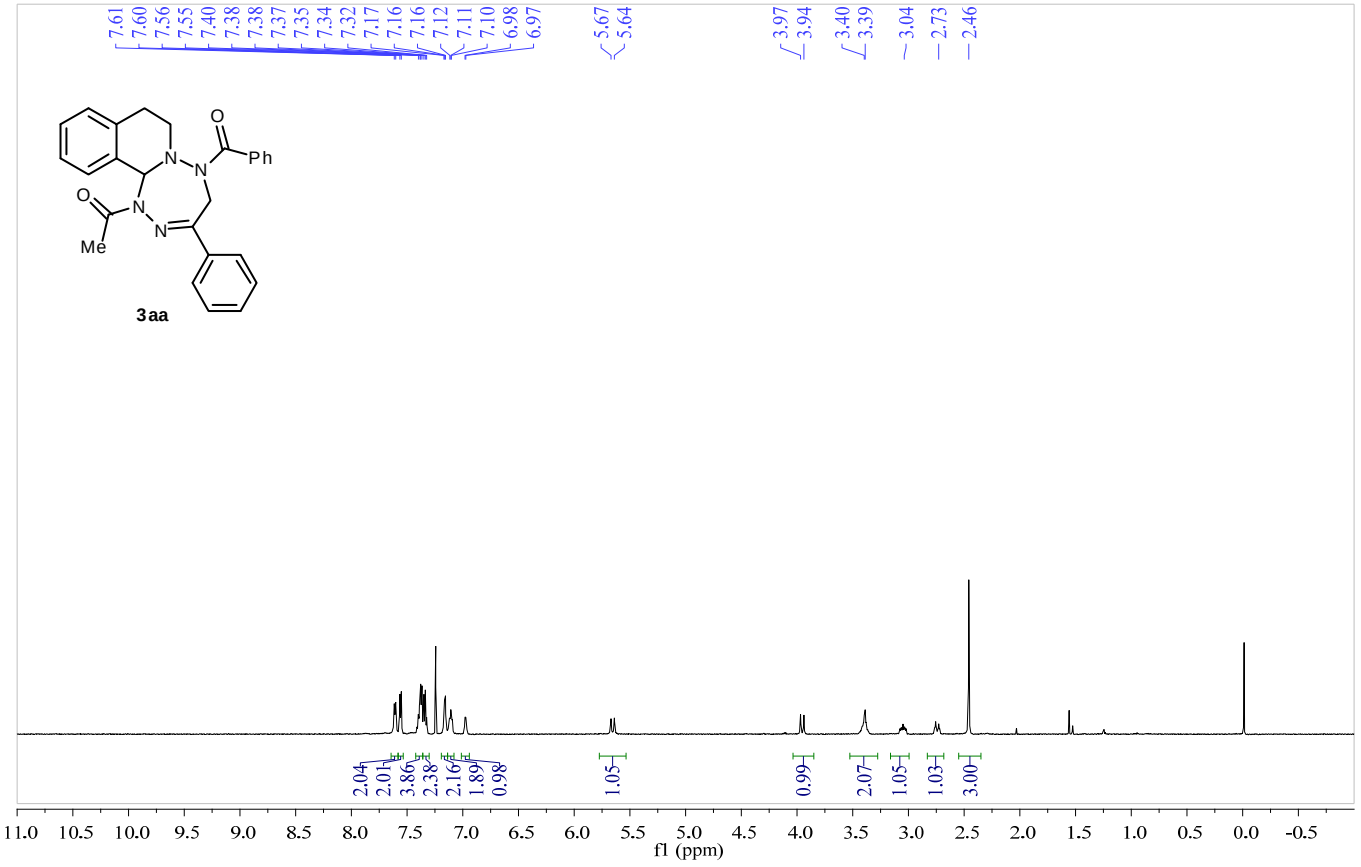


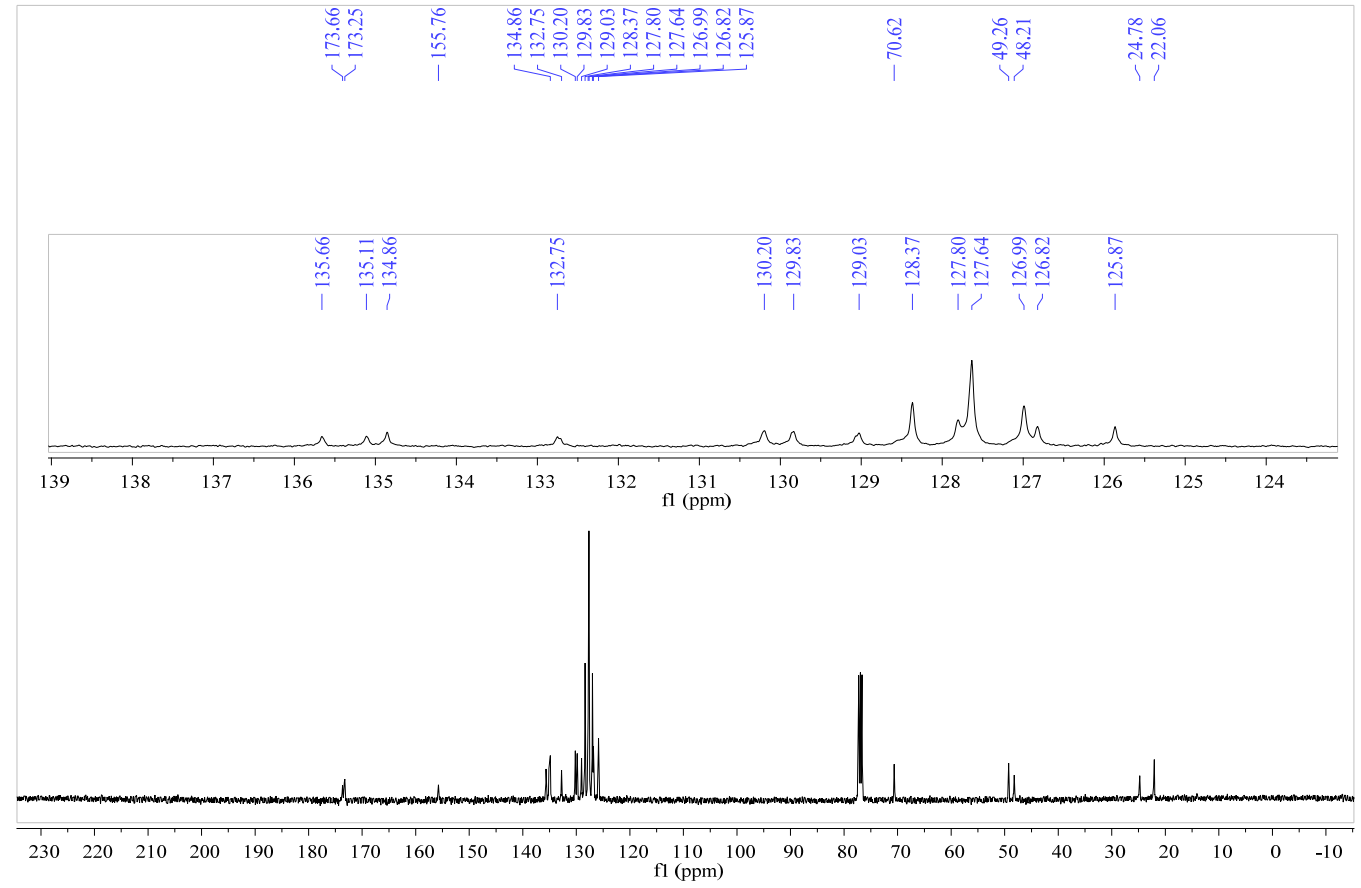
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of C,N-cyclic azomethine imine 2g



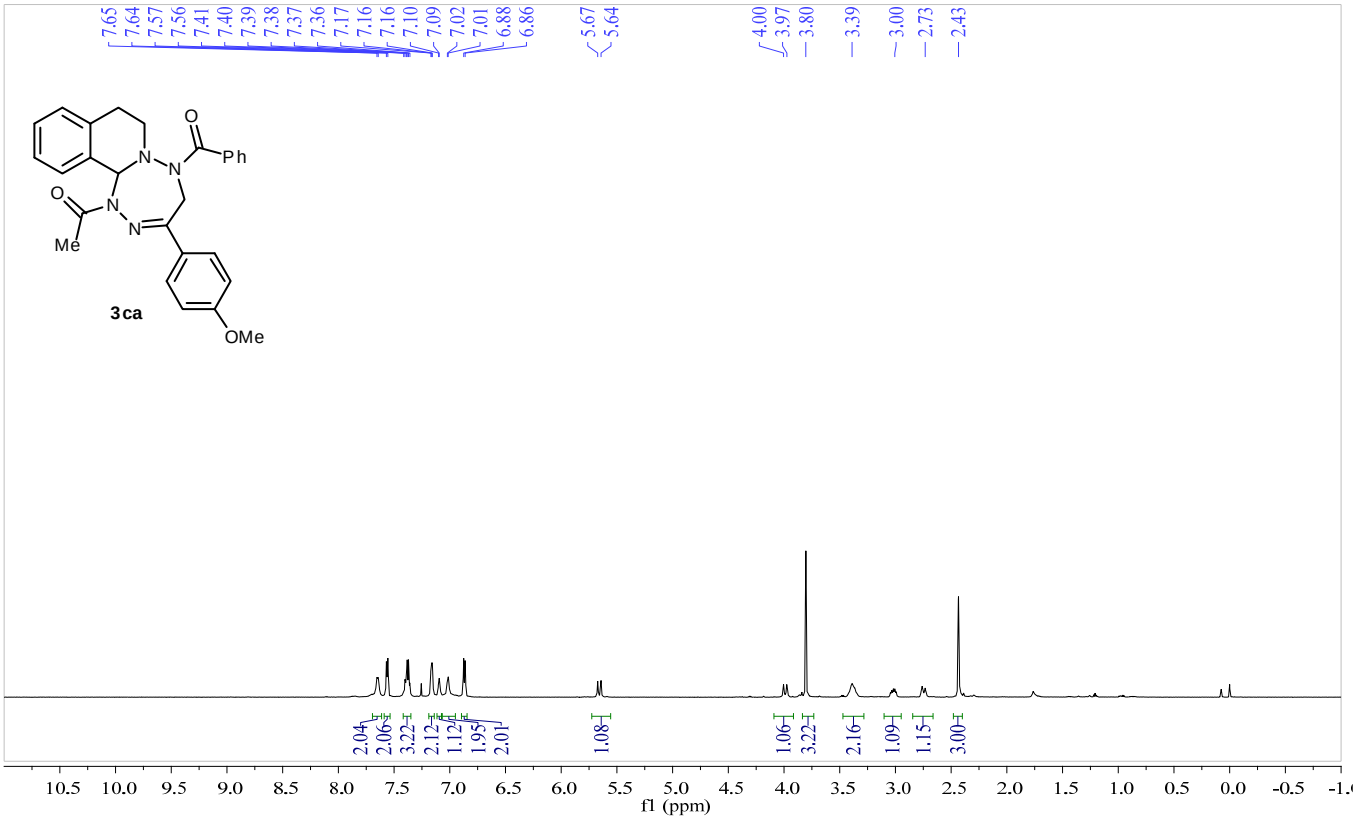


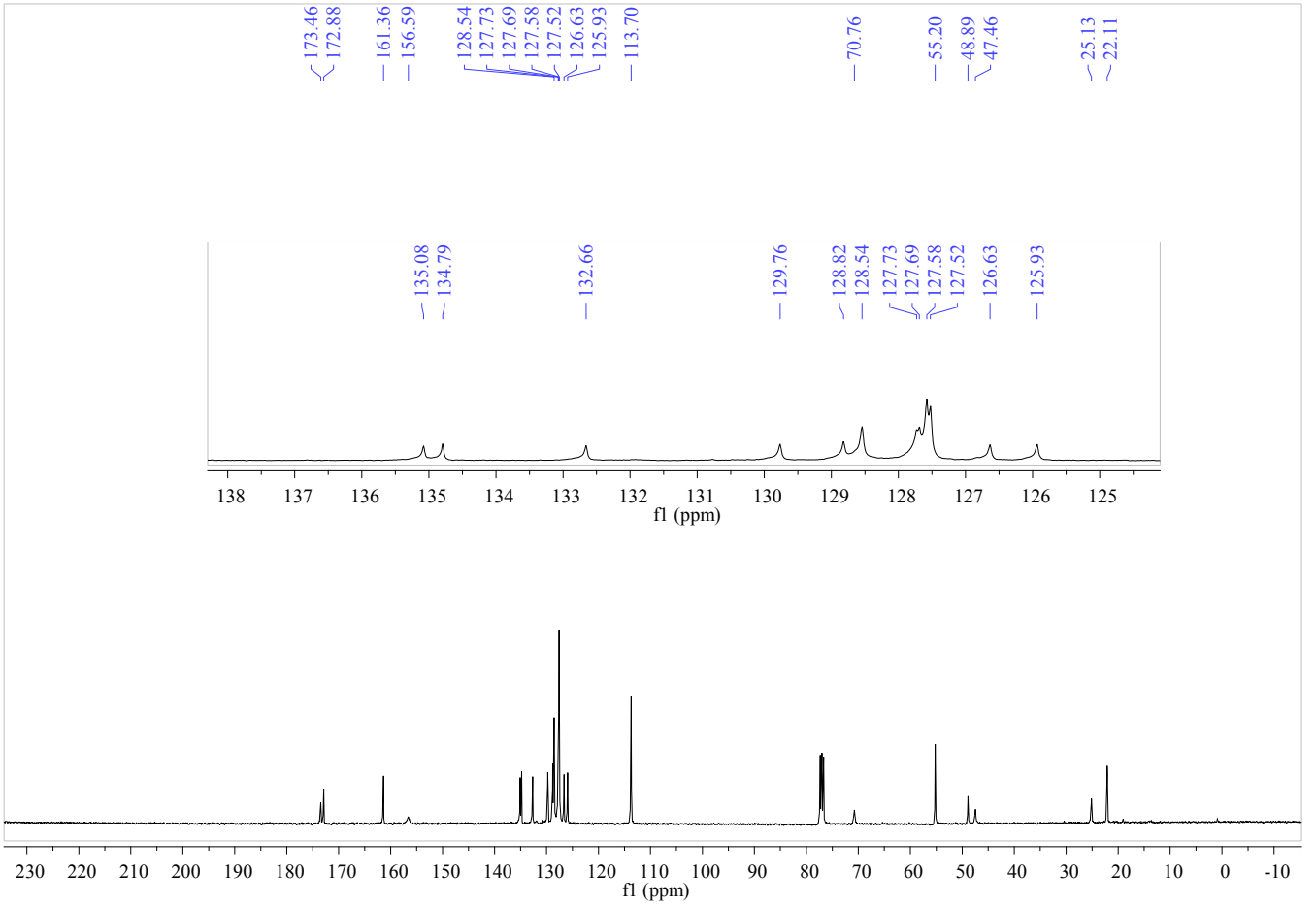
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3aa



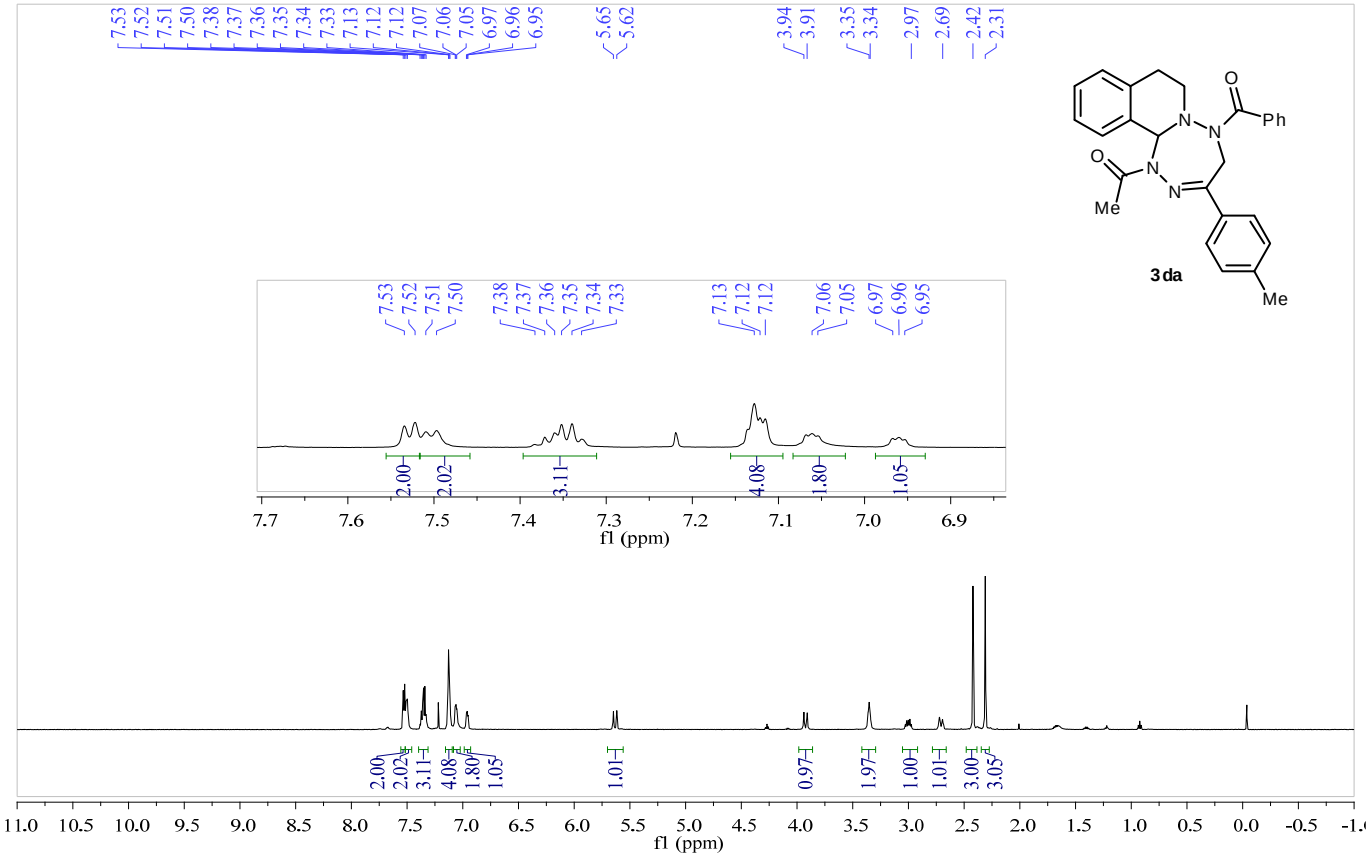


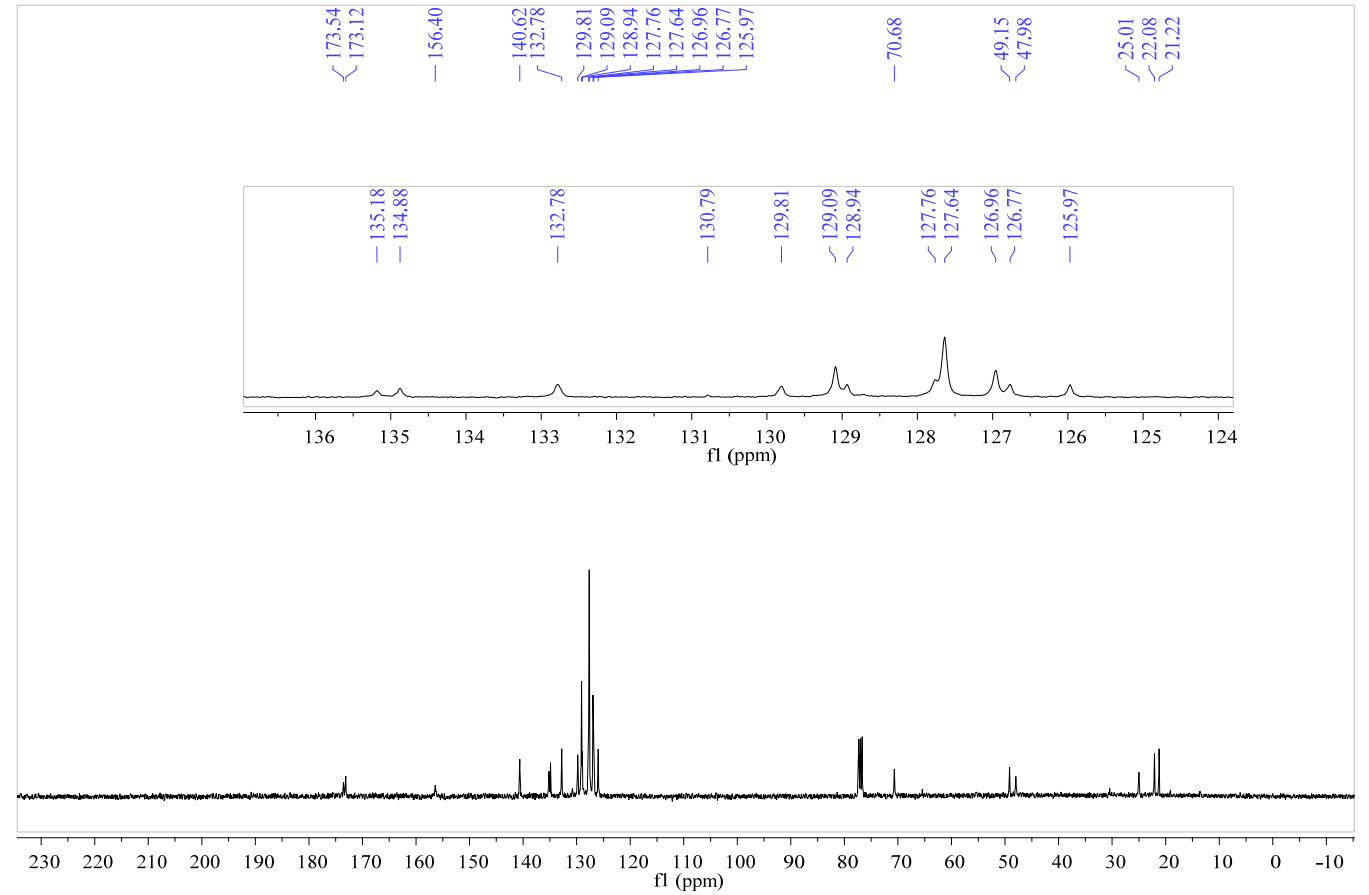
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3ca



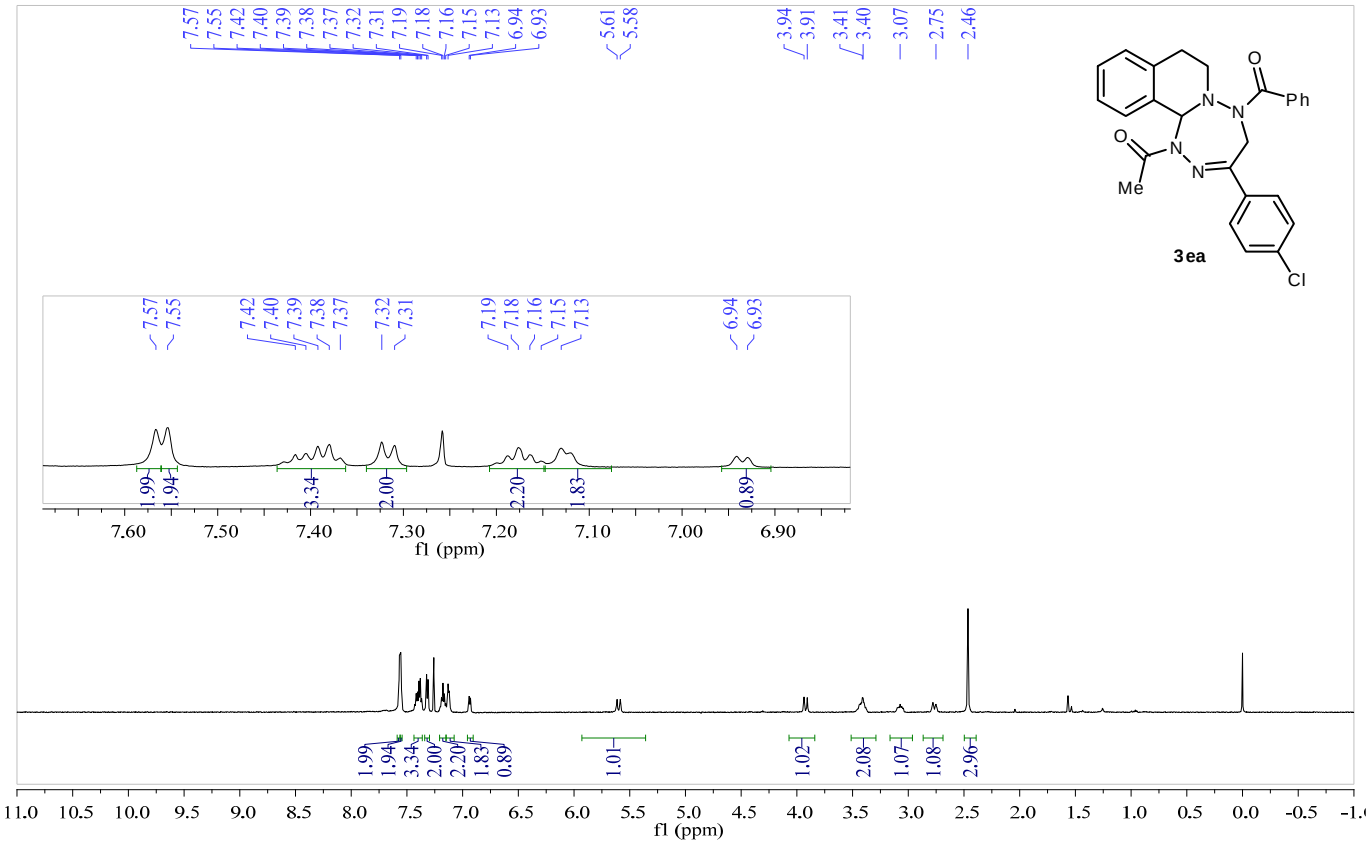


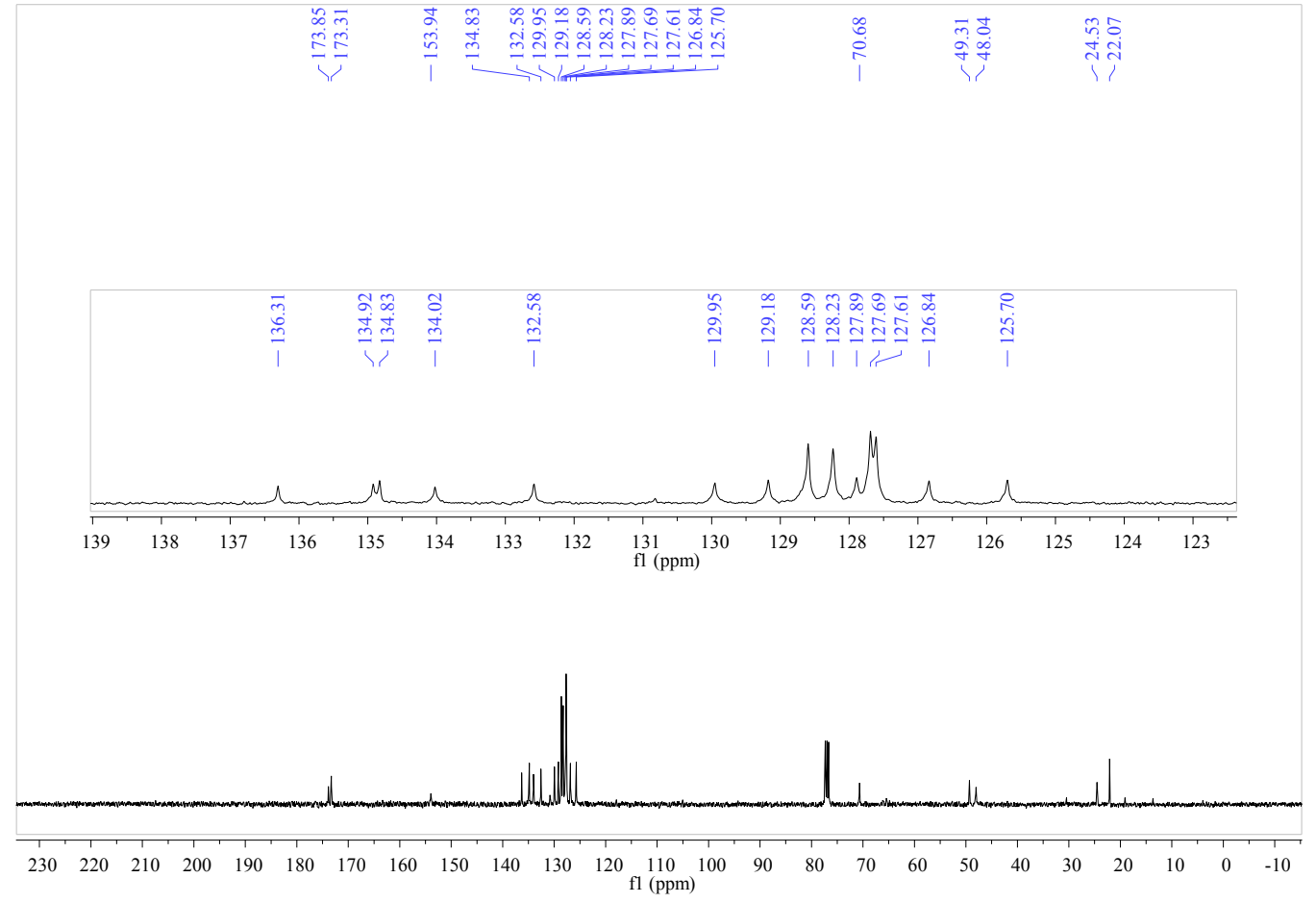
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3da



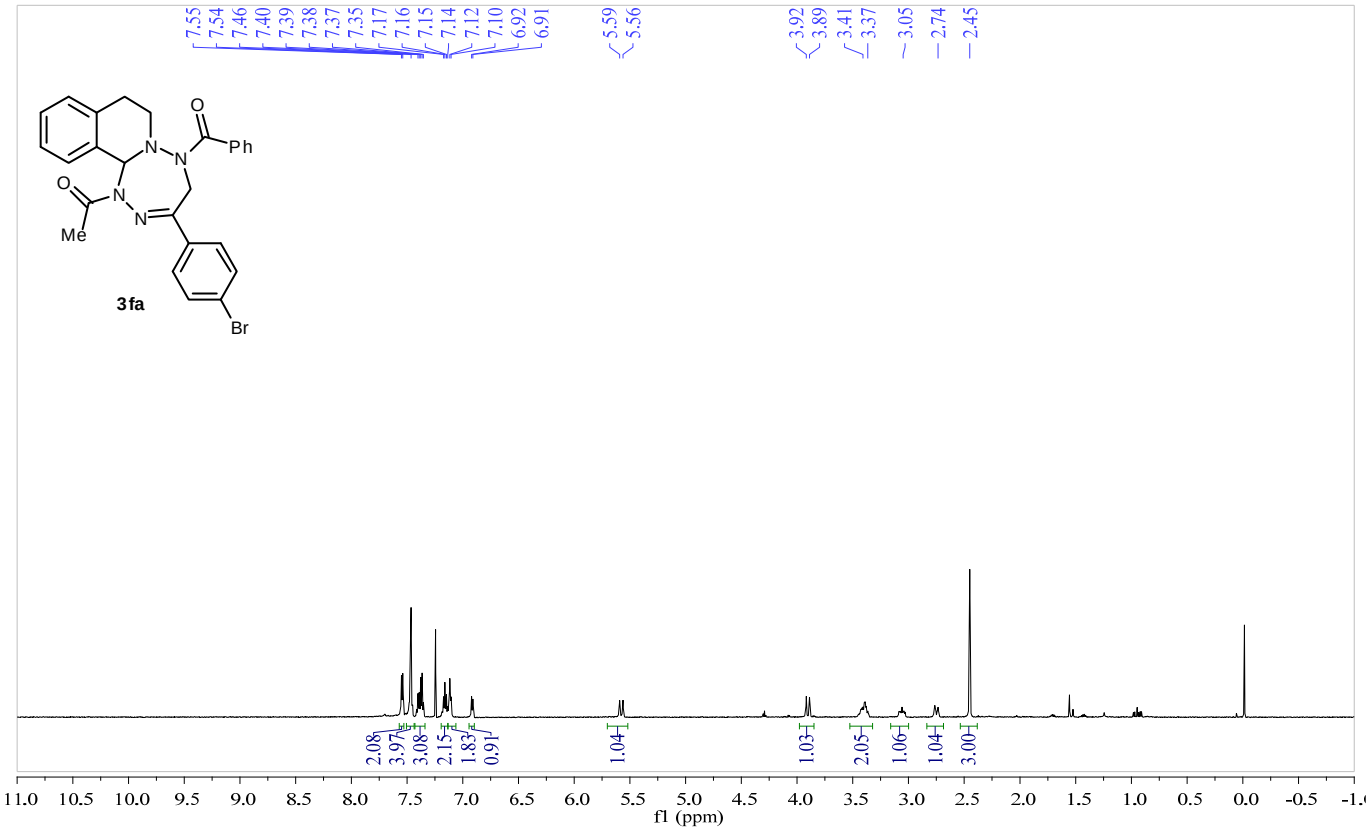


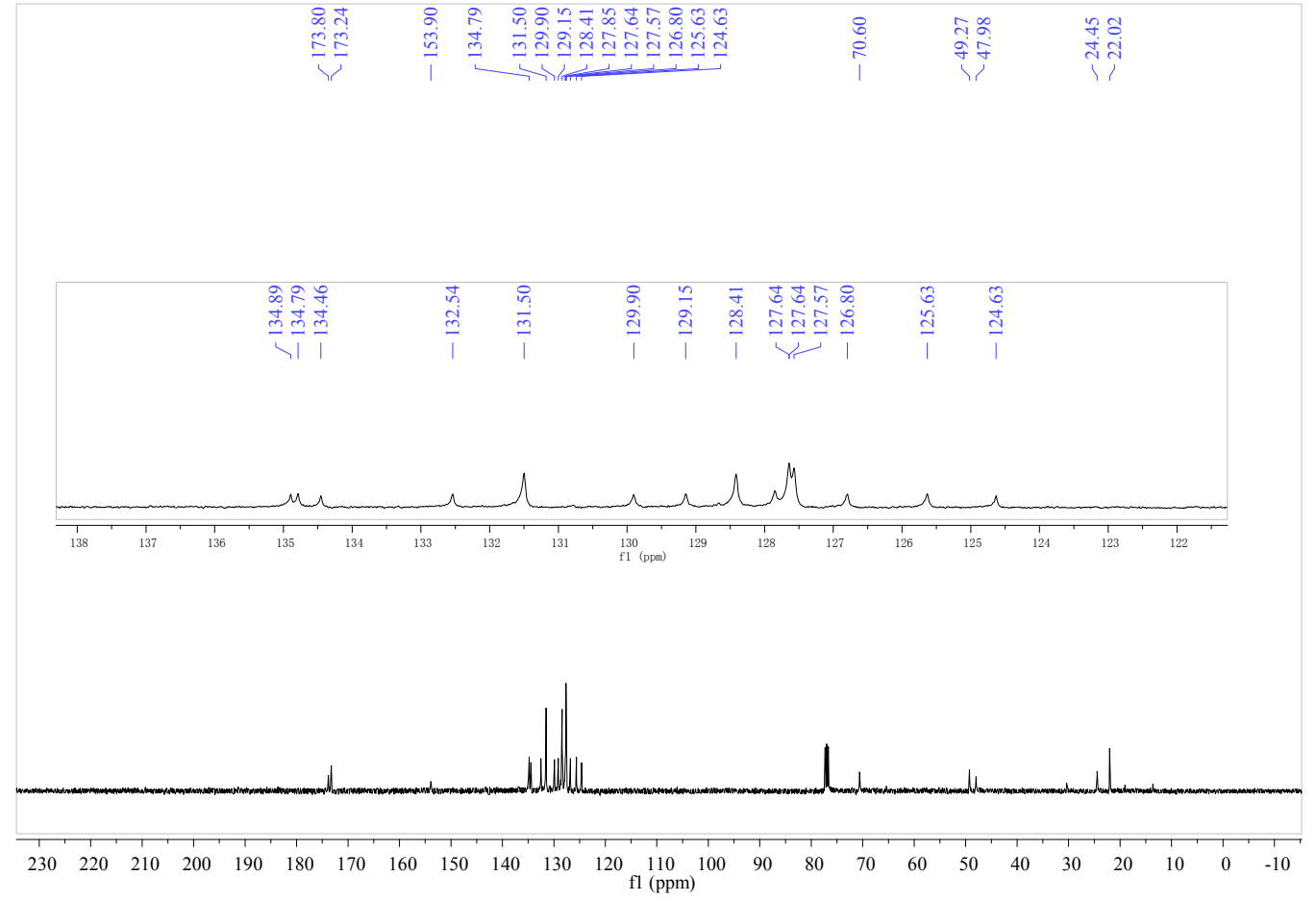
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3ea



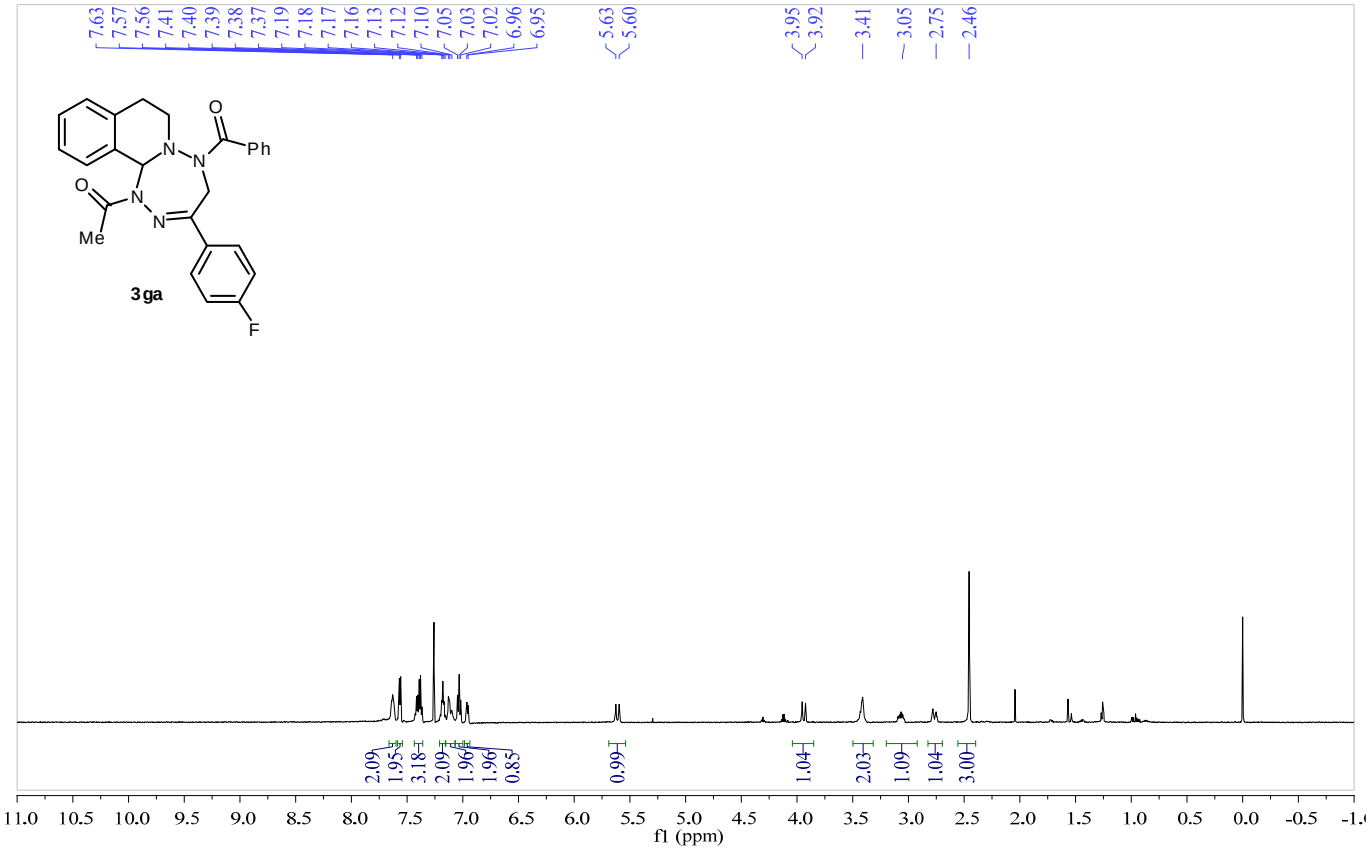


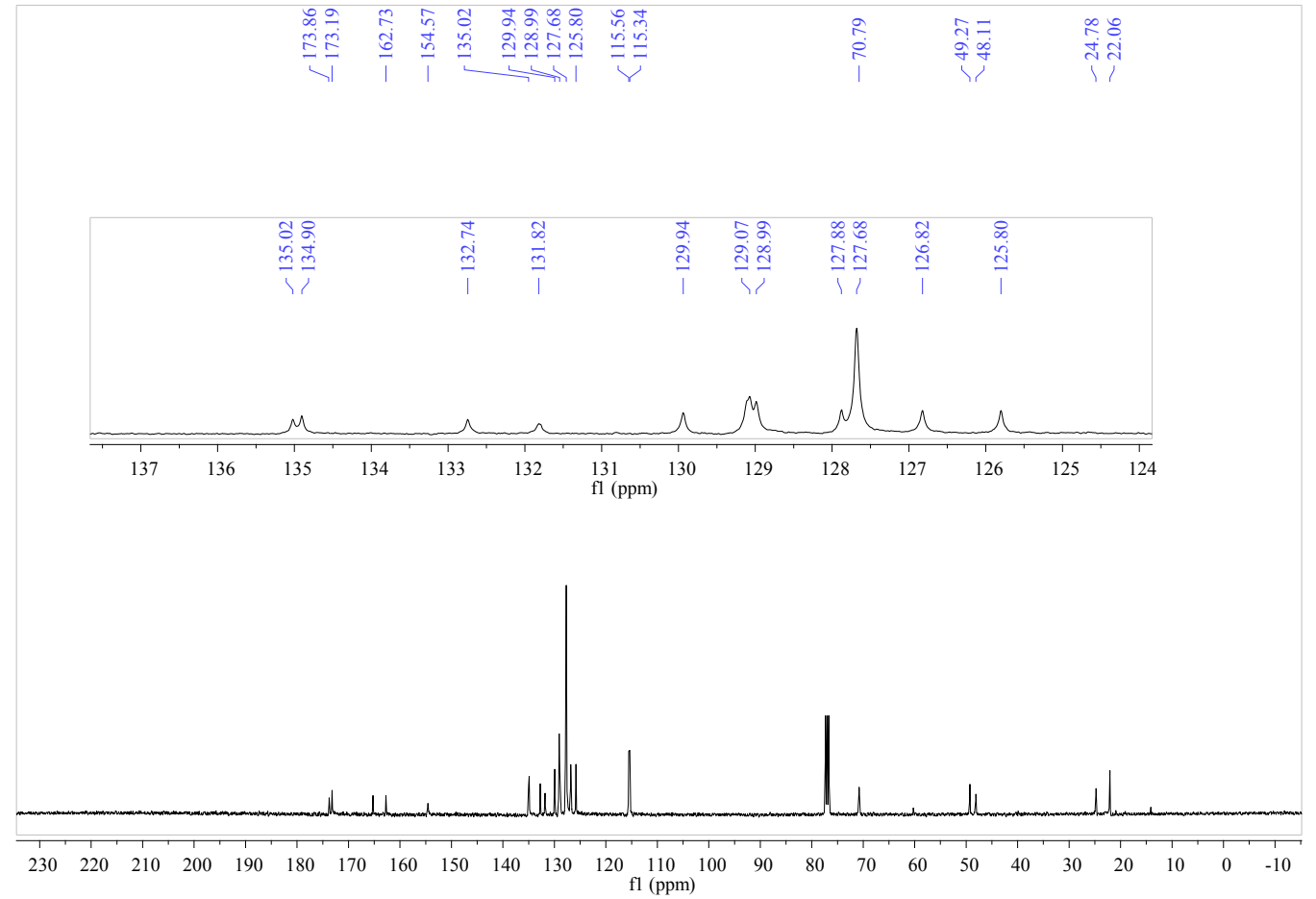
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3fa



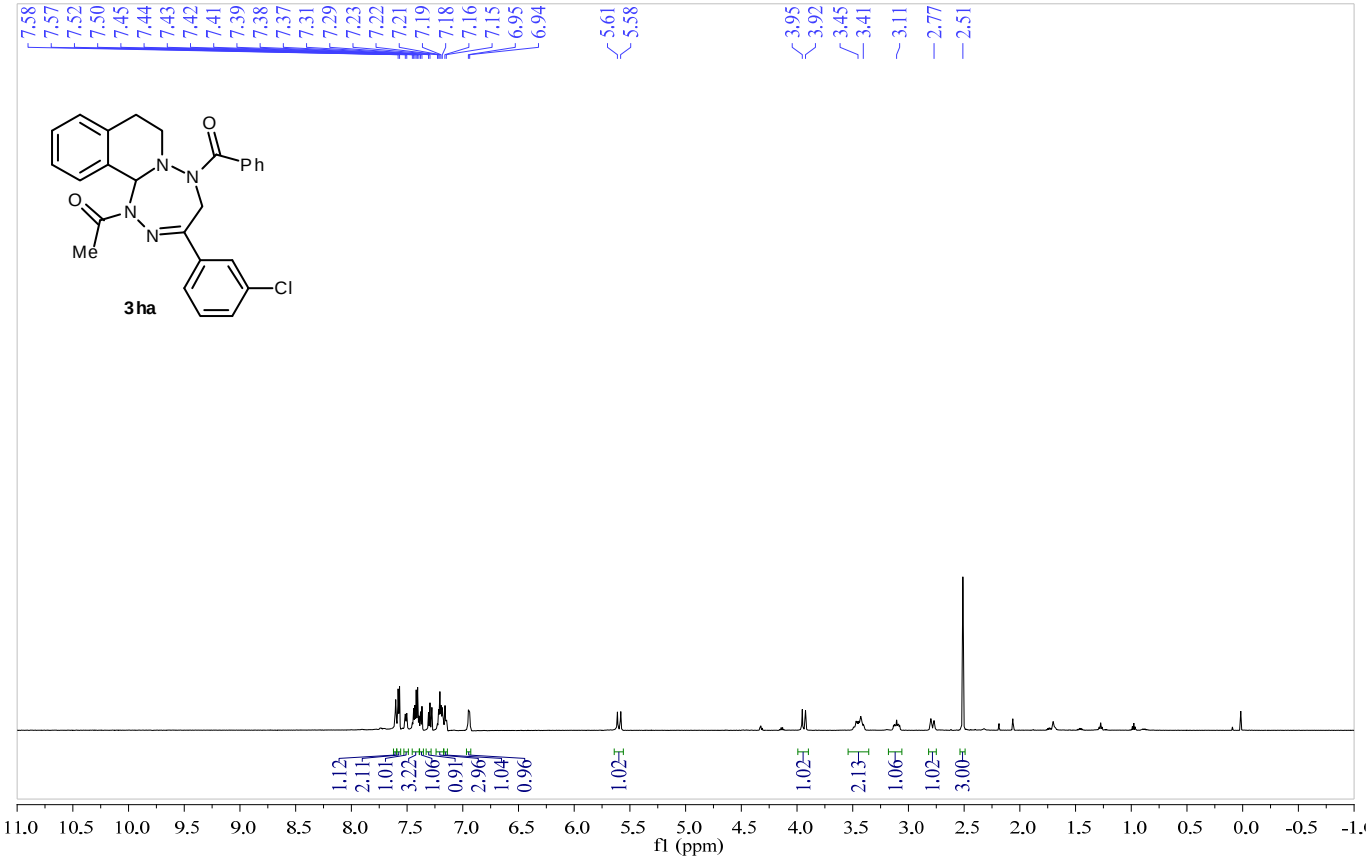


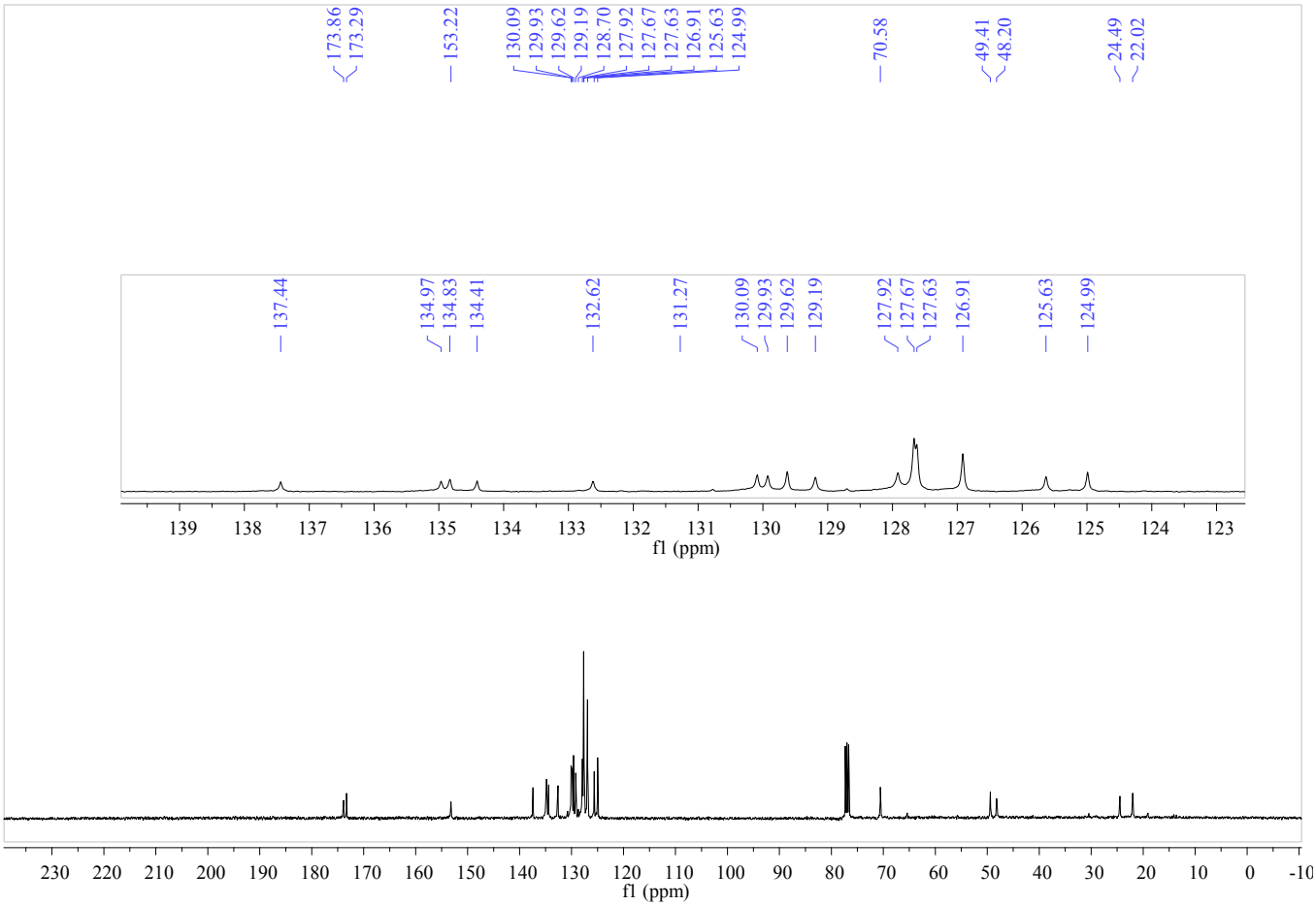
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3ga



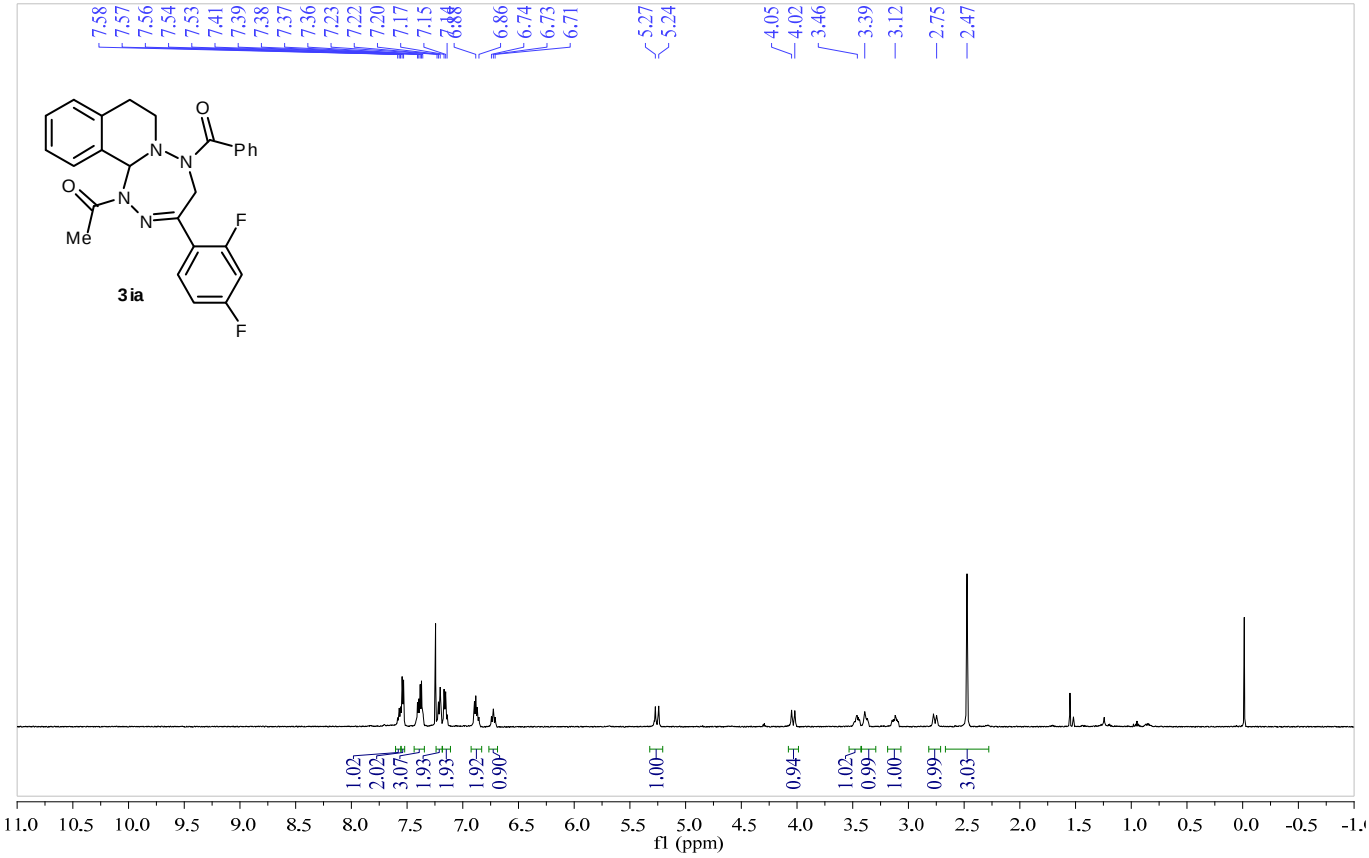


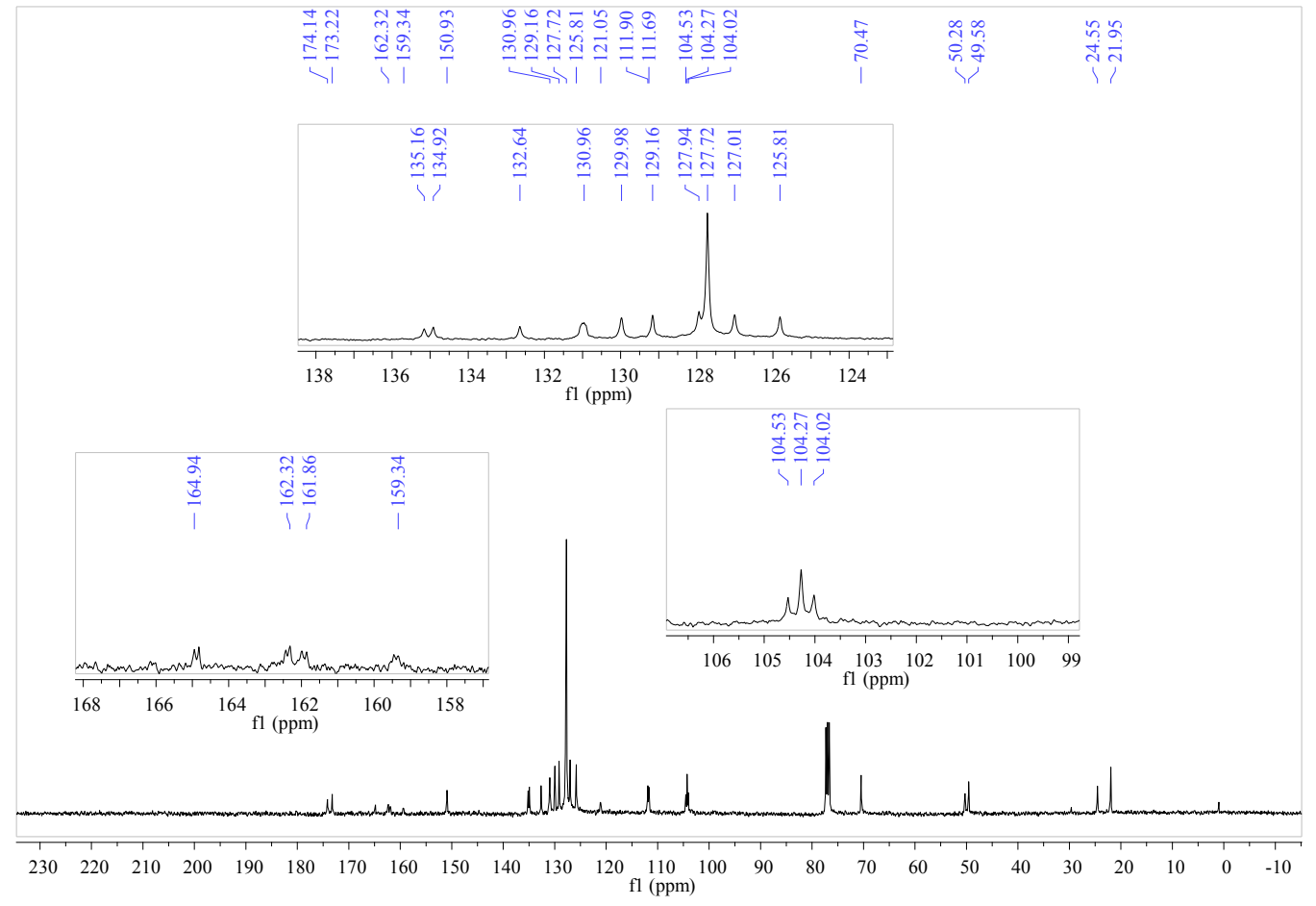
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3ha



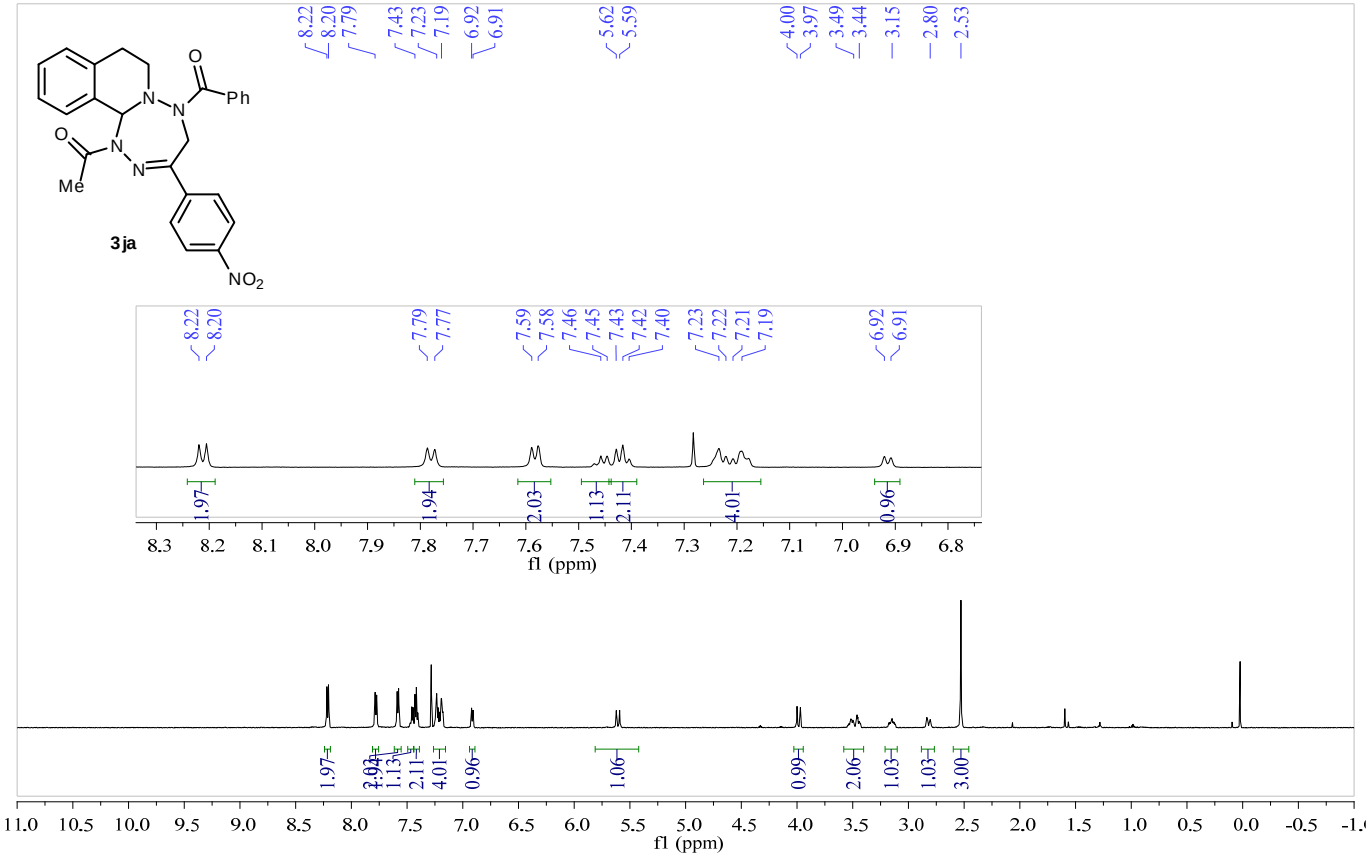


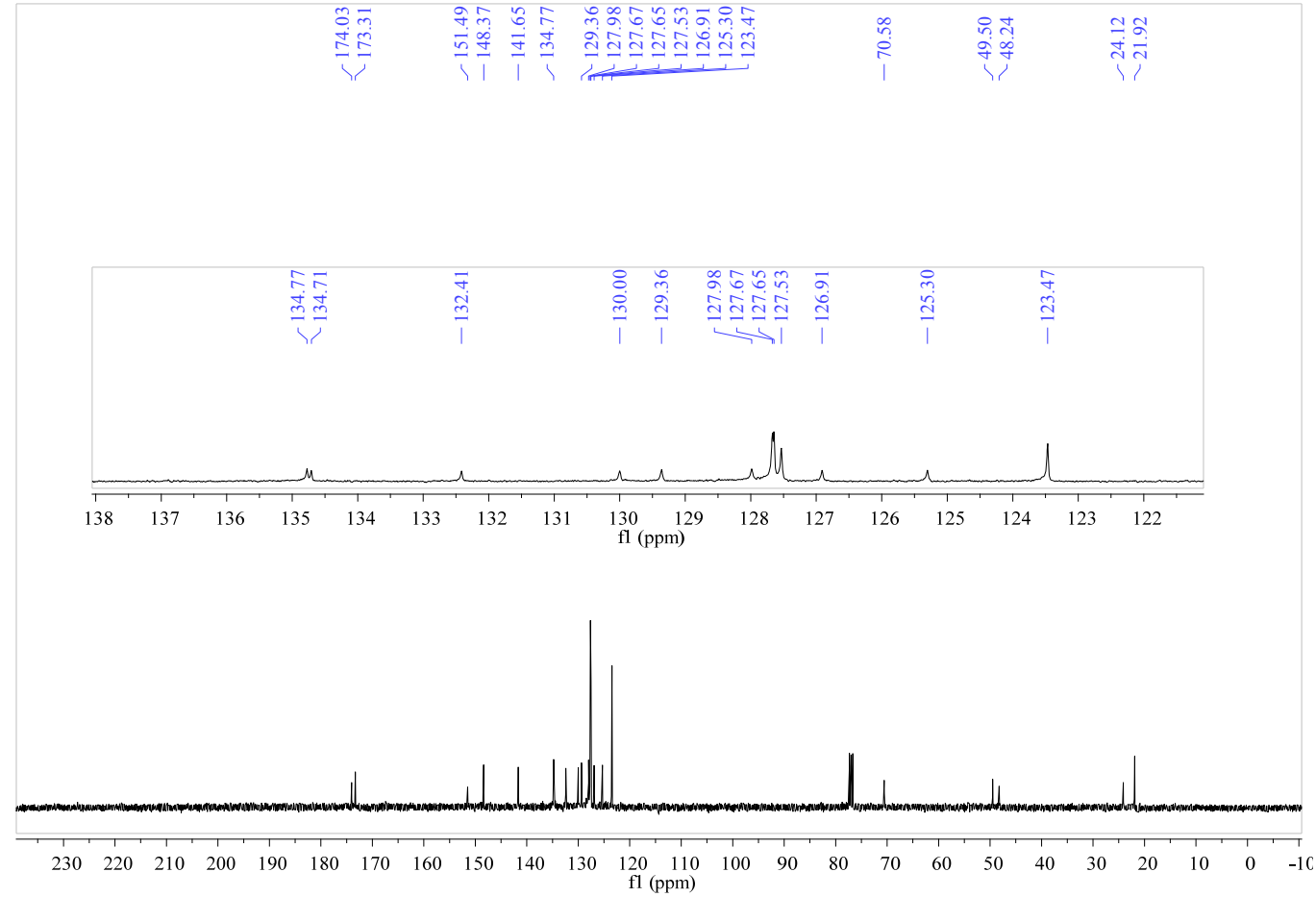
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3ia



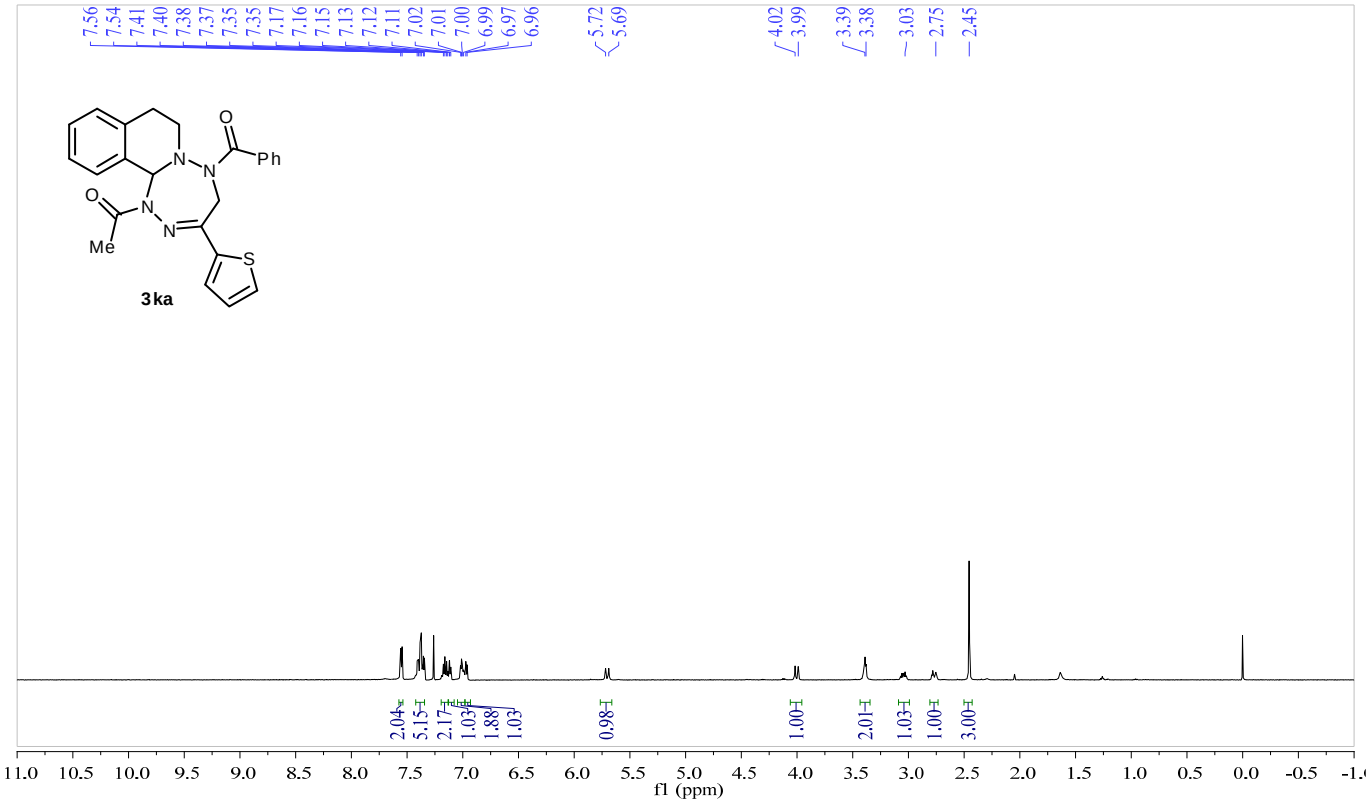


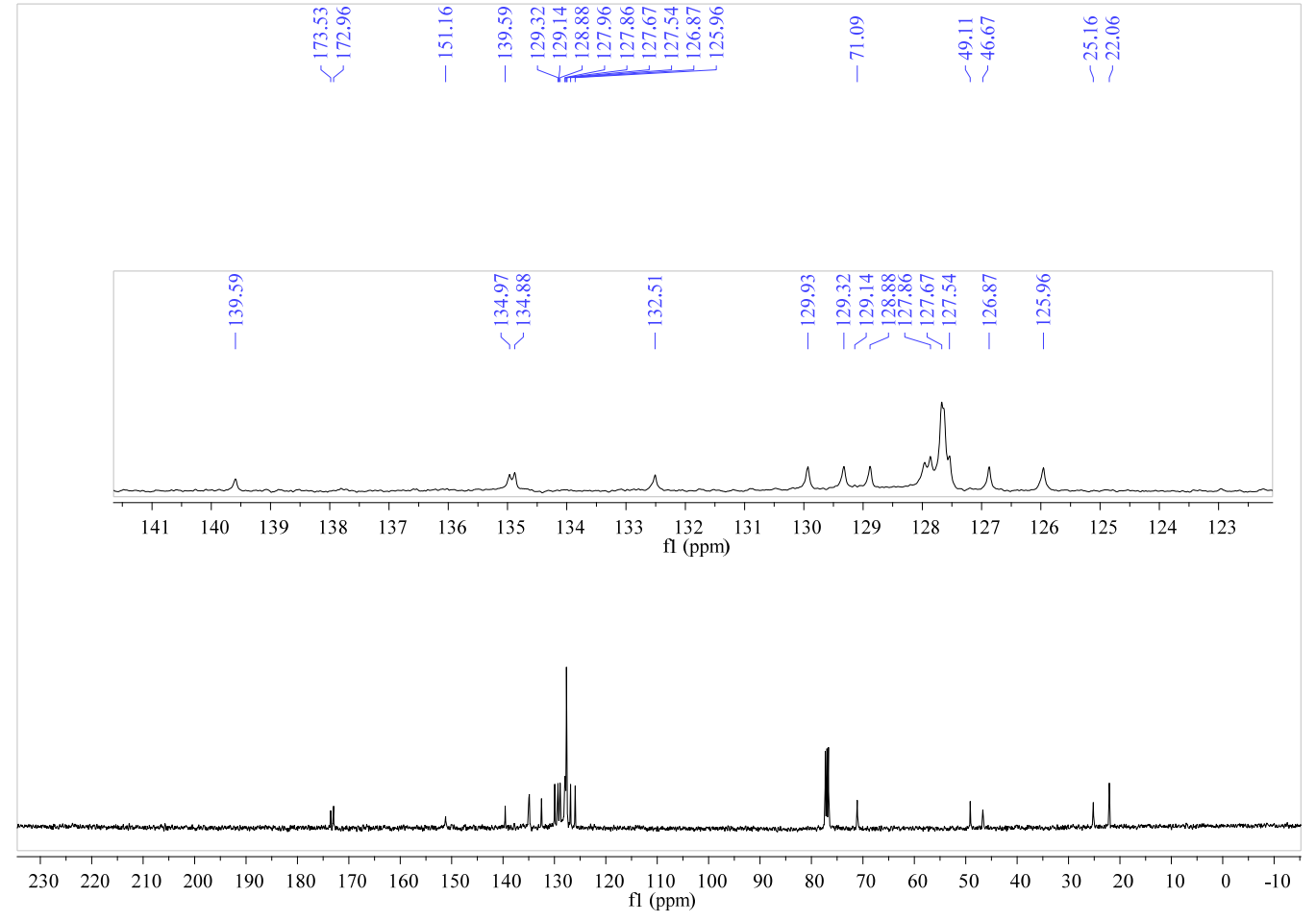
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3ja



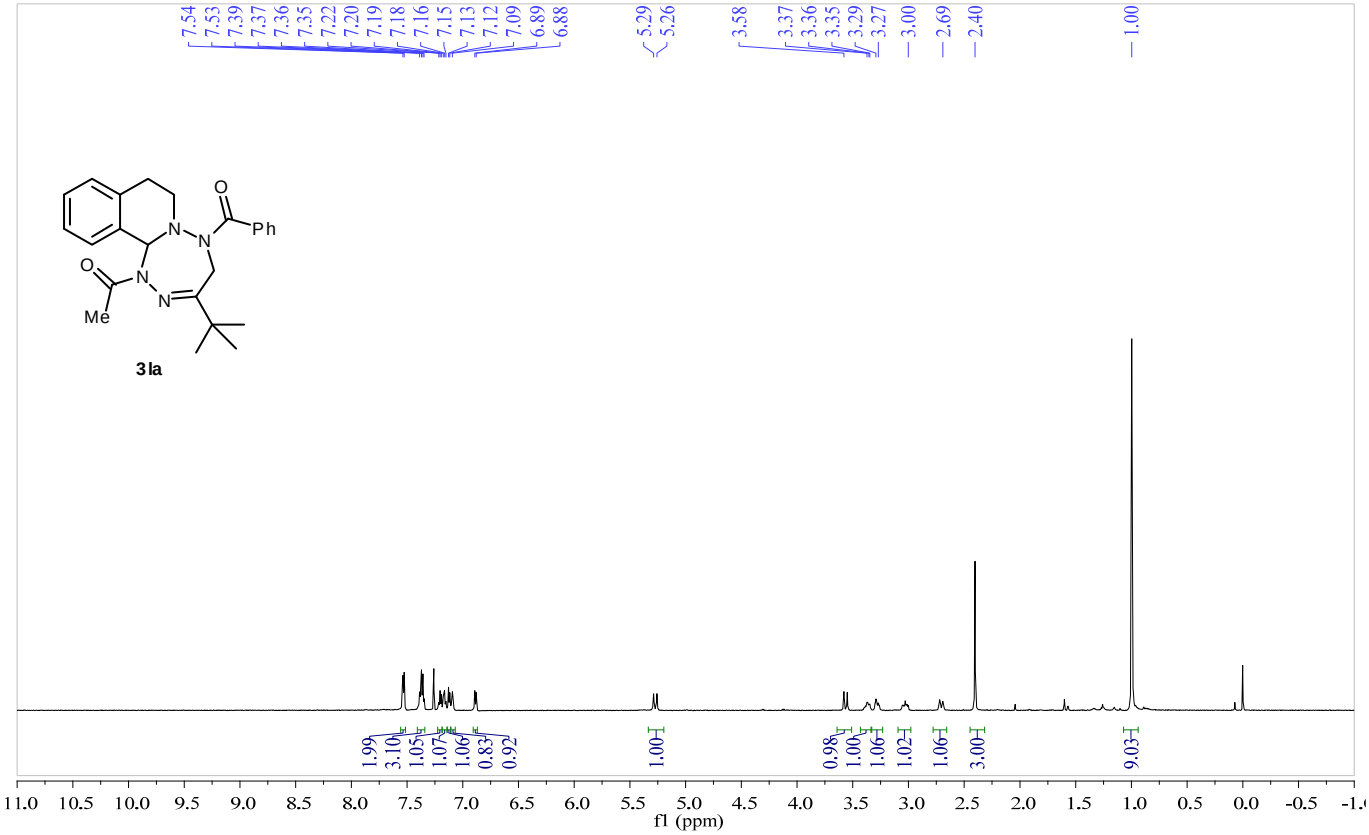


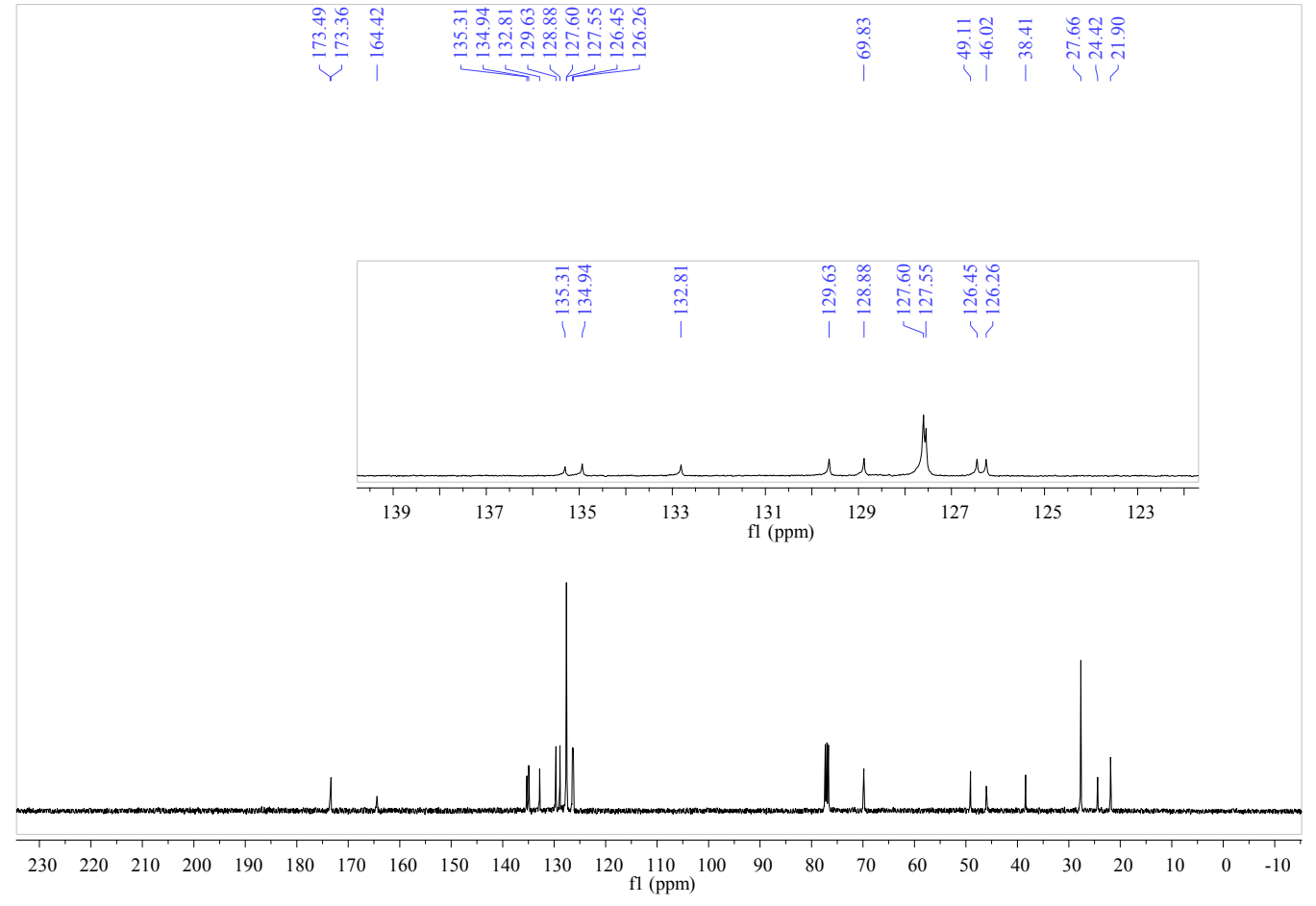
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3ka



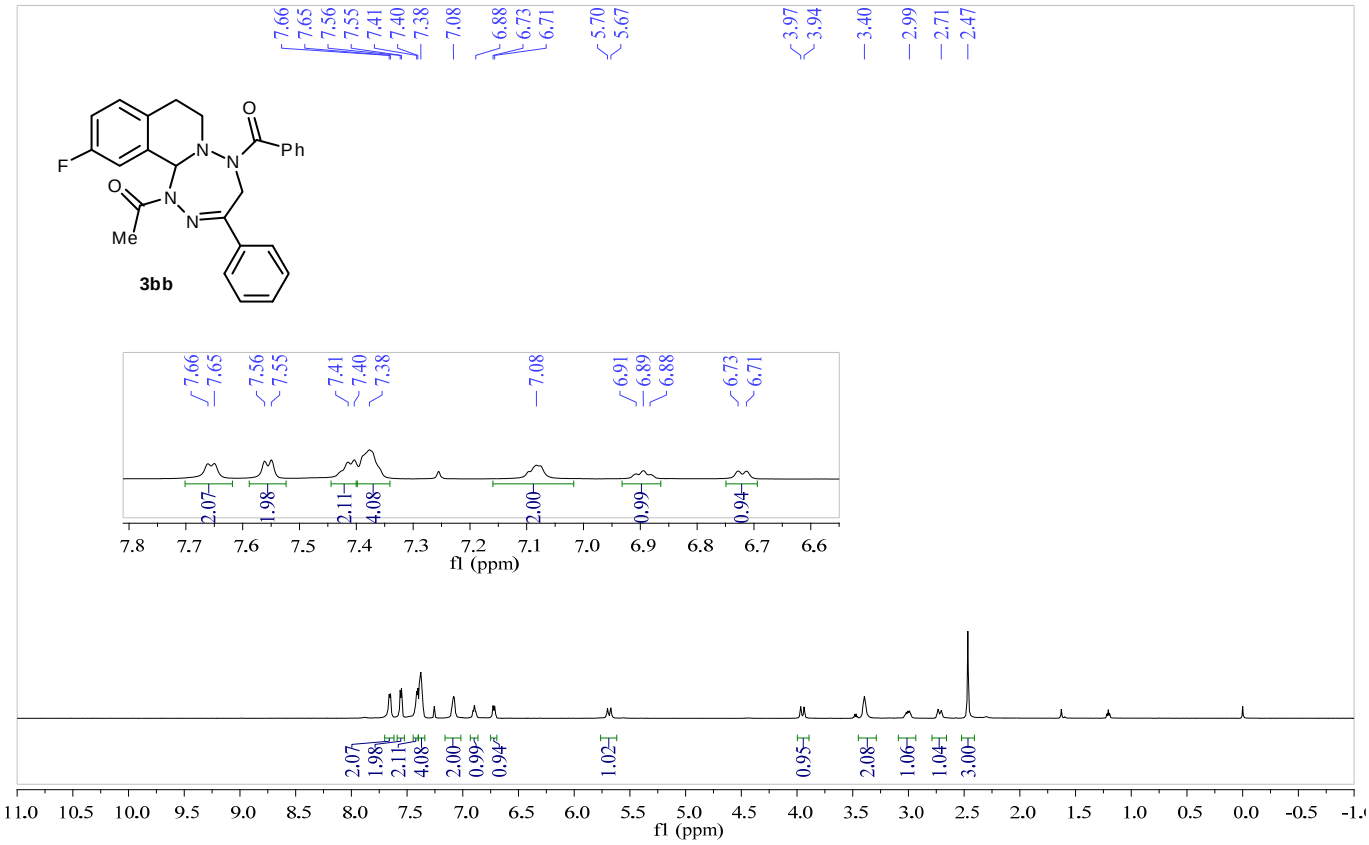


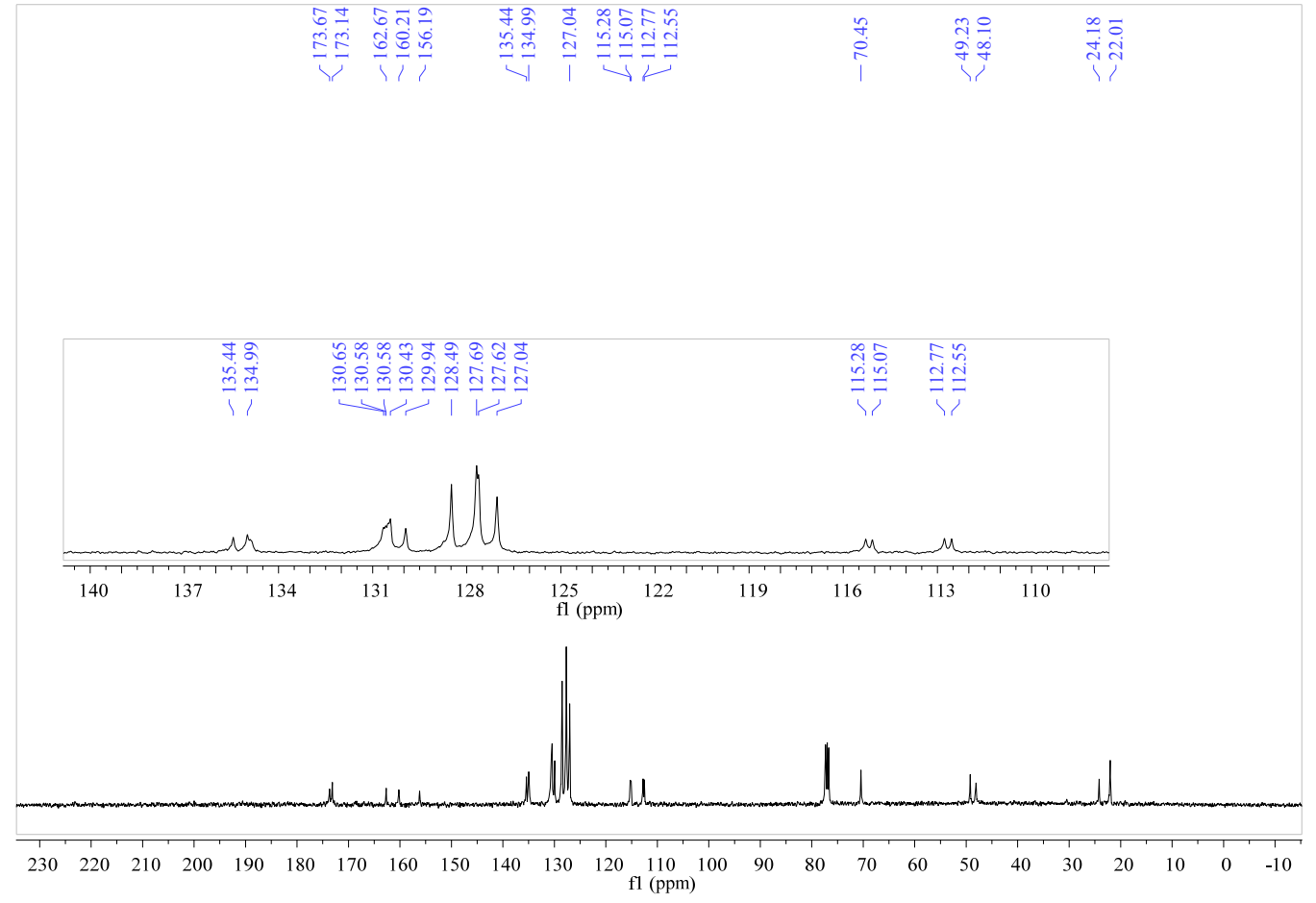
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3la



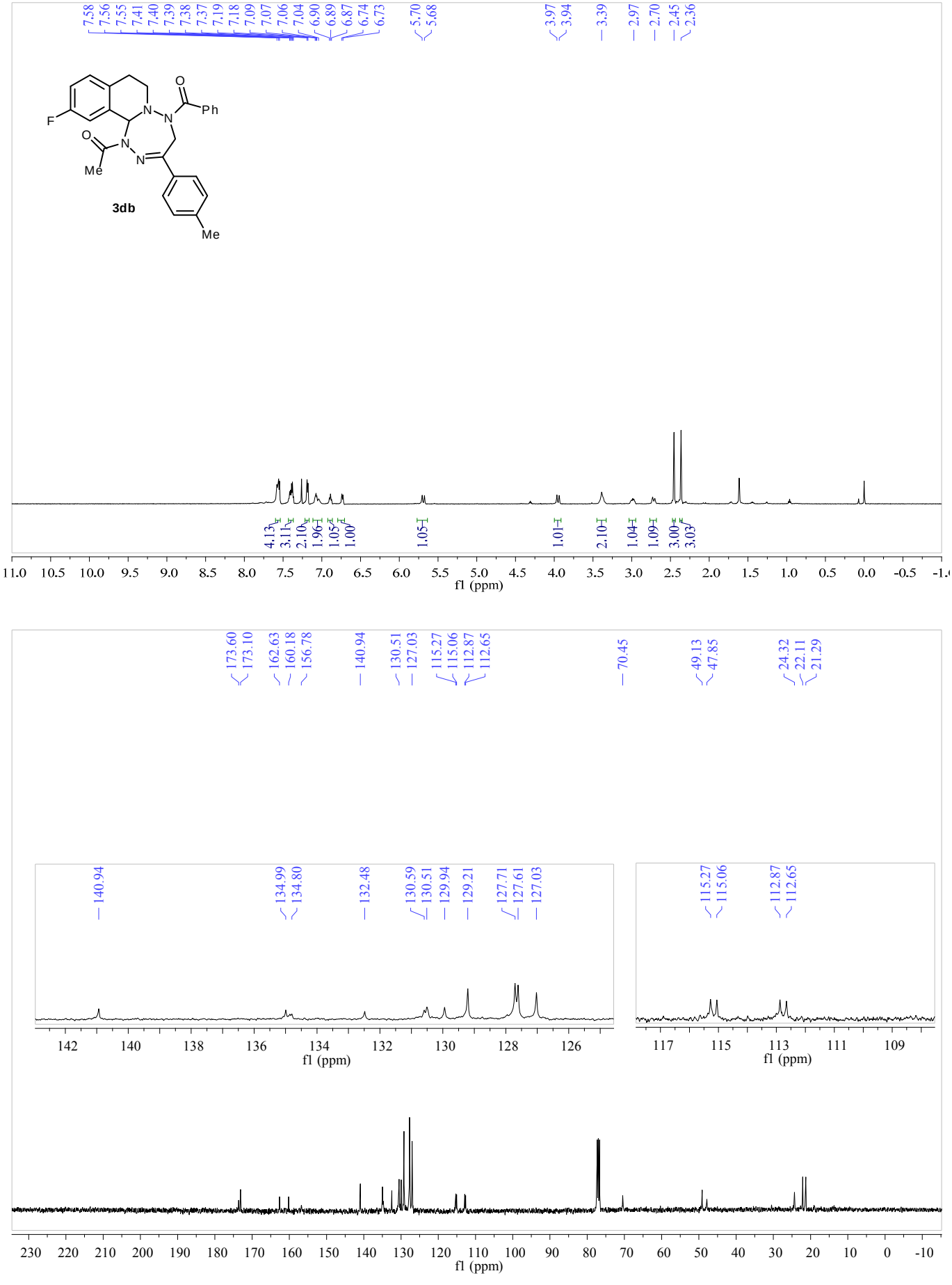


^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3bb

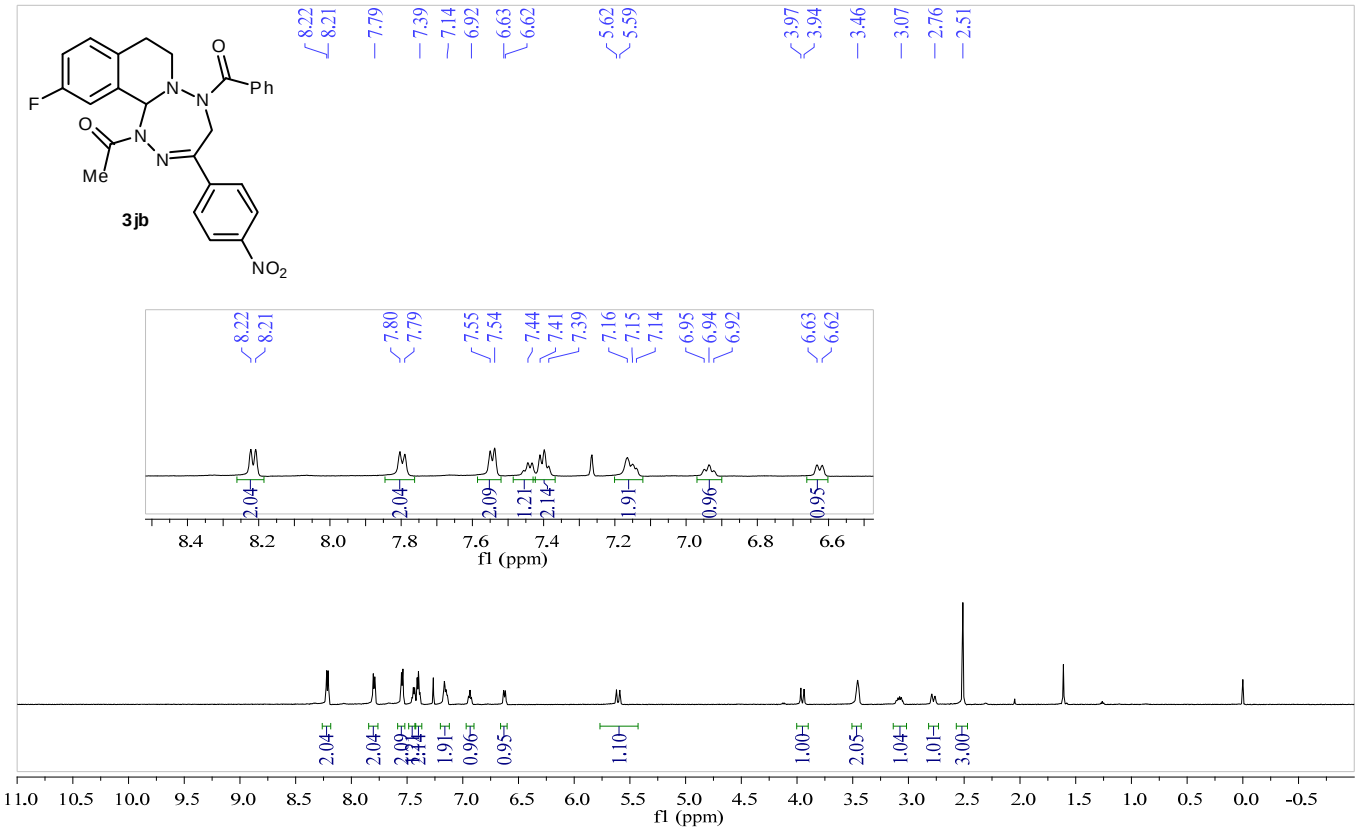


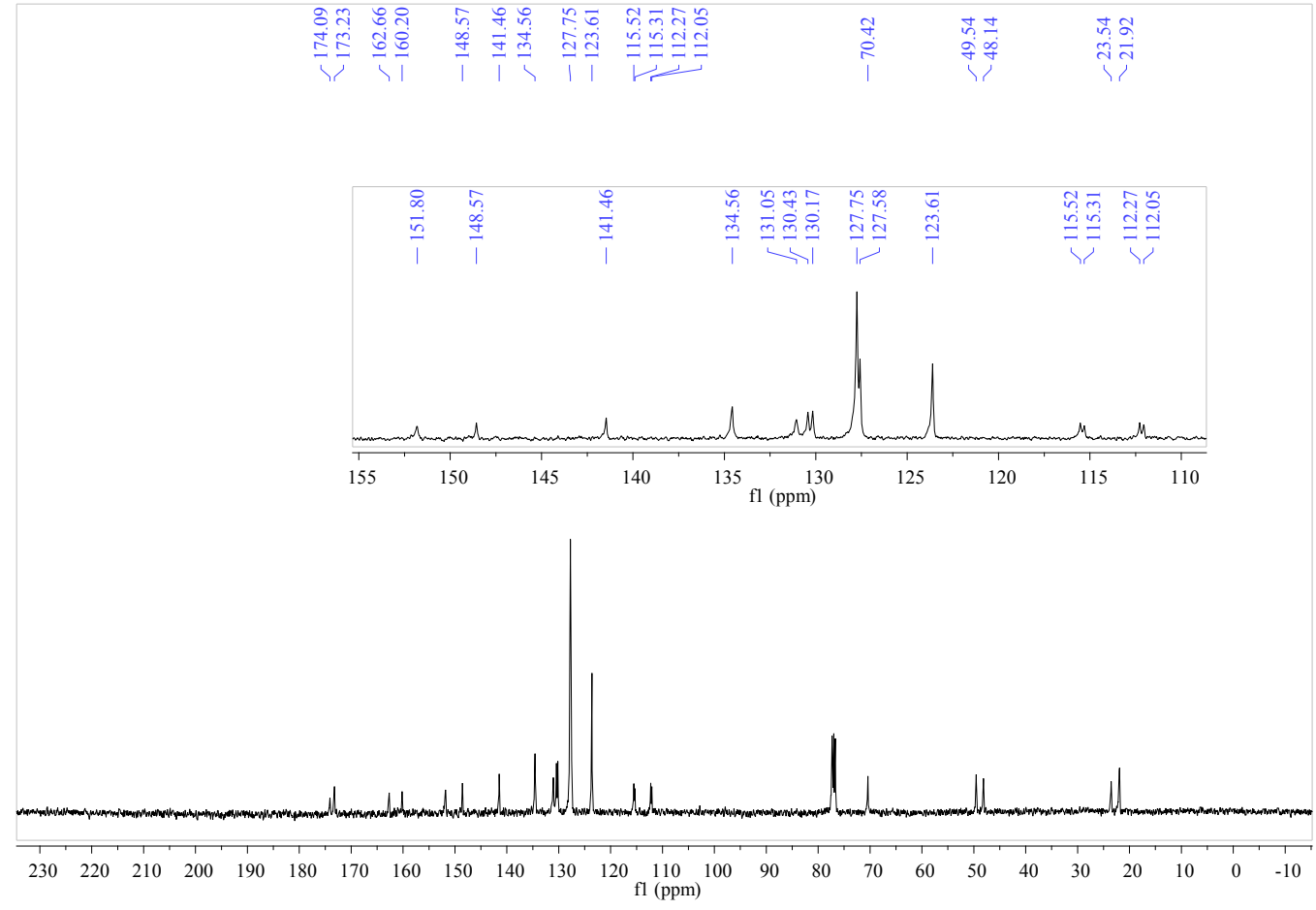


^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3db

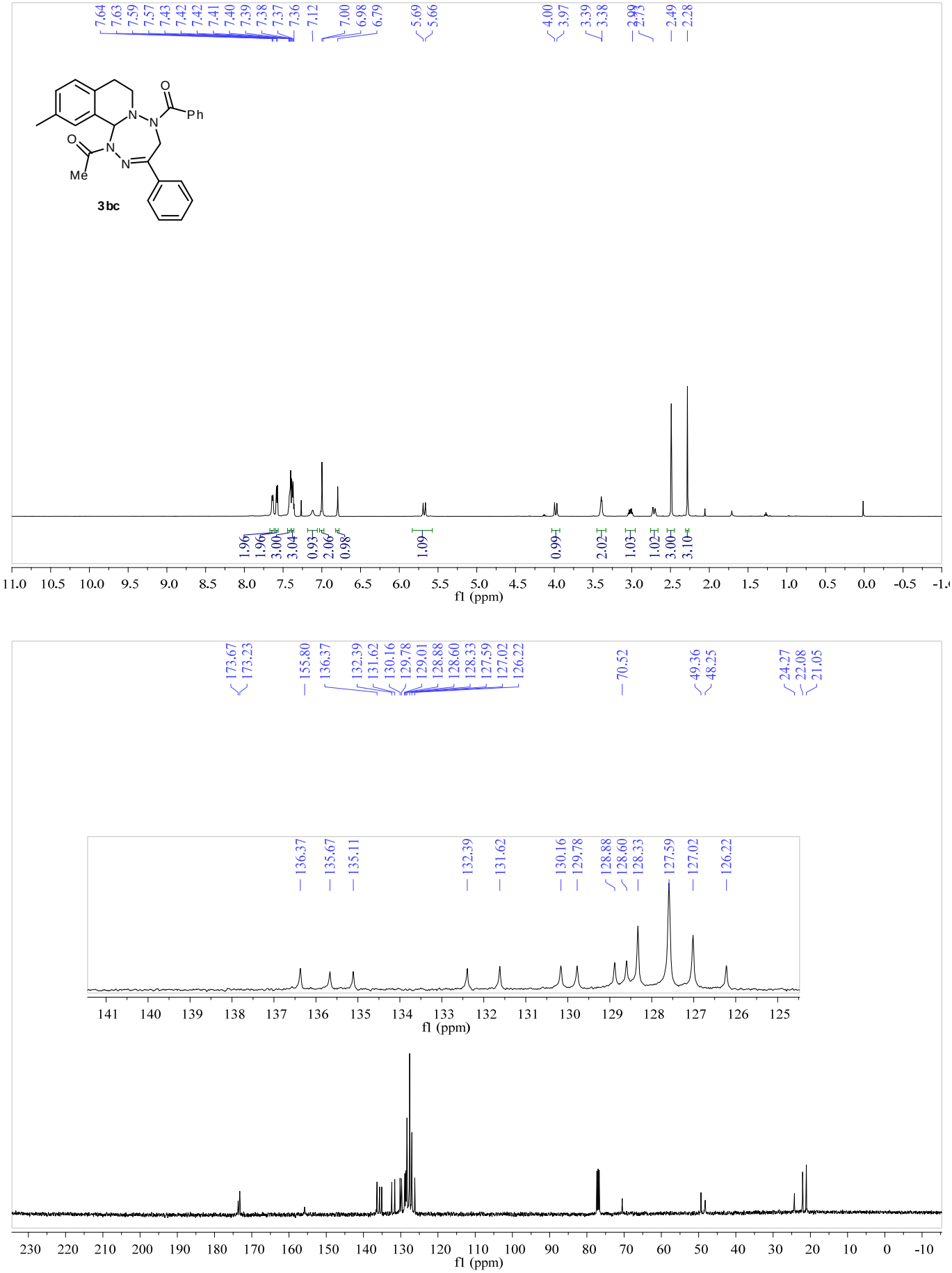


^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3jb

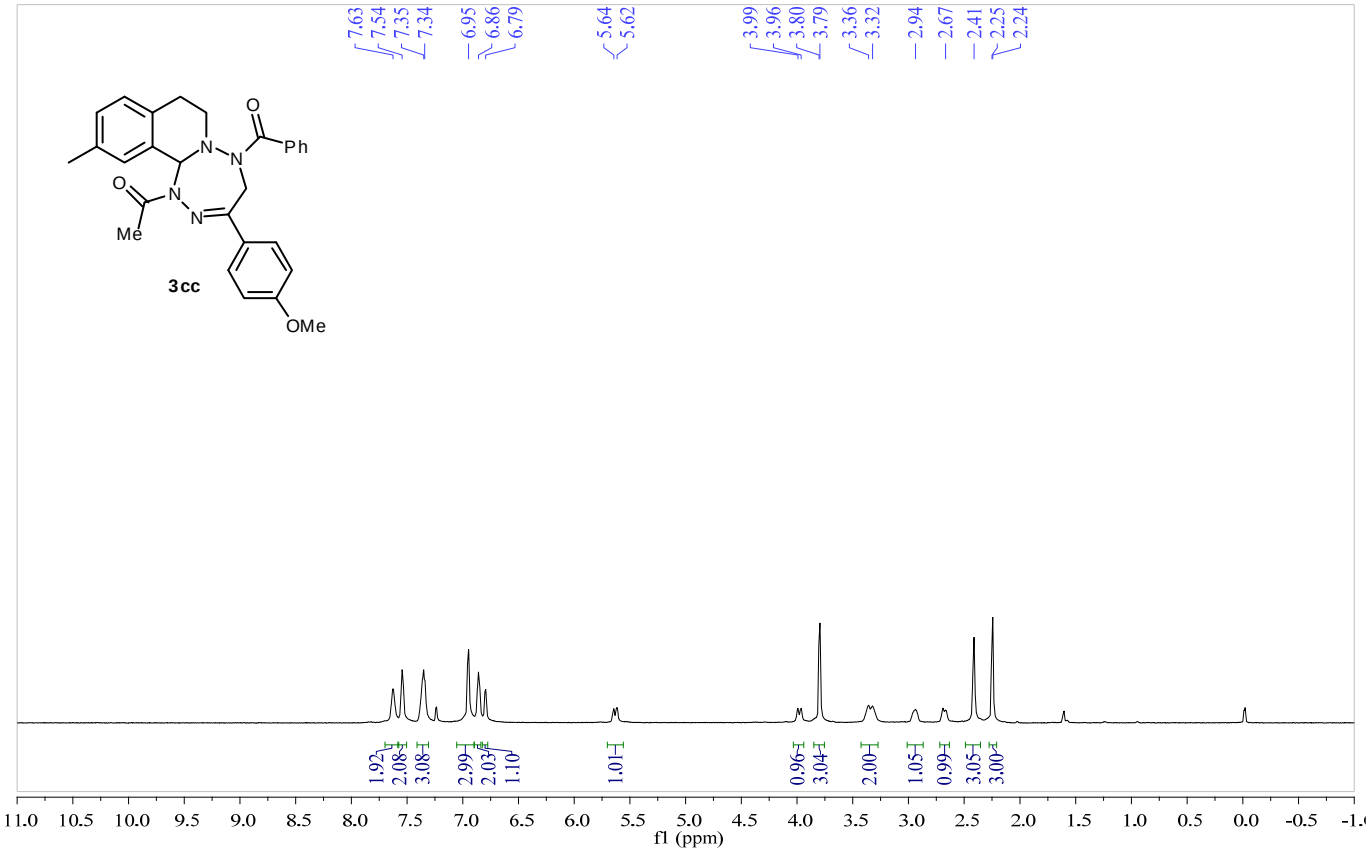


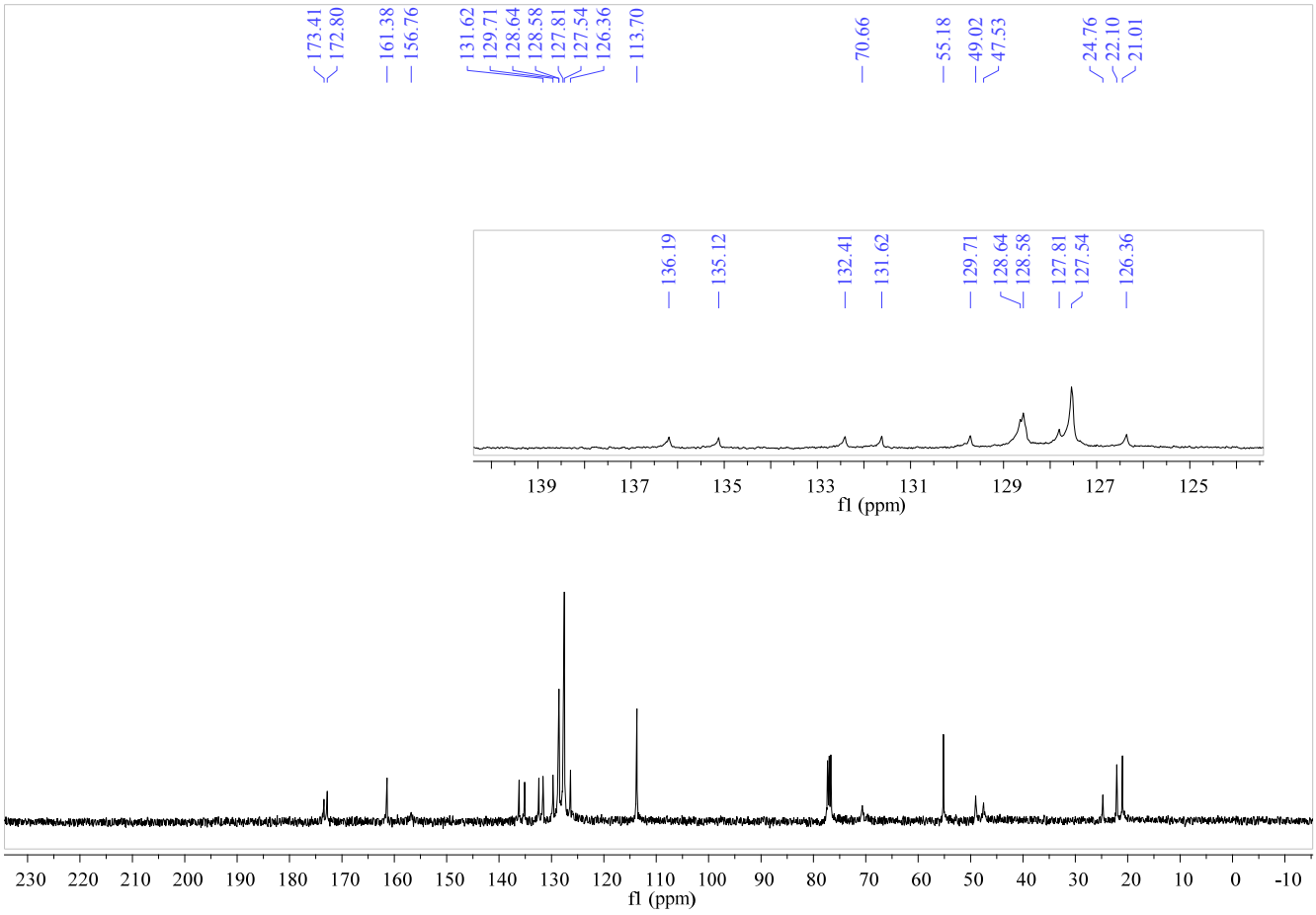


^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3bc

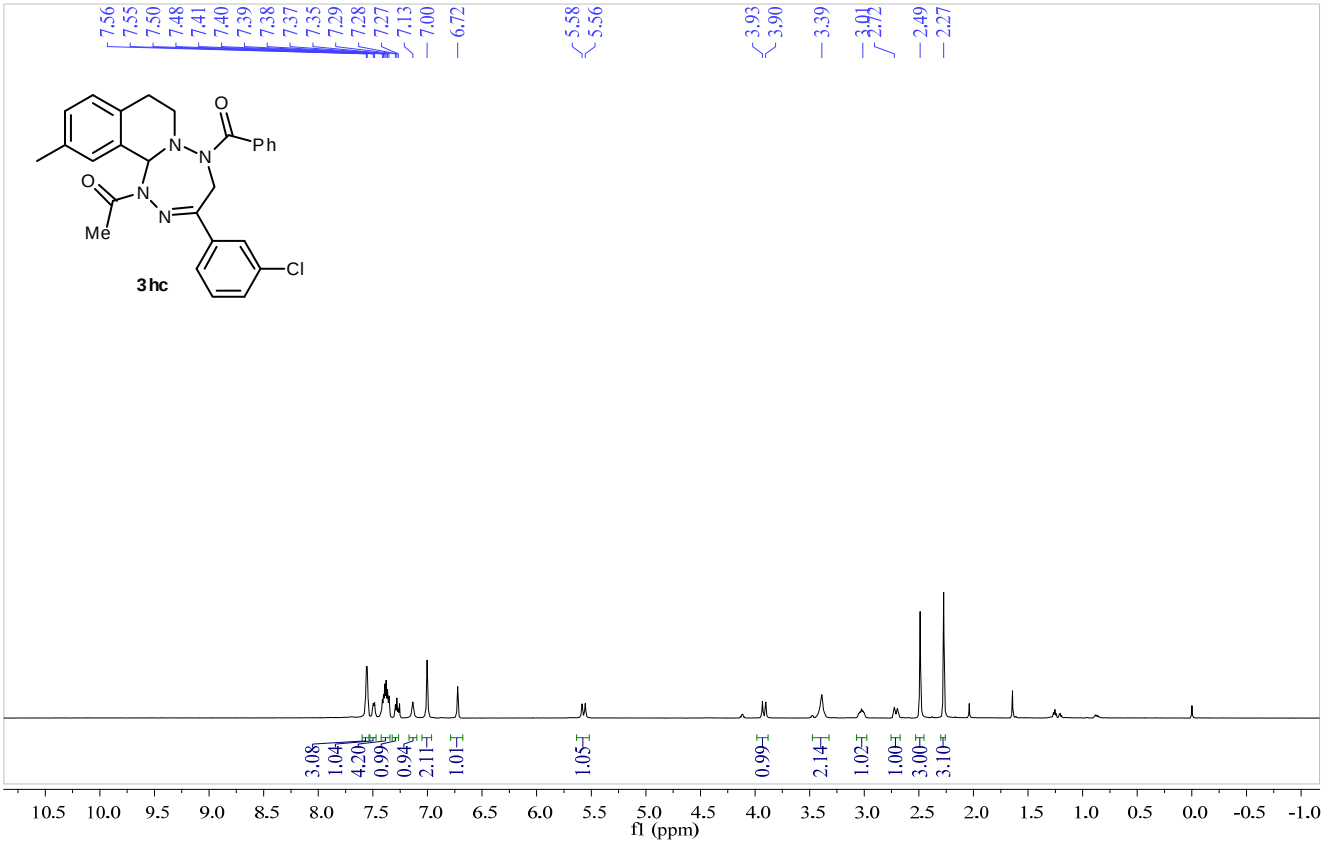


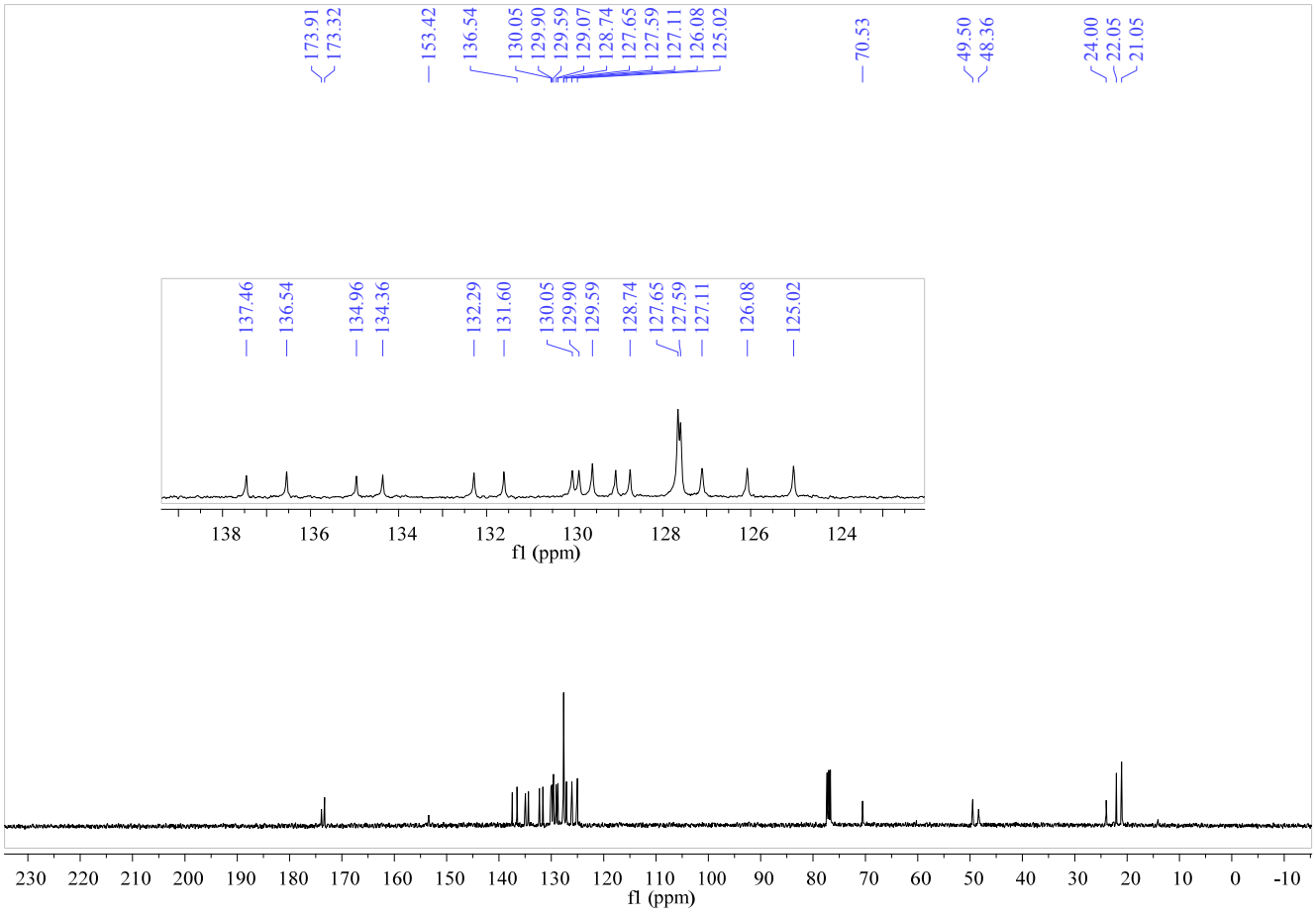
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3cc



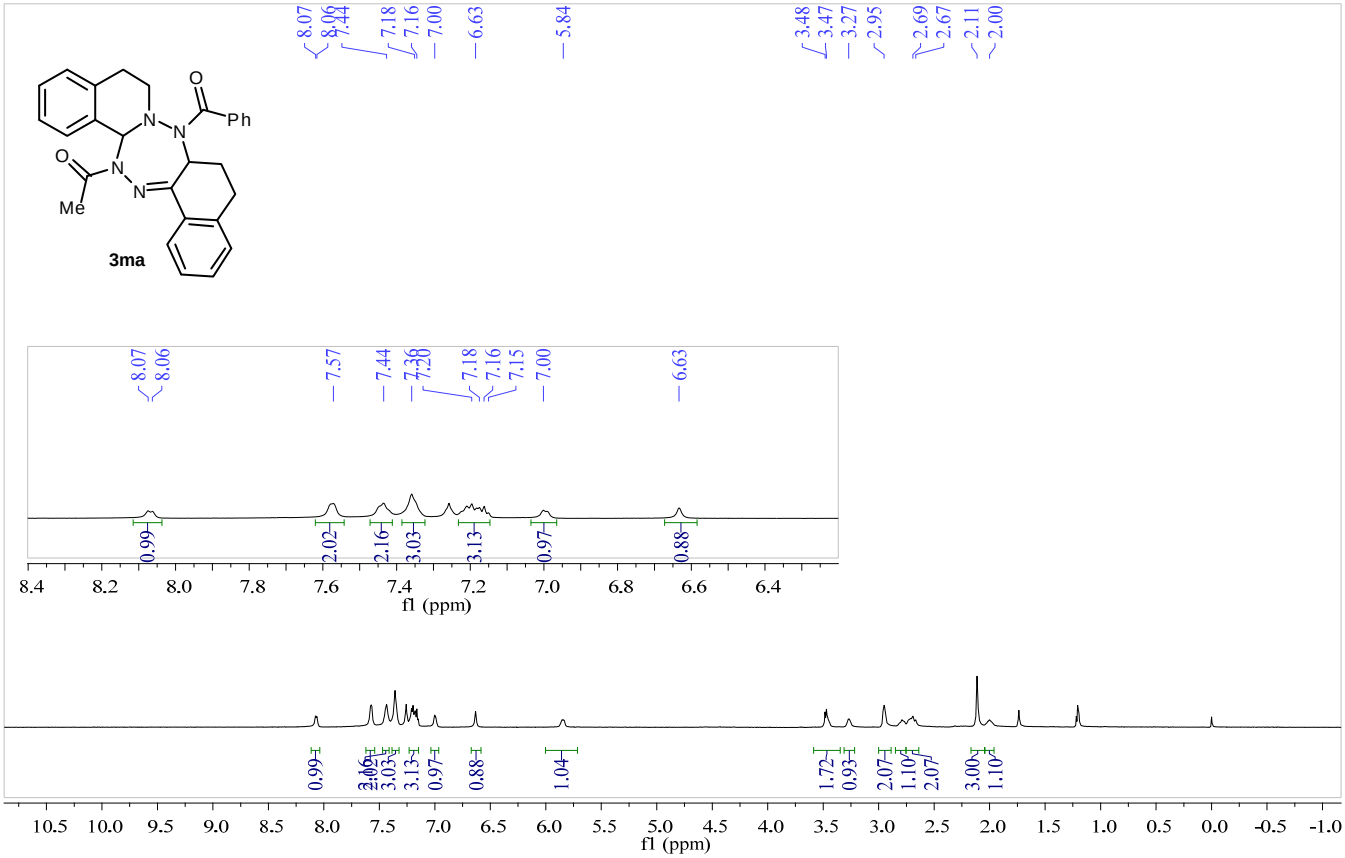


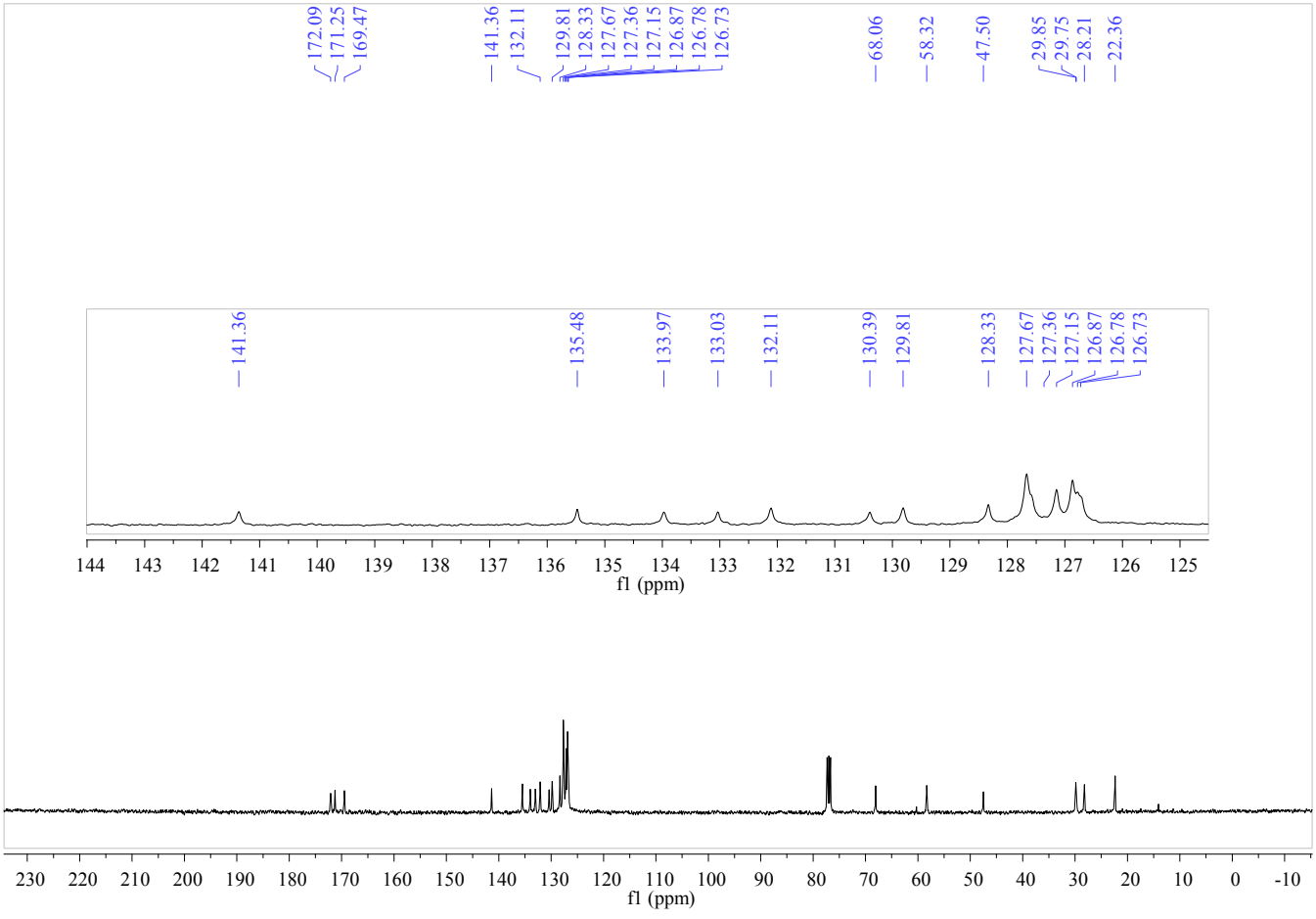
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3hc



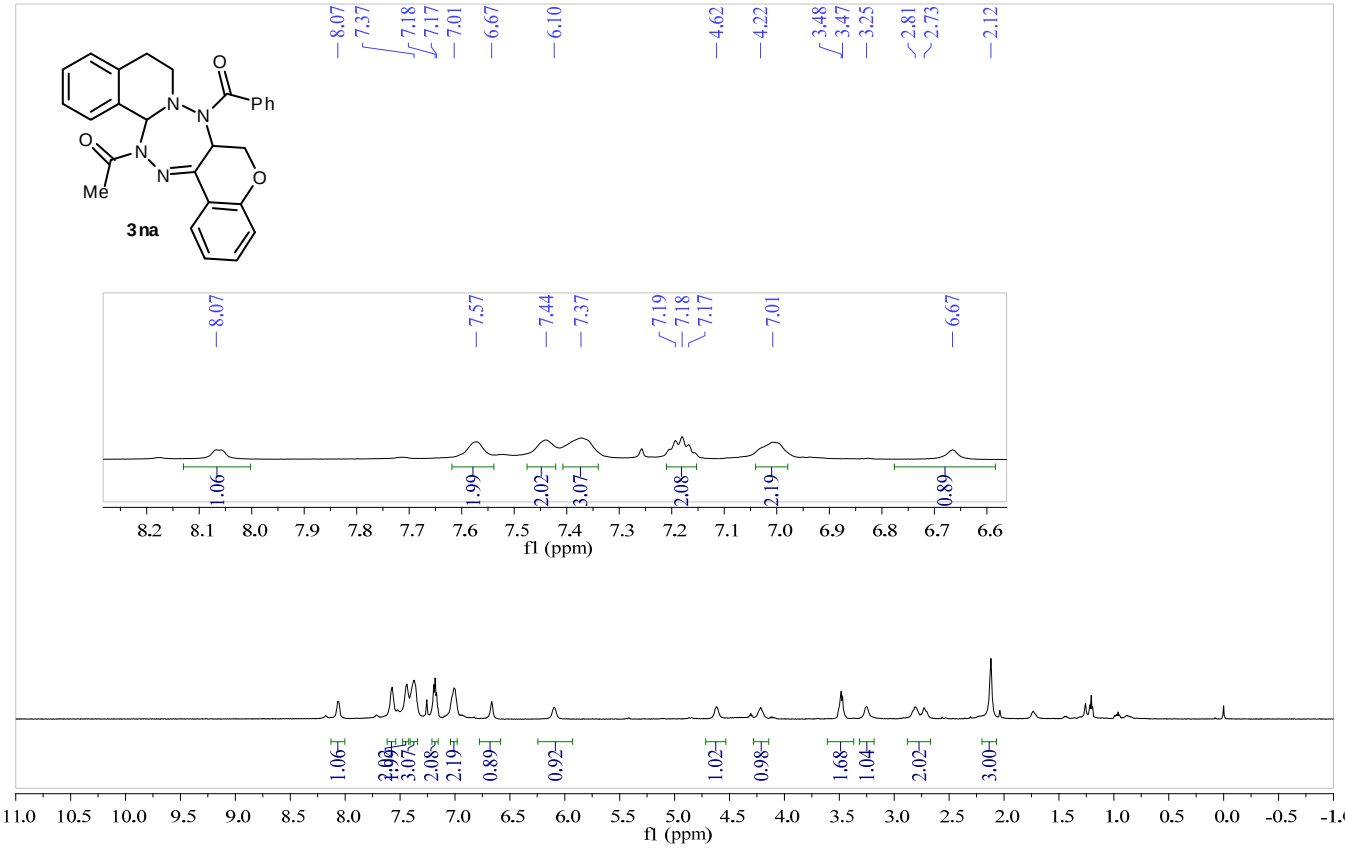


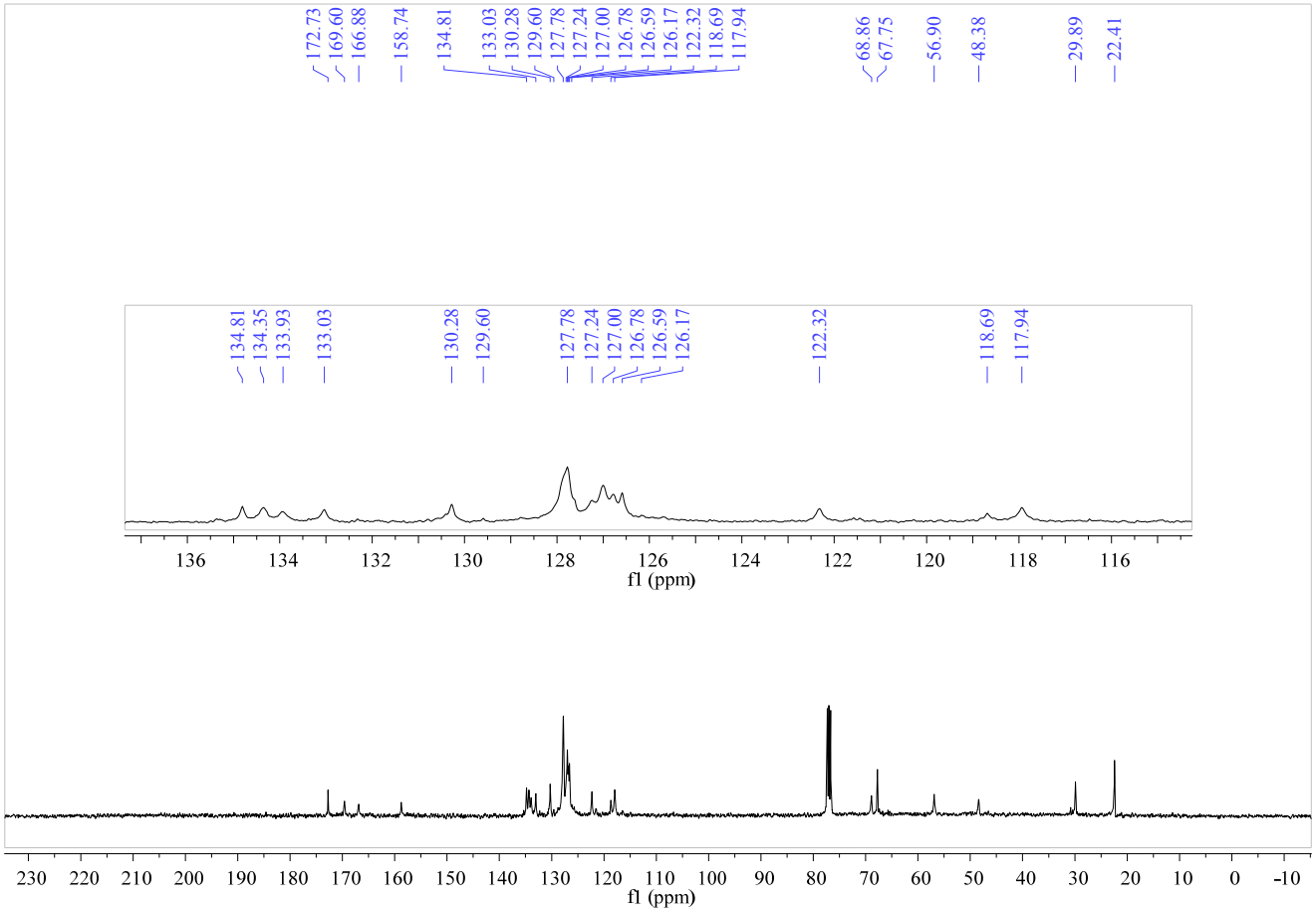
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3ma



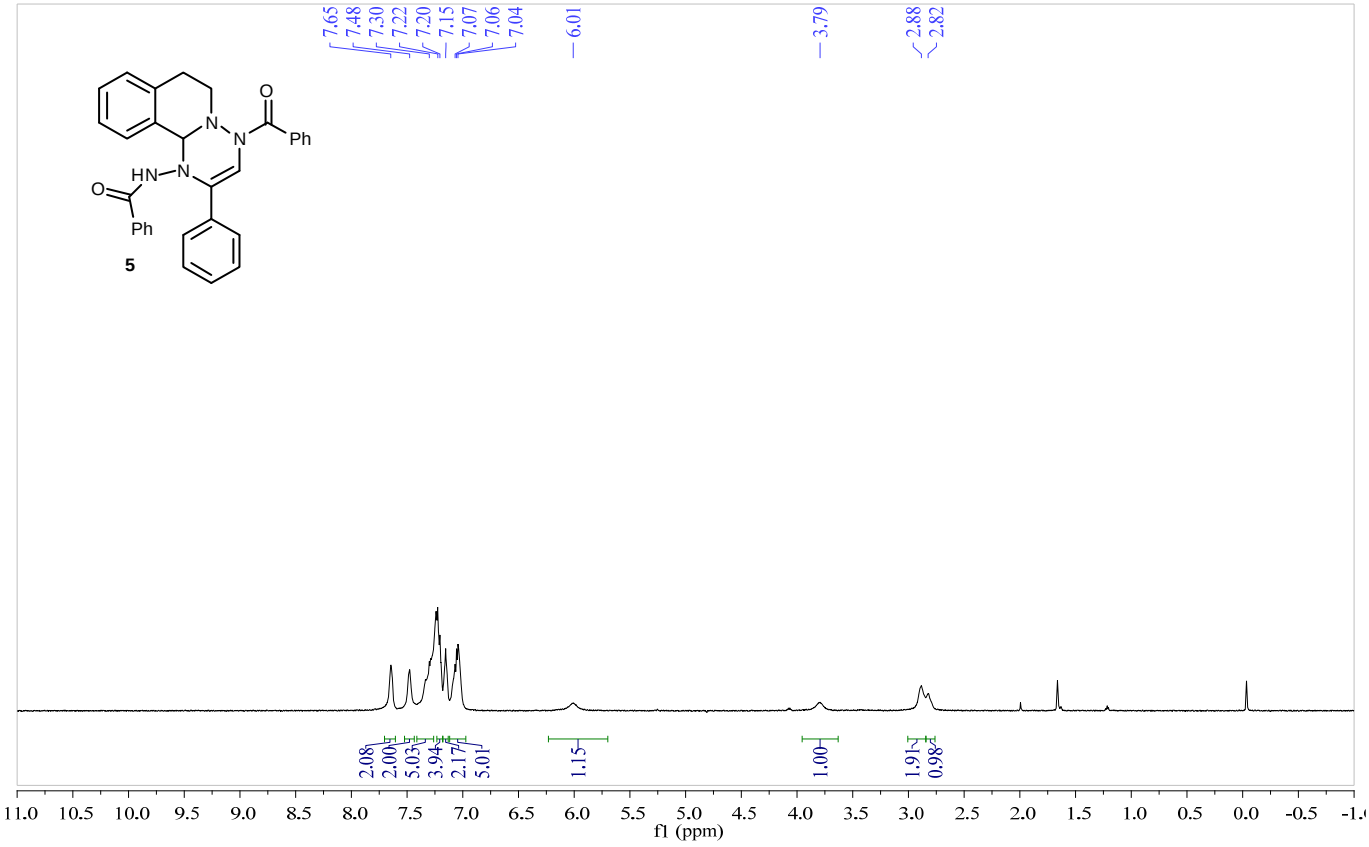


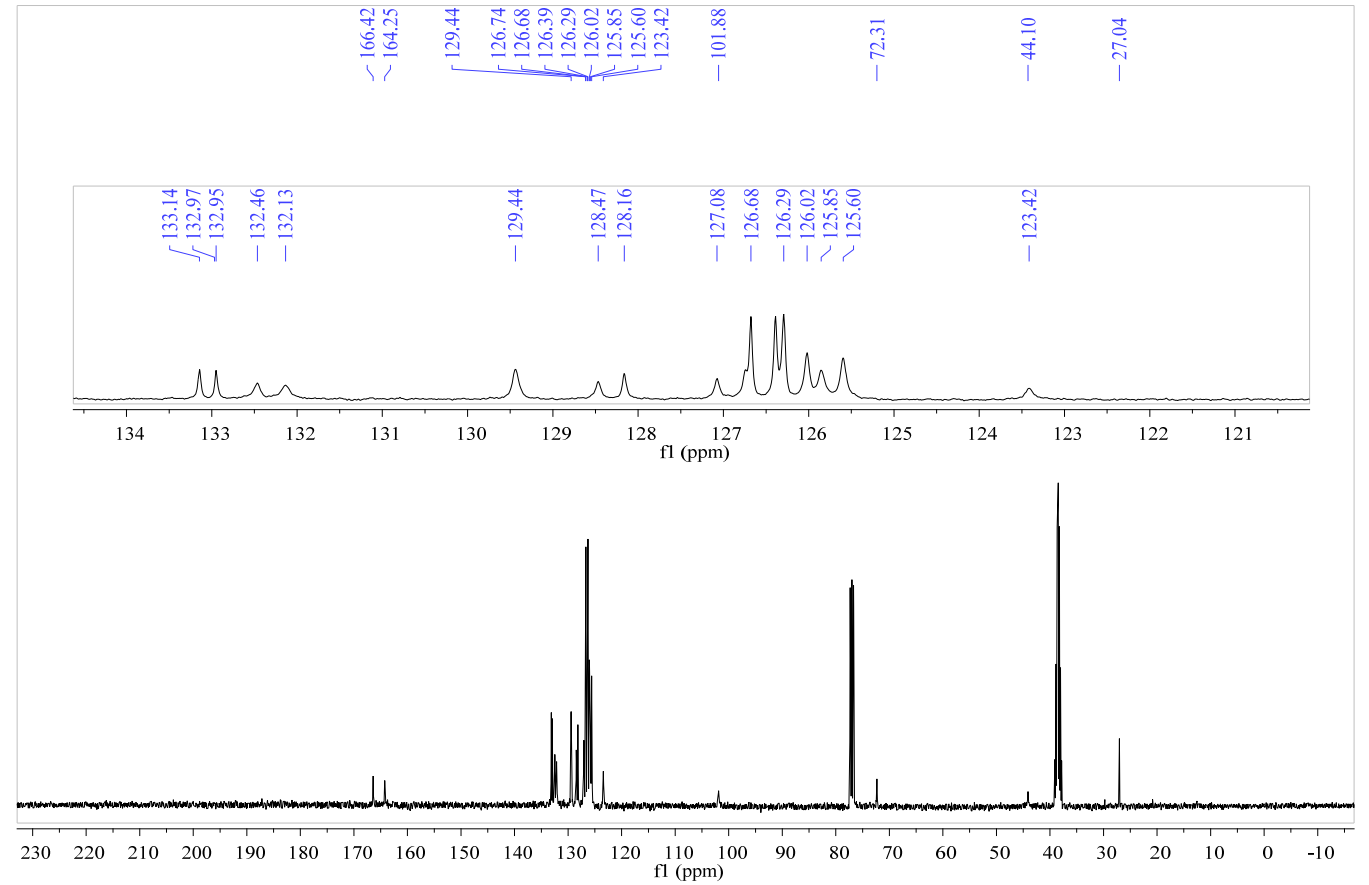
^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3na



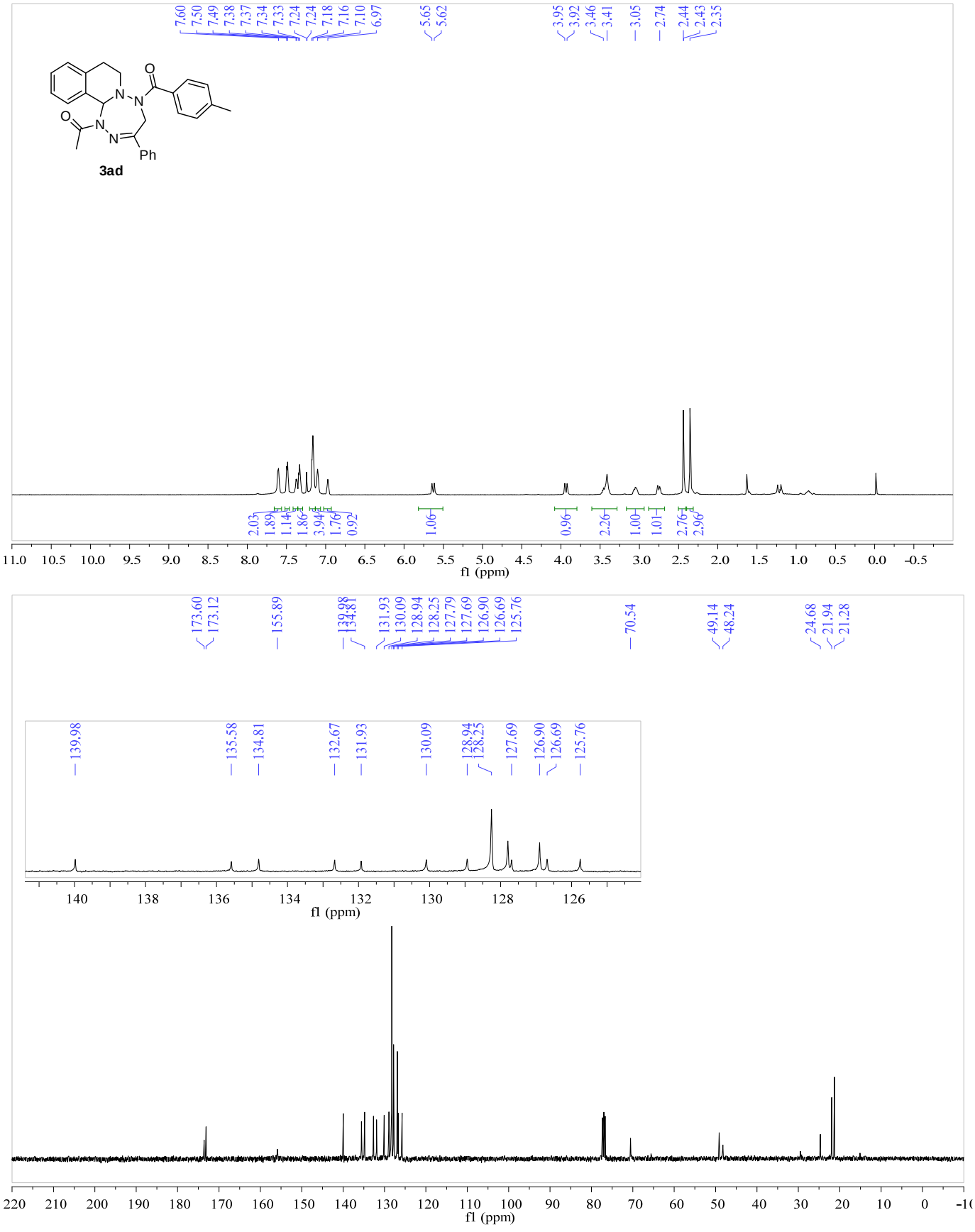


^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$/DMSO-d^6) spectrum of cyclic product 5

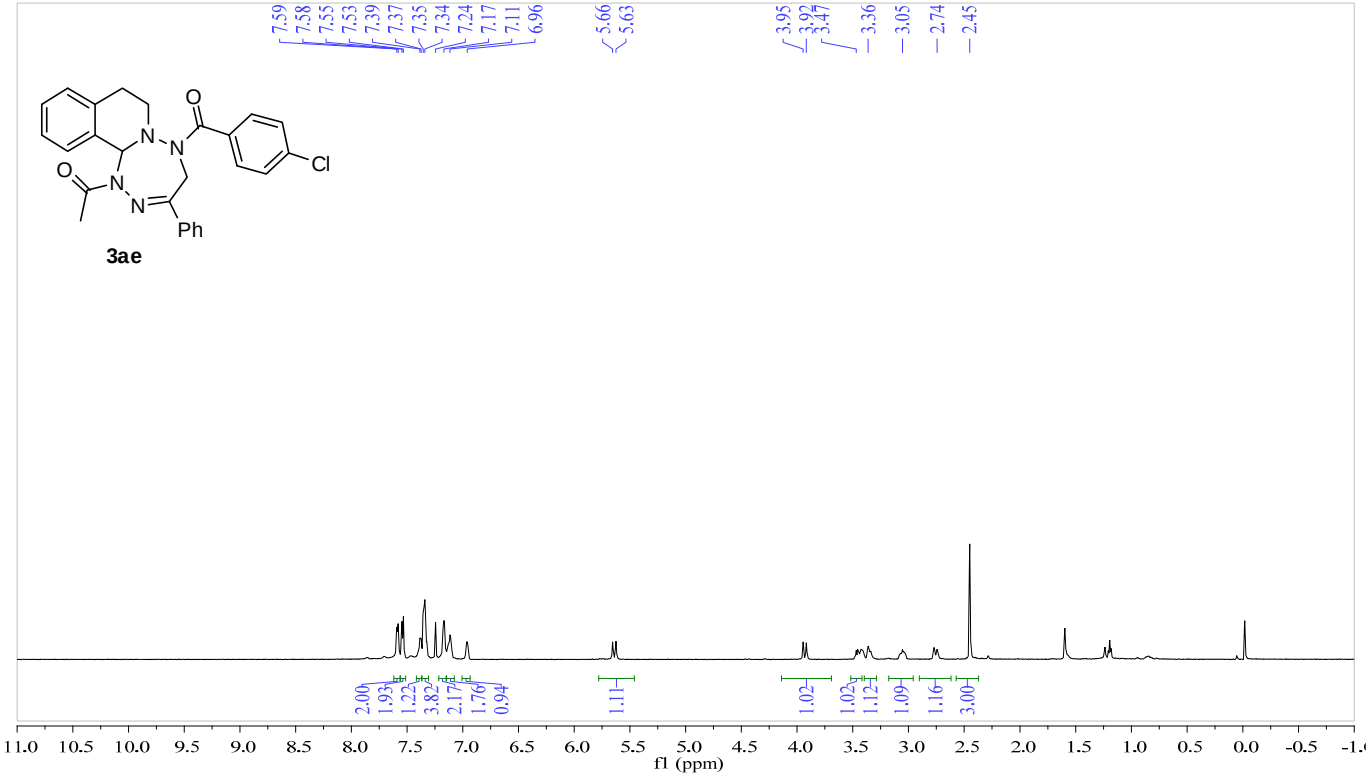


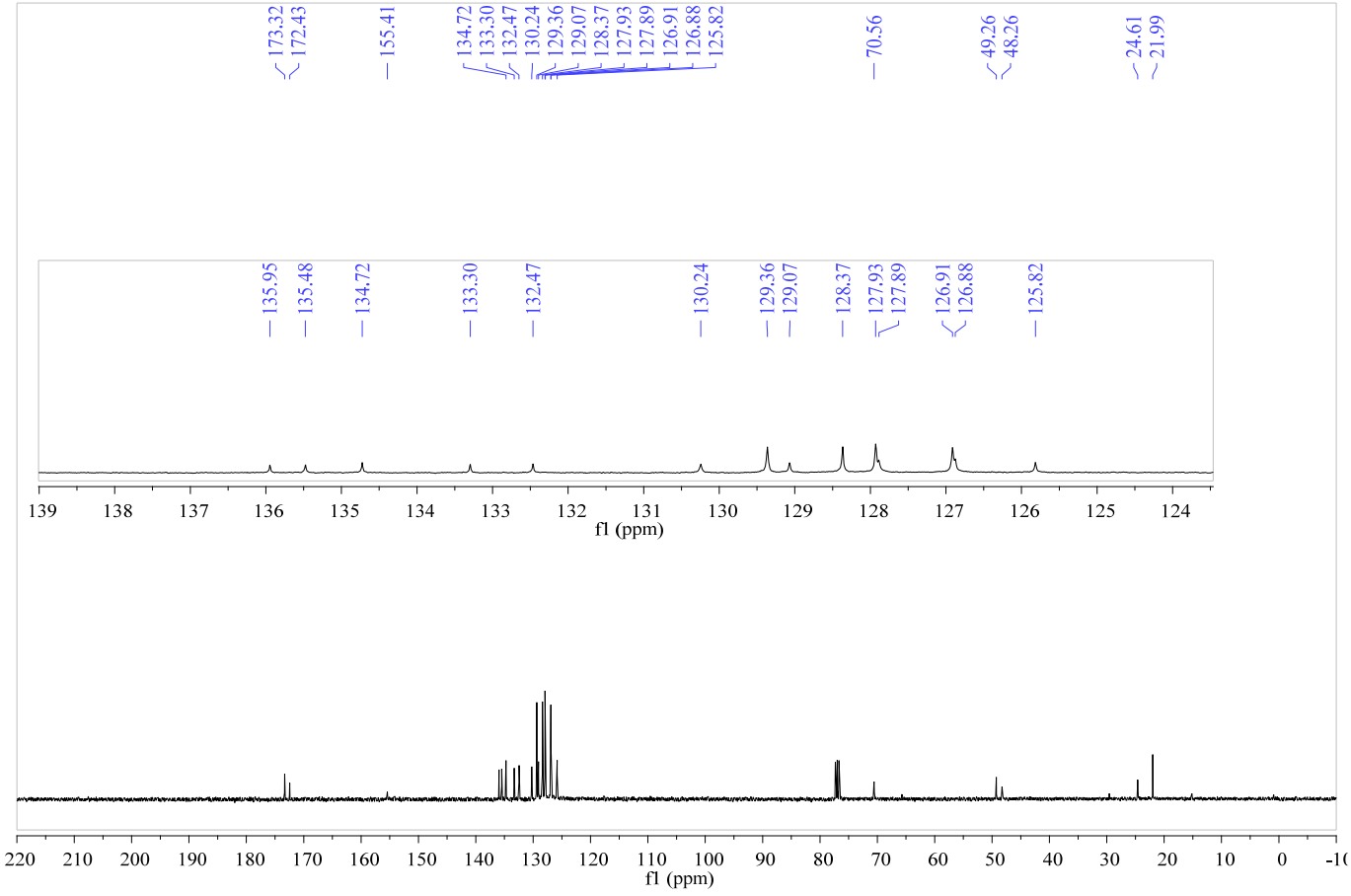


^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3ad



^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3ae





^{1}H NMR (600 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of cyclic product 3af