Synthesis of Polycyclic Imidazolidinones via Amine Redox-Annulation

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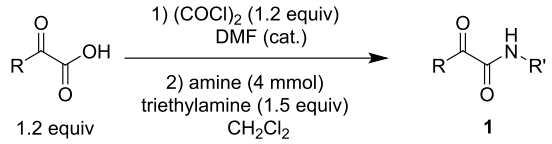
Supporting Information

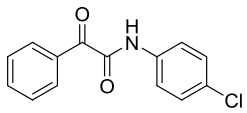
General Information: Reagents and solvents were purchased from commercial sources and purified by distillation or recrystallization prior to use. Purification of reaction products was carried out by flash column chromatography using EM Reagent silica gel 60 (230–400 mesh). Analytical thin layer chromatography was performed on EM Reagent 0.25 mm silica gel 60 F_{254} plates. Visualization was accomplished with UV light, and potassium permanganate, Dragendorff-Munier, and anisaldehyde stains, followed by heating. Infrared spectra were recorded on an ATI Mattson Genesis Series FT-Infrared spectrophotometer. Microwave reactions were carried out in a CEM Discover reactor. Silicon carbide (SiC) passive heating elements were purchased from Anton Paar. Melting points were recorded on a Thomas Hoover capillary melting point apparatus and are uncorrected. Proton nuclear magnetic resonance spectra (^{1}H-NMR) were recorded on a Varian VNMRS-500 MHz instrument and are reported in ppm using solvent as an internal standard ($CDCl_3$ at 7.26 ppm). Data are reported as app = apparent, s = singlet, d = doublet, t = triplet, br = broad, dd = doublet of doublets, ddd = doublet of doublet of doublets, m = multiplet, comp = complex; and coupling constant(s) in Hz. Proton-decoupled carbon nuclear magnetic resonance spectra (^{13}C-NMR) were recorded on a Varian VNMRS-500 MHz instrument and are reported in ppm using solvent as an internal standard ($CDCl_3$ at 77.16 ppm). Mass spectra were recorded on a Finnigan LCQ-DUO mass spectrometer or on a Finnigan 2001 Fourier Transform Ion Cyclotron Resonance Mass Spectrometer. 2-Oxo-2-(*p*-tolyl)acetic acid,[1] 2-(4-chlorophenyl)-2-oxoacetic acid,[1] and 2-(2-fluorophenyl)-2-oxoacetic acid[2] were prepared according to literature procedures.

General Procedure A for the preparation of α-ketoamides.

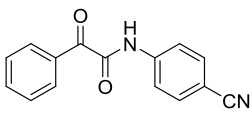


This method was adopted from a literature procedure.[3] To an ice-cooled solution of α-ketoacid (4.8 mmol, 1.2 equiv) in dry CH_2Cl_2 (10 mL, 0.4 M) was slowly added oxalyl chloride (0.406 mL, 4.8 mmol, 1.2 equiv), followed by two drops of DMF. The reaction mixture was allowed to warm to room temperature and stirred for 3 h. The resulting mixture was then cooled in an ice bath, and a solution of triethylamine (0.835 mL, 6 mmol, 1.5 equiv) and the amine (4 mmol, 1 equiv) in CH_2Cl_2 (5 mL) was added dropwise. The reaction mixture was allowed to warm to room temperature and stirred for 15 h. Subsequently, the reaction mixture was diluted with CH_2Cl_2 (20 mL) and washed with a 1N HCl solution (20 mL), water (20 mL), brine (20 mL) and dried over Na_2SO_4. Solvent was then removed under reduced pressure and the residue purified by silica gel chromatography. Compounds **1a**,[3] **1l**,[4] and **1n**[5] were previously reported and their published characterization data matched our own in all respects.

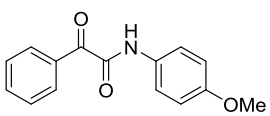
***N*-(4-chlorophenyl)-2-oxo-2-phenylacetamide (1b):** Following the general procedure A, product **1b** was obtained from 2-oxo-2-phenylacetic acid and 4-chloroaniline as a yellow solid in 33% yield (334 mg). R_f = 0.55 in hexane/EtOAc 80:20 v/v; mp = 142–144 °C; IR (KBr) 3342, 3102, 1697, 1663, 1590, 1533, 1494, 1447, 1402, 1280, 1170, 1088, 1012, 992, 880, 817, 798, 739, 689, 683, 498, 447, 419; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.99 (br s, 1H), 8.45–8.36 (comp, 2H), 7.70–7.62 (comp, 3H), 7.55–7.48 (comp, 2H), 7.38–7.33 (comp, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 187.2, 158.9, 135.4, 134.9, 133.1, 131.6, 130.5, 129.4, 128.7, 121.3; *m/z* (ESI–MS) (^{35}Cl) 260.0 $[M + H]^+$, (^{37}Cl) 261.9 $[M + H]^+$.



***N*-(4-cyanophenyl)-2-oxo-2-phenylacetamide (1c):** Following the general procedure A, product **1c** was obtained from 2-oxo-2-phenylacetic acid and 4-aminobenzonitrile as a yellow solid in 58% yield (581 mg). R_f = 0.31 in hexane/EtOAc 80:20 v/v; mp = 181–183 °C; IR (KBr) 3328, 2226, 1701, 1671, 1660, 1604, 1584, 1523, 1446, 1410, 1282, 1164, 988, 880, 850, 827, 746, 690, 666, 545, 458; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 9.16 (br s, 1H), 8.43–8.41 (comp, 2H), 7.86–7.83 (comp, 2H), 7.72–7.67 (comp, 3H), 7.56–7.51 (comp, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 186.5, 159.0, 140.7, 135.2, 133.6, 132.8, 131.7, 128.9, 120.1, 118.7, 108.6; *m/z* (ESI–MS) 250.8 $[M + H]^+$.

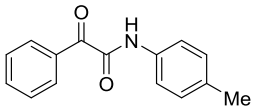


***N*-(4-methoxyphenyl)-2-oxo-2-phenylacetamide (1d):** Following the general procedure A, product **1d** was obtained from 2-oxo-2-phenylacetic acid and 4-methoxyaniline as a yellow solid in 43% yield (435 mg). R_f = 0.42 in hexane/EtOAc 80:20 v/v; mp = 99–101 °C; IR (KBr) 3367, 3343, 2991, 2838, 1685, 1664, 1654, 1593, 1541, 1535, 1509, 1448, 1408, 1303, 1277, 1246, 1165, 1035, 987, 880, 824, 744, 686, 671, 509469; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.89 (br s, 1H), 8.44–8.38 (comp, 2H), 7.67–7.59 (comp, 3H), 7.53–7.47 (comp, 2H), 6.96–6.89 (comp,

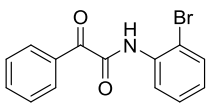


2H), 3.81 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 187.7, 158.8, 157.2, 134.7, 133.3, 131.6, 129.9, 128.6, 121.6, 114.5, 55.6; *m/z* (ESI–MS) 255.8 $[M + H]^+$.

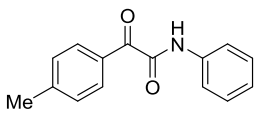
2-Oxo-2-phenyl-*N*-(*p*-tolyl)acetamide (**1e**)**:** Following the general procedure A, product **1e** was obtained from 2-oxo-2-phenylacetic acid and *p*-toluidine as a yellow solid in 40% yield (383 mg). R_f = 0.58 in hexane/EtOAc 80:20 v/v; mp = 112–113 °C; IR (KBr)3341, 3056, 2913, 1698, 1671, 1592, 1538, 1445, 1408, 1283, 1170, 988, 878, 821, 741, 702, 688, 669, 506, 497; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.90 (br s, 1H), 8.44–8.39 (comp, 2H), 7.69–7.63 (m, 1H), 7.62–7.57 (comp, 2H), 7.55–7.46 (comp, 2H), 7.20 (app d, *J* = 8.2 Hz, 2H), 2.35 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 187.6, 158.9, 135.2, 134.7, 134.2, 133.3, 131.6, 129.9, 128.7, 120.0, 21.1; *m/z* (ESI–MS) 239.9 $[M + H]^+$.



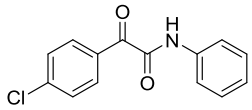
***N*-(2-bromophenyl)-2-oxo-2-phenylacetamide (1f):** Following the general procedure A, product **1f** was obtained from 2-oxo-2-phenylacetic acid and 2-bromoaniline as a yellow solid in 42% yield (501 mg). R_f = 0.64 in hexane/EtOAc 80:20 v/v; mp = 53–55 °C; IR (KBr) 3333, 3322, 3059, 1690, 1671, 1591, 1574, 1523, 1438, 1285, 1273, 1238, 1022, 1004, 990, 881, 749, 737, 681, 657, 561, 508, 433; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 9.59 (br s, 1H), 8.51 (dd, *J* = 8.2, 1.6 Hz, 1H), 8.46–8.35 (comp, 2H), 7.72–7.64 (m, 1H), 7.61 (dd, *J* = 8.1, 1.5 Hz, 1H), 7.52 (app t, *J* = 7.8 Hz, 2H), 7.39 (ddd, *J* = 8.3, 7.8, 1.5 Hz, 1H), 7.07 (app td, *J* = 7.7, 1.6 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 186.8, 159.0, 134.8, 133.1, 132.8, 131.6, 128.7, 128.6, 126.3, 121.6, 114.4; *m/z* (ESI–MS) (^{79}Br) 304.1 $[M + H]^+$, (^{81}Br) 305.9 $[M + H]^+$.



2-Oxo-*N*-phenyl-2-(*p*-tolyl)acetamide (1g): Following the general procedure A, product **1g** was obtained from 2-oxo-2-(*p*-tolyl)acetic acid and aniline as a white solid in 71% yield (680 mg). R_f = 0.59 in hexane/EtOAc 80:20 v/v; mp = 122–124 °C; IR (KBr) 3321, 3059, 1691, 1654, 1604, 1593, 1534, 1498, 1441, 1282, 1162, 1003, 981, 900, 878, 848, 770, 751, 687, 546, 496; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.96 (br s, 1H), 8.37–8.33 (comp, 2H), 7.72–7.68 (comp, 2H), 7.42–7.37 (comp, 2H), 7.31 (dd, *J* = 8.1, 0.5 Hz, 2H), 7.21–7.18 (m, 1H), 2.45 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 186.8, 159.1, 146.0, 136.7, 131.6, 130.6, 129.3, 129.2, 125.2, 119.9, 21.9; *m/z* (ESI–MS) 239.9 $[M + H]^+$.



2-(4-Chlorophenyl)-2-oxo-*N*-phenylacetamide (1h): Following the general procedure A, product **1h** was obtained from 2-(4-chlorophenyl)-2-oxoacetic acid and aniline as a white solid in 78% yield (810 mg). R_f = 0.62 in hexane/EtOAc 80:20 v/v; mp = 127–129 °C; IR (KBr) 3338, 3101, 1685, 1655, 1586, 1534, 1499, 1446, 1401, 1279, 1166, 1089, 986, 900, 875, 853, 788, 747, 685, 600, 565, 495; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.95 (br s, 1H), 8.44–8.40 (comp, 2H), 7.71–7.67 (comp, 2H), 7.51–7.47 (comp, 2H), 7.43–7.37 (comp, 2H), 7.23–7.19 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 186.2, 158.7, 141.7, 136.6, 133.1, 131.6, 129.4, 129.1, 125.6, 120.1; *m/z* (ESI–MS) (^{35}Cl) 259.9 $[M + H]^+$, (^{37}Cl) 262.0 $[M + H]^+$.



2-(2-Fluorophenyl)-2-oxo-*N*-phenylacetamide (1i): Following the general procedure A, product **1i** was obtained from 2-(2-fluorophenyl)-2-oxoacetic acid and aniline as a white solid in 90% yield (874 mg). R_f = 0.44 in hexane/EtOAc 80:20 v/v; mp = 78–80 °C; IR (KBr) 3267, 3144, 3076, 1670, 1649, 1609, 1600, 1551, 1497, 1479, 1458, 1448, 1298, 1265, 1216, 1192, 1102, 1033, 1007, 884, 839, 755, 690, 655, 538, 524, 495; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.76 (br s, 1H), 8.00–7.94 (m, 1H), 7.70 (app dt, *J* = 8.8, 1.7 Hz, 2H), 7.64–7.57 (m, 1H), 7.43–7.37 (comp, 2H), 7.31–7.26 (m, 1H), 7.23–7.16 (comp, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 188.0, 162.0 (d, J_{C-F} = 259.1 Hz), 158.4, 136.6, 135.6 (d, J_{C-F} = 9.1 Hz), 132.3 (d, J_{C-F} = 1.4 Hz), 129.4, 125.6, 124.3 (d, J_{C-F} = 3.8 Hz), 122.7 (d, J_{C-F} = 11.3 Hz), 120.0, 116.8 (d, J_{C-F} = 21.5 Hz); *m/z* (ESI–MS) 244.1 [M + H]$^+$.

***N*-butyl-2-oxo-2-phenylacetamide (1j):** Following the general procedure A, product **1j** was obtained from 2-oxo-2-phenylacetic acid and butan-1-amine as a colorless oil in 41% yield (336 mg). R_f = 0.54 in hexane/EtOAc 80:20 v/v; IR (KBr) 3313, 3070, 2960, 2933, 2873, 1667, 1597, 1526, 1449, 1376, 1287, 1220, 1178, 1068, 998, 920, 835, 747, 688, 672; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.30–8.26 (comp, 2H), 7.62–7.54 (m, 1H), 7.49–7.36 (comp, 2H), 7.21 (br s, 1H), 3.35 (td, *J* = 7.2, 6.1 Hz, 2H), 1.55 (tt, *J* = 7.7, 6.3 Hz, 2H), 1.36 (tq, *J* = 7.6, 7.4 Hz, 2H), 0.91 (t, *J* = 7.4 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 188.1, 162.0, 134.3, 133.4, 131.1, 128.4, 39.2, 31.3, 20.1, 13.7; *m/z* (ESI–MS) 266.2 [M + H]$^+$.

***N*-cyclohexyl-2-oxo-2-phenylacetamide (1k):** Following the general procedure A, product **1k** was obtained from 2-oxo-2-phenylacetic acid and cyclohexanamine as a white solid in 48% yield (446 mg). R_f = 0.57 in hexane/EtOAc 80:20 v/v; mp = 112–114 °C; IR (KBr) 3287, 2933, 2854, 1685, 1648, 1596, 1546, 1450, 1247, 1216, 1179, 1153, 1089, 1008, 905, 835, 750, 693, 670, 548, 440; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.32 (app d, *J* = 7.7 Hz, 2H), 7.60 (app t, *J* = 7.5 Hz, 1H), 7.46 (app t, *J* = 7.7 Hz, 2H), 6.98 (br s, 1H), 3.91–3.77 (m, 1H), 2.05–1.91 (comp, 2H), 1.83–1.69 (comp, 2H), 1.68–1.58 (m, 1H), 1.47–1.34 (comp, 2H), 1.33–1.12 (comp, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 188.2, 161.0, 134.4, 133.6, 131.3, 128.5, 48.6, 32.8, 25.5, 24.8; *m/z* (ESI–MS) 232.1 [M + H]$^+$.

2-Oxo-*N*-(*p*-tolyl)propanamide (1m): Following the general procedure A, product **1m** was obtained from pyruvic acid and *p*-toluidine as a white solid in 58% yield (411 mg). R_f = 0.61 in hexane/EtOAc 75:25 v/v; mp = 110–112 °C; IR (KBr) 3335, 2914, 1726, 1701, 1685, 1613, 1592, 1542, 1515, 1413, 1354, 1320, 1258, 1139, 1014, 976, 818, 733, 692, 607, 520, 493, 429; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.68 (br s, 1H), 7.53–7.46 (comp, 2H), 7.20–7.14 (comp, 2H), 2.56 (s, 3H), 2.33 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 197.6, 157.6, 135.2, 133.9, 129.8, 119.8, 24.2, 21.1.; *m/z* (ESI–MS) 178.3 [M + H]$^+$.

(*S*)-2-Oxo-2-phenyl-*N*-(1-phenylethyl)acetamide (1o): Following the general procedure A, product **1o** was obtained from 2-oxo-2-phenylacetic acid and (*S*)-1-phenylethan-1-amine as a white solid in 55% yield (552 mg). R_f = 0.60 in hexane/EtOAc 75:25 v/v; mp = 112–113 °C; IR (KBr) 3262, 2983, 2360, 2342, 1685, 1636, 1596, 1561, 1491, 1448, 1221, 1173, 1003, 853, 758, 699, 669; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.36–8.34 (m, 1H), 8.34–8.33 (m, 1H), 7.66–7.59 (m, 1H),

7.51–7.44 (comp, 2H), 7.41–7.36 (comp, 4H), 7.36–8.33 (m, 1H), 7.32–7.28 (m, 1H), 5.24–5.16 (m, 1H), 1.61 (d, J = 7.0 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 187.8, 160.9, 142.4, 134.5, 133.5, 131.4, 129.0, 128.6, 127.8, 126.3, 49.3, 21.8; *m/z* (ESI–MS) 253.9 $[M + H]^+$.

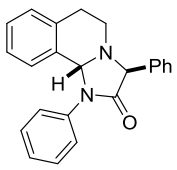
General Procedure B for the redox-annulation of α-ketoamides with secondary amines:

To a solution of the α-ketoamide **1** (0.5 mmol, 1 equiv), benzoic acid (12 mg, 0.1 mmol, 0.2 equiv) in toluene (0.1 M, 5 mL) was added the secondary amine (0.75 mmol, 1.5 equiv). The resulting mixture was heated under reflux until **1** was consumed as judged by TLC. The reaction mixture was then allowed to cool to room temperature, diluted with EtOAc (15 mL), and washed with saturated $NaHCO_3$ (2 x 20 mL). The combined aqueous layers were extracted with EtOAc (2 x 15 mL), and the combined organic layers washed with brine (40 mL) and dried over anhydrous Na_2SO_4. Solvent was then removed under reduced pressure and the residue purified by silica gel chromatography.

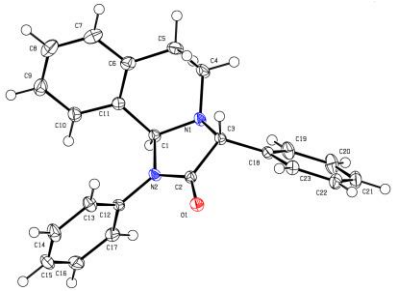
General Procedure C for the redox-annulation of 2-oxo-*N*,2-diphenylacetamide with piperidine, morpholine, and thiomorpholine:

A 10 mL microwave reaction tube was charged with a 10 x 8 mm SiC passive heating element, 2-oxo-*N*,2-diphenylacetamide (**1a**) (112.5 mg, 0.5 mmol, 1 equiv), benzoic acid (12 mg, 0.1 mmol, 0.2 equiv), toluene (0.25 M, 2 mL) and the corresponding amine (1.5 mmol, 3 equiv). The reaction tube was sealed with a Teflon-lined snap cap, and heated in a microwave reactor at 220 °C (200 W, 70–100 psi) for the indicated time. After cooling with compressed air flow, the reaction mixture was diluted with EtOAc (15 mL), and washed with saturated $NaHCO_3$ (2 x 20 mL). The combined aqueous layers were extracted with EtOAc (2 x 15 mL), and the combined organic layers washed with brine (40 mL), and dried over anhydrous Na_2SO_4. Solvent was then removed under reduced pressure and the residue purified by silica gel chromatography.

(±)-**2a:** To a solution of 2-oxo-*N*,2-diphenylacetamide (450.5 mg, 2 mmol, 1 equiv) and benzoic acid (48.8 mg, 0.4 mmol, 0.2 equiv) in toluene (0.1 M, 20 mL) were added 1,2,3,4-tetrahydroisoquinoline (0.38 mL, 3 mmol, 1.5 equiv). The resulting mixture was heated under reflux for 15 h. The reaction mixture was then allowed to cool to room temperature, diluted with EtOAc (40 mL), and washed with saturated $NaHCO_3$ (2 x 40 mL). The combined aqueous layers were extracted with EtOAc (2 x 40 mL), and the combined organic layers washed with brine (80 mL) and dried over anhydrous Na_2SO_4. Solvent was then removed under reduced pressure and the residue purified by silica gel chromatography. Product (±)-**2a** was obtained as a white solid in 93% yield (632.5 mg). R_f = 0.40 in hexane/EtOAc 75:25 v/v; mp = 159–162 °C; IR (KBr) 3067, 2939, 2919, 2859, 1702, 1490, 1411, 1279, 1146, 744, 698; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.60–7.56 (comp, 2H), 7.47–7.39 (comp, 6H), 7.38–7.33 (m, 1H), 7.30–7.26 (m, 1H), 7.25–7.20 (comp, 2H), 7.01 (app td, J = 7.4, 1.7 Hz, 1H), 6.76 (app d, J = 7.8 Hz, 1H), 6.28 (s, 1H), 4.57 (s, 1H), 3.34 (ddd, J = 12.9, 8.3, 4.4 Hz, 1H), 3.18–3.04 (comp, 2H), 2.76 (app dt, J = 15.8, 4.8 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 170.9, 137.4, 136.4, 135.5, 133.5, 129.4, 128.9, 128.7, 128.4, 128.3, 128.2, 127.3, 126.5, 126.4, 124.8, 74.6, 67.3, 45.0, 24.5; *m/z* (ESI–MS) 341.2 $[M + H]^+$.



X-ray quality crystals of (±)-**2a** were obtained from hexane/ethyl acetate through slow diffusion at room temperature.



The requisite CIF has been submitted to the journal and deposited with the CCDC (deposition # 1548697).

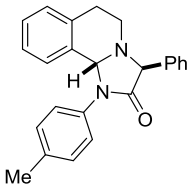
(±)-**2b:** Following the general procedure B, **1b** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 10 h. Product (±)-**2b** was obtained as a pale yellow solid in 88% yield (165 mg). R_f = 0.46 in hexane/EtOAc 75:25 v/v; mp = 138–140 °C; IR (KBr) 3064, 3034, 2929, 1715, 1497, 1453, 1387, 1324, 1153, 1091, 830, 748, 733, 694, 508; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.58–7.54 (comp, 2H), 7.44–7.32 (comp, 7H), 7.28–7.21 (comp, 2H), 7.04 (app td, *J* = 7.8, 1.6 Hz, 1H), 6.75 (d, *J* = 7.8 Hz, 1H), 6.23 (s, 1H), 4.54 (s, 1H), 3.33 (ddd, *J* = 12.8, 8.2, 4.5 Hz, 1H), 3.12 (ddd, *J* = 16.0, 8.2, 4.5 Hz, 1H), 3.05 (ddd, *J* = 12.8, 5.8, 4.6 Hz, 1H), 2.77 (app dt, *J* = 16.0, 5.1 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.0, 136.2, 136.1, 135.7, 133.2, 131.9, 129.5, 129.0, 128.8, 128.42, 128.37, 128.3, 127.2, 126.5, 125.9, 74.6, 67.4, 45.2, 24.6; *m/z* (ESI–MS) (^{35}Cl) 375.1 [M + H]$^+$, (^{37}Cl) 377.2 [M + H]$^+$.

(±)-**2c:** Following the general procedure B, **1c** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 25 h. Product (±)-**2c** was obtained as a yellow solid in 86% yield (158 mg). R_f = 0.31 in hexane/EtOAc 75:25 v/v; mp = 164–167 °C; IR (KBr) 3062, 3027, 2929, 2849, 2224, 1724, 1602, 1510, 1455, 1383, 1304, 1181, 1156, 841, 749, 696, 633, 550; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.78–7.74 (comp, 2H), 7.72–7.68 (comp, 2H), 7.52–7.48 (comp, 2H), 7.43–7.39 (comp, 2H), 7.38–7.34 (m, 1H), 7.29 (app t, *J* = 7.4 Hz, 1H), 7.24 (d, *J* = 6.9 Hz, 1H), 7.10 (app td, *J* = 7.6, 1.2 Hz, 1H), 6.83 (d, *J* = 7.7 Hz, 1H), 6.30 (s, 1H), 4.42 (s, 1H), 3.42 (ddd, *J* = 12.5, 7.8, 4.6 Hz, 1H), 3.08 (ddd, *J* = 16.1, 7.8, 4.5 Hz, 1H), 2.88 (ddd, *J* = 12.4, 6.8, 4.5 Hz, 1H), 2.79 (ddd, *J* = 16.2, 6.6, 4.8 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.3, 142.0, 136.0, 135.8, 133.4, 133.2, 128.8, 128.68, 128.66, 128.4, 126.9, 126.2, 122.5, 118.7, 110.2, 108.8, 73.5, 67.8, 45.2, 24.8; *m/z* (ESI–MS) 366.1 [M + H]$^+$.

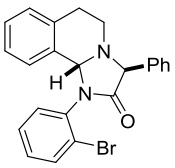
(±)-**2d:** Following the general procedure B, **1d** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 14 h. Product (±)-**2d** was obtained as a yellow solid in 85% yield (157 mg). R_f = 0.23 in hexane/EtOAc 75:25 v/v; mp = 140–142 °C; IR (KBr) 3064, 3032, 2999, 2964, 2839, 1701, 1513, 1455, 1401, 1295, 1251, 1217, 1155, 1030, 835, 748, 706, 588; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.62–7.58 (comp, 2H), 7.43–7.39 (comp, 2H), 7.36–7.32 (m, 1H), 7.24–7.18 (comp, 4H), 7.01–6.96 (m, 1H), 6.93–6.90 (comp, 2H), 6.67 (d, *J* = 7.8 Hz, 1H), 6.14 (s, 1H), 4.60 (s, 1H), 3.82 (s, 3H), 3.27 (ddd, *J* = 14.2, 9.1, 4.3 Hz, 1H), 3.17–3.09 (comp, 2H), 2.79–2.74 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.2, 158.3, 136.6, 135.5, 133.2, 129.9,

129.0, 128.7, 128.3, 128.24, 128.16, 127.9, 127.3, 126.2, 114.7, 75.4, 67.3, 55.6, 45.3, 24.8.; *m/z* (ESI–MS) 371.2 $[M + H]^+$.

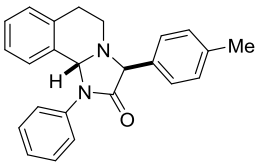
(±)-**2e:** Following the general procedure B, **1e** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 8 h. Product (±)-**2e** was obtained as a yellow solid in 92% yield (163 mg). R_f = 0.46 in hexane/EtOAc 75:25 v/v; mp = 150–153 °C; IR (KBr) 3062, 3024, 2937, 2835, 1702, 1601, 1516, 1451, 1404, 1213, 1147, 1020, 823, 747, 727, 692, 507; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.62–7.55 (comp, 2H), 7.45–7.39 (comp, 2H), 7.37–7.32 (m, 1H), 7.31–7.15 (comp, 6H), 7.01 (app td, *J* = 7.7, 1.9 Hz, 1H), 6.76 (d, *J* = 7.7 Hz, 1H), 6.23 (s, 1H), 4.58 (s, 1H), 3.35–3.25 (m, 1H), 3.18–3.05 (comp, 2H), 2.80–2.69 (m, 1H), 2.38 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 170.9, 136.6, 136.4, 135.5, 134.7, 133.5, 130.0, 128.8, 128.7, 128.4, 128.2, 128.1, 127.5, 126.3, 125.0, 74.8, 67.3, 45.1, 24.6, 21.2; *m/z* (ESI–MS) 355.2 $[M + H]^+$.



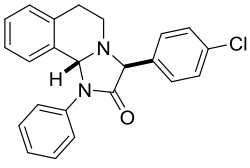
(±)-**2f:** Following the general procedure B, **1f** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 36 h. Product (±)-**2f** was obtained as a yellow solid in 70% yield (147 mg). R_f = 0.28 in hexane/EtOAc 75:25 v/v; mp = 60–62 °C; IR (KBr) 3064, 3029, 2929, 2837, 1713, 1586, 1477, 1450, 1399, 1347, 1213, 1153, 1070, 902, 823, 749, 731, 698; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.77–7.75 (comp, 2H), 7.65–7.63 (m, 1H), 7.44–7.41 (comp, 2H), 7.36–7.33 (m, 1H), 7.27–7.19 (comp, 4H), 6.93–6.89 (comp, 2H), 6.39 (d, *J* = 7.7 Hz, 1H), 6.19 (s, 1H), 4.69 (s, 1H), 3.37–3.35 (m, 1H), 3.22 (ddd, *J* = 12.3, 6.8, 4.3 Hz, 1H), 3.12 (ddd, *J* = 16.3, 6.8, 4.0 Hz, 1H), 2.94 (ddd, *J* = 16.3, 7.4, 4.4 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.9, 135.9, 135.8, 135.3, 133.6, 132.3, 131.6, 131.0, 130.0, 129.0, 128.7, 128.6, 128.31, 128.27, 128.0, 127.9, 125.8, 75.2, 68.3, 46.6, 26.2; *m/z* (ESI–MS) (^{79}Br) 419.0 $[M + H]^+$, (^{81}Br) 421.1 $[M + H]^+$.



(±)-**2g:** Following the general procedure B, **1g** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 14 h. Product (±)-**2g** was obtained as a yellow solid in 96% yield (160 mg). R_f = 0.48 in hexane/EtOAc 75:25 v/v; mp = 120–123 °C; IR (KBr) 3062, 3039, 2922, 2889, 1716, 1593, 1497, 1453, 1396, 1297, 1155, 1123, 810, 749, 730, 690, 503; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.46–7.36 (comp, 6H), 7.29–7.26 (m, 1H), 7.25–7.19 (comp, 4H), 7.00 (app td, *J* = 7.4, 1.7 Hz, 1H), 6.74 (d, *J* = 7.8 Hz, 1H), 6.25 (s, 1H), 4.51 (s, 1H), 3.32 (ddd, *J* = 12.8, 8.3, 4.4 Hz, 1H), 3.15–3.03 (comp, 2H), 2.75 (app dt, *J* = 15.9, 5.0 Hz, 1H), 2.37 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.2, 138.1, 137.6, 135.6, 133.6, 133.5, 129.5, 129.4, 128.8, 128.4, 128.2, 127.4, 126.45, 126.38, 124.8, 74.6, 67.2, 45.1, 24.6, 21.4; *m/z* (ESI–MS) 355.3 $[M + H]^+$.



(±)-**2h:** Following the general procedure B, **1h** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 13 h. Product (±)-**2h** was obtained as a light yellow solid in 97% yield (181 mg). R_f = 0.50 in hexane/EtOAc 75:25 v/v; mp = 125–127 °C; IR (KBr) 3064, 3039, 2932, 2867, 1702, 1596, 1499, 1389, 1296, 1204, 1156, 1085, 818, 742, 690; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.54–7.50 (comp, 2H), 7.43–7.40 (comp, 4H), 7.40–7.36 (comp, 2H), 7.30–7.25 (m, 1H), 7.24–7.19 (comp, 2H), 7.00 (app td, *J* = 7.8, 1.6 Hz, 1H), 6.74 (d, *J* = 7.8 Hz, 1H), 6.25 (s, 1H), 4.53 (s, 1H), 3.32 (ddd, *J* = 13.0, 8.6, 4.4 Hz, 1H), 3.11 (ddd, *J* = 16.1, 8.6, 4.6 Hz, 1H), 3.03 (ddd, *J* = 12.9, 5.6, 4.5 Hz, 1H), 2.75 (app dt, *J* = 16.2, 4.9 Hz, 1H); ^{13}C



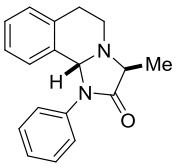
NMR (125 MHz, $CDCl_3$) δ 170.3, 137.2, 135.3, 134.9, 134.0, 133.3, 129.6, 129.3, 129.2, 128.7, 128.1, 127.1, 126.5, 126.3, 124.6, 74.5, 66.5, 45.0, 24.4; *m/z* (ESI–MS) (^{35}Cl) 375.1 $[M + H]^+$, (^{37}Cl) 377.1 $[M + H]^+$.

(±)-**2i:** Following the general procedure B, **1i** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 17 h. Product (±)-**2i** was obtained as a yellow solid in 80% yield (144 mg). R_f = 0.41 in hexane/EtOAc 75:25 v/v; mp = 58–61 °C; IR (KBr) 3062, 2924, 2844, 1717, 1596, 1492, 1458, 1391, 1231, 1154, 752, 731, 692; ^1H NMR ($CDCl_3$, 500 MHz) δ 7.58–7.49 (comp, 3H), 7.48–7.42 (comp, 2H), 7.36–7.27 (comp, 2H), 7.25–7.18 (comp, 3H), 7.15–7.09 (m, 1H), 7.04–6.99 (m, 1H), 6.79 (d, *J* = 7.8 Hz, 1H), 6.35 (s, 1H), 4.98 (s, 1H), 3.31 (ddd, *J* = 13.2, 9.9, 3.9 Hz, 1H), 3.22 (ddd, *J* = 14.7, 9.8, 4.4 Hz, 1H), 3.06 (app dt, *J* = 12.9, 4.4 Hz, 1H), 2.68 (app dt, *J* = 16.3, 4.1 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 170.0, 162.2 (d, *J* = 247.8 Hz), 137.5, 135.4, 133.8, 130.14 (d, *J* = 3.7 Hz), 130.10, 130.0, 129.5, 129.2, 129.0, 128.1, 127.0, 126.5 (d, *J* = 6.7 Hz) 124.6, 124.5 (d, *J* = 3.5 Hz), 124.3 (d, *J* = 12.5 Hz), 115.8 (d, *J* = 21.7 Hz), 74.7, 60.7, 44.7, 23.7; *m/z* (ESI–MS) 359.2 $[M + H]^+$.

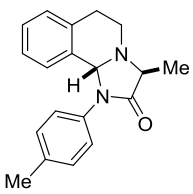
(±)-**2j:** Following the general procedure B, **1j** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 15 h. Product (±)-**2j** was obtained as a yellow solid in 86% yield (137 mg). R_f = 0.40 in hexane/EtOAc 75:25 v/v; mp = 45–48 °C; IR (KBr) 3059, 3024, 2959, 2934, 2871, 2859, 1695, 1596, 1452, 1432, 1343, 1308, 1211, 1148, 749, 729, 696, 568; ^1H NMR ($CDCl_3$, 500 MHz) δ 7.58–7.55 (comp, 2H), 7.41–7.37 (comp, 2H), 7.34–7.30 (comp, 2H), 7.29–7.27 (comp, 2H), 7.23 (d, *J* = 6.9 Hz, 1H), 5.72 (d, *J* = 1.4 Hz, 1H), 4.47 (s, 1H), 3.70 (ddd, *J* = 14.2, 9.1, 6.9 Hz, 1H), 3.20 (ddd, *J* = 14.0, 8.9, 5.0 Hz, 1H), 3.15–3.10 (m, 1H), 3.08–3.02 (comp, 2H), 2.71 (dt, *J* = 6.5, 4.4 Hz, 1H), 1.62–1.55 (m, 1H), 1.50–1.42 (m, 1H), 1.37–1.27 (comp, 2H), 0.91 (t, *J* = 7.4 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.7, 137.4, 136.7, 133.0, 129.3, 128.6, 128.3, 127.9, 127.78, 127.76, 126.4, 73.1, 67.5, 46.7, 40.6, 29.4, 25.4, 20.1, 13.8; *m/z* (ESI–MS) 321.2 $[M + H]^+$.

(±)-**2k:** Following the general procedure B, **1k** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 22 h. Product (±)-**2k** was obtained as a light yellow solid in 72% yield (125 mg). R_f = 0.47 in hexane/EtOAc 75:25 v/v; mp = 99–102 °C; IR (KBr) 3069, 3022, 2938, 2851, 1693, 1606, 1496, 1453, 1367, 1151, 890, 746, 698, 570; ^1H NMR ($CDCl_3$, 500 MHz) δ 7.48 (app d, *J* = 7.5 Hz, 2H), 7.40–7.34 (comp, 3H), 7.33–7.27 (comp, 3H), 7.21 (app d, *J* = 7.6 Hz, 1H), 5.62 (s, 1H), 4.30 (s, 1H), 3.49 (app tt, *J* = 12.0, 3.7 Hz, 1H), 3.16 (ddd, *J* = 12.4, 8.4, 3.9 Hz, 1H), 3.03 (ddd, *J* = 15.7, 8.3, 4.1 Hz, 1H), 2.89 (ddd, *J* = 12.3, 6.0, 4.3 Hz, 1H), 2.68 (ddd, *J* = 15.9, 6.0, 4.0 Hz, 1H), 2.27–2.17 (m, 1H), 2.00–1.92 (m, 1H), 1.86–1.75 (comp, 3H), 1.70–1.56 (comp, 2H), 1.31–1.10 (comp, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 172.4, 137.6, 137.1, 134.5, 128.8, 128.6, 128.3, 128.03, 127.98, 127.2, 126.5, 110.2, 74.4, 68.4, 56.0, 46.9, 29.9, 29.6, 26.4, 26.3, 25.7, 25.4; *m/z* (ESI–MS) 347.3 $[M + H]^+$.

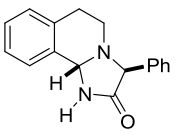
(±)-**2l:** Following the general procedure B, **1l** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 2.5 h. Product (±)-**2l** was obtained as a pale yellow solid in 62% yield (88 mg). R_f = 0.22 in hexane/EtOAc 50:50 v/v; mp = 142–144 °C; IR (KBr) 3057, 2949, 2922, 2812, 1694, 1595, 1500, 1412, 1318, 1221, 1151, 757, 732, 695; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.44–7.36 (comp, 4H), 7.30–7.25 (m, 1H), 7.22–7.14 (comp, 2H), 6.97 (ddd, *J* = 7.8, 7.1, 1.7 Hz, 1H), 6.68 (app d, *J* = 7.8 Hz, 1H), 6.13 (s, 1H), 3.55 (qd, *J* = 6.6, 1.0 Hz, 1H), 3.33 (ddd, *J* = 12.9, 8.6, 4.4 Hz, 1H), 3.13 (ddd, *J* = 12.9, 5.8, 4.5 Hz, 1H), 3.05 (ddd, *J* = 16.4, 8.6, 4.6 Hz, 1H), 2.71 (app dt, *J* = 16.3, 5.1 Hz, 1H), 1.44 (d, *J* = 6.6 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 173.2, 137.4, 135.5, 133.4, 129.4, 128.7, 128.1, 127.3, 126.6, 126.3, 125.0, 74.6, 58.6, 45.2, 24.5, 15.6; *m/z* (ESI–MS) 279.1 $[M + H]^+$.



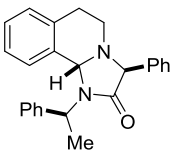
(±)-**2m**: Following the general procedure B, **1m** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 22 h. Product (±)-**2m** was obtained as a yellow solid in 71% yield (103 mg). R_f = 0.23 in hexane/EtOAc 50:50 v/v; mp = 145–147 °C; IR (KBr) 3064, 3034, 2922, 2862, 2812, 1693, 1515, 1453, 1312, 1221, 1152, 843, 820, 752, 728, 615, 503; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.25–7.14 (comp, 6H), 6.97 (ddd, *J* = 7.8, 7.1, 1.7 Hz, 1H), 6.68 (d, *J* = 7.8 Hz, 1H), 6.08 (s, 1H), 3.56 (q, *J* = 6.7 Hz, 1H), 3.30 (ddd, *J* = 12.9, 8.6, 4.3 Hz, 1H), 3.14 (ddd, *J* = 12.9, 5.8, 4.5 Hz, 1H), 3.04 (ddd, *J* = 16.4, 8.6, 4.5 Hz, 1H), 2.70 (ddd, *J* = 16.4, 5.8, 4.4 Hz, 1H), 2.37 (s, 3H), 1.43 (d, *J* = 6.7 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 173.2, 136.5, 135.5, 134.7, 133.4, 130.0, 128.7, 128.0, 127.6, 126.2, 125.2, 74.8, 58.6, 45.2, 24.5, 21.2, 15.6; *m/z* (ESI–MS) 293.2 $[M + H]^+$.



(±)-**2n:** Following the general procedure B, **1n** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 3 h. Product (±)-**2n** was obtained as a yellow oil in 53% yield (70 mg). R_f = 0.21 in hexane/EtOAc 65:25 v/v; IR (KBr) 3216, 3062, 2924, 2862, 1708, 1509, 1452, 1283, 1153, 1116, 1068, 746, 701, 605; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.61–7.53 (comp, 2H), 7.45–7.37 (comp, 2H), 7.38–7.31 (m, 1H), 7.30–7.26 (comp, 2H), 7.22–7.15 (comp, 2H), 6.95 (br s, 1H), 5.81 (s, 1H), 4.41 (s, 1H), 3.20 (ddd, *J* = 14.3, 10.0, 4.0 Hz, 1H), 3.15–3.02 (comp, 2H), 2.64 (app dt, *J* = 16.4, 4.4 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 174.8, 136.8, 135.2, 134.5, 129.1, 128.8, 128.3, 128.2, 127.8, 127.2, 126.8, 69.6, 65.4, 44.3, 23.9; *m/z* (ESI–MS) 265.2 $[M + H]^+$.



2o: Following the general procedure B, **1o** and 1,2,3,4-tetrahydroisoquinoline were heated at reflux for 36 h. Product **2o** was obtained as a yellow solid in 86% yield (158 mg) (1.3:1 mixture of diastereomers). R_f = 0.43 in hexane/EtOAc 75:25 v/v; IR (KBr) 3061, 3028, 2934, 2872, 1697, 1606, 1494, 1452, 1407, 1298, 1151, 750, 698; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.63–7.59 (comp, 2.01H), 7.53–7.48 (comp, 1.46H), 7.44–7.21 (comp, 13.89H), 7.17 (app d, *J* = 7.5 Hz, 1.74H), 7.15–7.11 (comp, 3.69H), 7.07 (app d, *J* = 7.6 Hz, 0.73H), 6.87 (app d, *J* = 7.6 Hz, 1.01H), 5.67 (s, 0.73H), 5.60 (s, 1.00H), 5.30 (q, *J* = 7.2 Hz, 1.19H), 4.95 (q, *J* = 7.3 Hz, 0.88H), 4.45 (s, 1.01H), 4.40 (s, 0.74H), 3.18–3.06 (comp, 2.01H), 3.05–2.91 (comp, 3.79H), 2.75–2.66 (comp, 1.95H), 1.87 (d, *J* = 7.3 Hz, 2.20 H), 1.43 (d, *J* = 7.2 Hz, 3.01 H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 173.2, 173.0, 141.6, 140.0, 137.7, 137.2, 137.1, 133.2, 132.8, 129.2, 128.8, 128.68, 128.67, 128.63, 128.58,



128.5, 128.3, 128.02, 127.96, 127.9, 127.6, 127.4, 127.32, 127.29, 126.8, 126.3, 126.1, 74.2, 74.0, 69.1, 68.5, 54.0, 51.4, 48.5, 47.2, 26.6, 26.0, 18.9, 17.6; *m/z* (ESI–MS) 369.2 $[M + H]^+$.

(±)-**3a:** Following the general procedure B, **1a** and 6,7-dimethoxy-1,2,3,4-tetrahydroisoquinoline were heated at reflux for 17 h. Product (±)-**3a** was obtained as a white solid in 84% yield (168 mg). R_f = 0.25 in hexane/EtOAc 65:35 v/v; mp = 182–184 °C; IR (KBr) 3064, 3009, 2957, 2929, 2917, 2844, 2829, 2784, 1694, 1518, 1505, 1495, 1452, 1358, 1255, 1206, 1133, 1094, 754, 696, 509; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.61–7.59 (comp, 2H), 7.44–7.40 (comp, 4H), 7.38–7.33 (comp, 3H), 7.29–7.26 (m, 1H), 6.64 (s, 1H), 6.21 (s, 1H), 6.09 (s, 1H), 4.64 (s, 1H), 3.86 (s, 3H), 3.30 (s, 3H), 3.25–3.17 (comp, 2H), 3.10–3.04 (m, 1H), 2.63 (app dt, *J* = 16.3, 4.4 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 170.8, 148.8, 147.1, 137.3, 136.5, 129.5, 128.7, 128.3, 127.3, 126.9, 126.3, 111.2, 110.2, 66.7, 56.0, 55.3, 44.7, 23.5; *m/z* (ESI–MS) 401.4 $[M + H]^+$.

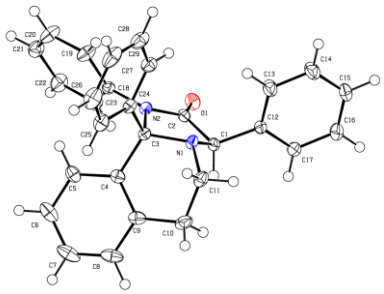
(±)-**3b:** Following the general procedure B, **1a** and 2,3,4,9-tetrahydro-1*H*-pyrido[3,4-*b*]indole were heated at reflux for 13 h. Product (±)-**3b** was obtained as a yellow solid in 91% yield (173 mg). R_f = 0.39 in hexane/EtOAc 75:25 v/v; mp = 210–213 °C; IR (KBr) 3375, 3059, 3027, 2957, 2925, 2839, 1710, 1596, 1492, 1456, 1384, 1293, 1195, 1137, 747, 695; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.79–7.76 (comp, 2H), 7.59–7.50 (comp, 5H), 7.46–7.41 (comp, 2H), 7.40–7.36 (comp, 2H), 7.35–7.31 (m, 1H), 7.21–7.17 (comp, 2H), 7.16–7.13 (m, 1H), 6.59 (d, *J* = 1.2 Hz, 1H), 4.69 (s, 1H), 3.37 (ddd, *J* = 14.1, 5.5, 1.2 Hz, 1H), 3.31 (ddd, *J* = 14.5, 11.1, 4.4 Hz, 1H), 3.12 (dddd, *J* = 16.5, 11.0, 5.6, 1.5 Hz, 1H), 2.73–2.67 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 170.1, 137.3, 136.5, 136.2, 130.8, 130.0, 128.8, 128.7, 128.5, 126.23, 126.16, 123.2, 122.0, 120.1, 119.0, 111.4, 110.5, 70.8, 65.0, 43.2, 15.8; *m/z* (ESI–MS) 380.2 $[M + H]^+$.

(±)-**3c:** Following the general procedure B, **1a** and 2,3-dihydro-1H-benzo[*de*]isoquinoline were heated at reflux for 48 h. Product (±)-**3c** was obtained as a yellow solid in 45% yield (85 mg). R_f = 0.50 in hexane/EtOAc 75:25 v/v; mp = 84–87 °C; IR (KBr) 3062, 3027, 2932, 2814, 1708, 1597, 1499, 1451, 1383, 1310, 1201, 1125, 908, 745, 503; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.82 (d, *J* = 8.2 Hz, 1H), 7.77 (d, *J* = 8.3 Hz, 1H), 7.71 (dd, *J* = 8.6, 1.0 Hz, 2H), 7.55–7.43 (comp, 5H), 7.42–7.35 (comp, 3H), 7.34–7.27 (comp, 2H), 7.25 (d, *J* = 7.0 Hz, 1H), 7.13 (d, *J* = 7.2 Hz, 1H), 6.81 (s, 1H), 4.61 (d, *J* = 16.8 Hz, 1H), 4.39 (s, 1H), 4.16 (d, *J* = 16.8 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.0, 138.3, 135.9, 132.9, 130.3, 129.6, 129.1, 128.8, 128.59, 128.57, 128.4, 128.1, 126.9, 126.2, 126.1, 125.8, 123.9, 122.9, 74.1, 66.9, 48.0; *m/z* (ESI–MS) 377.3 $[M + H]^+$

(±)-**3d:** Following the general procedure B, **1a** and 1-phenyl-1,2,3,4-tetrahydroisoquinoline were heated at reflux for 38 h. Product (±)-**3d** was obtained as a yellow solid in 89% yield (160 mg). R_f = 0.28 in hexane/EtOAc 80:20 v/v; mp = 209–212 °C; IR (KBr) 3067, 3022, 2927, 2832, 1710, 1598, 1498, 1449, 1366, 1347, 1207, 1128, 925, 749, 695, 507; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.73–7.59 (comp, 4H), 7.44 (dd, *J* = 8.3, 6.9 Hz, 2H), 7.41–7.34 (comp, 4H), 7.28 (app td, *J* = 7.4, 1.3 Hz, 1H), 7.26–7.22 (m, 1H), 7.21–7.14 (m, 3H), 7.01 (app td, *J* = 7.5, 1.7 Hz, 1H), 6.83 (ddd, *J* = 7.7, 1.7, 0.8 Hz, 2H), 6.64 (d, *J* = 7.9 Hz, 1H), 4.76 (s, 1H), 3.29 (ddd, *J* = 17.3, 10.8, 7.3 Hz, 1H), 3.13–2.94

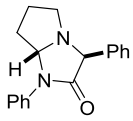
(comp, 2H), 2.55–2.42 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.6, 142.9, 137.0, 136.9, 135.0, 134.2, 130.4, 130.2, 129.8, 129.0, 128.9, 128.71, 128.67, 128.3, 128.04, 128.0, 127.5, 125.2, 110.2, 83.5, 63.8, 39.1, 22.2; *m/z* (ESI–MS) 417.2 $[M + H]^+$.

X-ray quality crystals of (±)-**3d** were obtained from hexane/ethyl acetate through slow diffusion at room temperature.



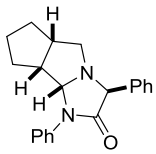
The requisite CIF has been submitted to the journal and deposited with the CCDC (deposition # 1548698).

(±)-**3e:** Following the general procedure B, **1a** and pyrrolidine were heated at reflux for 4 h.



Product (±)-**3e** was obtained as a pale yellow solid in 94% yield (131 mg). R_f = 0.35 in hexane/EtOAc 75:25 v/v; mp = 85–88 °C; IR (KBr) 3062, 2971, 2931, 2812, 1686, 1599, 1500, 1449, 1395, 1310, 1201, 1111, 1028, 878, 756, 740, 692; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.62–7.60 (comp, 2H), 7.52–7.51 (comp, 2H), 7.39–7.35 (comp, 4H), 7.31–7.29 (m, 1H), 7.20–7.16 (m, 1H), 5.55 (m, 1H), 4.56 (s, 1H), 3.43–3.39 (m, 1H), 2.97–2.92 (m, 1H), 2.35–2.29 (m, 1H), 2.00–1.94 (comp, 2H), 1.90–1.85 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.7, 138.2, 137.3, 129.1, 128.6, 127.8, 126.9, 125.2, 121.3, 80.0, 70.9, 55.6, 31.5, 24.2; *m/z* (ESI–MS) 279.1 $[M + H]^+$.

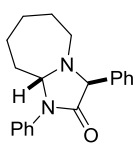
(±)-**3f:** Following the general procedure B, **1a** and octahydrocyclopenta[*c*]pyrrole were heated at reflux for 25 h.



Product (±)-**3f** was obtained as a yellow solid in 83% yield (132 mg). R_f = 0.57 in hexane/EtOAc 75:25 v/v; mp = 124–126 °C; IR (KBr) 3062, 3027, 2950, 2909, 2855, 1689, 1598, 1522, 1500, 1446, 1397, 1363, 1309, 1204, 1105, 1025, 903, 750, 688; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.65–7.61 (comp, 2H), 7.43–7.39 (comp, 2H), 7.39–7.32 (comp, 4H), 7.30–7.26 (m, 1H), 7.18–7.14 (m, 1H), 5.11 (d, *J* = 1.1 Hz, 1H), 4.49 (s, 1H), 3.24–3.16 (comp, 2H), 2.74–2.66 (comp, 2H), 1.97–1.90 (m, 1H), 1.88–1.80 (comp, 2H), 1.77–1.72 (m, 1H), 1.67–1.60 (comp, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 172.2, 137.6, 136.7, 129.0, 128.6, 127.7, 127.0, 125.1, 121.6, 87.8, 69.7, 61.3, 47.4, 42.9, 33.4, 33.1, 27.3; *m/z* (ESI–MS) 319.1 $[M + H]^+$.

(±)-**3g** and (±)-**3g'**:
Following the general procedure B, products (±)-**3g** and (±)-**3g'** were obtained from **1a** and azepane (reflux for 27 h) in a 1.4:1 ratio (126 mg, 82% combined yield).

Characterization data for the major diastereomer (±)-**3g:**



Product (±)-**3g** was obtained as a yellow oil in 48% yield (74 mg). R_f = 0.50 in hexane/EtOAc 75:25 v/v; IR (KBr) 3059, 3029, 2928, 2854, 1705, 1597, 1500, 1400, 1352, 1213, 1148, 1070, 970, 751, 695, 503; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.54–7.52 (comp, 2H), 7.49–7.47 (comp, 2H), 7.42–7.36 (comp, 4H), 7.33–7.30 (m, 1H), 5.44 (app dt, *J* = 7.8, 2.6 Hz, 1H),

4.52 (d, *J* = 2.5 Hz, 1H), 3.10 (dd, *J* = 12.8, 8.2 Hz, 1H), 2.69 (ddd, *J* = 12.8, 8.8, 0.9 Hz, 1H), 1.94–1.89 (m, 1H), 1.84–1.80 (comp, 4H), 1.62–1.56 (m, 1H), 1.53–1.44 (comp, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 169.9, 137.8, 136.8, 129.2, 128.5, 128.3, 128.1, 125.7, 123.1, 79.6, 69.6, 50.1, 30.1, 28.9, 25.8, 25.4; *m/z* (ESI–MS) 307.1 $[M + H]^+$.

Characterization data for the minor diastereomer (±)-**3g':** Product (±)-**3g'** was obtained as a yellow solid in 34% yield (52 mg). R_f = 0.42 in hexane/EtOAc 75:25 v/v; mp = 122–125 °C; IR (KBr) 3057, 3029, 2929, 2857, 1716, 1595, 1495, 1406, 1386, 1356, 1224, 1170, 1025, 993, 759, 694; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.59–7.54 (comp, 2H), 7.48–7.29 (m, 6H), 7.25–7.20 (m, 1H), 4.88 (app dt, *J* = 9.3, 2.0 Hz, 1H), 4.31 (d, *J* = 1.8 Hz, 1H), 3.05 (app dt, *J* = 11.9, 7.3 Hz, 1H), 2.73 (app dt, *J* = 12.0, 6.0 Hz, 1H), 2.24–2.11 (m, 1H), 1.96–1.59 (comp, 6H), 1.57–1.44 (comp, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 169.7, 138.2, 136.5, 129.0, 128.5, 128.08, 128.06, 126.3, 124.6, 78.2, 71.1, 52.3, 35.8, 28.6, 26.2, 25.3; *m/z* (ESI–MS) 307.2 $[M + H]^+$.

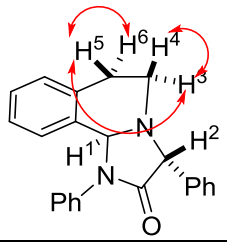
(±)-**3h:** Following the general procedure C, **1a** and piperidine were heated at 220 ºC under microwave irradiation for 0.5 h. Product (±)-**3h** was obtained as a yellow oil in 75% yield (109 mg) (2.9:1 mixture of diastereomers). R_f = 0.40 in hexane/EtOAc 75:25 v/v; IR (KBr) 3056, 3027, 2939, 2857, 1703, 1597, 1501, 1452, 1404, 1319, 1231, 1153, 752, 695; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.56 (dd, *J* = 7.8, 0.9 Hz, 0.49H), 7.50–7.46 (comp, 1.62H), 7.43–7.32 (comp, 5.49H), 7.30–7.25 (comp, 1.77H), 7.24–7.17 (comp, 1.08H), 5.04 (dd, *J* = 9.5, 3.2 Hz, 0.34H), 4.67 (s, 0.32H), 4.29 – 4.25 (m, 1.00H), 4.01 (d, *J* = 2.0 Hz, 0.97H), 3.00–2.92 (comp, 1.25H), 2.81–2.74 (m, 0.36H), 2.44–2.36 (comp, 1.08H), 2.15–2.08 (comp, 1.03H), 2.05–2.00 (m, 0.37H), 1.99–1.92 (comp, 1.04H), 1.89–1.82 (m, 0.38H), 1.71–1.62 (comp, 2.30H), 1.56–1.39 (comp, 2.97H), 1.28–1.24 (m, 0.33H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.3, 136.4, 136.0, 129.3, 129.0, 128.9, 128.8, 128.7, 128.6, 128.3, 126.2, 125.6, 124.4, 122.3, 110.2, 77.5, 74.5, 69.4, 66.4, 48.0, 45.8, 29.4, 28.0, 25.4, 23.2, 22.2, 20.9; *m/z* (ESI–MS) 293.1 $[M + H]^+$.

(±)-**3i:** Following the general procedure C, **1a** and morpholine were heated at 220 ºC under microwave irradiation for 0.5 h. Product (±)-**3i** was obtained as a yellow oil in 69% yield (102 mg) (1.3:1 mixture of diastereomers). R_f = 0.17 in hexane/EtOAc 75:25 v/v; IR (KBr) 3065, 3029, 2962, 2917, 2856, 1719, 1597, 1499, 1395, 1355, 1154, 1105, 970, 908, 754, 728, 696, 510; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.58–7.48 (comp, 5.47H), 7.46–7.28 (comp, 10.41H), 7.26–7.18 (comp, 1.93H), 5.18 (dd, *J* = 8.0, 3.5 Hz, 1.01H), 4.75 (s, 1.00H), 4.54 (app dt, *J* = 9.0, 2.3 Hz, 0.74H), 4.19 (dd, *J* = 10.2, 2.4 Hz, 0.83H), 4.12 (s, 0.74H), 3.99 (dd, *J* = 12.1, 3.5 Hz, 1.19H), 3.92 (ddd, *J* = 11.2, 3.2, 1.2 Hz, 1.00H), 3.82–3.71 (comp, 2.21H), 3.67–3.59 (comp, 2.21H), 3.47 (app t, *J* = 9.6 Hz, 0.92H), 3.24 (ddd, *J* = 13.8, 10.0, 3.6 Hz, 1.26H), 2.94 (app dt, *J* = 14.0, 3.0 Hz, 1.13H), 2.84–2.72 (comp, 1.58H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.0, 170.9, 136.5, 135.7, 135.6, 135.3, 129.4, 129.2, 128.8, 128.69, 128.68, 128.6, 128.4, 128.3, 126.2, 125.9, 123.2, 122.0, 73.5, 70.8, 68.9, 68.6, 67.3, 66.1, 65.8, 62.3, 47.6, 45.7; *m/z* (ESI–MS) 295.1 $[M + H]^+$.

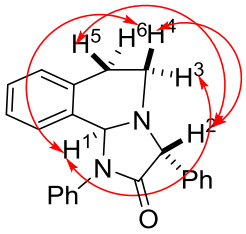
(±)-**3j:** Following the general procedure C, **1a** and thiomorpholine were heated at 220 ºC under microwave irradiation for 1 h. Product (±)-**3j** was obtained as a white solid in 67% yield (104 mg) (1.8:1 mixture of diastereomers). R_f = 0.37 in hexane/EtOAc 75:25 v/v; IR (KBr) 3062, 3032, 2914, 2819, 1719, 1596, 1495, 1394, 1364, 1191, 1121, 756, 743, 695; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.57–7.52 (comp, 2.08H), 7.46 (app dt, *J* = 8.0, 1.8 Hz, 1.15H), 7.44–7.32 (comp, 9.52H), 7.30–7.26 (comp, 1.27H), 7.26–7.21 (comp, 1.80H), 5.25 (ddd, *J* = 9.6, 3.4, 1.5 Hz, 1.08H), 4.73 (app dt, *J* = 9.6, 2.2 Hz, 0.61H), 4.70 (d, *J* = 1.5 Hz, 1.00H), 4.14 (d, *J* = 2.1 Hz, 0.57H), 3.25–3.16 (comp, 1.85H), 3.11–3.03 (comp, 1.24H), 3.03–2.96 (m, 0.70H), 2.91–2.83 (m, 1.85H), 2.80 (app dt, *J* = 12.3, 1.6 Hz, 0.66H), 2.75–2.65 (comp, 3.07H), 2.42 (ddd, *J* = 13.0, 4.0, 2.4 Hz, 0.64H), 2.23–2.17 (comp, 1.19H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 170.2, 169.9, 135.9, 135.5, 129.8, 129.5, 129.3, 129.3, 128.9, 128.81, 128.77, 128.7, 128.6, 126.7, 126.2, 124.4, 122.6, 76.8, 73.9, 69.0, 66.2, 50.1, 47.3, 31.0, 28.6, 27.8, 23.3; *m/z* (ESI–MS) 310.9 $[M + H]^+$.

2D-NMR Analysis for (±)-2a, Selected Interactions (in $CDCl_3$)

GCOSY



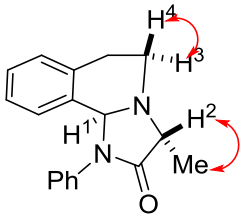
NOESY



Protons	Chemical Shifts (ppm)
H^1	6.28
H^2	4.57
H^3	3.35
H^4, H^5	3.18–3.04
H^6	2.76

2D-NMR Analysis for (±)-2l, Selected Interactions (in $CDCl_3$)

GCOSY



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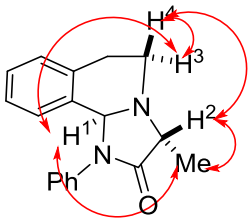
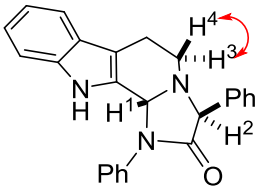


Protons	Chemical Shifts (ppm)
H^1	6.13
H^2	3.55
H^3	3.33
H^4	3.13
Me	1.44

2D-NMR Analysis for (±)-3b, Selected Interactions (in $CDCl_3$)

GCOSY



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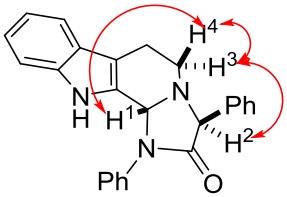
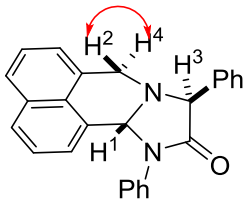


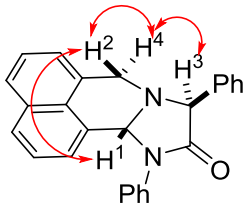
Protons	Chemical Shifts (ppm)
H^1	6.59
H^2	4.69
H^3, H^4	3.37
H^5, H^6	3.31

2D-NMR Analysis for (±)-3c, Selected Interactions (in $CDCl_3$)

GCOSY



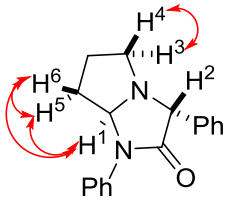
NOESY



Protons	Chemical Shifts (ppm)
H^1	6.82
H^2	4.61
H^3	4.40
H^4	4.17

2D-NMR Analysis for (±)-3e, Selected Interactions (in $CDCl_3$)

GCOSY



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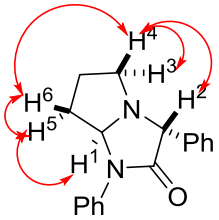
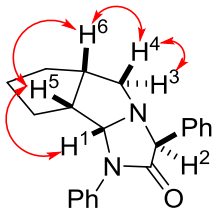


Protons	Chemical Shifts (ppm)
H^1	5.54
H^2	4.57
H^3	3.41
H^4	2.94
H^5	2.32
H^6	1.88

2D-NMR Analysis for (±)-3f, Selected Interactions (in $CDCl_3$)

GCOSY



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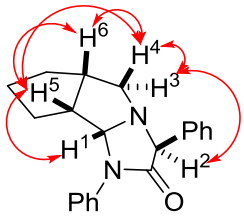
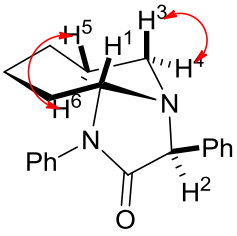


Protons	Chemical Shifts (ppm)
H^1	5.11
H^2	4.49
H^3, H^4	3.24–3.16
H^5, H^6	2.74–2.66

2D-NMR Analysis for (±)-3g, Selected Interactions (in $CDCl_3$)

GCOSY



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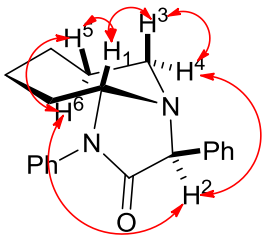
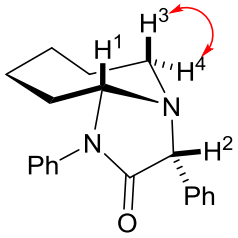


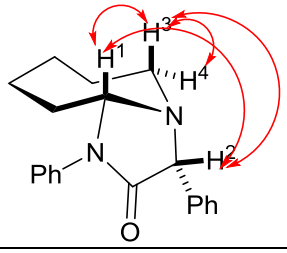
Protons	Chemical Shifts (ppm)
H^1	5.44
H^2	4.54
H^3	3.11
H^4	2.69
H^5	1.96–1.87
H^6	1.53–1.40

2D-NMR Analysis for (±)-3g', Selected Interactions (in $CDCl_3$)

GCOSY



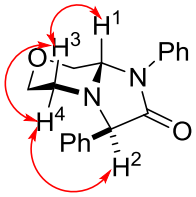
NOESY



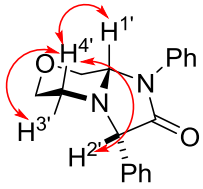
Protons	Chemical Shifts (ppm)
H^1	4.89
H^2	4.32
H^3	3.06
H^4	2.73

2D-NMR Analysis for (±)-3i, Selected Interactions (in $CDCl_3$)

NOSEY for major



NOSEY for minor



Protons assigned to the major product

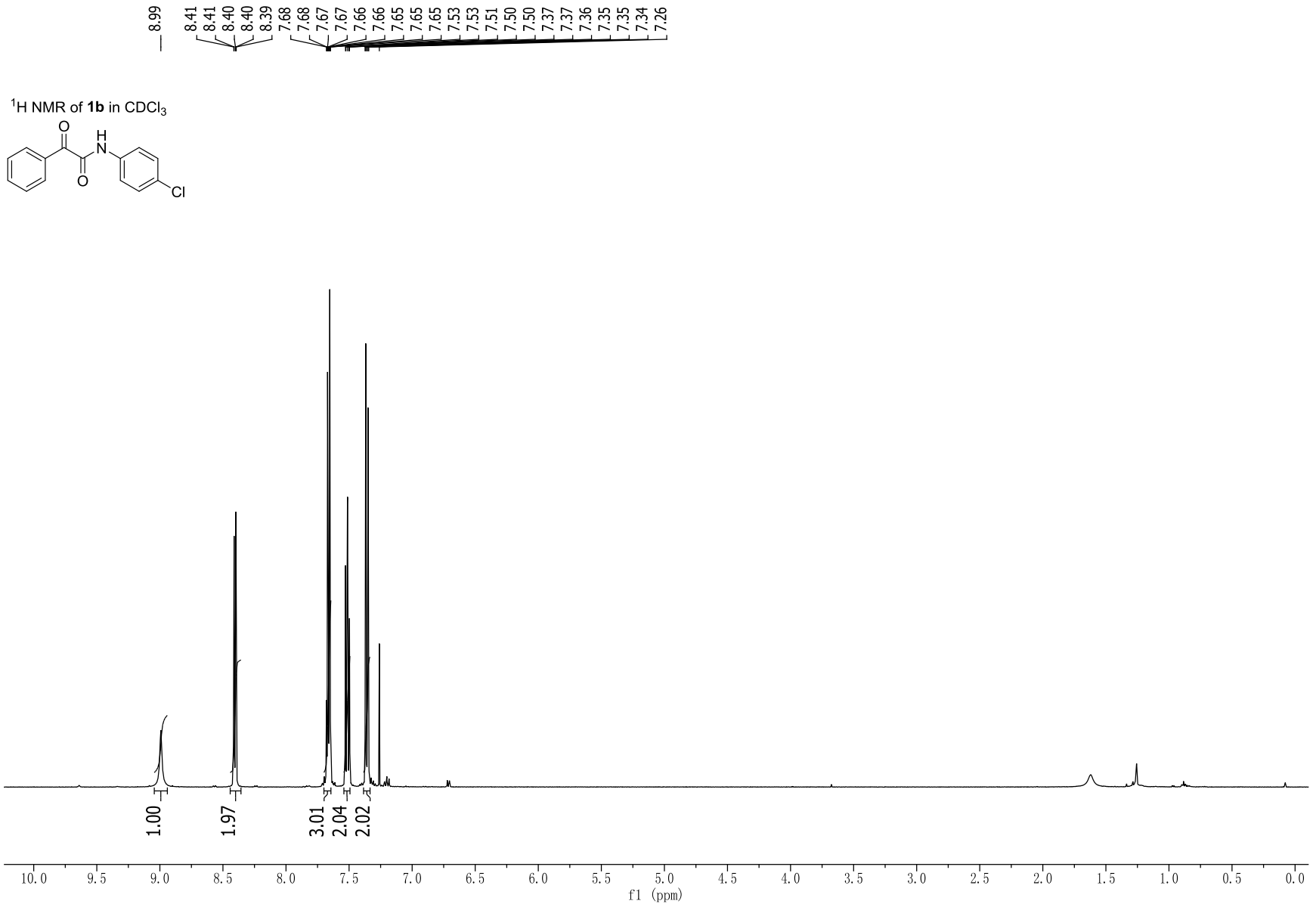
Protons	Chemical Shifts (ppm)
H^1	5.18
H^2	4.76
H^3	3.23
H^4	2.93

Protons assigned to the minor product

Protons	Chemical Shifts (ppm)
$H^{1'}$	4.54
$H^{2'}$	4.13
$H^{3'}$	2.81
$H^{4'}$	2.76

References:

(1) Jiang, L.; Ma, N.; Qiu, J.; Zhang, R. *J Chem Res*, **2013**, *37*, 143.
(2) Zhuang, J.; Wang, C.; Xie, F.; Zhang, W. *Tetrahedron* **2009**, *65*, 9797.
(3) Ishida, N.; Necas, D.; Masuda, Y.; Murakami, M. *Angew. Chem., Int. Ed*. **2015**, *54*, 7418.
(4) Goudedranche, S.; Pierrot, D.; Constantieux, T.; Bonne, D.; Rodriguez, J. *Chem. Commun*. **2014**, *50*, 15605.
(5) Arcus, C.; Prydal, B. *J. Chem. Soc*. **1954**, 4018.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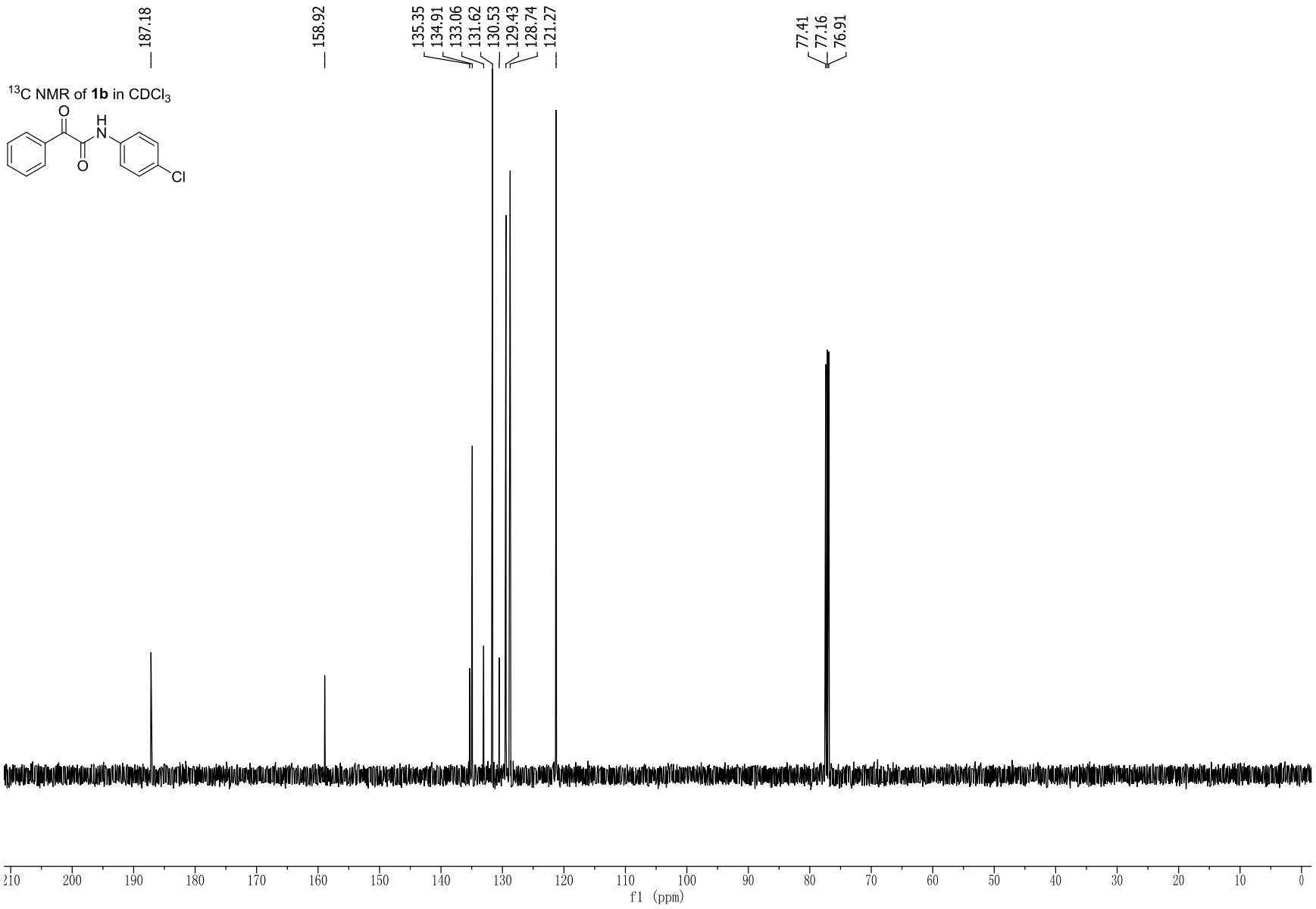
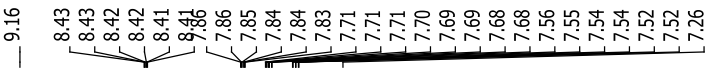
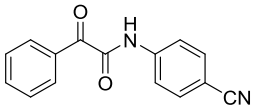
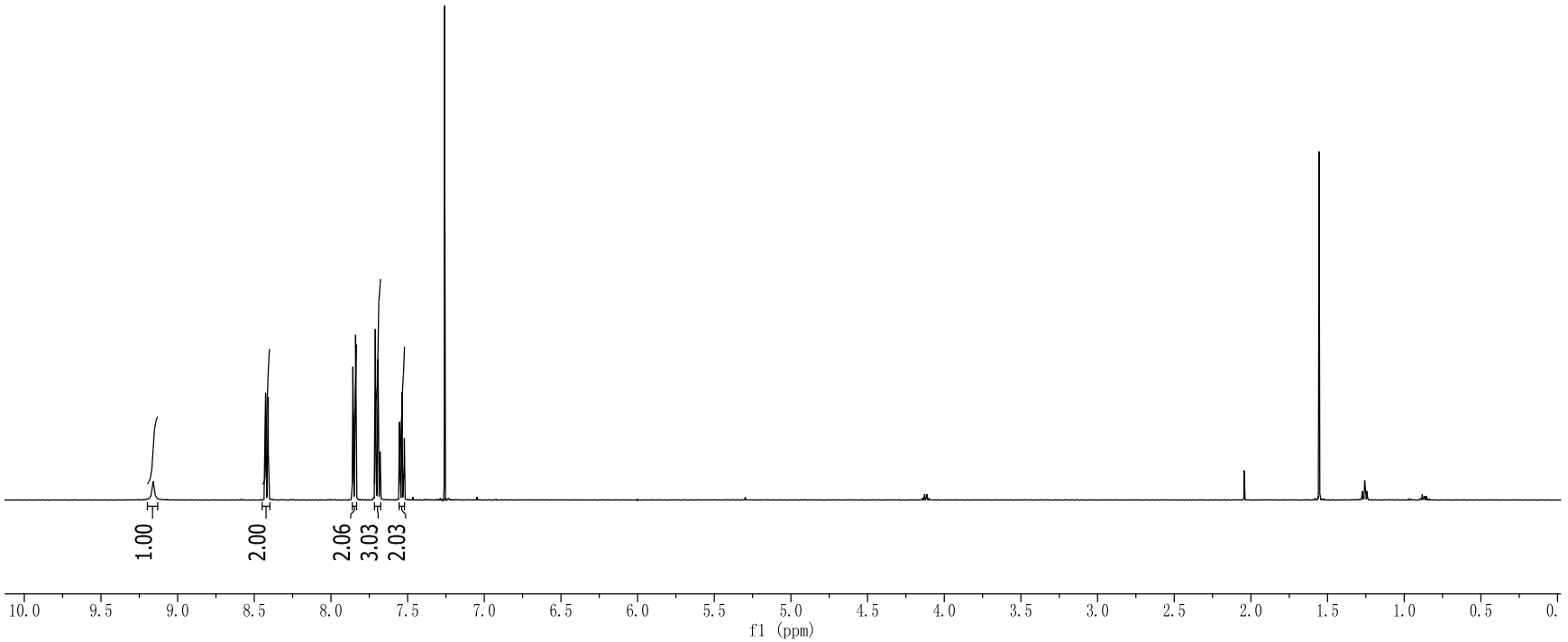
8.41
8.41
8.40
8.40
8.39
7.68
7.68
7.67
7.67
7.66
7.66
7.65
7.65
7.65
7.53
7.53
7.51
7.50
7.50
7.37
7.37
7.36
7.35
7.35
7.34
7.26
1H NMR of 1b in CDCl3
O
H
N
O
Cl
1.00
1.97
3.01
2.04
2.02
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

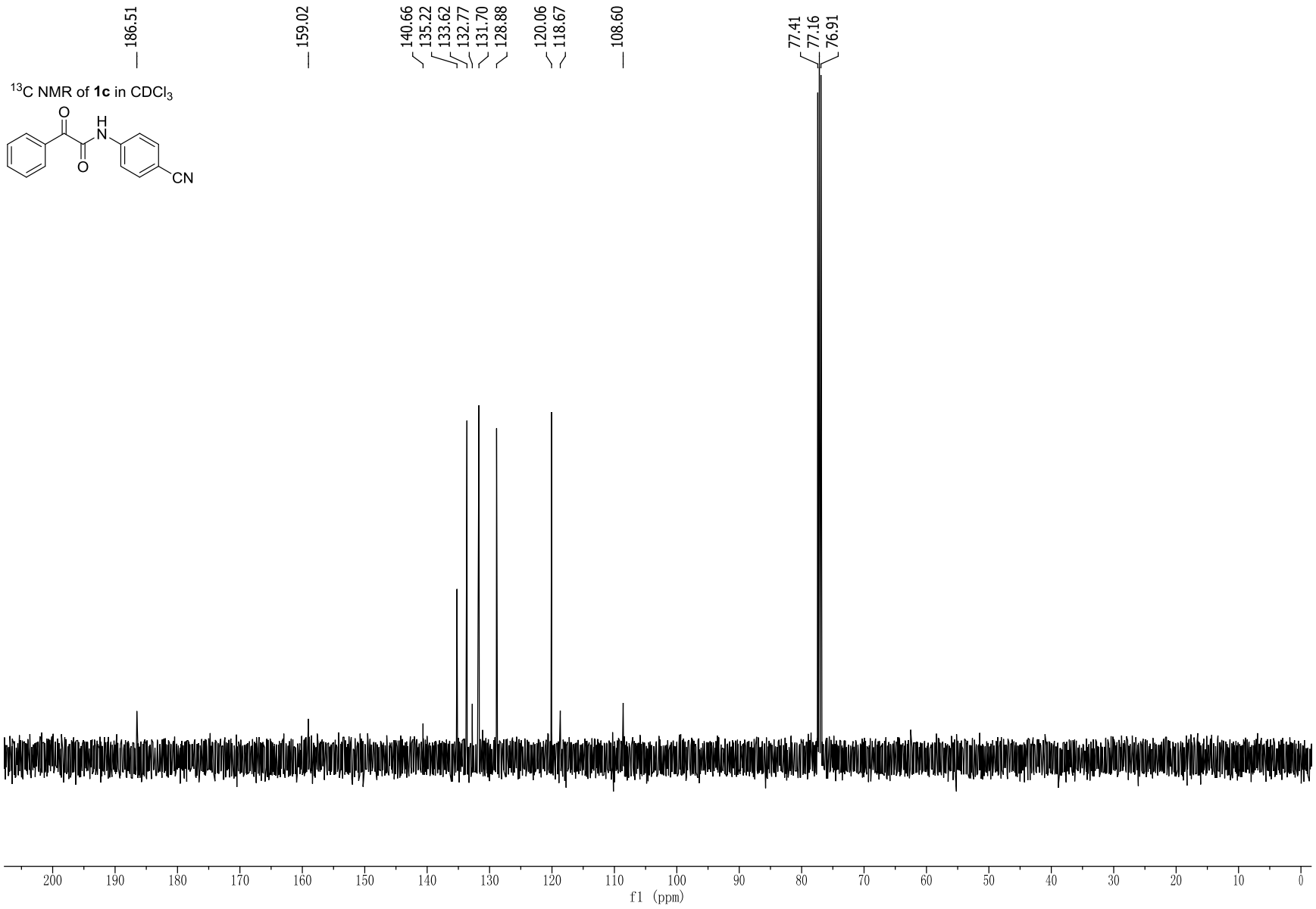

187.18
158.92
135.35
134.91
133.06
131.62
130.53
129.43
128.74
121.27
77.41
77.16
76.91
^{13}C NMR of **1b** in $CDCl_3$
O
H
N
O
Cl
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

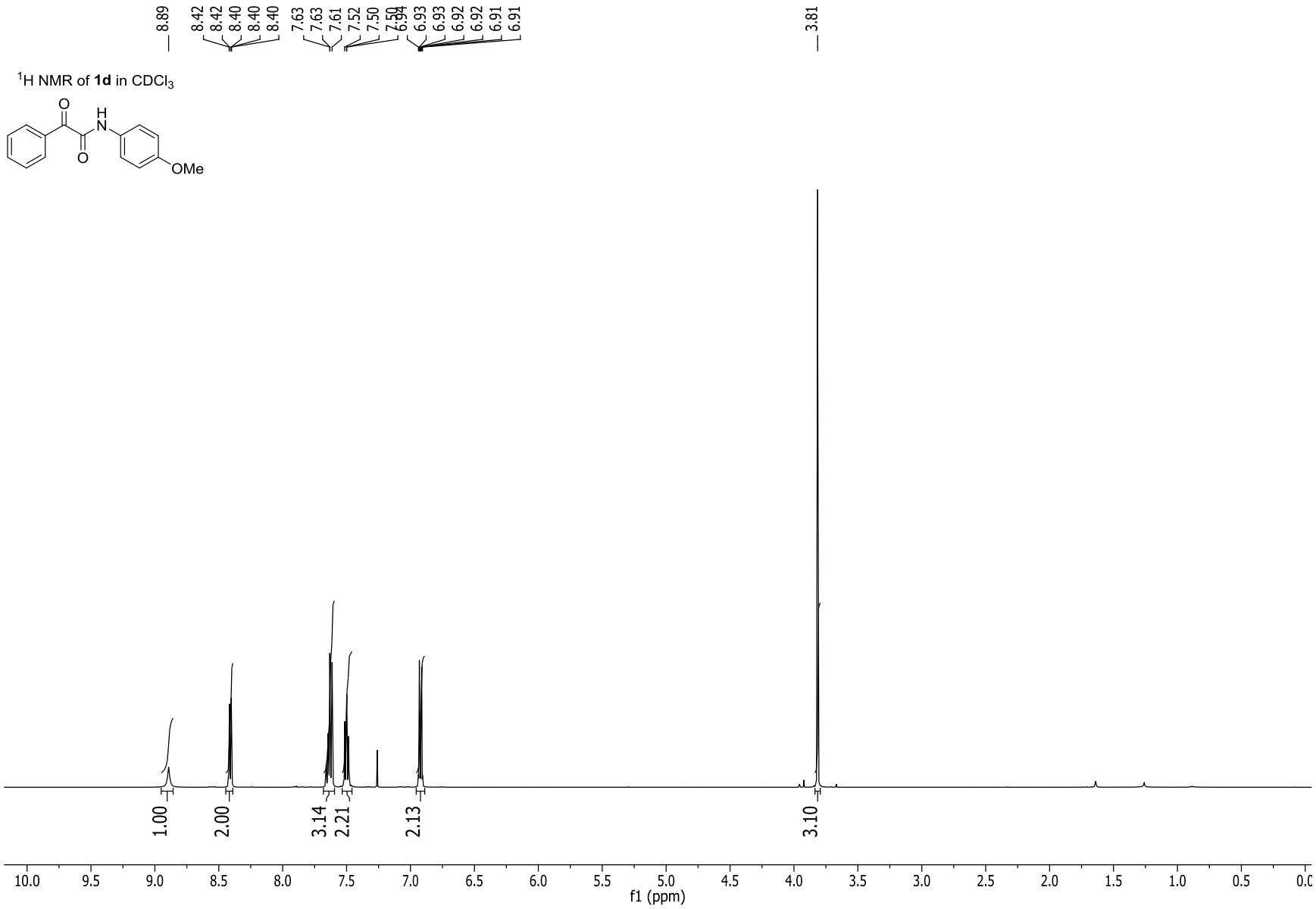


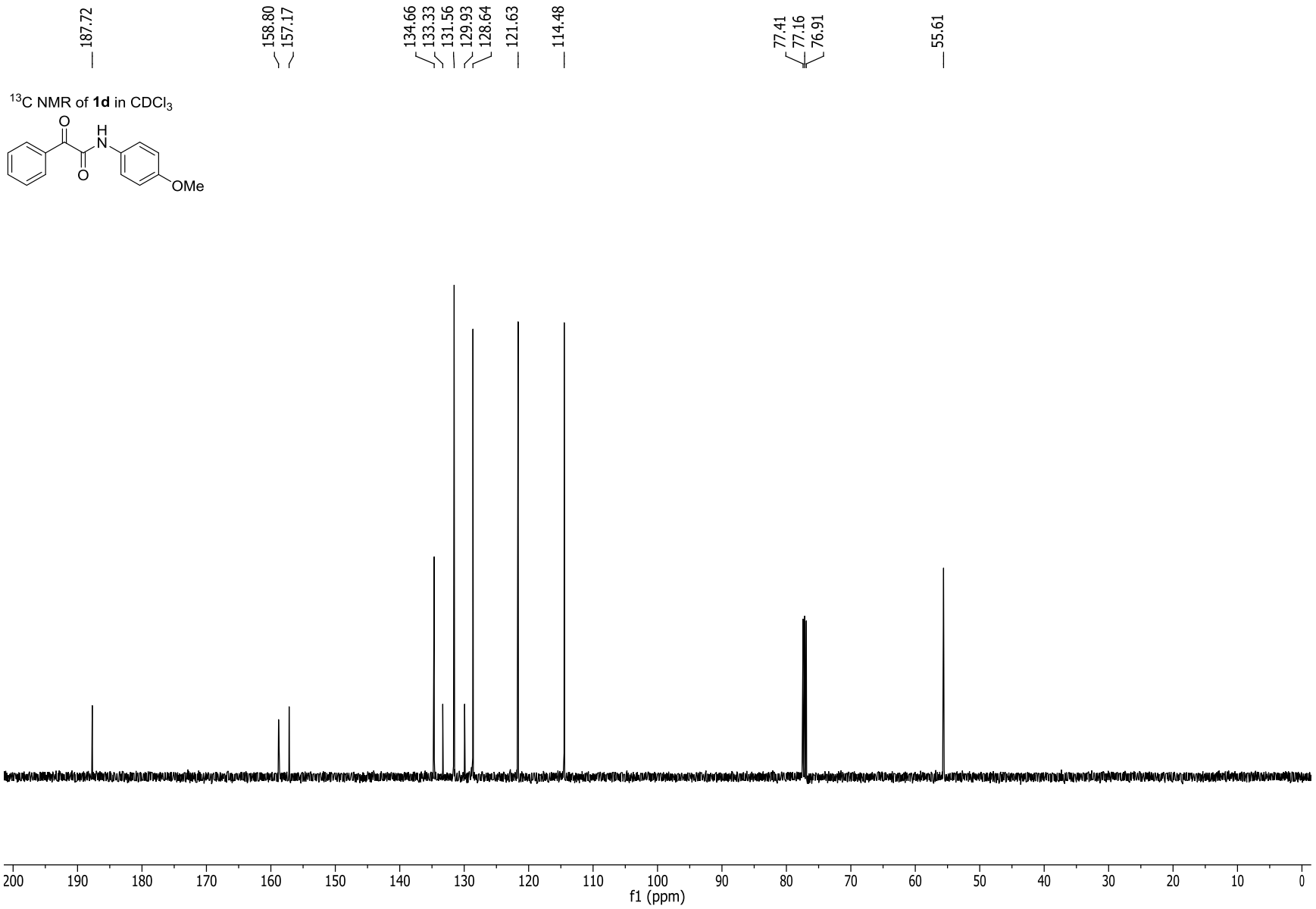
^{1}H NMR of **1c** in $CDCl_3$






186.51
159.02
140.66
135.22
133.62
132.77
131.70
128.88
120.06
118.67
108.60
77.41
77.16
76.91
^{13}C NMR of **1c** in $CDCl_3$
O
H
N
O
CN
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


8.89
8.42
8.42
8.40
8.40
8.40
7.63
7.63
7.61
7.52
7.50
7.50
6.94
6.93
6.93
6.92
6.92
6.91
6.91
3.81
1H NMR of 1d in CDCl3
O
H
N
O
OMe
1.00
2.00
3.14
2.21
2.13
3.10
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)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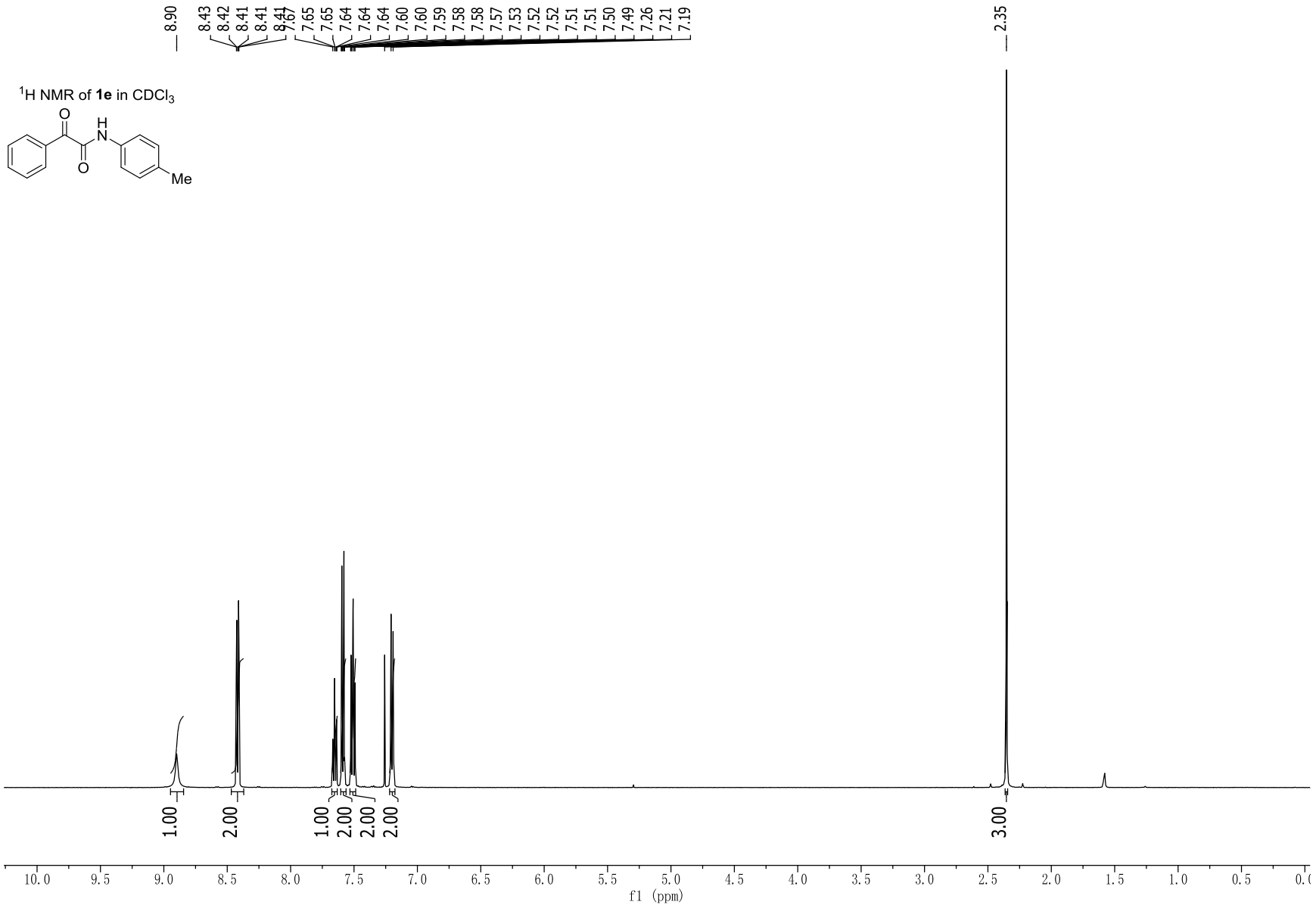
157.17
134.66
133.33
131.56
129.93
128.64
121.63
114.48
77.41
77.16
76.91
55.61
13C NMR of 1d in CDCl3
O
H
N
O
OMe
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)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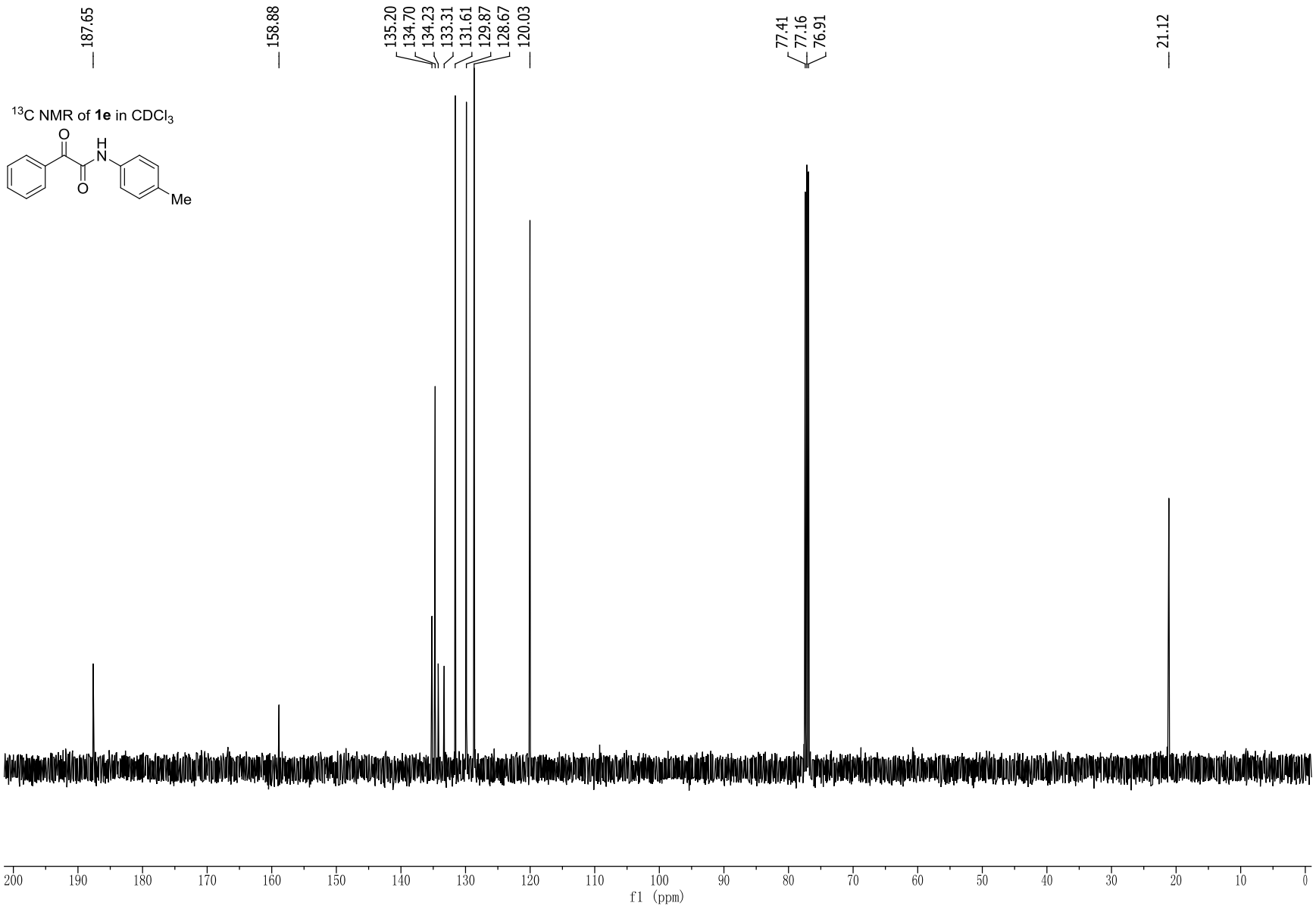
8.41
8.41
7.67
7.65
7.65
7.64
7.64
7.64
7.60
7.60
7.59
7.58
7.58
7.57
7.53
7.52
7.52
7.51
7.51
7.50
7.49
7.26
7.21
7.19
2.35
1H NMR of 1e in CDCl3
O
H
N
O
Me
1.00
2.00
1.00
2.00
2.00
2.00
3.00
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
f1 (ppm)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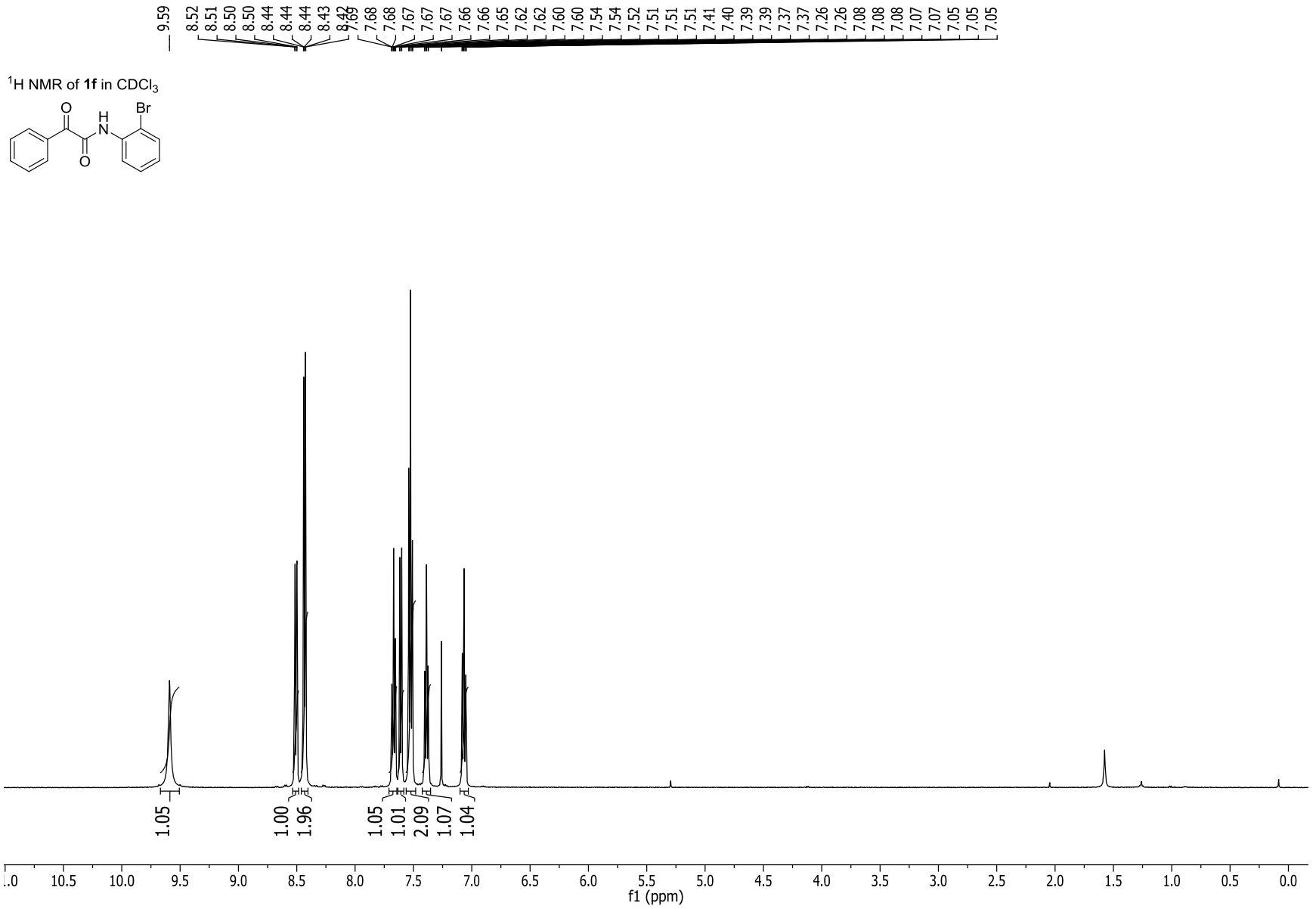
135.20
134.70
134.23
133.31
131.61
129.87
128.67
120.03
77.41
77.16
76.91
21.12
13C NMR of 1e in CDCl3
O
H
N
O
Me
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)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8.50
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8.44
8.44
8.43
8.42
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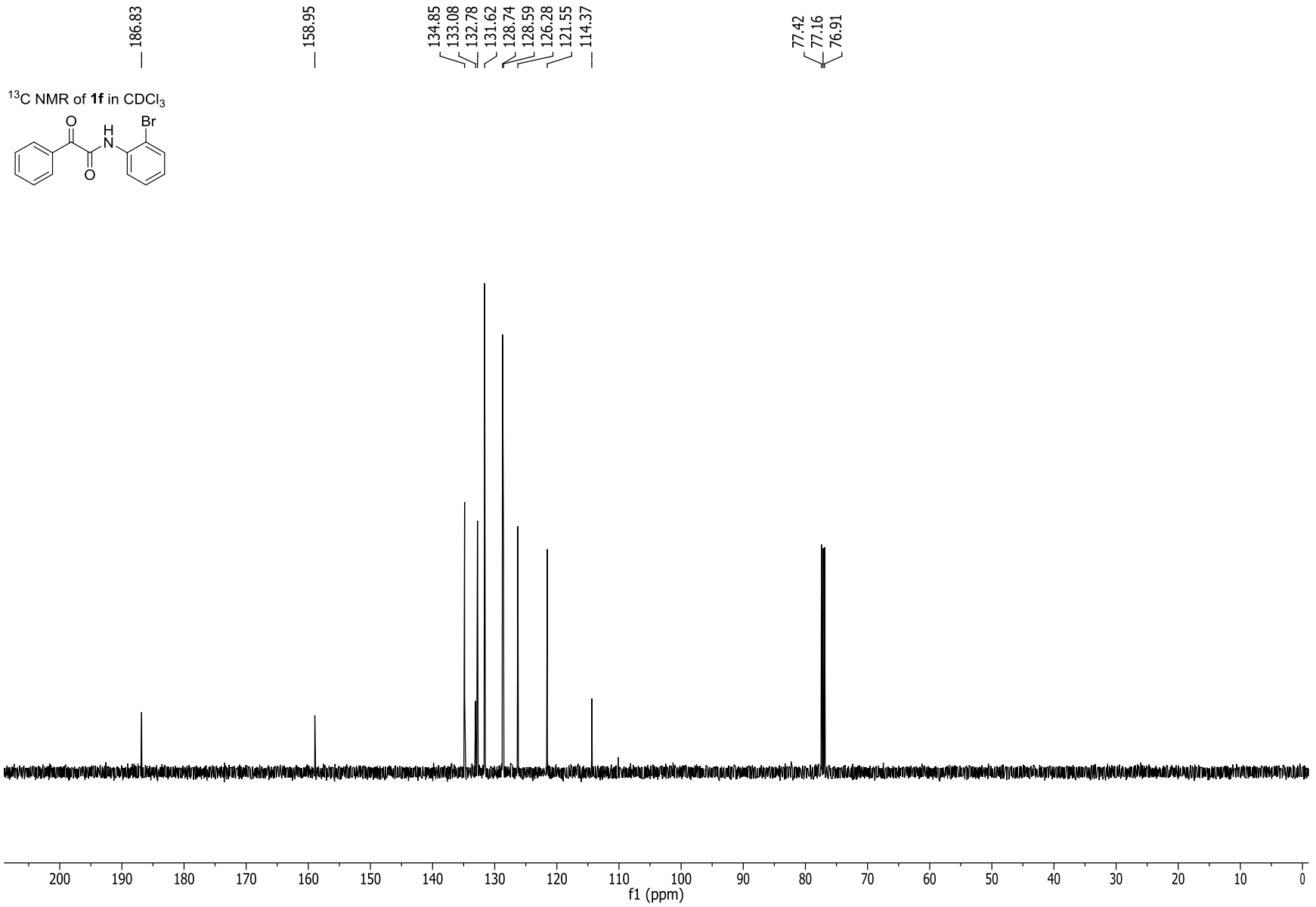
7.68
7.68
7.67
7.67
7.67
7.66
7.66
7.65
7.62
7.62
7.60
7.60
7.54
7.54
7.52
7.51
7.51
7.51
7.41
7.40
7.39
7.39
7.37
7.37
7.26
7.26
7.08
7.08
7.08
7.07
7.07
7.05
7.05
7.05
1H NMR of 1f in CDCl3
O
H
N
Br
O
1.05
1.00
1.96
1.05
1.01
2.09
1.07
1.04
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)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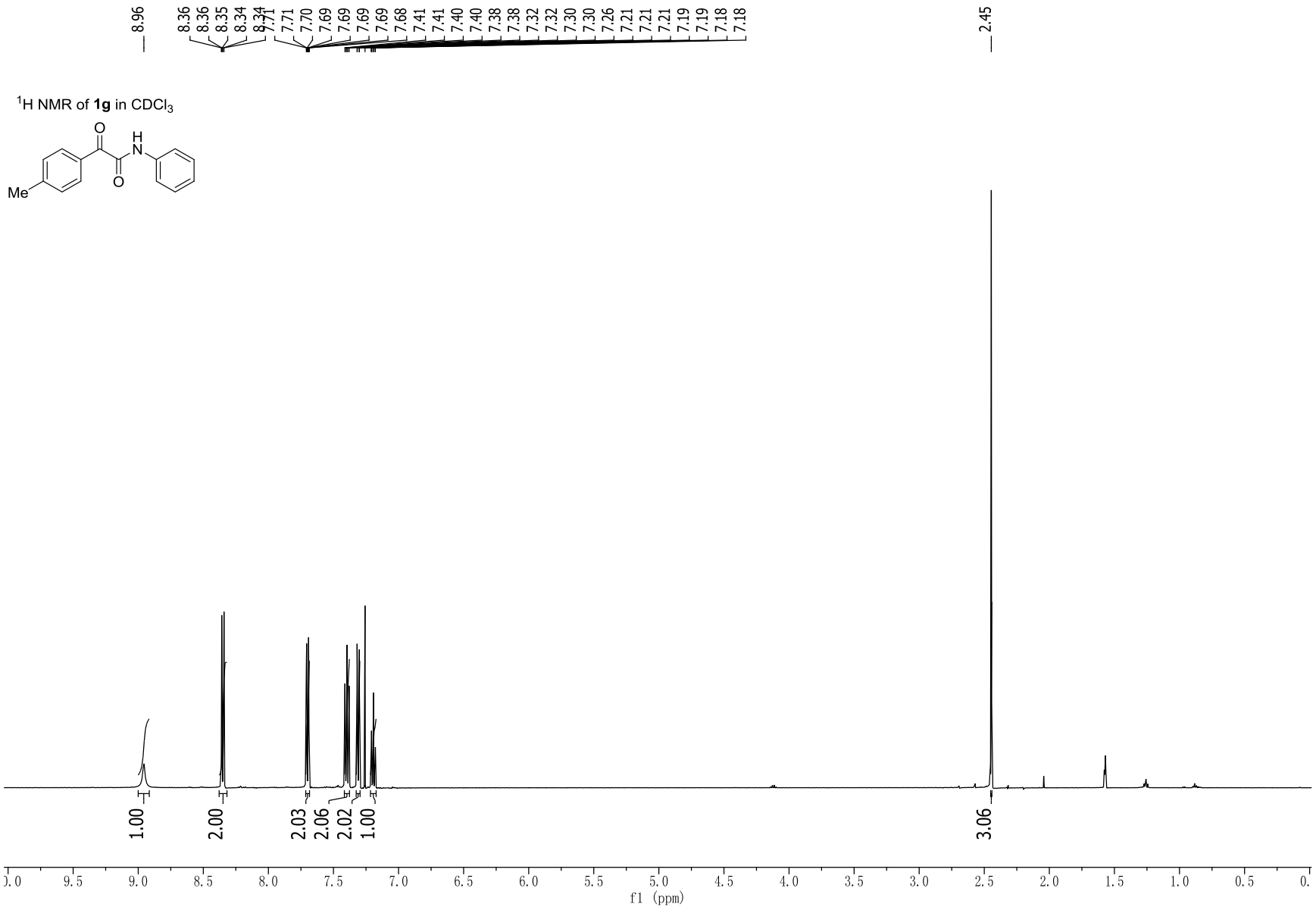
133.08
132.78
131.62
128.74
128.59
126.28
121.55
114.37
77.42
77.16
76.91
13C NMR of 1f in CDCl3
O
H
N
Br
O
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)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8.34
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7.71
7.70
7.69
7.69
7.69
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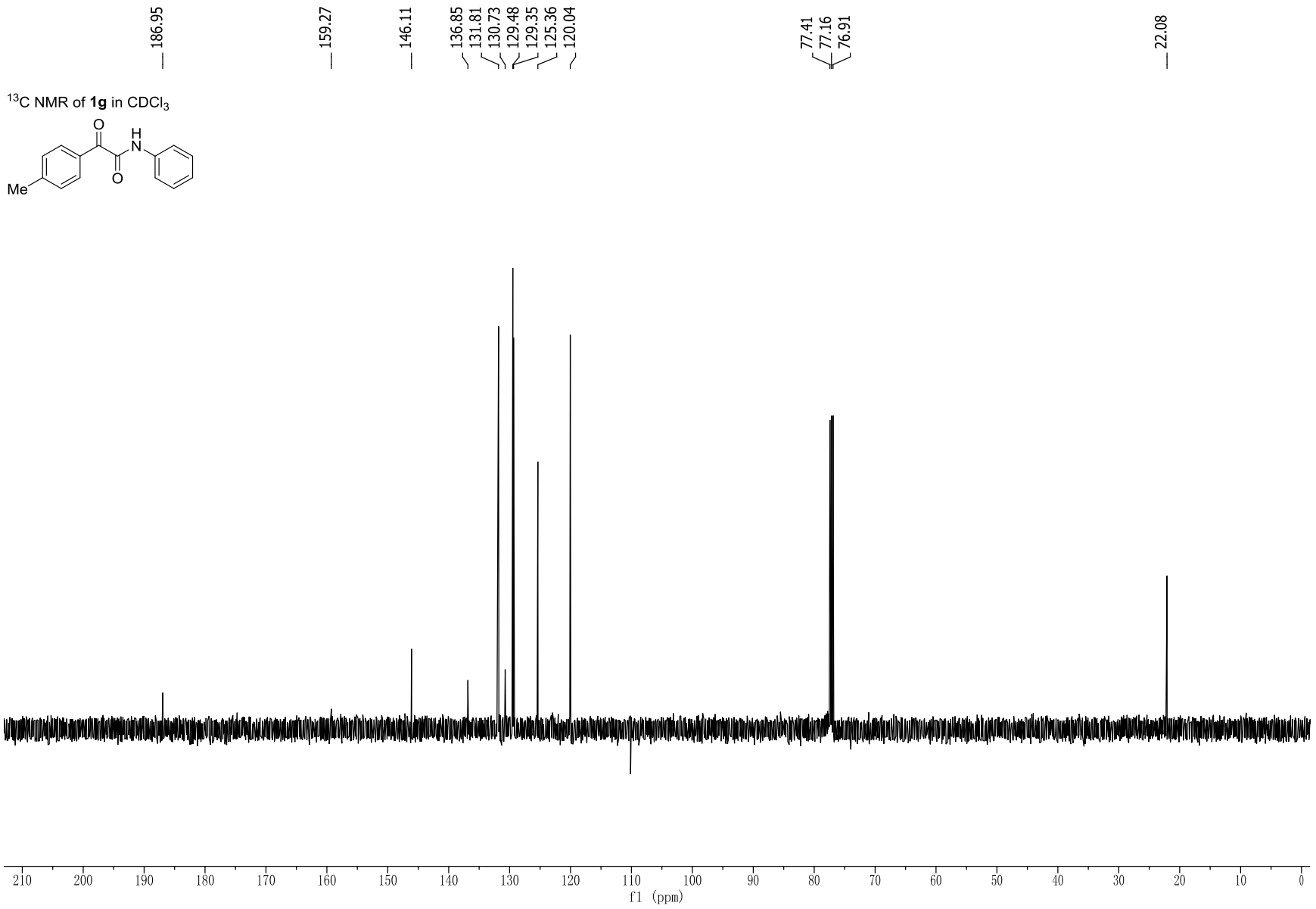
7.68
7.41
7.41
7.40
7.40
7.38
7.38
7.32
7.32
7.30
7.30
7.26
7.21
7.21
7.21
7.19
7.19
7.18
7.18
2.45
1H NMR of 1g in CDCl3
O
N
H
O
Me
1.00
2.00
2.03
2.06
2.02
1.00
3.06
f1 (ppm)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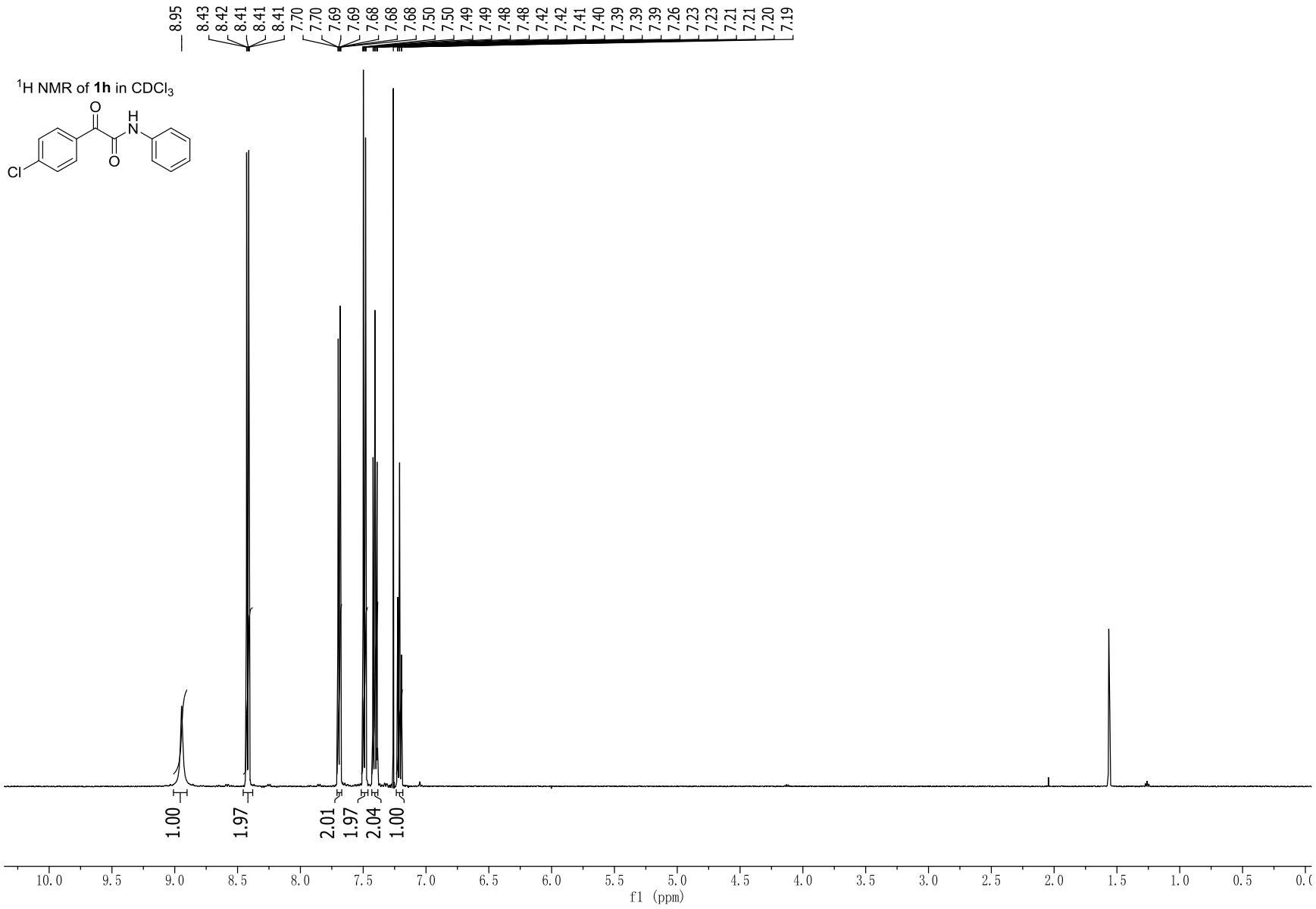
159.27
146.11
136.85
131.81
130.73
129.48
129.35
125.36
120.04
77.41
77.16
76.91
22.08
13C NMR of 1g in CDCl3
O
H
N
O
Me
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


8.95
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8.41
7.70
7.70
7.69
7.69
7.68
7.68
7.68
7.50
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7.49
7.49
7.48
7.48
7.42
7.42
7.41
7.40
7.39
7.39
7.39
7.26
7.23
7.23
7.21
7.21
7.20
7.19
1H NMR of 1h in CDCl3
O
H
N
O
Cl
1.00
1.97
2.01
1.97
2.04
1.00
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
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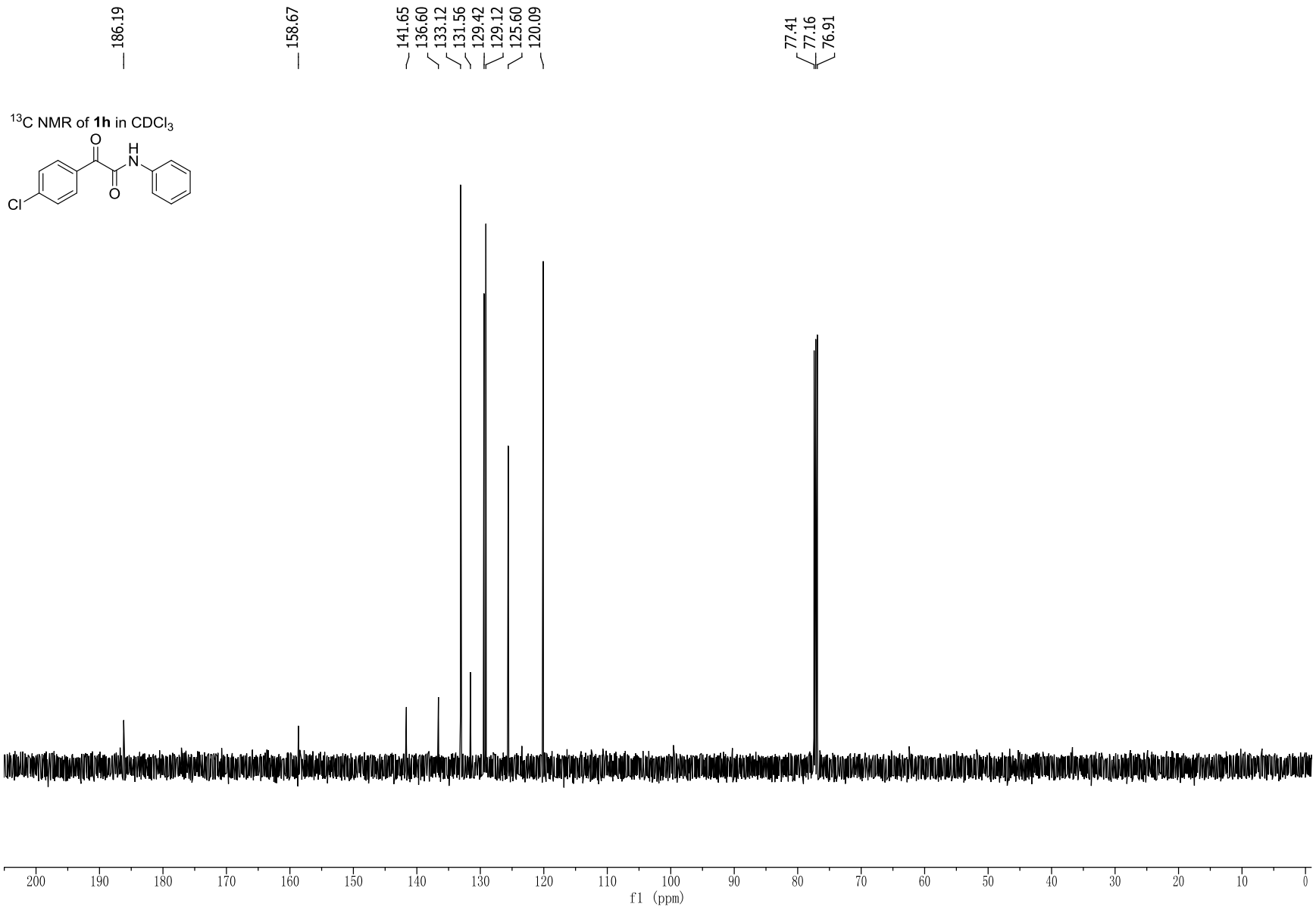
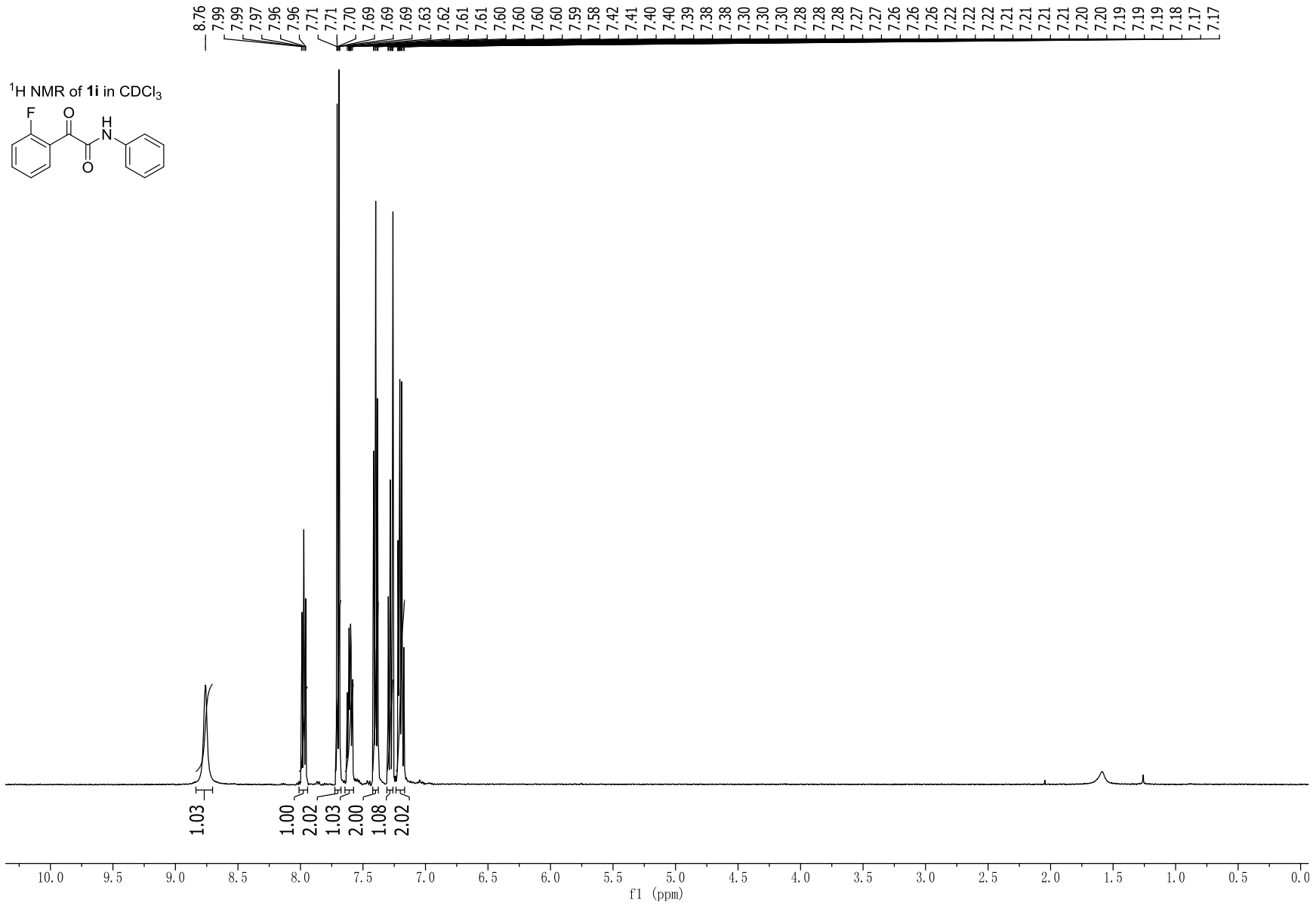
3.0
2.5
2.0
1.5
1.0
0.5
f1 (ppm)


186.19
158.67
141.65
136.60
133.12
131.56
129.42
129.12
125.60
120.09
77.41
77.16
76.91
13C NMR of 1h in CDCl3
O
H
N
O
Cl
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


^{1}H NMR of **1i** in $CDCl_3$
F
O
H
N
O
8.76
7.99
7.99
7.97
7.96
7.96
7.71
7.71
7.70
7.69
7.69
7.69
7.63
7.62
7.61
7.61
7.60
7.60
7.60
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7.58
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7.38
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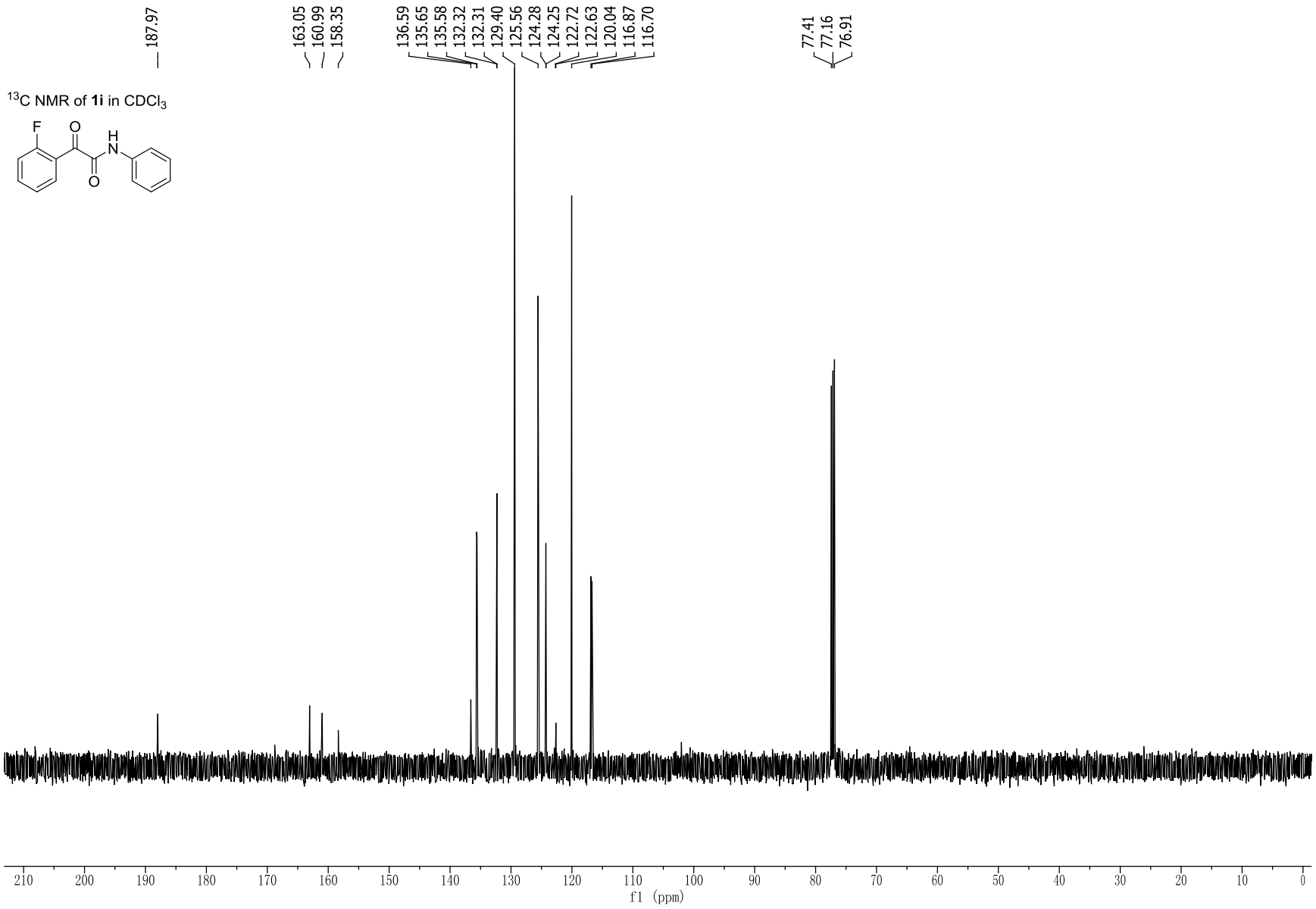
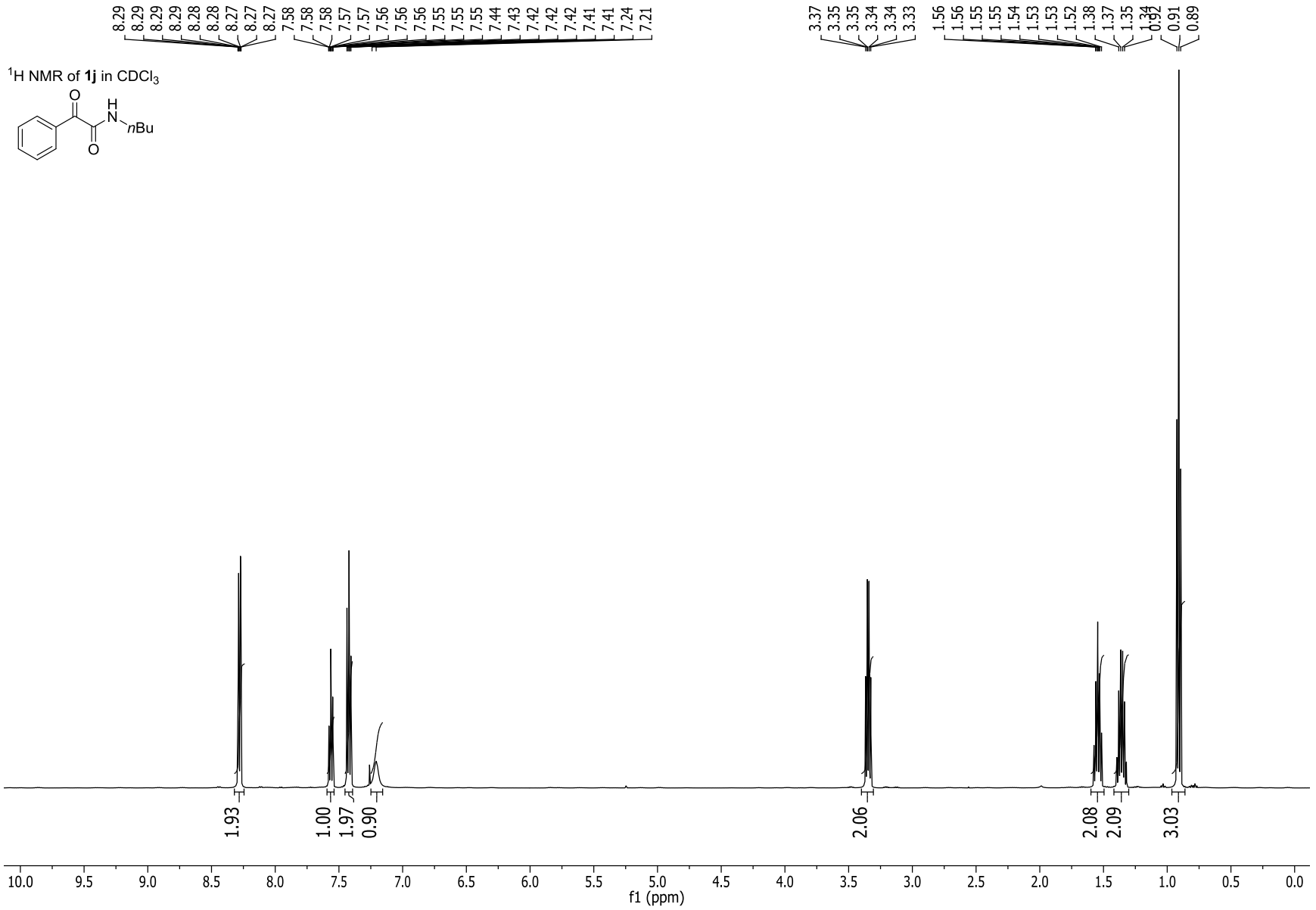
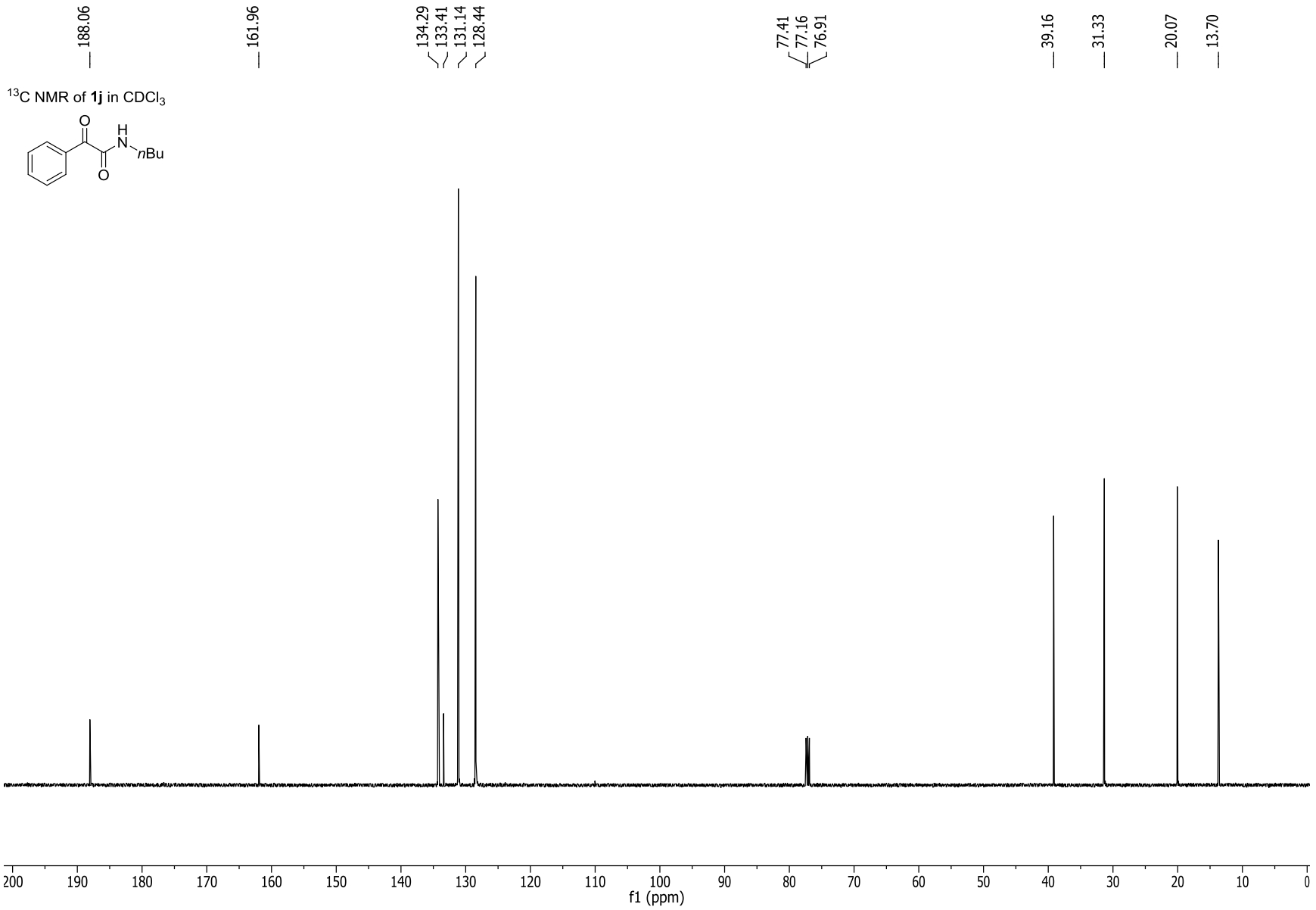
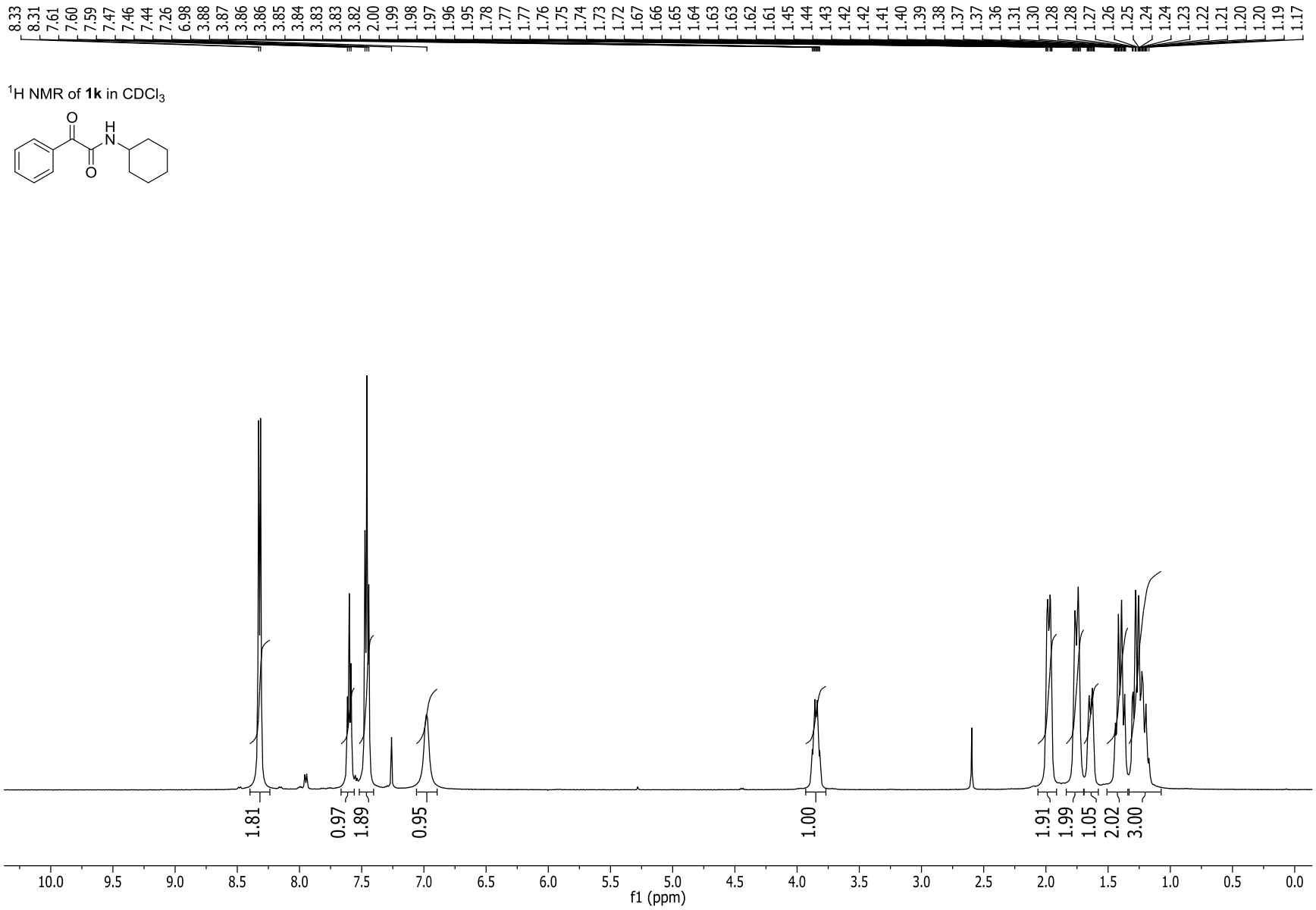
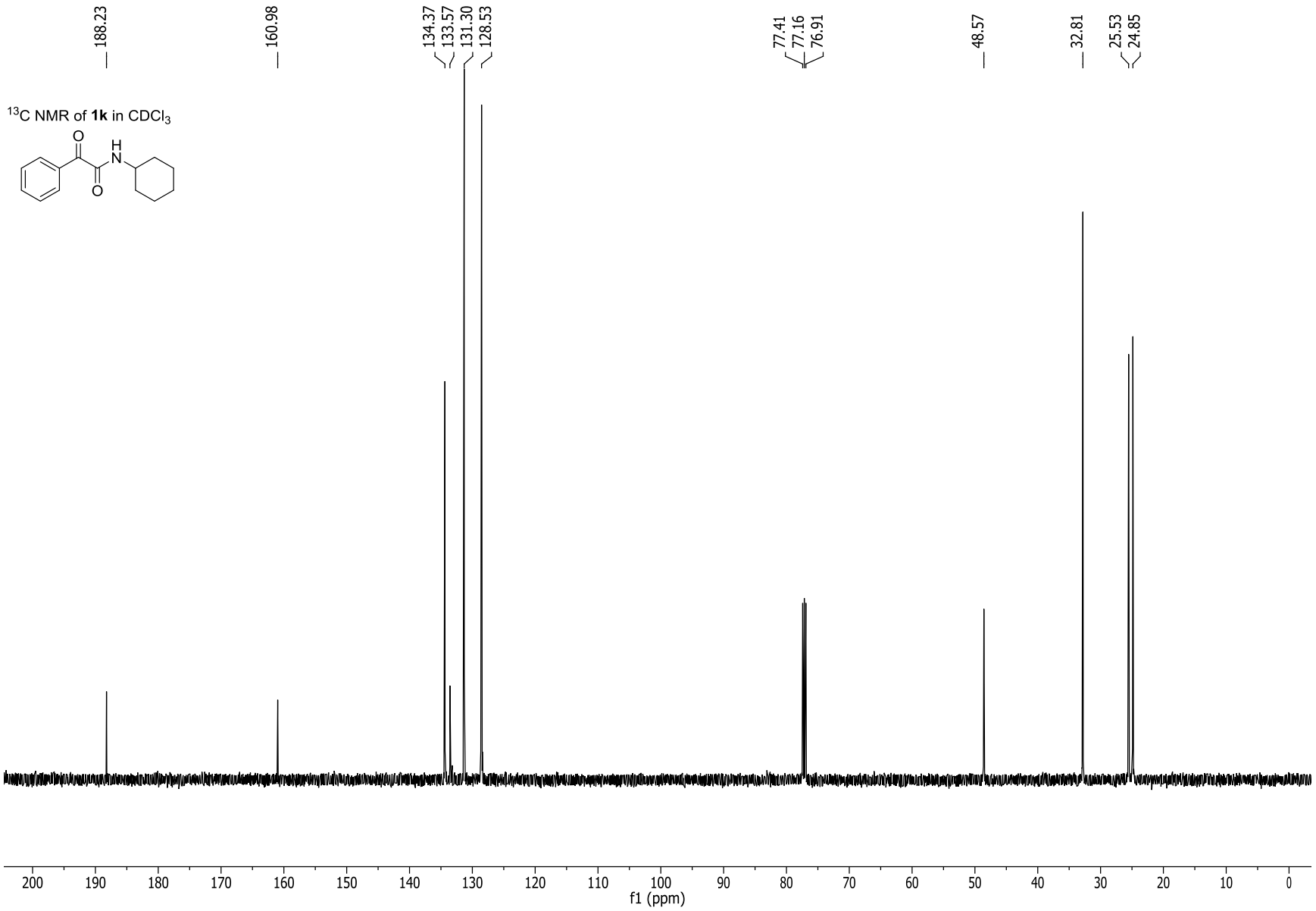
7.28
7.27
7.27
7.26
7.26
7.26
7.22
7.22
7.22
7.21
7.21
7.21
7.21
7.20
7.20
7.19
7.19
7.19
7.18
7.17
7.17
1.03
1.00
2.02
1.03
2.00
1.08
2.02
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

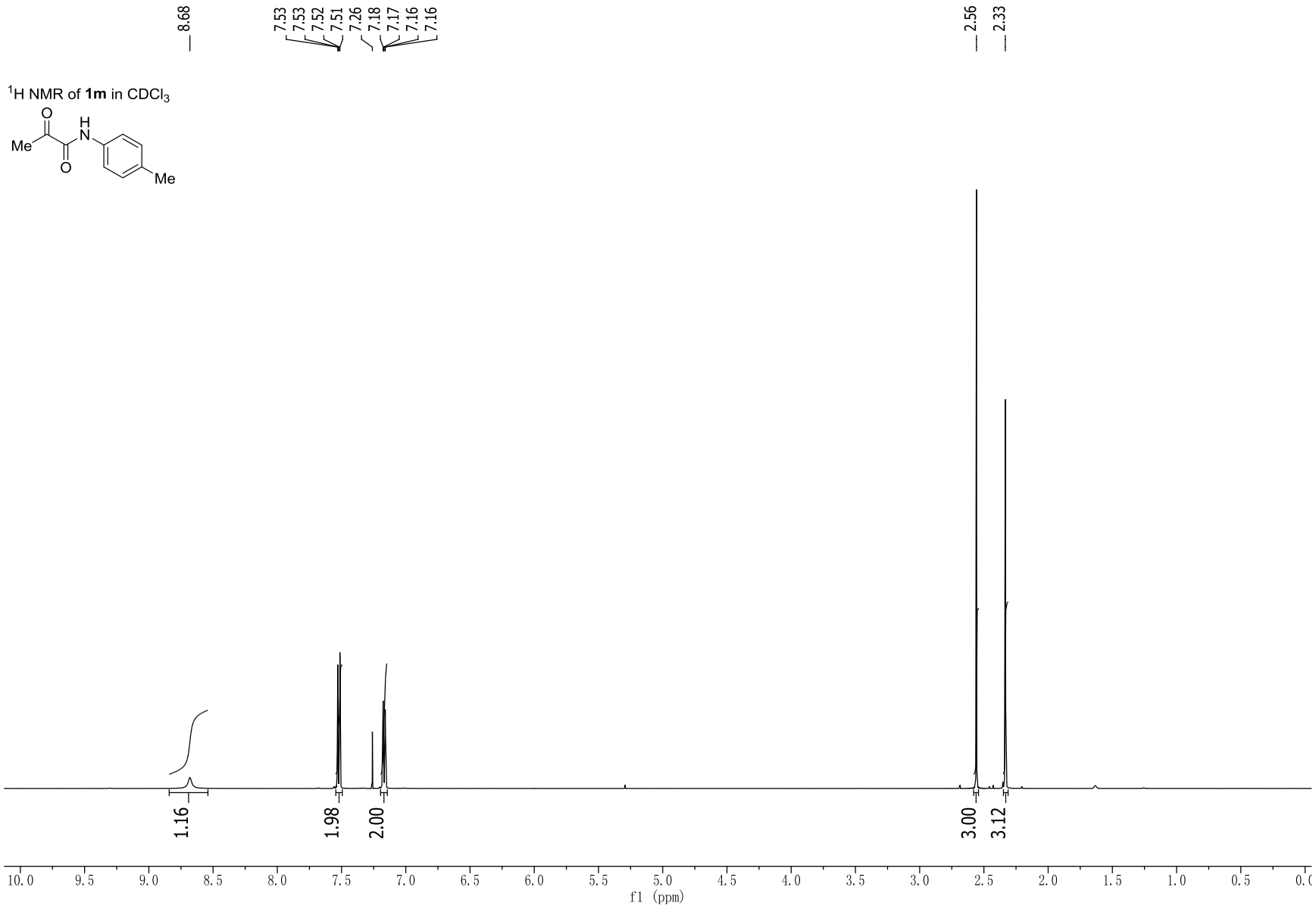

187.97
163.05
160.99
158.35
136.59
135.65
135.58
132.32
132.31
129.40
125.56
124.28
124.25
122.72
122.63
120.04
116.87
116.70
77.41
77.16
76.91
13C NMR of 1i in CDCl3
F
O
H
N
O
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

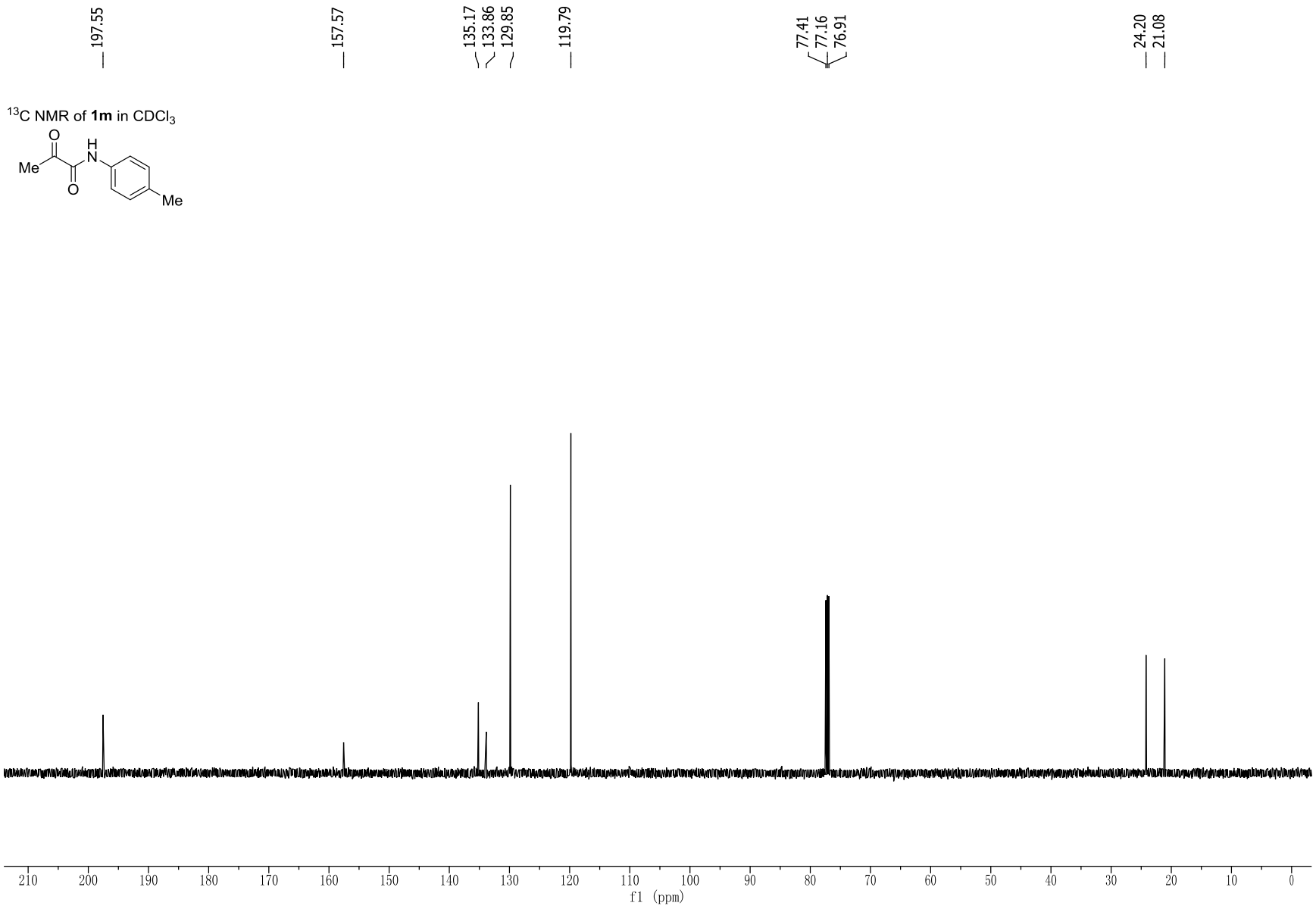

^{1}H NMR of **1j** in $CDCl_3$
nBu
8.29 8.29 8.29 8.29 8.28 8.28 8.27 8.27 8.27 7.58 7.58 7.58 7.57 7.57 7.56 7.56 7.56 7.55 7.55 7.55 7.44 7.43 7.42 7.42 7.42 7.41 7.41 7.24 7.21
3.37 3.35 3.35 3.34 3.34 3.33
1.56 1.56 1.55 1.55 1.54 1.53 1.53 1.52 1.38 1.37 1.35 1.34 0.92 0.91 0.89
1.93 1.00 1.97 0.90 2.06 2.08 2.09 3.03
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)

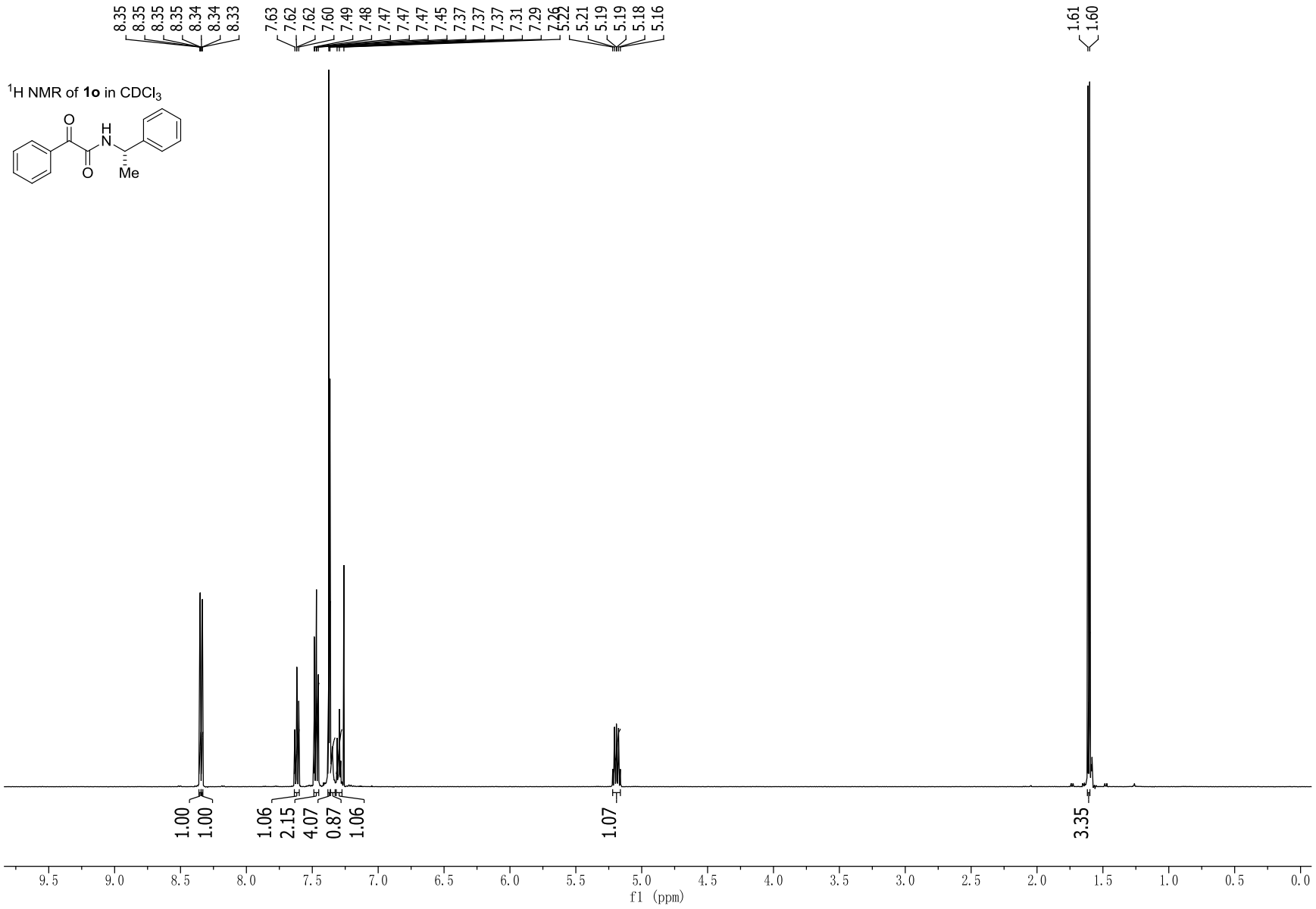

188.06
161.96
134.29
133.41
131.14
128.44
77.41
77.16
76.91
39.16
31.33
20.07
13.70
^{13}C NMR of **1j** in $CDCl_3$
O
H
N
*n*Bu
O
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

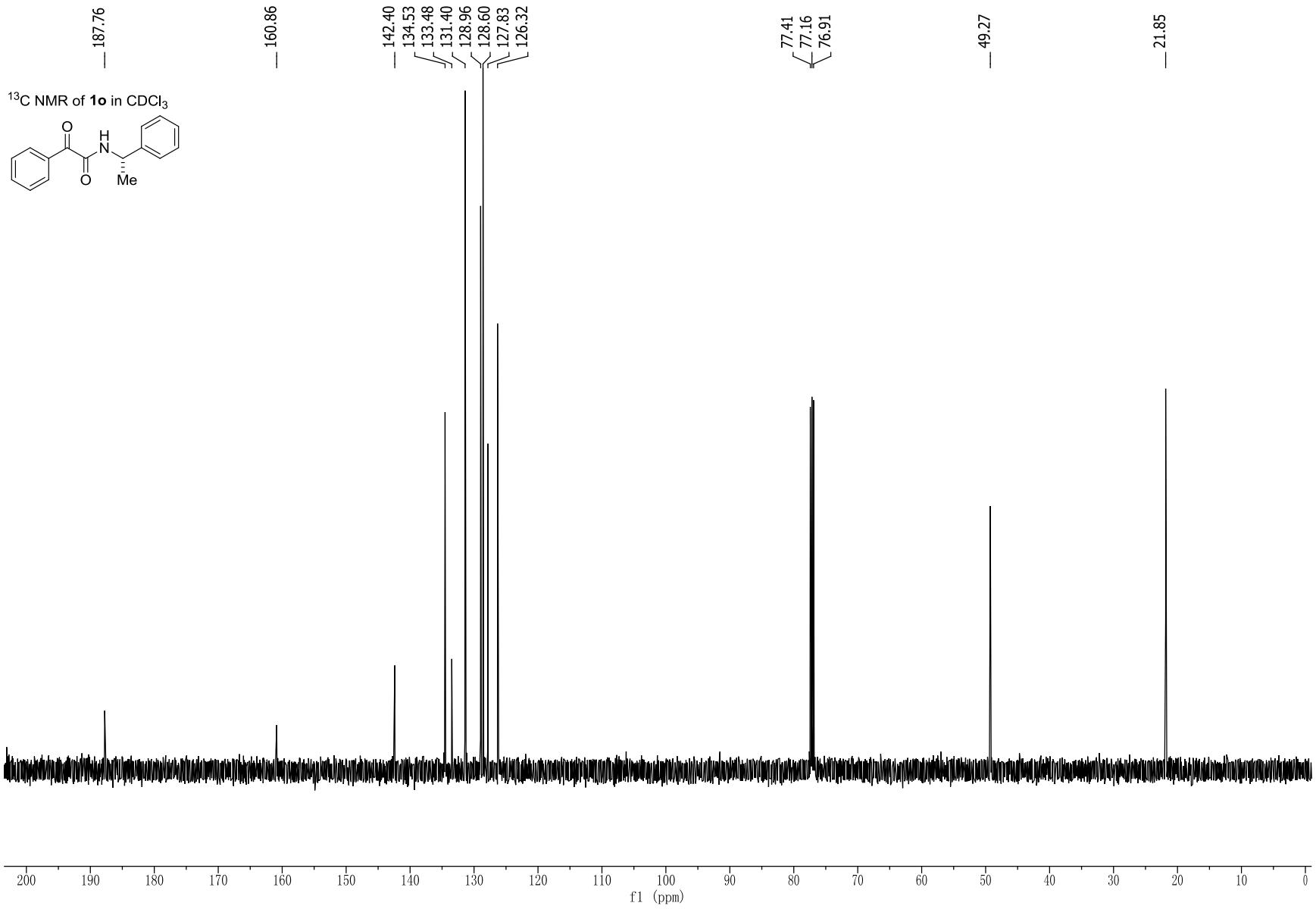

8.33 8.31 7.61 7.60 7.59 7.47 7.46 7.44 7.26 6.98 3.88 3.87 3.86 3.86 3.85 3.84 3.83 3.83 3.82 2.00 1.99 1.98 1.97 1.96 1.95 1.78 1.77 1.77 1.76 1.75 1.74 1.73 1.72 1.67 1.66 1.65 1.64 1.63 1.63 1.62 1.61 1.45 1.44 1.43 1.42 1.42 1.41 1.40 1.39 1.38 1.37 1.37 1.36 1.31 1.30 1.28 1.28 1.27 1.26 1.25 1.24 1.24 1.23 1.22 1.21 1.20 1.20 1.19 1.17
^{1}H NMR of **1k** in $CDCl_3$
O
H
N
O
1.81
0.97
1.89
0.95
1.00
1.91
1.99
1.05
2.02
3.00
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)

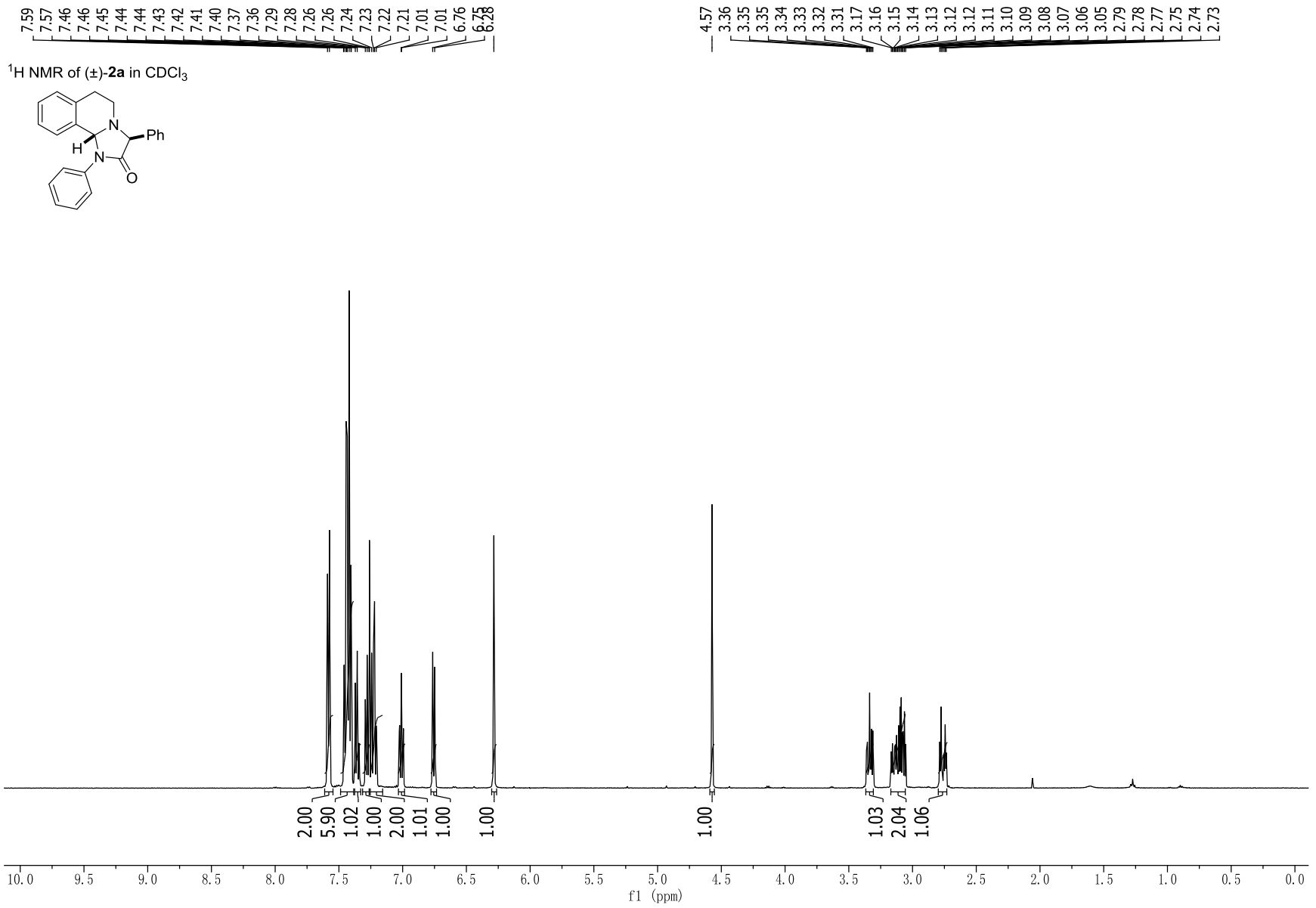

188.23
160.98
134.37
133.57
131.30
128.53
77.41
77.16
76.91
48.57
32.81
25.53
24.85
^{13}C NMR of **1k** in $CDCl_3$
O
H
N
O
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

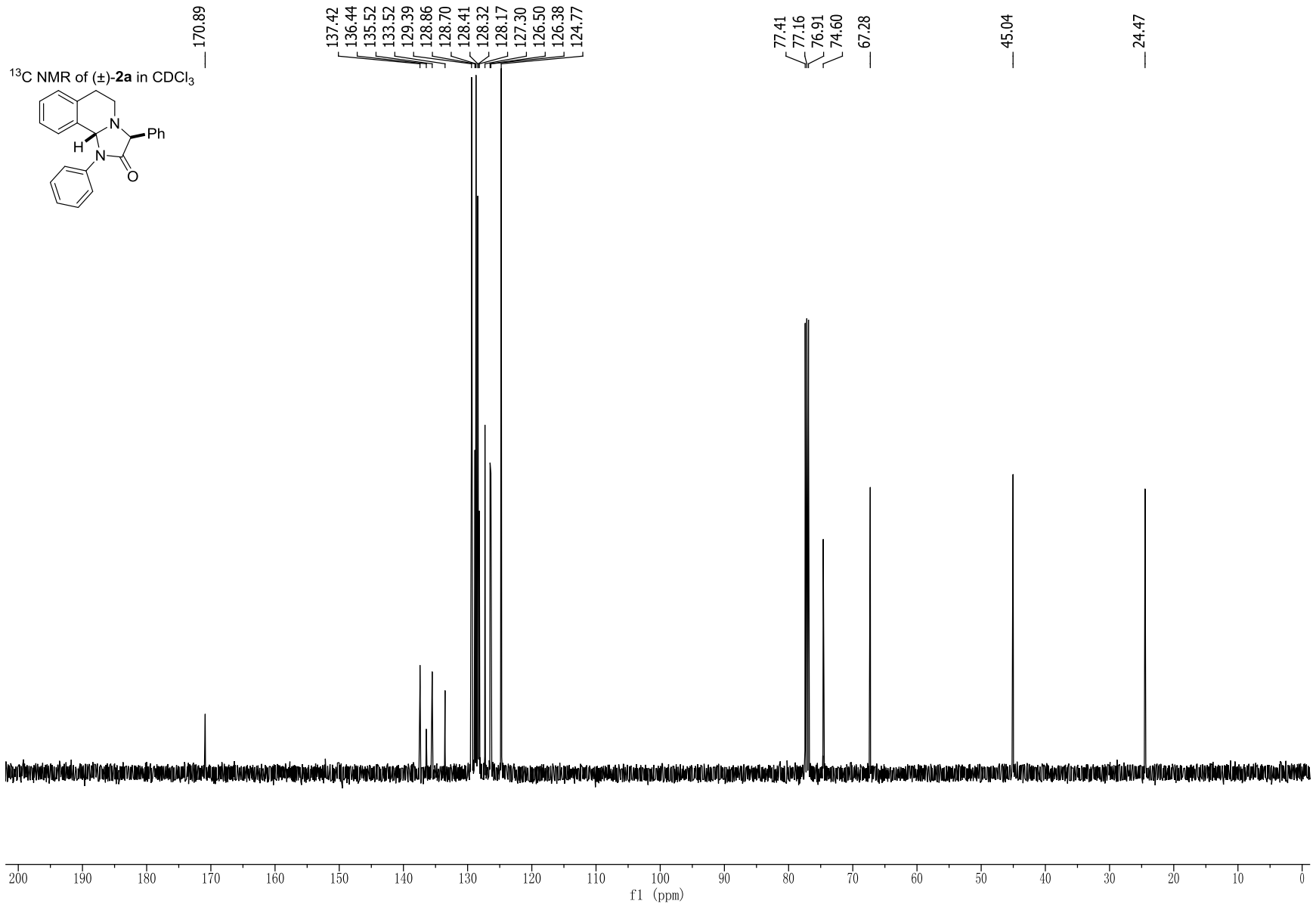

8.68
7.53
7.53
7.52
7.51
7.26
7.18
7.17
7.16
7.16
2.56
2.33
^{1}H NMR of **1m** in $CDCl_3$
Me
O
H
N
O
Me
1.16
1.98
2.00
3.00
3.12
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
f1 (ppm)


197.55
157.57
135.17
133.86
129.85
119.79
77.41
77.16
76.91
24.20
21.08
13C NMR of 1m in CDCl3
O
H
N
Me
O
Me
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

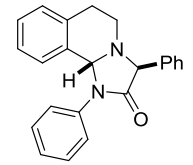

8.35
8.35
8.35
8.35
8.34
8.34
8.33
7.63
7.62
7.62
7.60
7.49
7.48
7.47
7.47
7.47
7.45
7.37
7.37
7.37
7.31
7.29
7.26
5.22
5.21
5.19
5.19
5.18
5.16
1.61
1.60
1H NMR of 1o in CDCl3
O
H
N
O
Me
1.00
1.00
1.06
2.15
4.07
0.87
1.06
1.07
3.35
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


187.76
160.86
142.40
134.53
133.48
131.40
128.96
128.60
127.83
126.32
77.41
77.16
76.91
49.27
21.85
13C NMR of 1o in CDCl3
O
H
N
O
Me
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

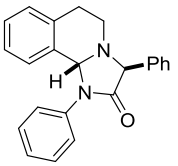

7.59
7.57
7.46
7.46
7.45
7.44
7.44
7.43
7.42
7.41
7.40
7.37
7.36
7.29
7.28
7.26
7.26
7.24
7.23
7.22
7.21
7.01
7.01
6.76
6.75
6.28
4.57
3.36
3.35
3.35
3.34
3.33
3.32
3.31
3.17
3.16
3.15
3.14
3.13
3.12
3.12
3.11
3.10
3.09
3.08
3.07
3.06
3.05
2.79
2.78
2.77
2.75
2.74
2.73
1H NMR of (±)-2a in CDCl3
Ph
H
N
N
O
2.00
5.90
1.02
1.00
2.00
1.01
1.00
1.00
1.00
1.03
2.04
1.06
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)

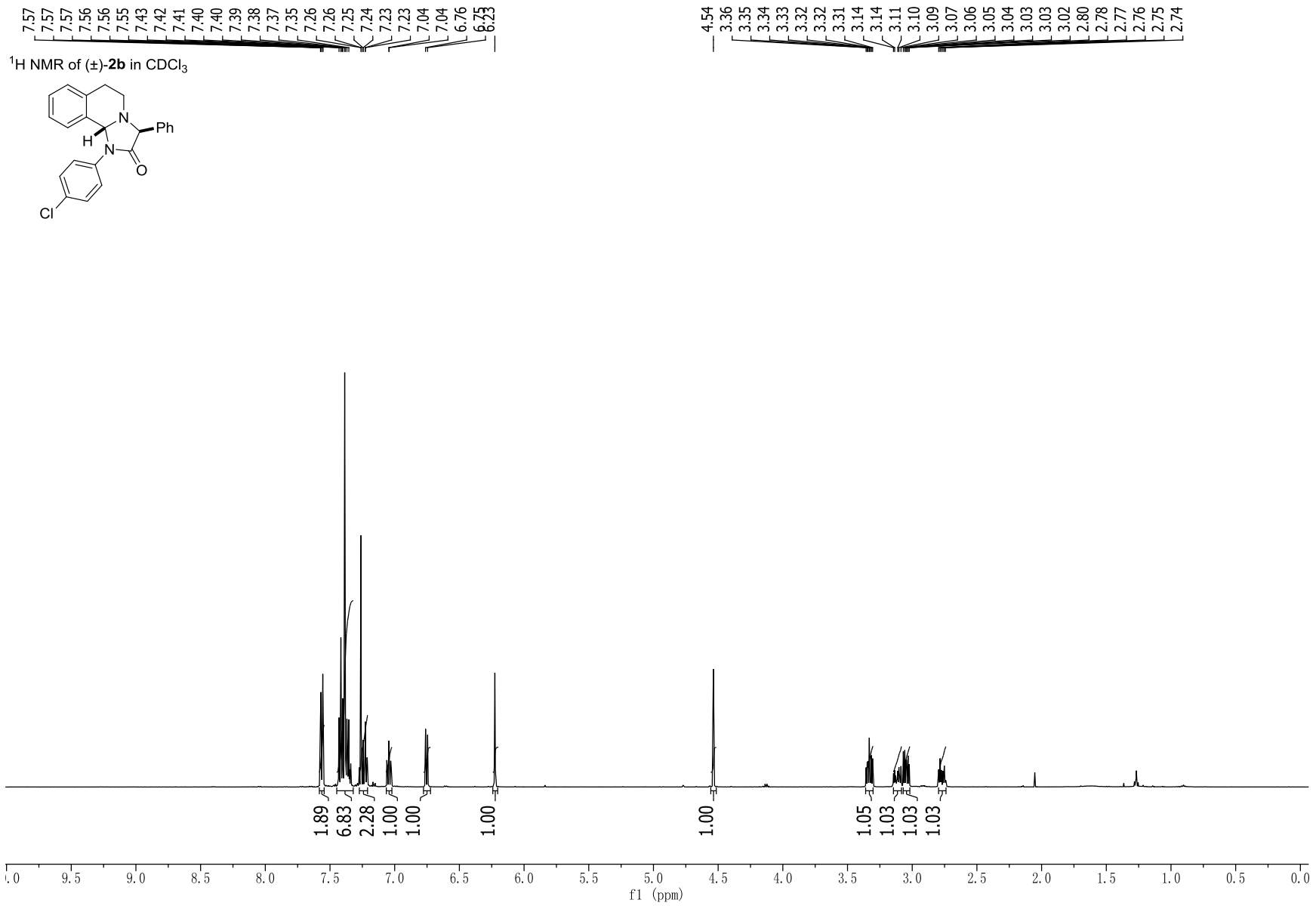

^{13}C NMR of (±)-**2a** in $CDCl_3$
170.89
137.42
136.44
135.52
133.52
129.39
128.86
128.70
128.41
128.32
128.17
127.30
126.50
126.38
124.77
77.41
77.16
76.91
74.60
67.28
45.04
24.47
Ph
H
N
N
O
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

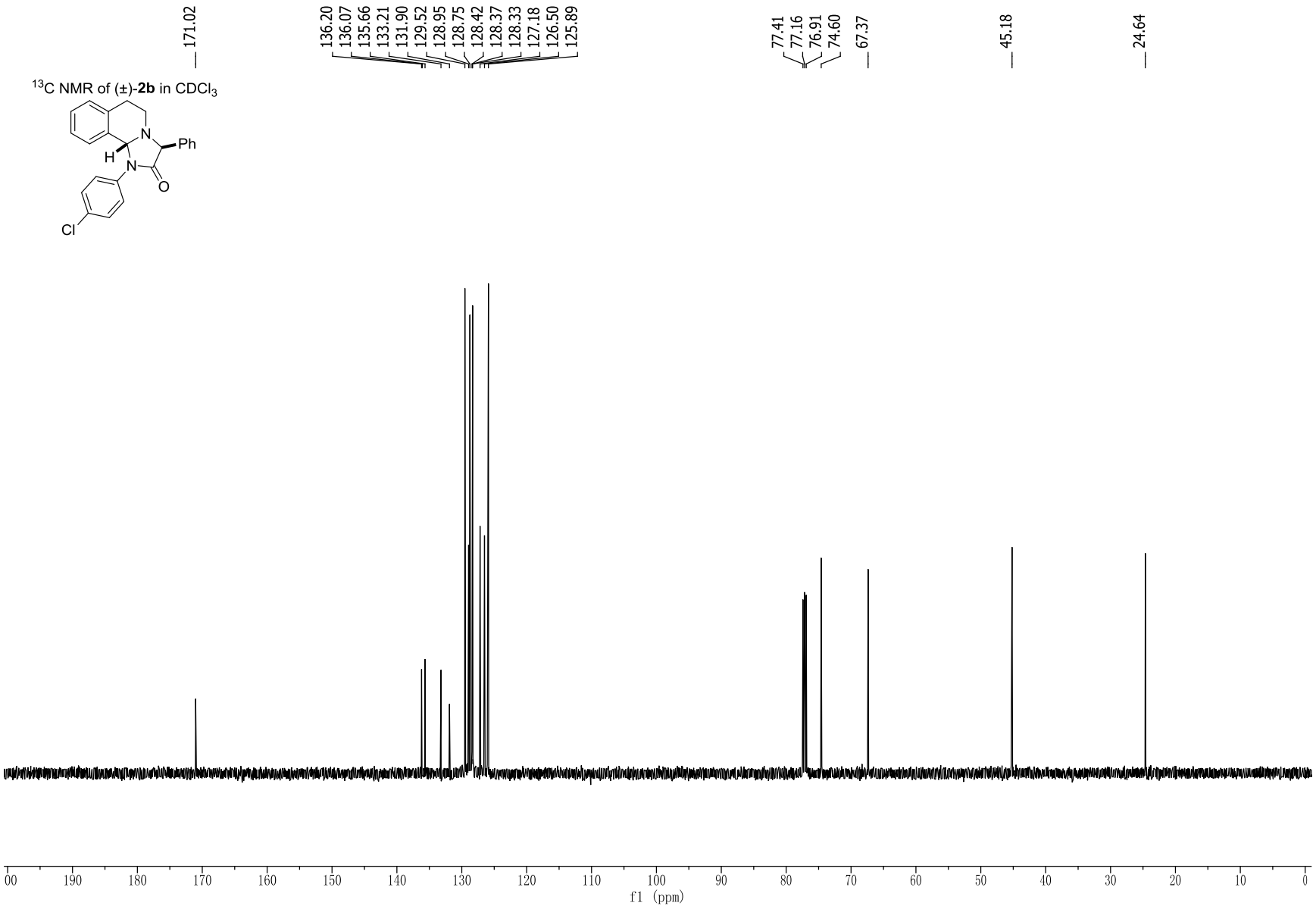
GCOSY of (±)-**2a** in $CDCl_3$

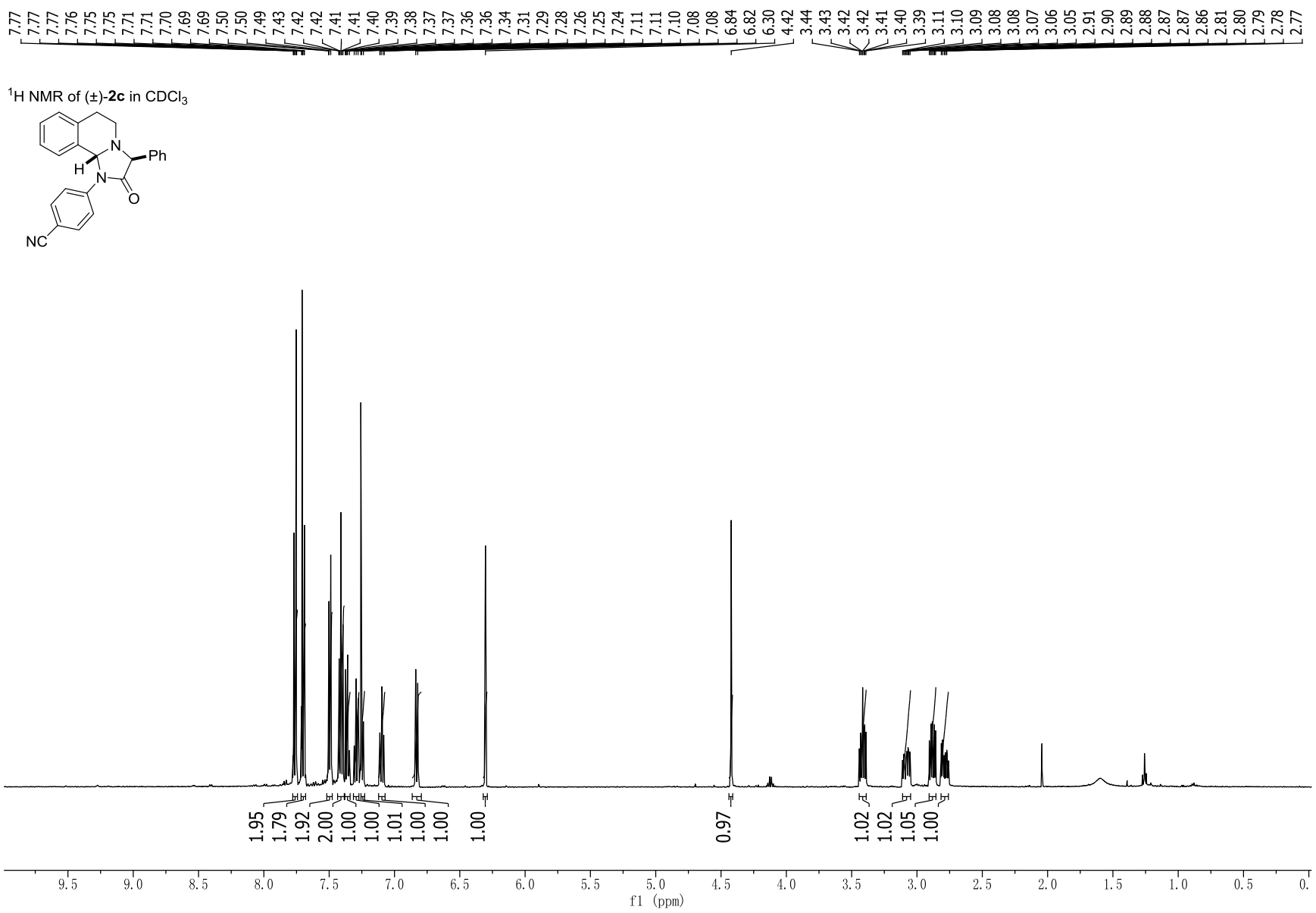


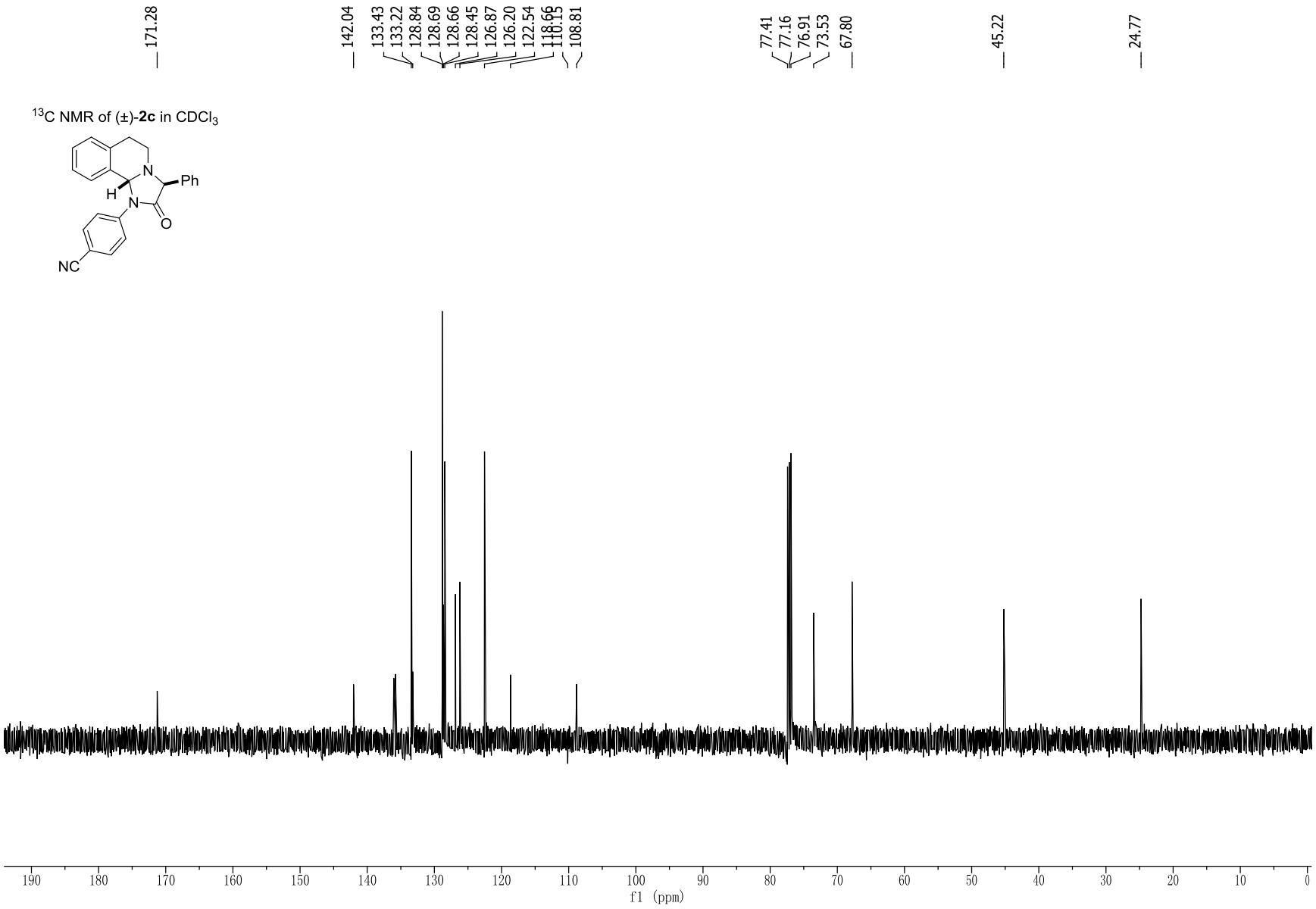
NOESY of (±)-**2a** in $CDCl_3$

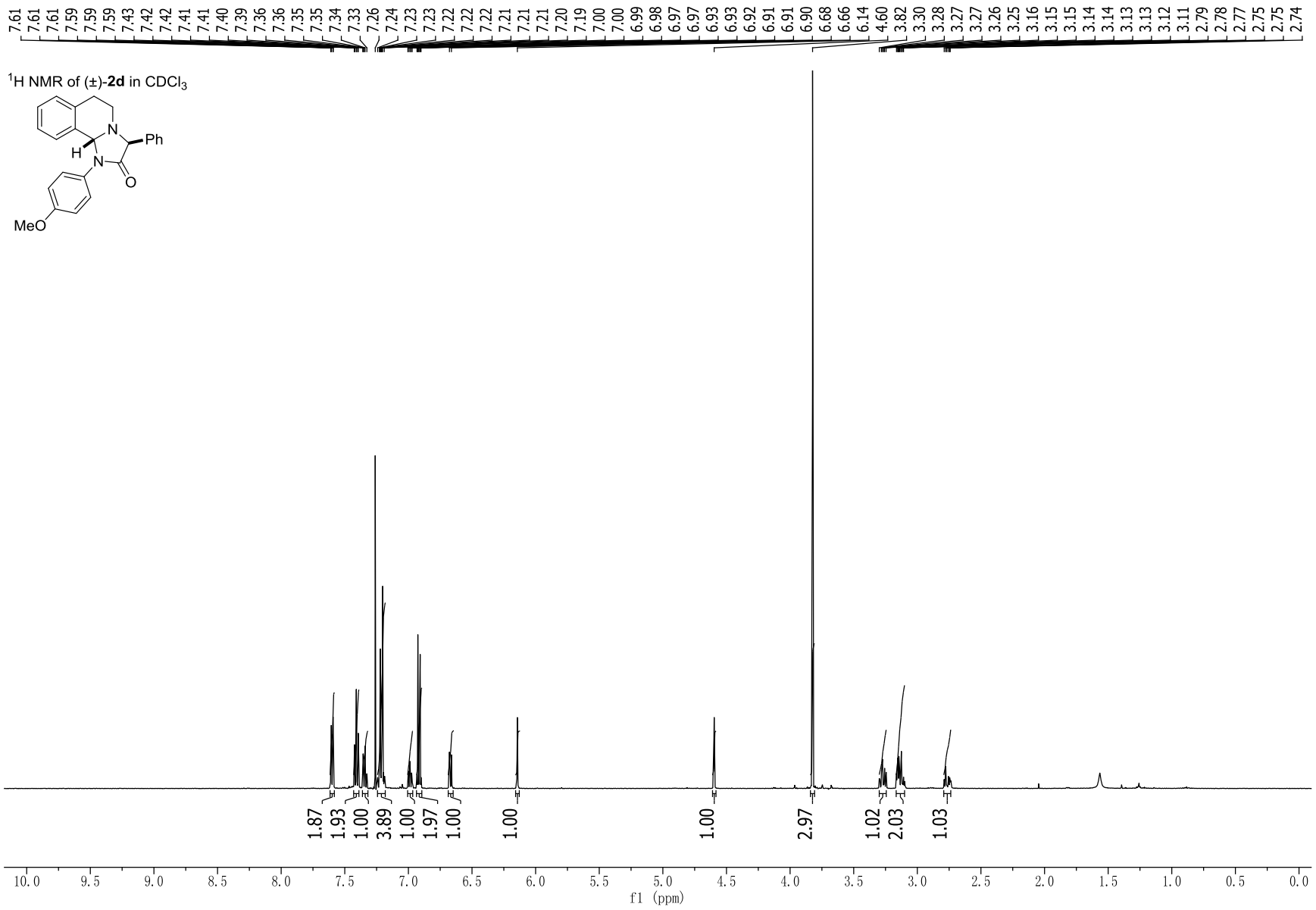


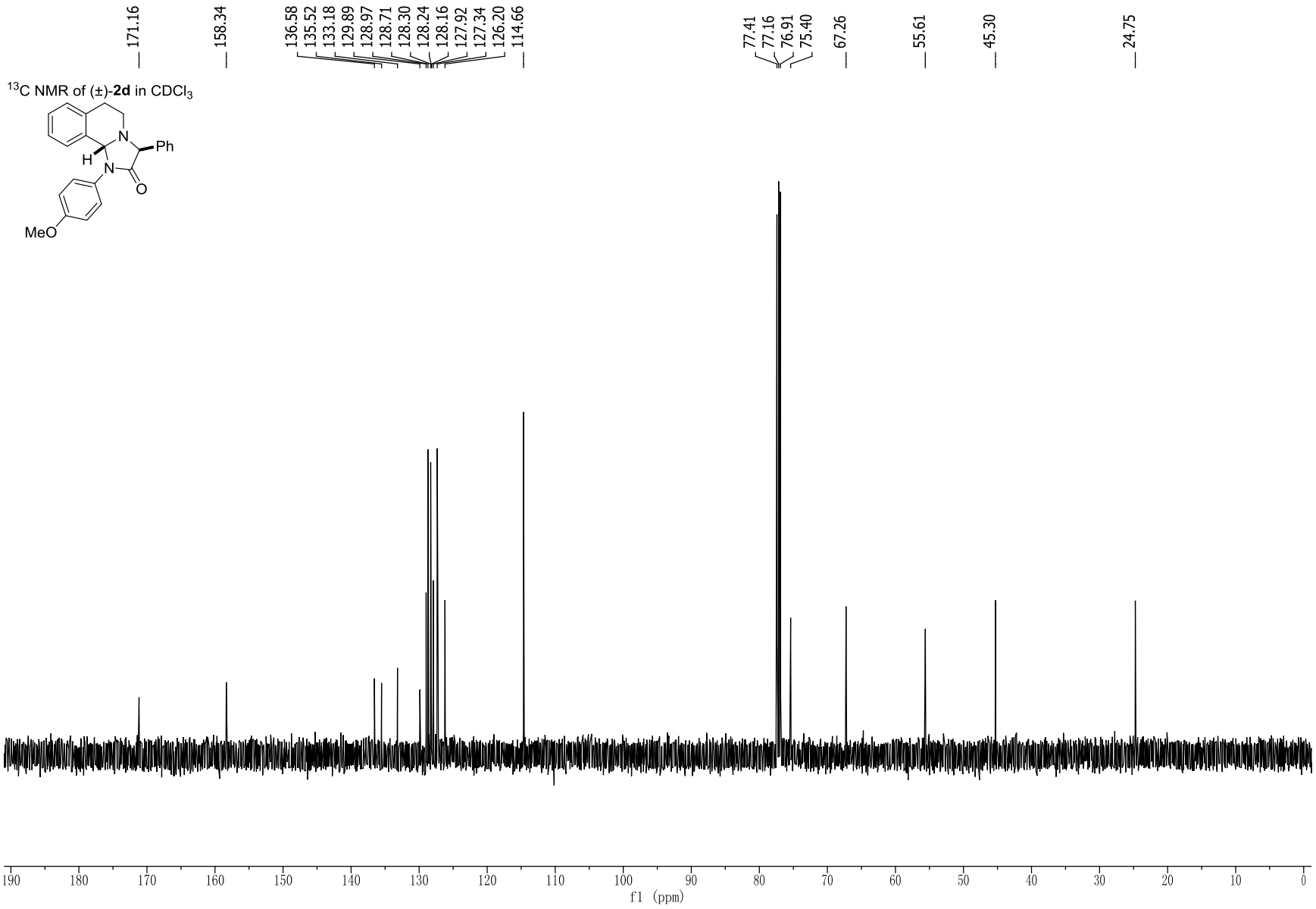

7.57
7.57
7.57
7.56
7.56
7.55
7.43
7.42
7.41
7.40
7.40
7.39
7.38
7.37
7.35
7.26
7.26
7.25
7.24
7.23
7.23
7.04
7.04
6.76
6.75
6.73
4.54
3.36
3.35
3.34
3.33
3.32
3.32
3.31
3.14
3.14
3.11
3.10
3.09
3.07
3.06
3.05
3.04
3.03
3.03
3.02
2.80
2.78
2.77
2.76
2.75
2.74
1H NMR of (±)-2b in CDCl3
N
H
N
O
Ph
Cl
1.89
6.83
2.28
1.00
1.00
1.00
1.00
1.05
1.03
1.03
1.03
f1 (ppm)

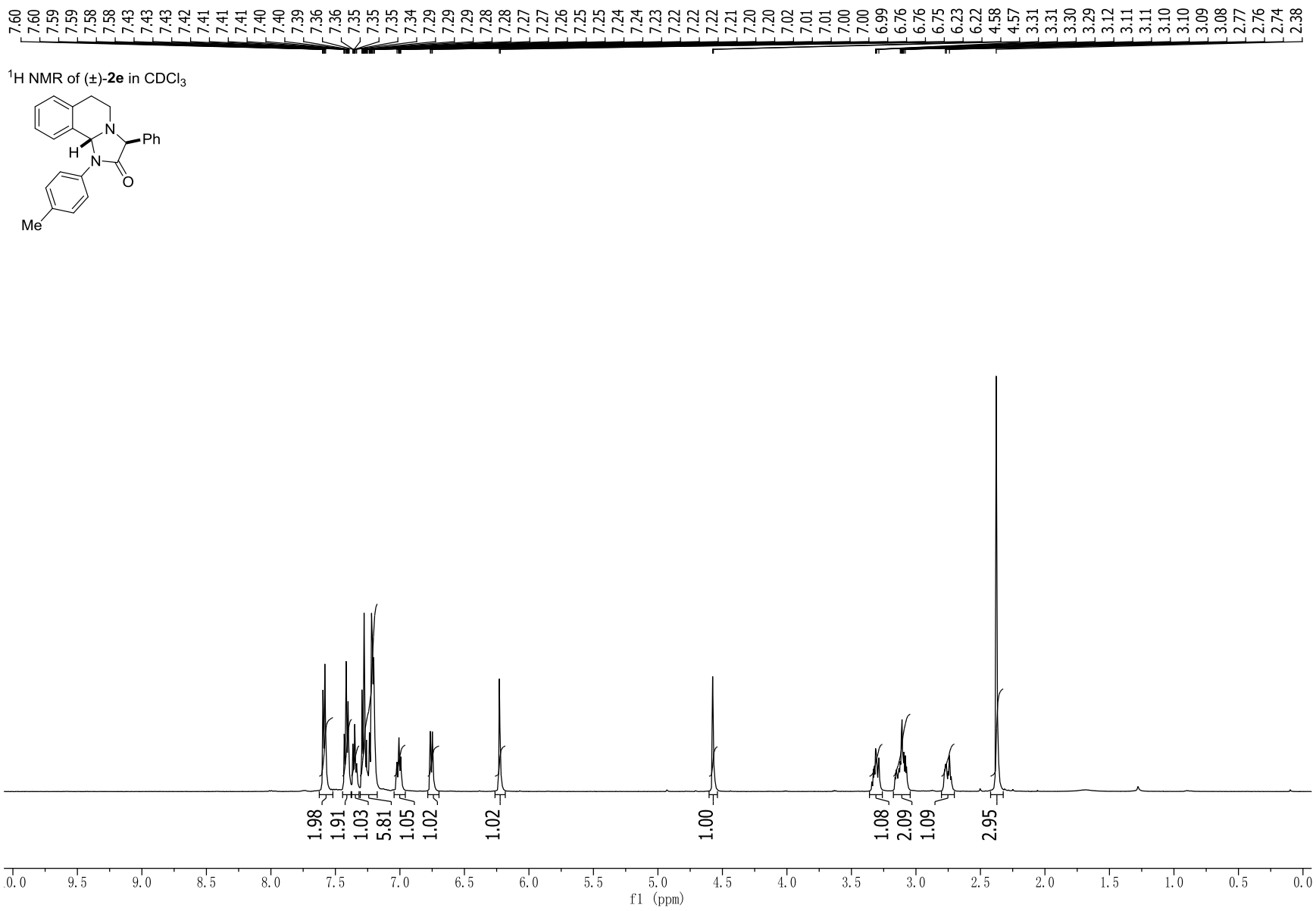

171.02
136.20
136.07
135.66
133.21
131.90
129.52
128.95
128.75
128.42
128.37
128.33
127.18
126.50
125.89
77.41
77.16
76.91
74.60
67.37
45.18
24.64
13C NMR of (±)-2b in CDCl3
Ph
H
N
O
Cl
f1 (ppm)

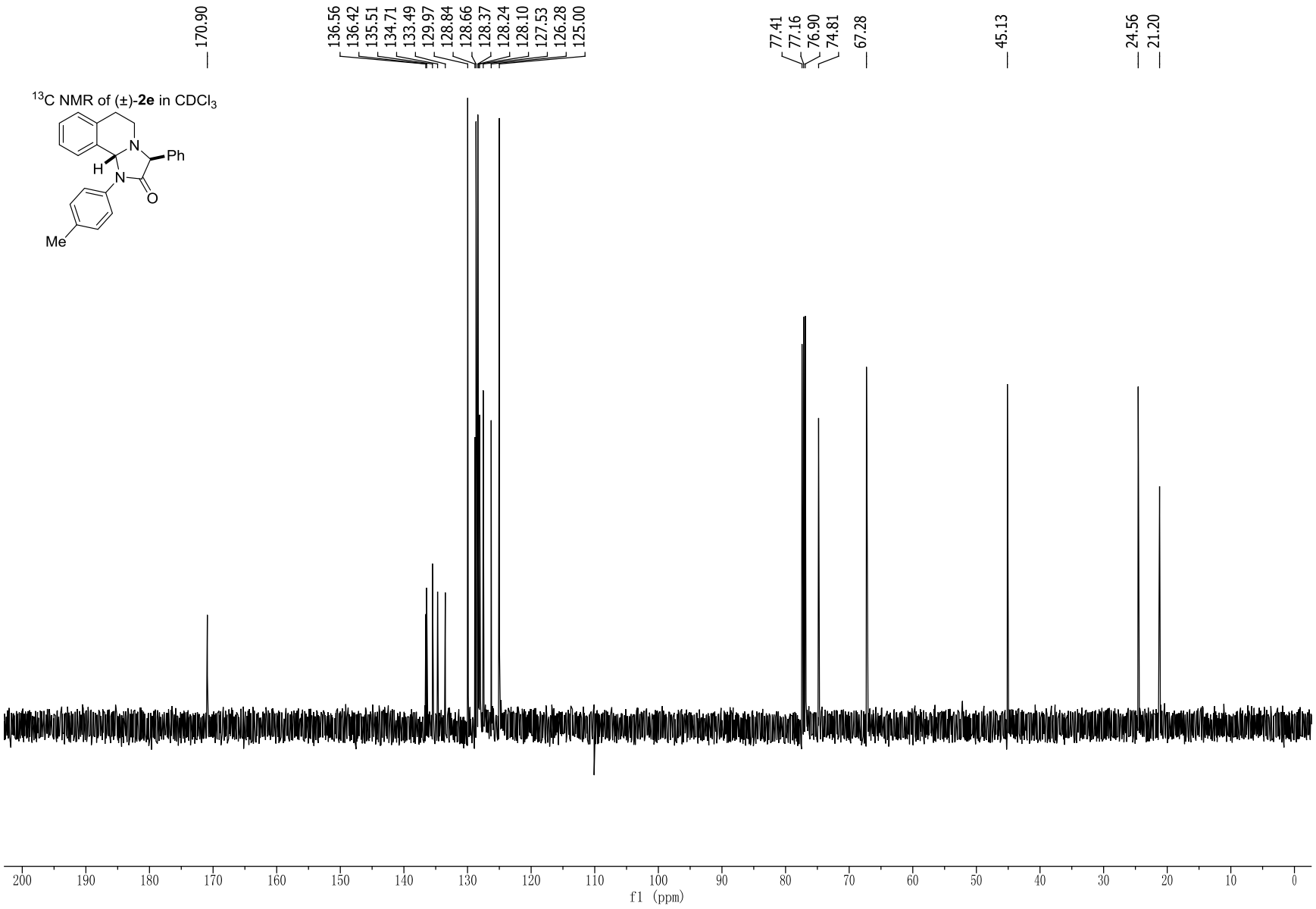

7.77 7.77 7.77 7.76 7.75 7.75 7.71 7.71 7.70 7.69 7.69 7.50 7.50 7.49 7.43 7.42 7.42 7.41 7.41 7.40 7.39 7.38 7.37 7.37 7.36 7.36 7.34 7.31 7.29 7.28 7.26 7.25 7.24 7.11 7.11 7.10 7.08 7.08 6.84 6.82 6.30 4.42 3.44 3.43 3.42 3.42 3.41 3.40 3.39 3.11 3.10 3.09 3.08 3.08 3.07 3.06 3.05 2.91 2.90 2.89 2.88 2.87 2.87 2.86 2.81 2.80 2.79 2.78 2.77
1H NMR of (±)-2c in CDCl3
Ph
N
H
N
O
NC
1.95 1.79 1.92 2.00 1.00 1.00 1.01 1.00 1.00 1.00 0.97 1.02 1.02 1.05 1.00
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.
f1 (ppm)

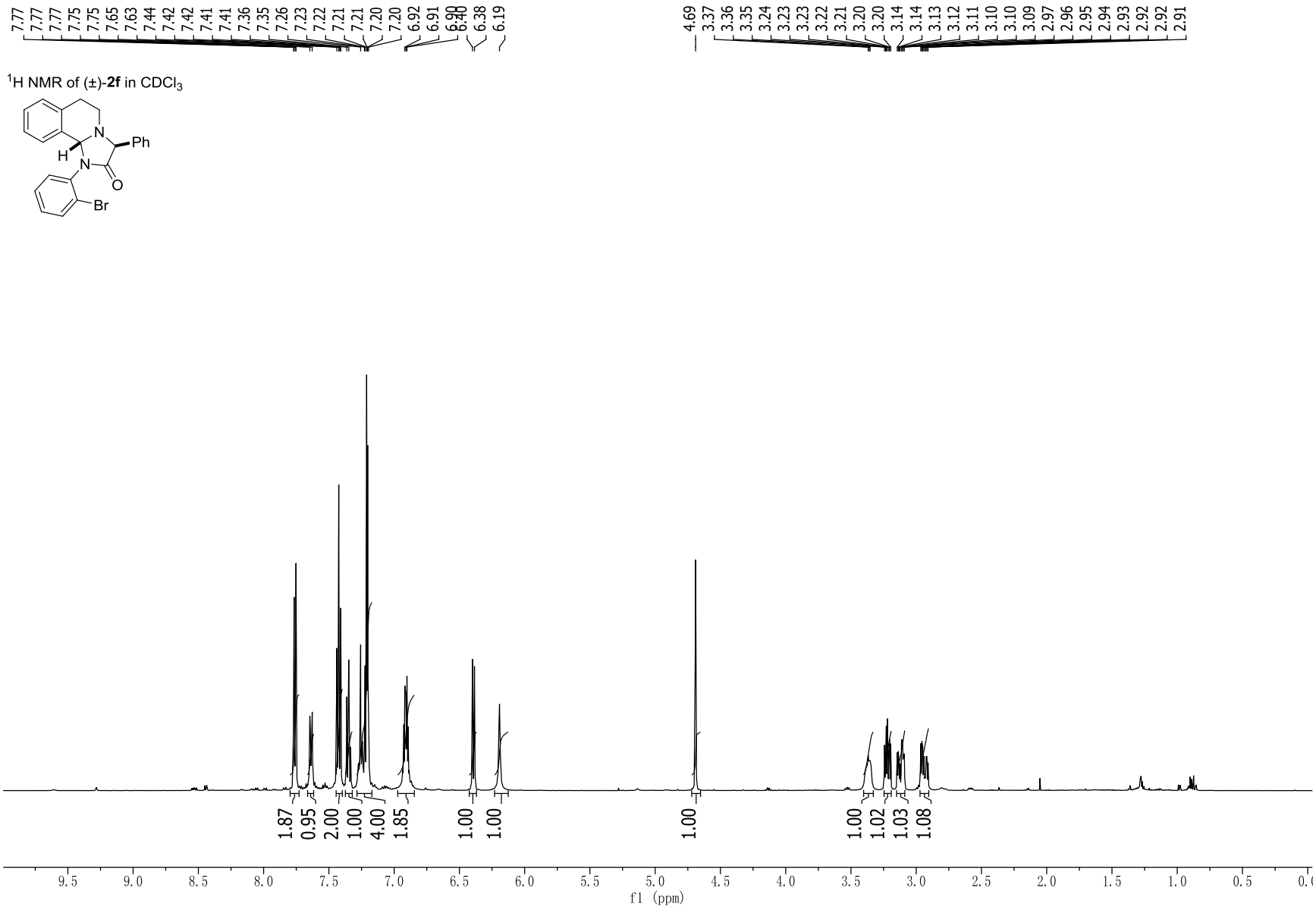

171.28
142.04
133.43
133.22
128.84
128.69
128.66
128.45
126.87
126.20
122.54
118.66
110.15
108.81
77.41
77.16
76.91
73.53
67.80
45.22
24.77
13C NMR of (±)-2c in CDCl3
Ph
H
N
N
O
NC
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

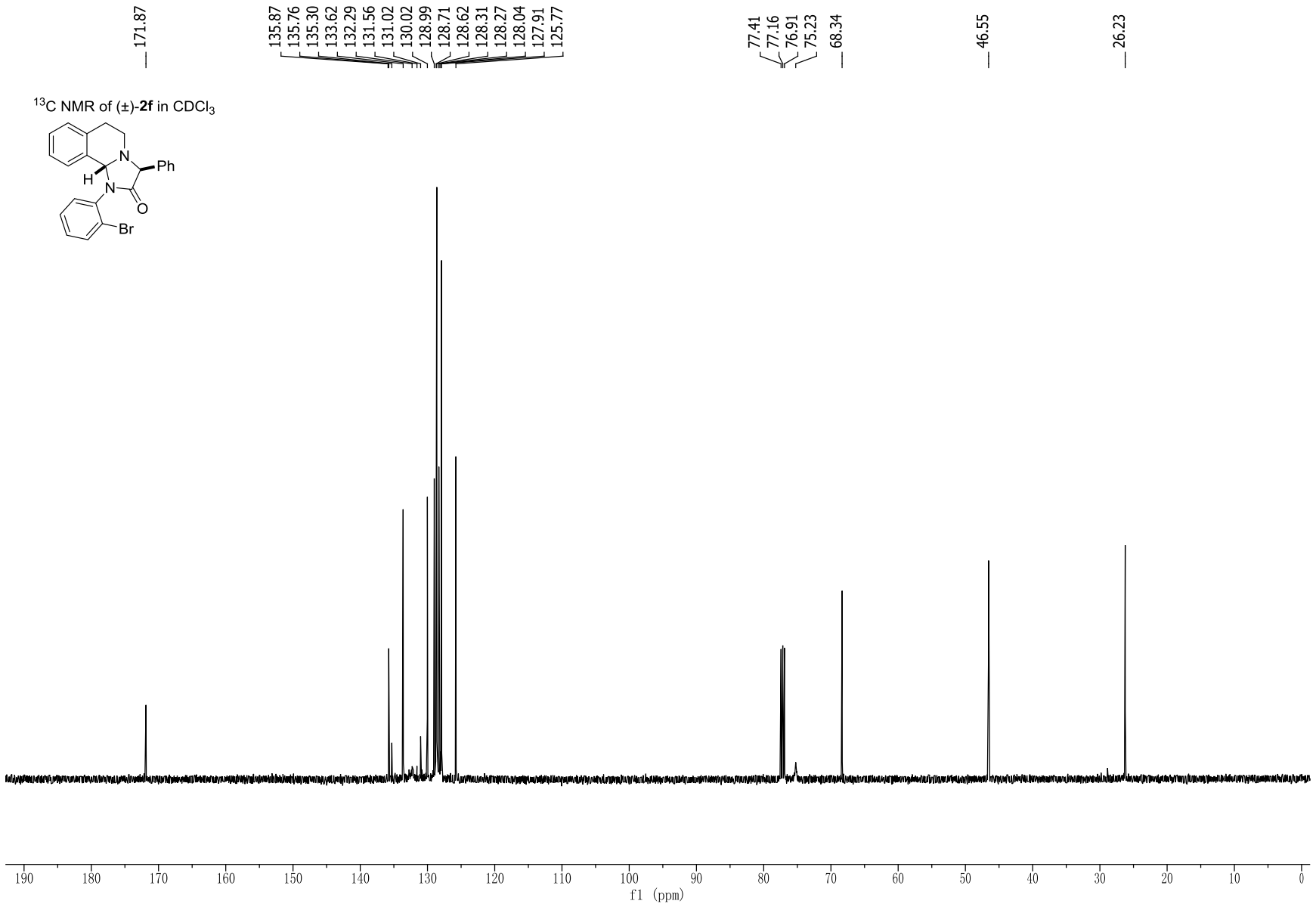

7.61 7.61 7.61 7.59 7.59 7.59 7.43 7.42 7.42 7.41 7.41 7.40 7.39 7.36 7.36 7.35 7.35 7.34 7.33 7.26 7.24 7.23 7.23 7.22 7.22 7.22 7.21 7.21 7.21 7.20 7.19 7.00 7.00 6.99 6.98 6.97 6.97 6.93 6.93 6.92 6.91 6.91 6.90 6.68 6.66 6.14 4.60 3.82 3.30 3.28 3.27 3.27 3.26 3.25 3.16 3.15 3.15 3.14 3.14 3.13 3.13 3.12 3.11 2.79 2.78 2.77 2.75 2.75 2.74
1H NMR of (±)-2d in CDCl3
Ph
N
H
N
O
MeO
1.87 1.93 1.00 3.89 1.00 1.97 1.00 1.00 1.00 2.97 1.02 2.03 1.03
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)

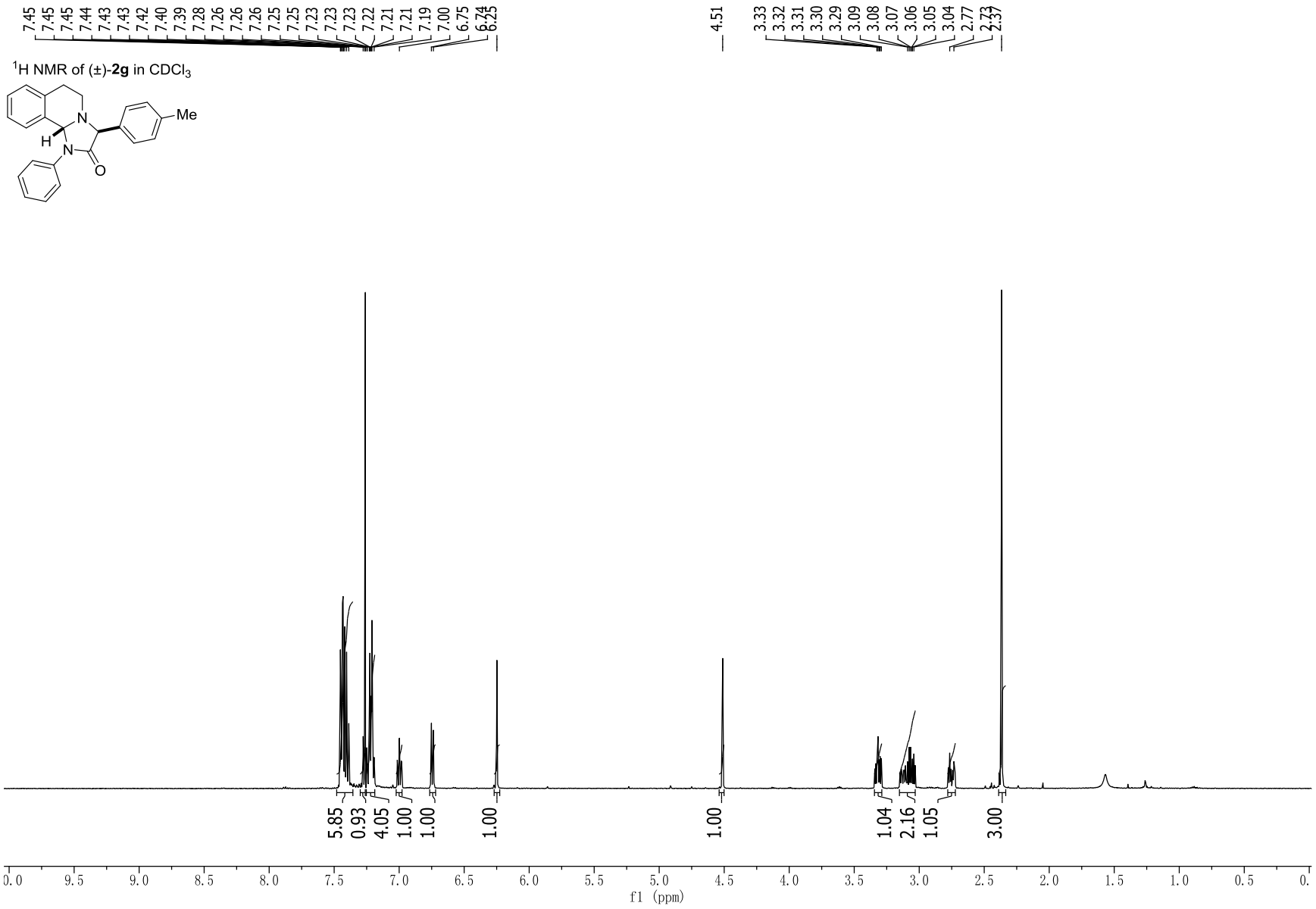

171.16
158.34
136.58
135.52
133.18
129.89
128.97
128.71
128.30
128.24
128.16
127.92
127.34
126.20
114.66
77.41
77.16
76.91
75.40
67.26
55.61
45.30
24.75
13C NMR of (±)-2d in CDCl3
N
H
N
O
Ph
MeO
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

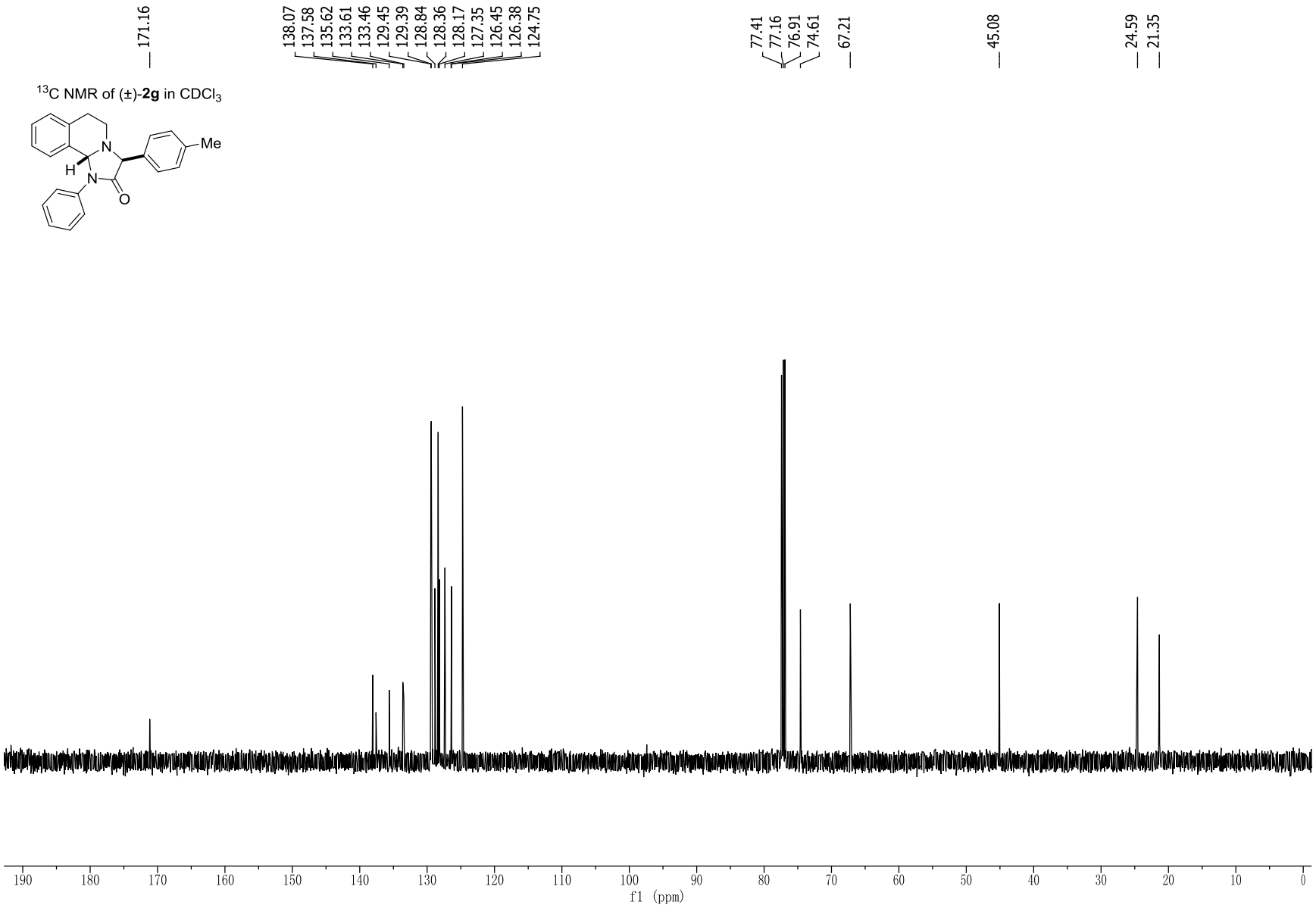

7.60 7.60 7.59 7.59 7.58 7.58 7.43 7.43 7.43 7.42 7.41 7.41 7.41 7.40 7.40 7.39 7.36 7.36 7.35 7.35 7.35 7.34 7.29 7.29 7.29 7.28 7.28 7.27 7.27 7.26 7.25 7.25 7.24 7.24 7.23 7.22 7.22 7.22 7.21 7.20 7.20 7.02 7.01 7.01 7.00 7.00 6.99 6.76 6.76 6.75 6.23 6.22 4.58 4.57 3.31 3.31 3.30 3.29 3.12 3.11 3.11 3.10 3.10 3.09 3.08 2.77 2.76 2.74 2.38
1H NMR of (±)-2e in CDCl3
N
Ph
H
N
O
Me
1.98 1.91 1.03 5.81 1.05 1.02 1.02 1.00 1.08 2.09 1.09 2.95
f1 (ppm)

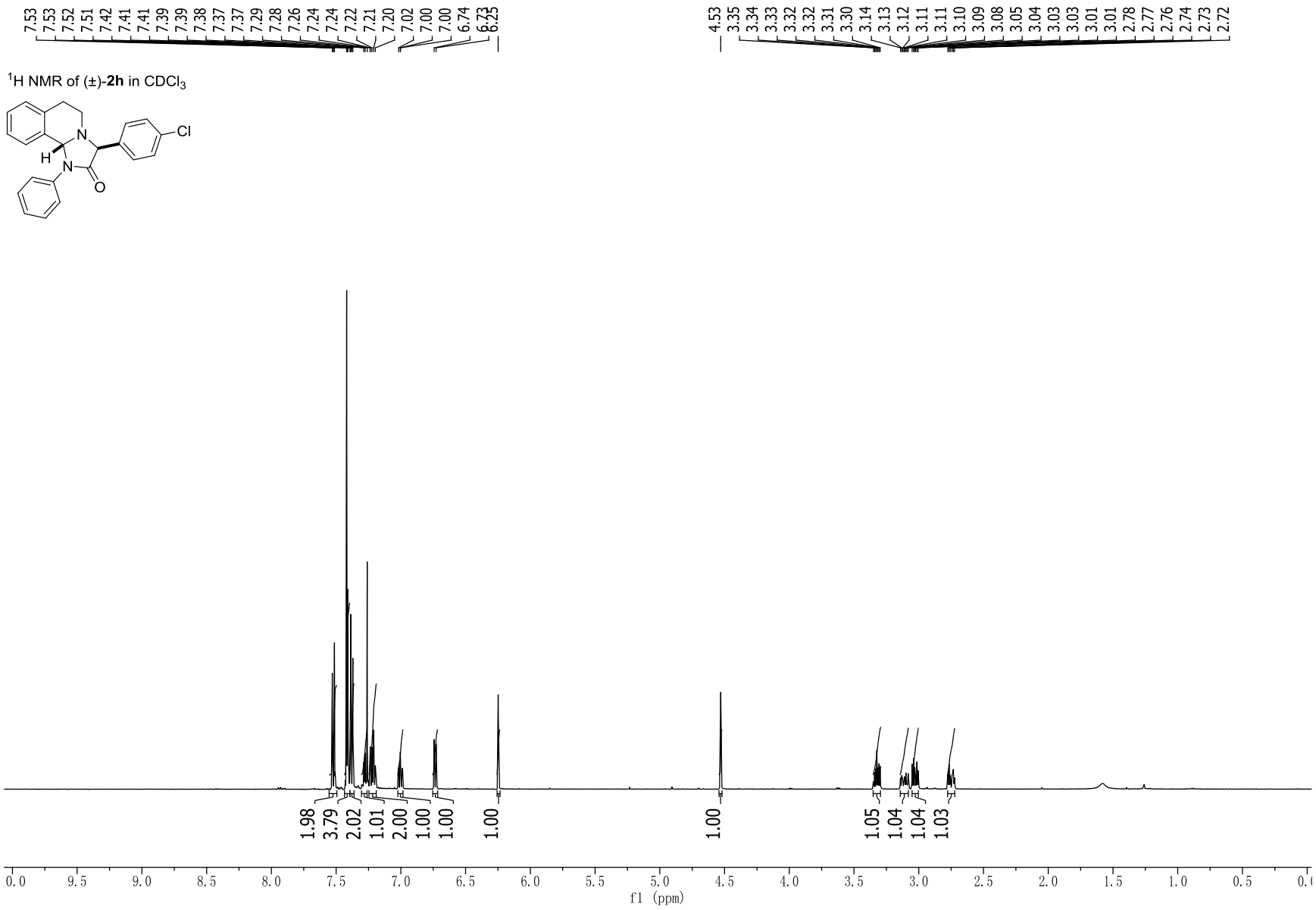

170.90
136.56
136.42
135.51
134.71
133.49
129.97
128.84
128.66
128.37
128.24
128.10
127.53
126.28
125.00
77.41
77.16
76.90
74.81
67.28
45.13
24.56
21.20
13C NMR of (±)-2e in CDCl3
N
Ph
H
N
O
Me
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

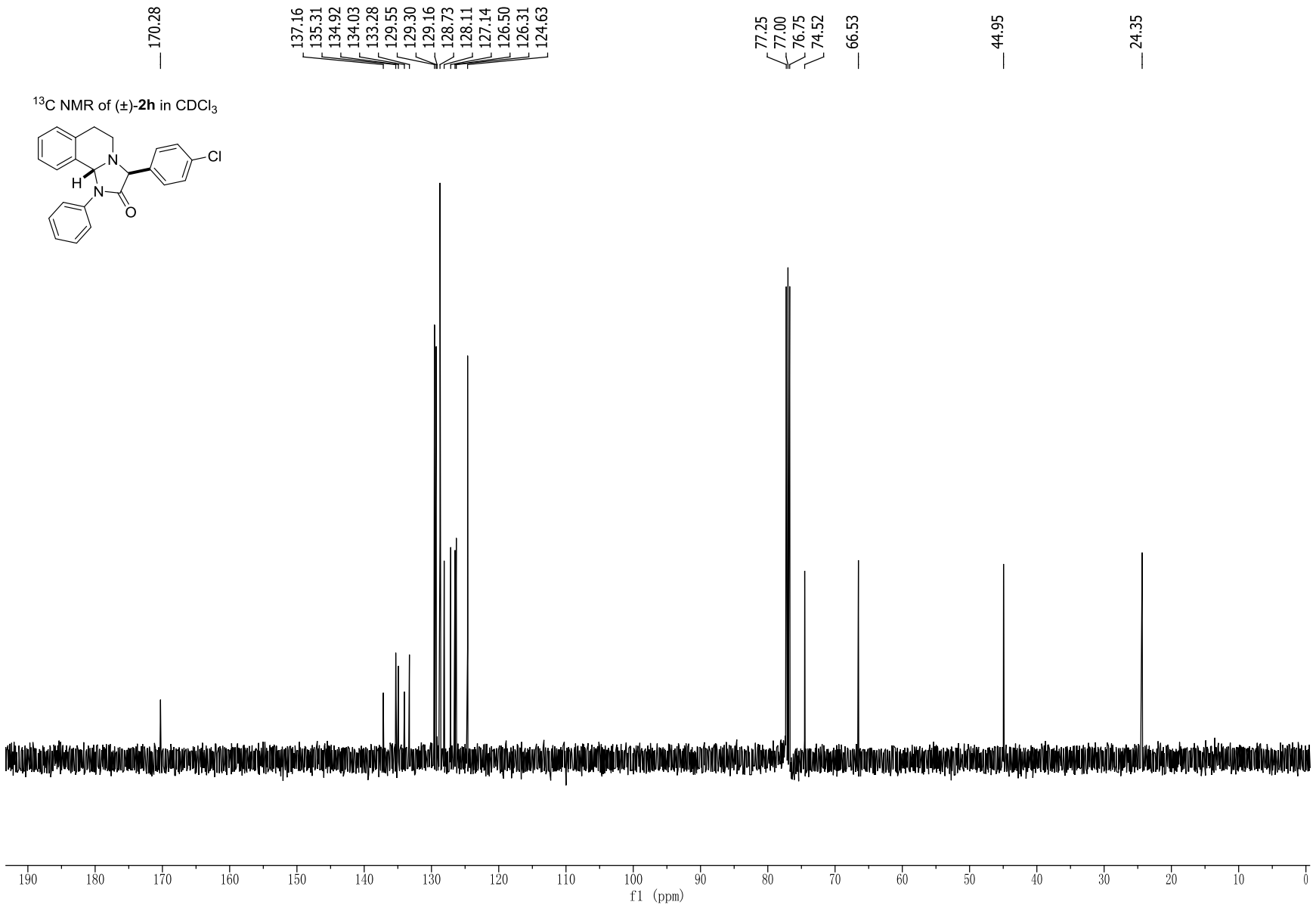

7.77
7.77
7.77
7.75
7.75
7.65
7.63
7.44
7.42
7.42
7.41
7.41
7.36
7.35
7.26
7.23
7.22
7.21
7.21
7.20
7.20
6.92
6.91
6.90
6.40
6.38
6.19
4.69
3.37
3.36
3.35
3.24
3.23
3.23
3.22
3.21
3.20
3.20
3.14
3.14
3.13
3.12
3.11
3.10
3.10
3.09
2.97
2.96
2.95
2.94
2.93
2.92
2.92
2.91
1H NMR of (±)-2f in CDCl3
N
Ph
H
N
O
Br
1.87
0.95
2.00
1.00
4.00
1.85
1.00
1.00
1.00
1.00
1.02
1.03
1.08
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
f1 (ppm)

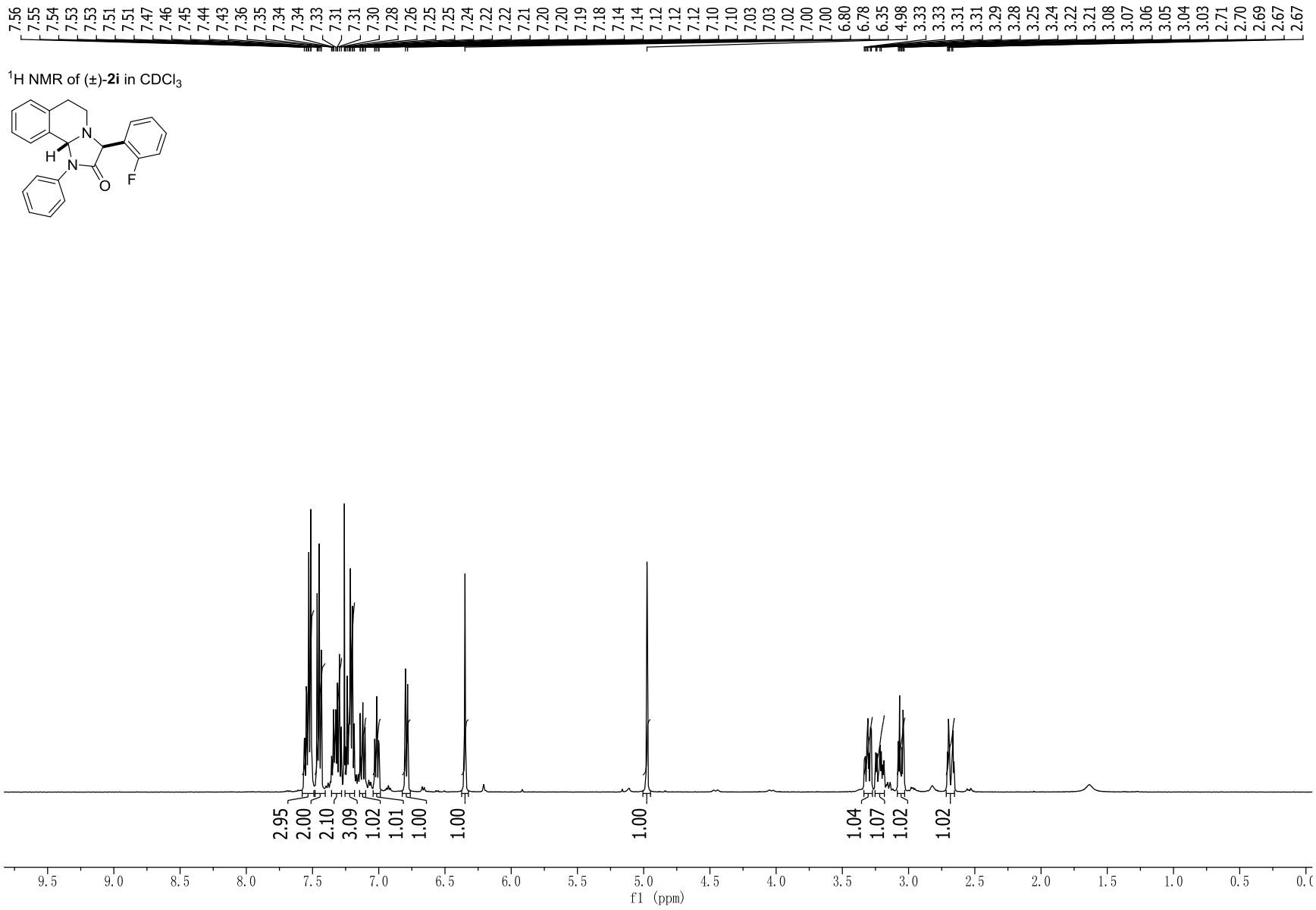

171.87
135.87
135.76
135.30
133.62
132.29
131.56
131.02
130.02
128.99
128.71
128.62
128.31
128.27
128.04
127.91
125.77
77.41
77.16
76.91
75.23
68.34
46.55
26.23
13C NMR of (±)-2f in CDCl3
N
Ph
H
N
O
Br
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

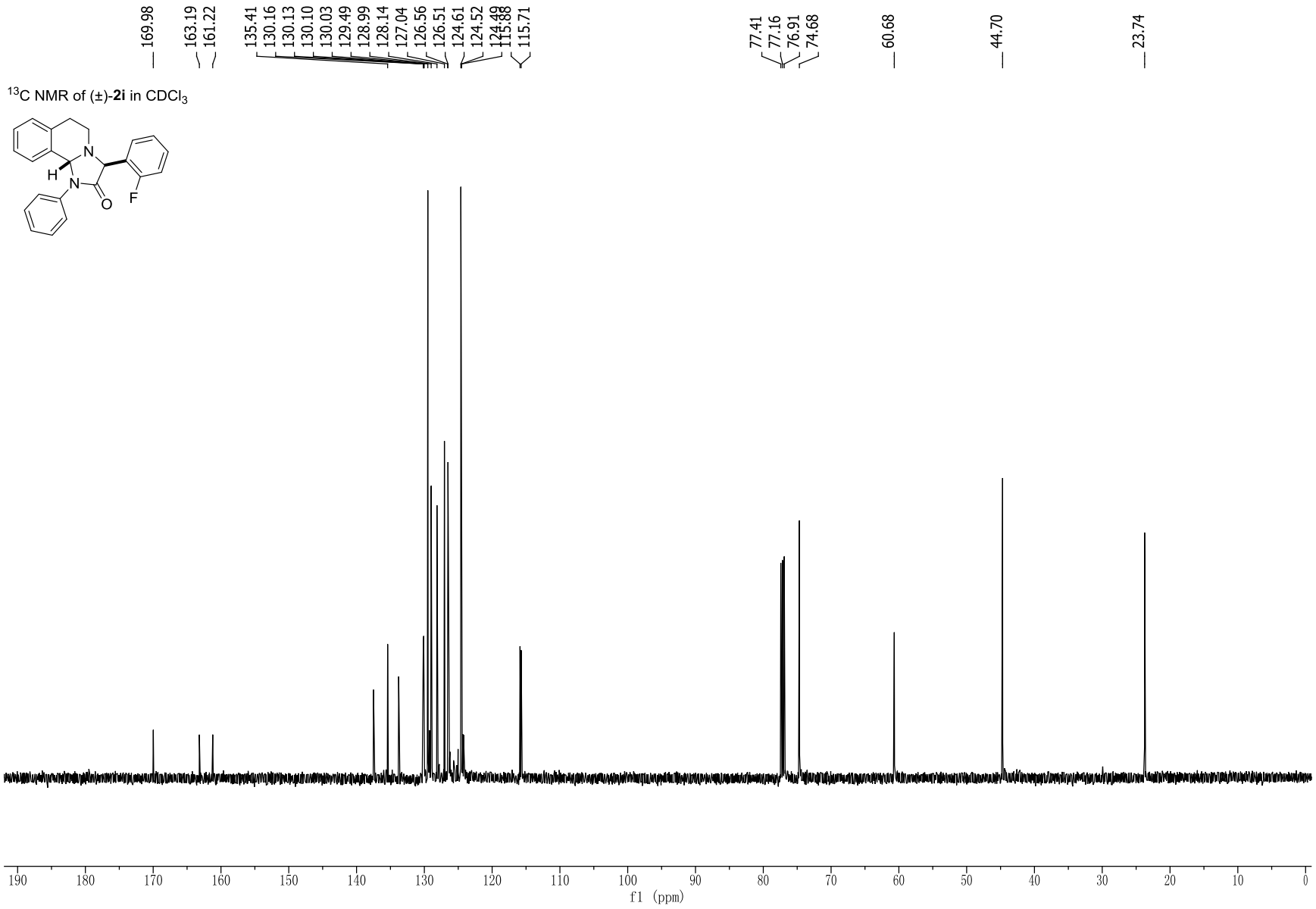

7.45
7.45
7.45
7.44
7.43
7.43
7.42
7.40
7.39
7.28
7.26
7.26
7.26
7.25
7.25
7.23
7.23
7.23
7.22
7.21
7.21
7.19
7.00
6.75
6.74
6.25
4.51
3.33
3.32
3.31
3.30
3.29
3.09
3.08
3.07
3.06
3.05
3.04
2.77
2.73
2.37
1H NMR of (±)-2g in CDCl3
Me
H
N
N
O
5.85
0.93
4.05
1.00
1.00
1.00
1.00
1.04
2.16
1.05
3.00
f1 (ppm)

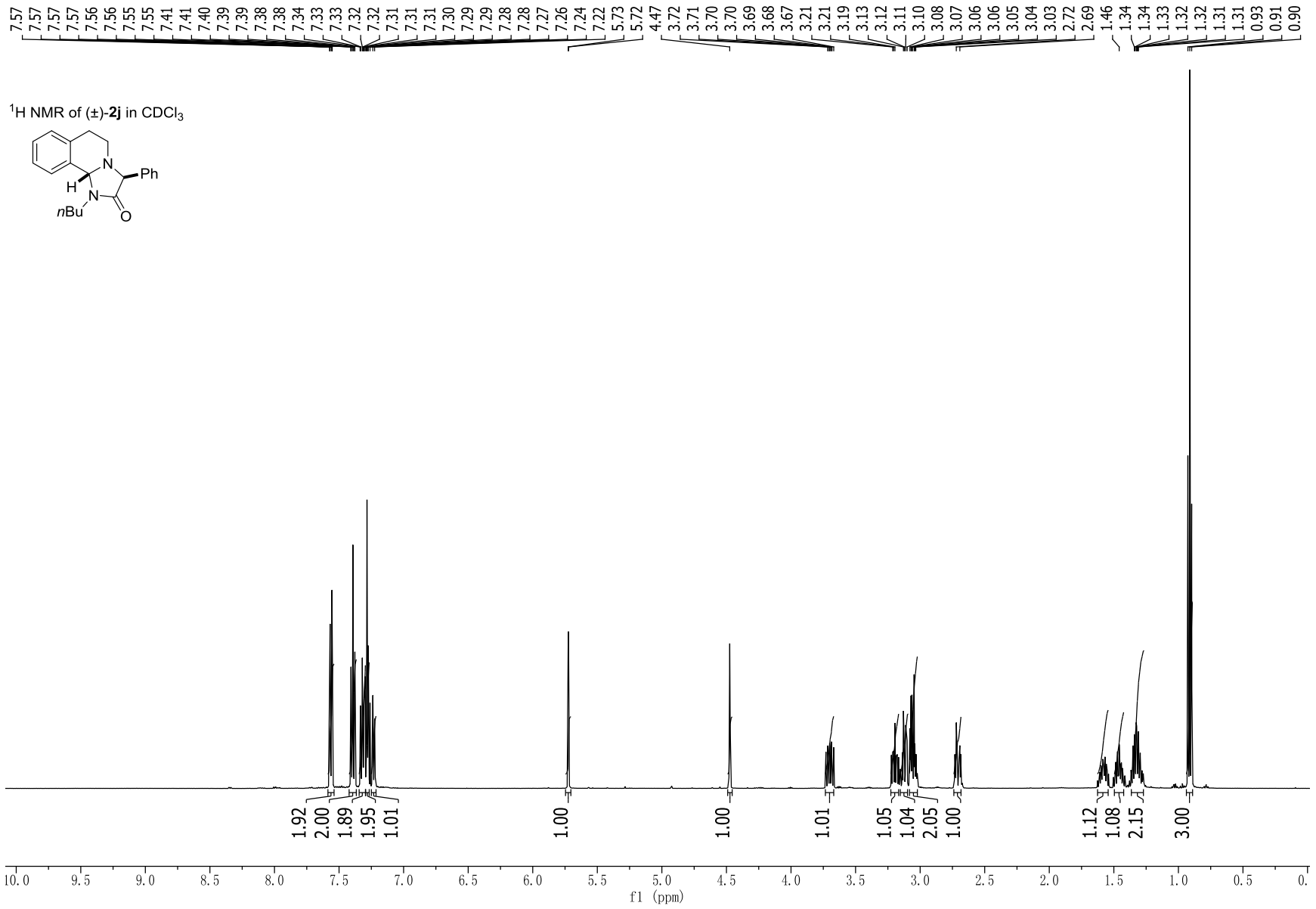

171.16
138.07
137.58
135.62
133.61
133.46
129.45
129.39
128.84
128.36
128.17
127.35
126.45
126.38
124.75
77.41
77.16
76.91
74.61
67.21
45.08
24.59
21.35
13C NMR of (±)-2g in CDCl3
Me
H
N
N
O
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

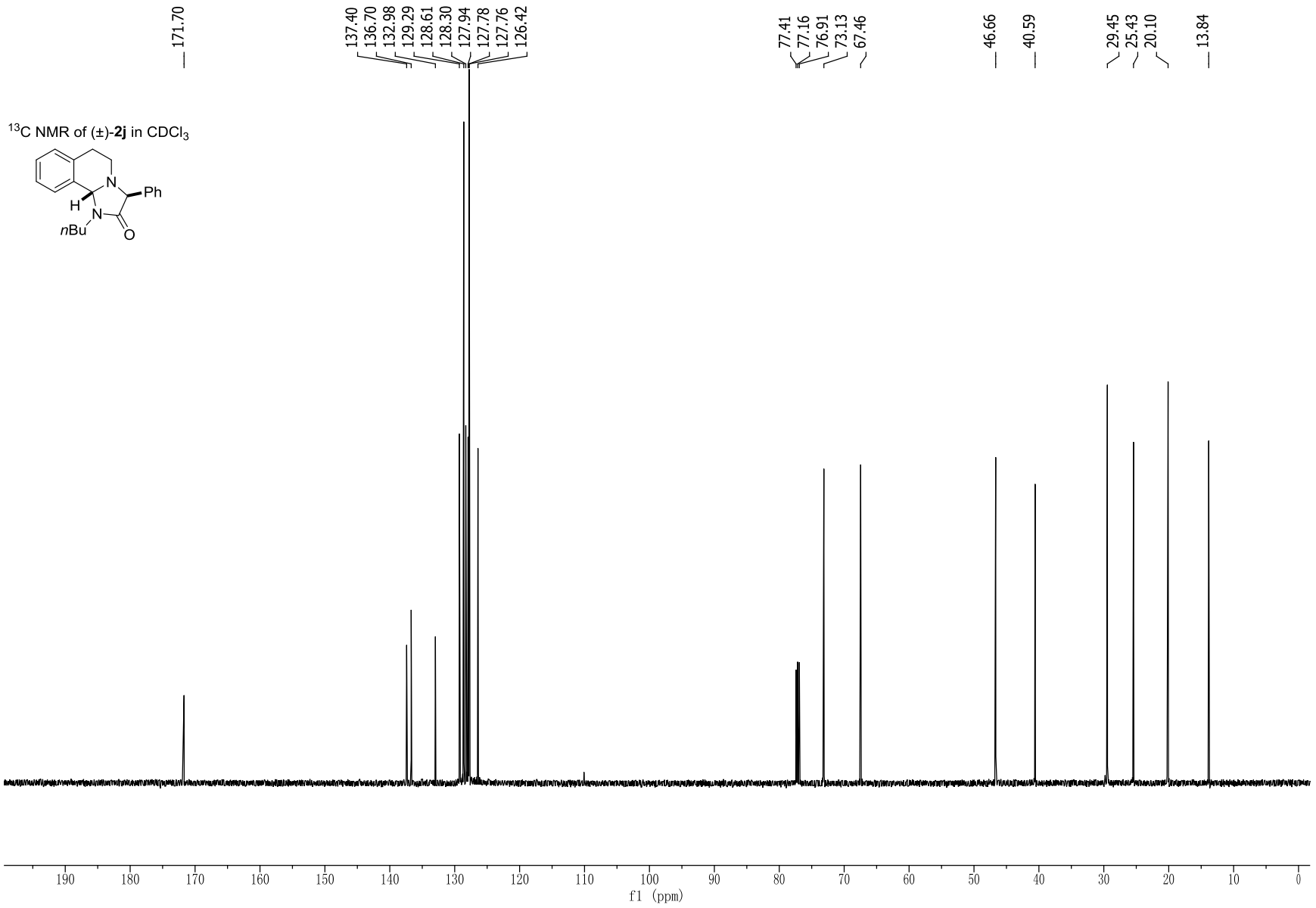

7.53
7.53
7.52
7.51
7.42
7.41
7.41
7.39
7.39
7.38
7.37
7.37
7.29
7.28
7.26
7.24
7.24
7.22
7.21
7.20
7.02
7.00
7.00
6.74
6.73
6.73
4.53
3.35
3.34
3.33
3.32
3.32
3.31
3.30
3.14
3.13
3.12
3.11
3.11
3.10
3.09
3.08
3.05
3.04
3.03
3.03
3.01
3.01
2.78
2.77
2.76
2.74
2.73
2.72
1H NMR of (±)-2h in CDCl3
Cl
H
N
N
O
1.98
3.79
2.02
1.01
2.00
1.00
1.00
1.00
1.00
1.05
1.04
1.04
1.03
f1 (ppm)

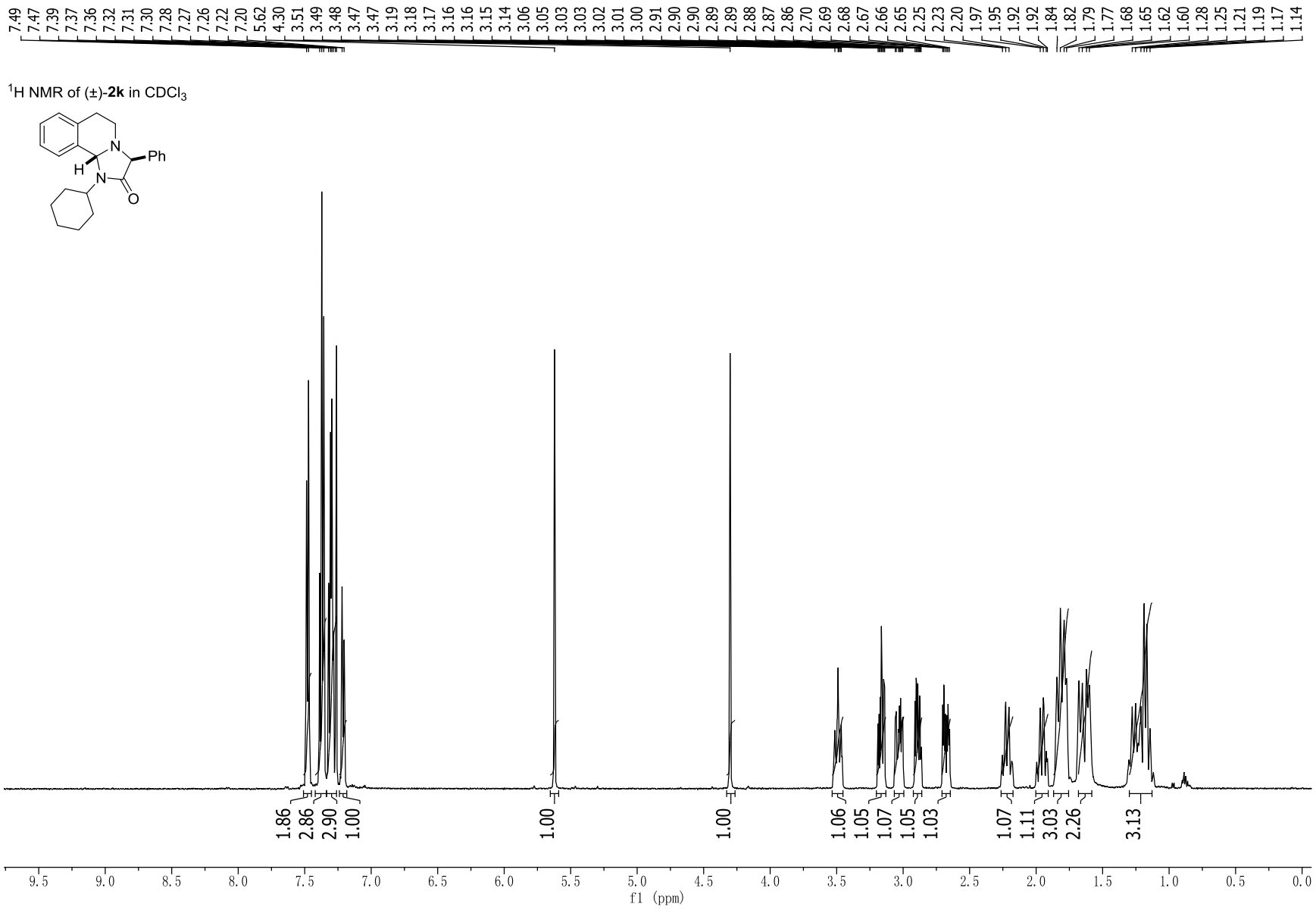

170.28
137.16
135.31
134.92
134.03
133.28
129.55
129.30
129.16
128.73
128.11
127.14
126.50
126.31
124.63
77.25
77.00
76.75
74.52
66.53
44.95
24.35
13C NMR of (±)-2h in CDCl3
Cl
H
N
N
O
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

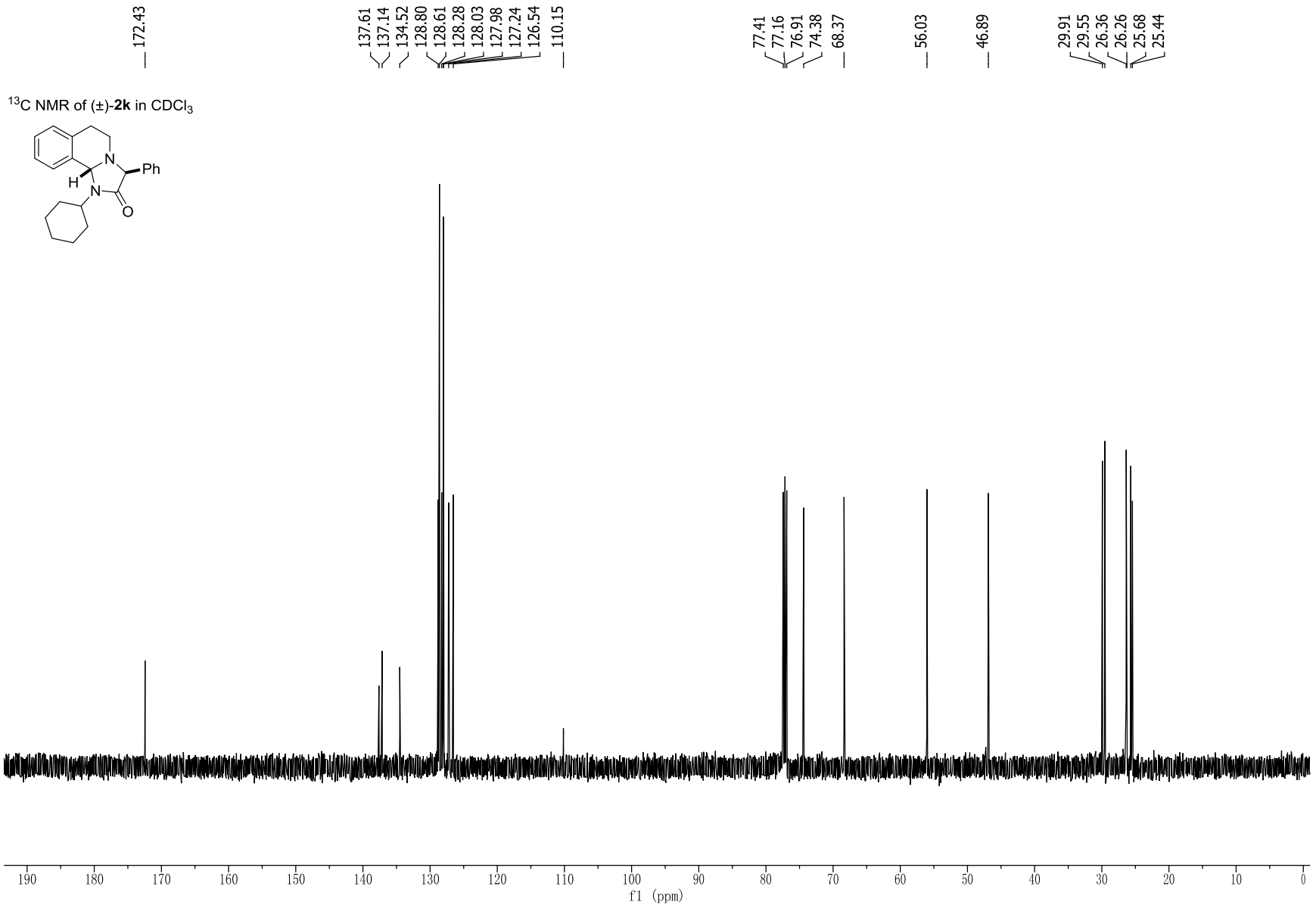

7.56 7.55 7.54 7.53 7.53 7.51 7.51 7.47 7.46 7.45 7.44 7.43 7.36 7.35 7.34 7.34 7.33 7.31 7.31 7.30 7.28 7.26 7.25 7.25 7.24 7.22 7.22 7.21 7.20 7.20 7.19 7.18 7.14 7.14 7.12 7.12 7.12 7.10 7.10 7.03 7.03 7.02 7.00 7.00 6.80 6.78 6.35 4.98 3.33 3.33 3.31 3.31 3.29 3.28 3.25 3.24 3.22 3.21 3.08 3.07 3.06 3.05 3.04 3.03 2.71 2.70 2.69 2.67 2.67
1H NMR of (±)-2i in CDCl3
H
N
N
O
F
2.95 2.00 2.10 3.09 1.02 1.01 1.00 1.00 1.00 1.04 1.07 1.02 1.02
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
f1 (ppm)

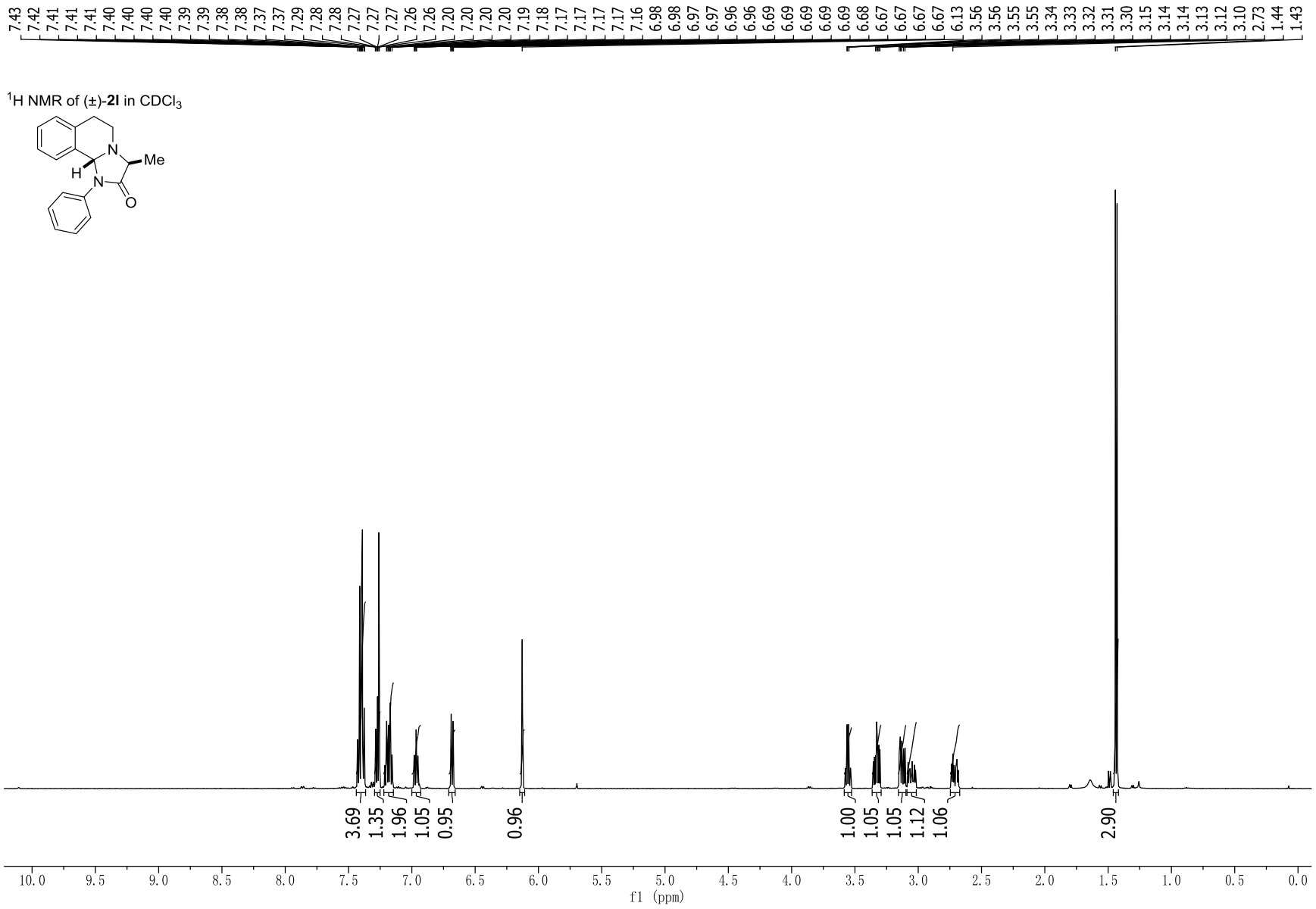

169.98
163.19
161.22
135.41
130.16
130.13
130.10
130.03
129.49
128.99
128.14
127.04
126.56
126.51
124.61
124.52
124.49
115.88
115.71
77.41
77.16
76.91
74.68
60.68
44.70
23.74
13C NMR of (±)-2i in CDCl3
N
H
N
O
F
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

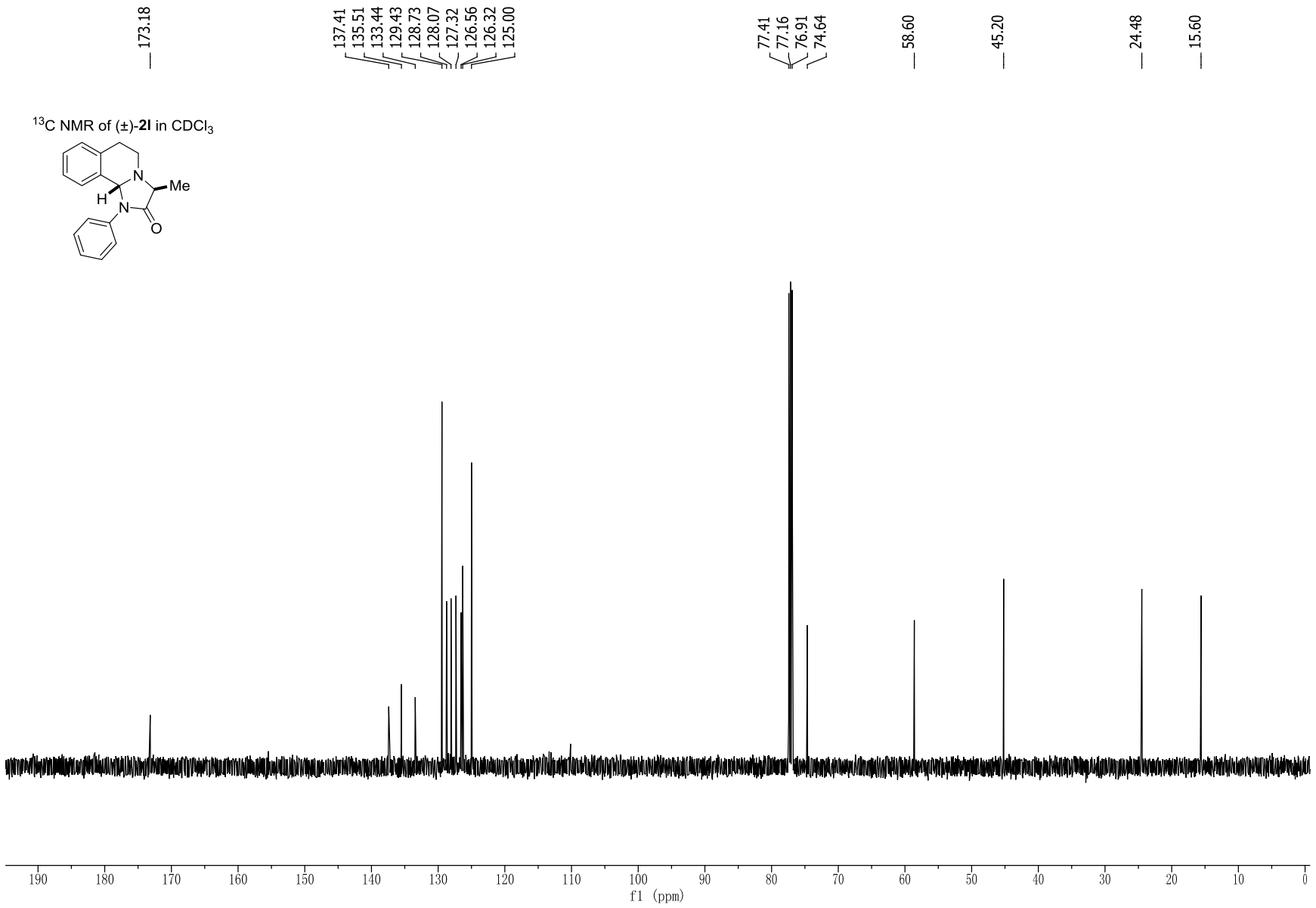

^{1}H NMR of (±)-**2j** in $CDCl_3$
Ph
H
N
N
*n*Bu
O
7.57 7.57 7.57 7.57 7.56 7.56 7.55 7.55 7.41 7.41 7.40 7.39 7.39 7.38 7.38 7.34 7.33 7.33 7.32 7.32 7.31 7.31 7.31 7.30 7.29 7.29 7.28 7.28 7.27 7.26 7.24 7.22 5.73 5.72 4.47 3.72 3.71 3.70 3.70 3.69 3.68 3.67 3.21 3.21 3.19 3.13 3.12 3.11 3.10 3.08 3.07 3.06 3.06 3.05 3.04 3.03 2.72 2.69 1.46 1.34 1.34 1.33 1.32 1.32 1.31 1.31 0.93 0.91 0.90
1.92 2.00 1.89 1.95 1.01 1.00 1.00 1.01 1.05 1.04 2.05 1.00 1.12 1.08 2.15 3.00
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.
f1 (ppm)


171.70
137.40
136.70
132.98
129.29
128.61
128.30
127.94
127.78
127.76
126.42
77.41
77.16
76.91
73.13
67.46
46.66
40.59
29.45
25.43
20.10
13.84
^{13}C NMR of (±)-**2j** in $CDCl_3$
N
Ph
H
N
*n*Bu
O
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

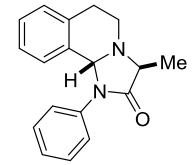

7.49 7.47 7.39 7.37 7.36 7.32 7.31 7.30 7.28 7.27 7.26 7.22 7.20 5.62 4.30 3.51 3.49 3.48 3.47 3.47 3.19 3.18 3.17 3.16 3.16 3.15 3.14 3.06 3.05 3.03 3.03 3.02 3.01 3.00 2.91 2.90 2.90 2.89 2.89 2.88 2.87 2.86 2.70 2.69 2.68 2.67 2.66 2.65 2.25 2.23 2.20 1.97 1.95 1.92 1.92 1.84 1.82 1.79 1.77 1.68 1.65 1.62 1.60 1.28 1.25 1.21 1.19 1.17 1.14
1H NMR of (±)-2k in CDCl3
N
Ph
H
N
O
1.86 2.86 2.90 1.00 1.00 1.00 1.06 1.05 1.07 1.05 1.03 1.07 1.11 3.03 2.26 3.13
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)


172.43
137.61
137.14
134.52
128.80
128.61
128.28
128.03
127.98
127.24
126.54
110.15
77.41
77.16
76.91
74.38
68.37
56.03
46.89
29.91
29.55
26.36
26.26
25.68
25.44
13C NMR of (±)-2k in CDCl3
Ph
H
N
N
O
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

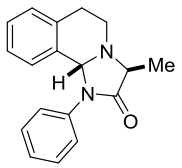

7.43 7.42 7.41 7.41 7.41 7.40 7.40 7.40 7.40 7.39 7.39 7.38 7.38 7.37 7.37 7.29 7.28 7.28 7.27 7.27 7.27 7.26 7.26 7.20 7.20 7.20 7.20 7.19 7.18 7.17 7.17 7.17 7.17 7.16 6.98 6.98 6.97 6.97 6.96 6.96 6.69 6.69 6.69 6.69 6.68 6.67 6.67 6.67 6.67 6.13 3.56 3.56 3.55 3.55 3.34 3.33 3.32 3.31 3.30 3.15 3.14 3.14 3.13 3.12 3.10 2.73 1.44 1.43
^{1}H NMR of (±)-**2l** in $CDCl_3$
N
Me
H
N
O
3.69 1.35 1.96 1.05 0.95 0.96 1.00 1.05 1.05 1.12 1.06 2.90
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)

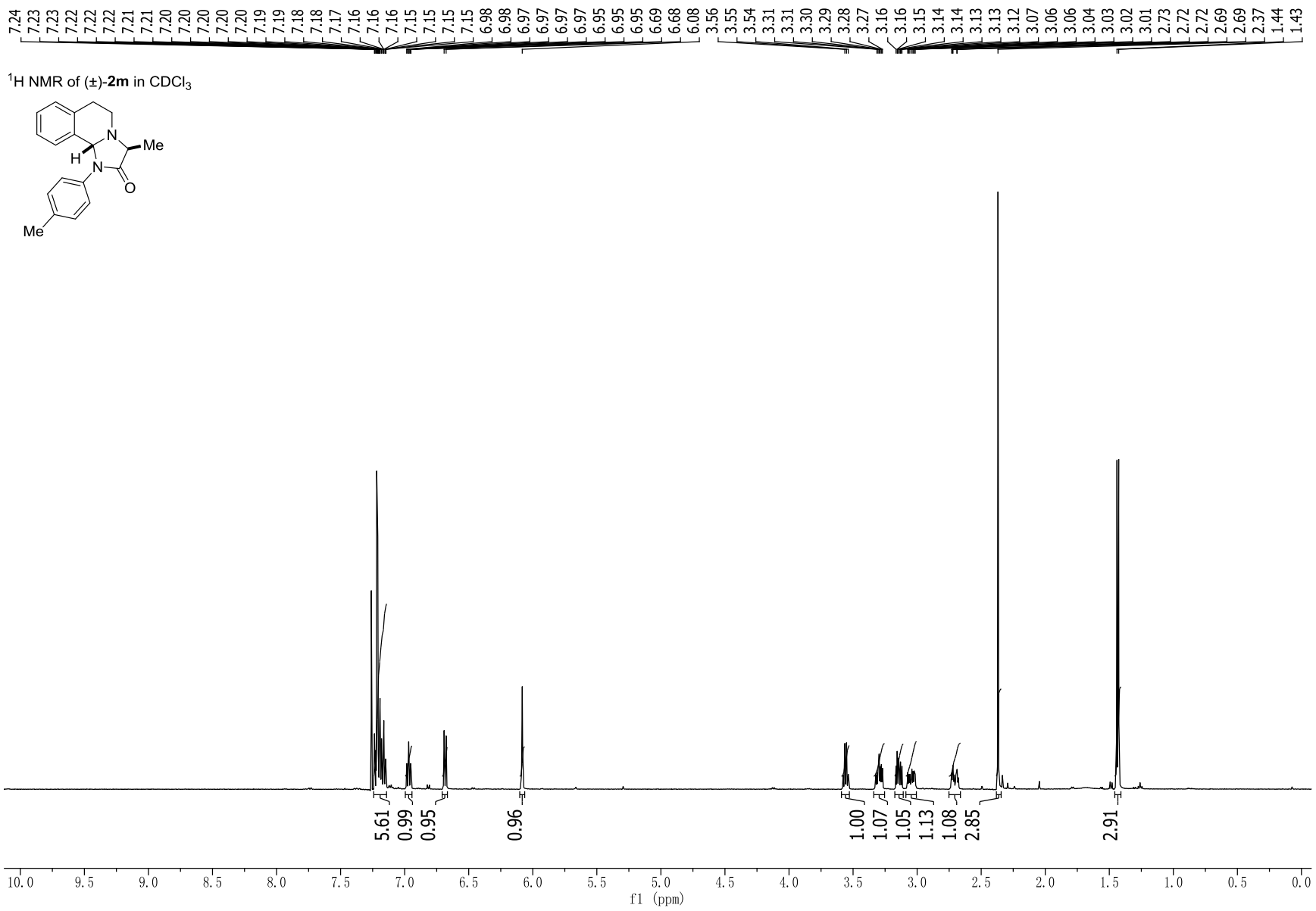

173.18
137.41
135.51
133.44
129.43
128.73
128.07
127.32
126.56
126.32
125.00
77.41
77.16
76.91
74.64
58.60
45.20
24.48
15.60
13C NMR of (±)-2l in CDCl3
N
Me
H
N
O
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

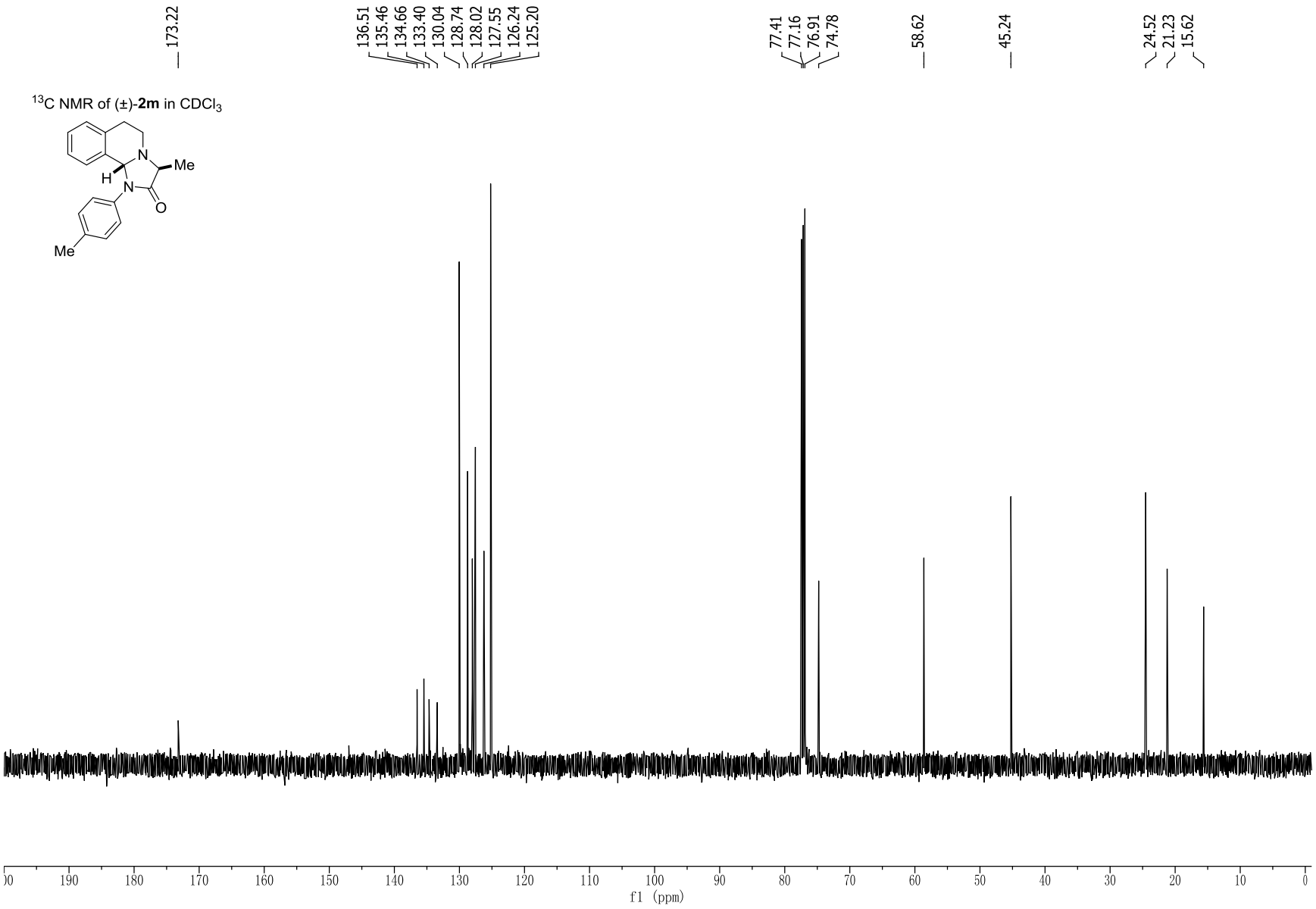
GCOSY of (±)-**2l** in $CDCl_3$

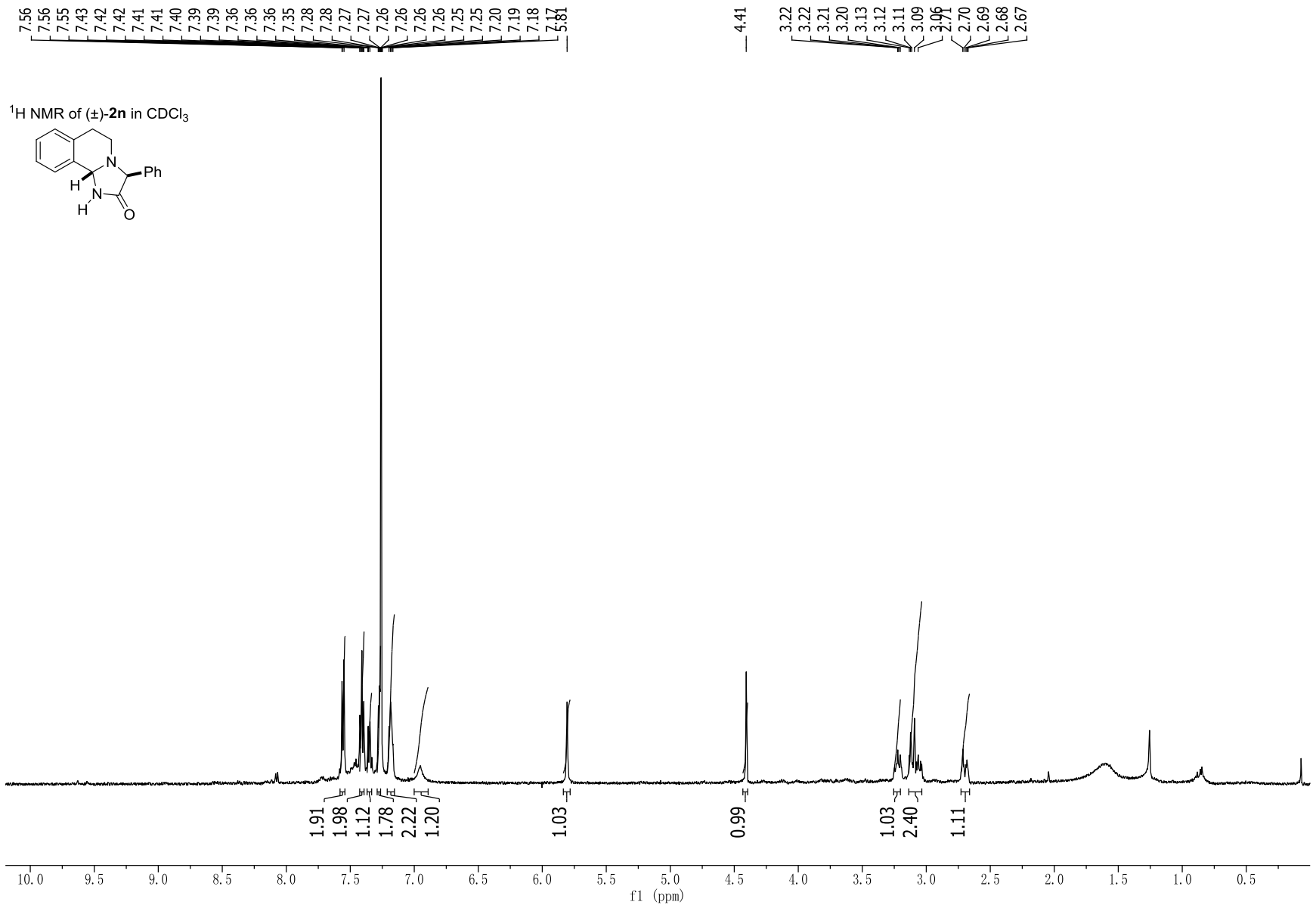


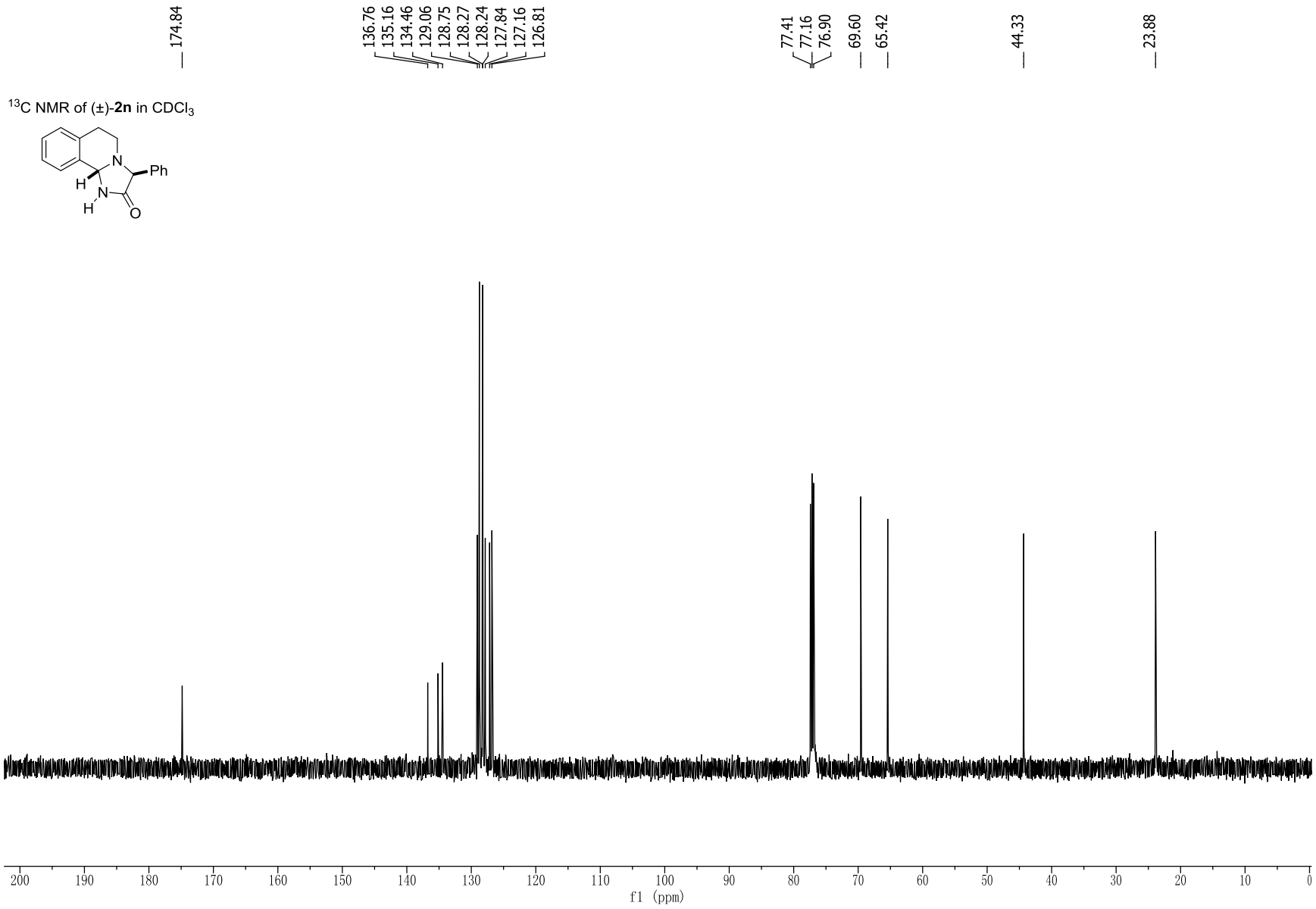
NOESY of (±)-**2l** in $CDCl_3$

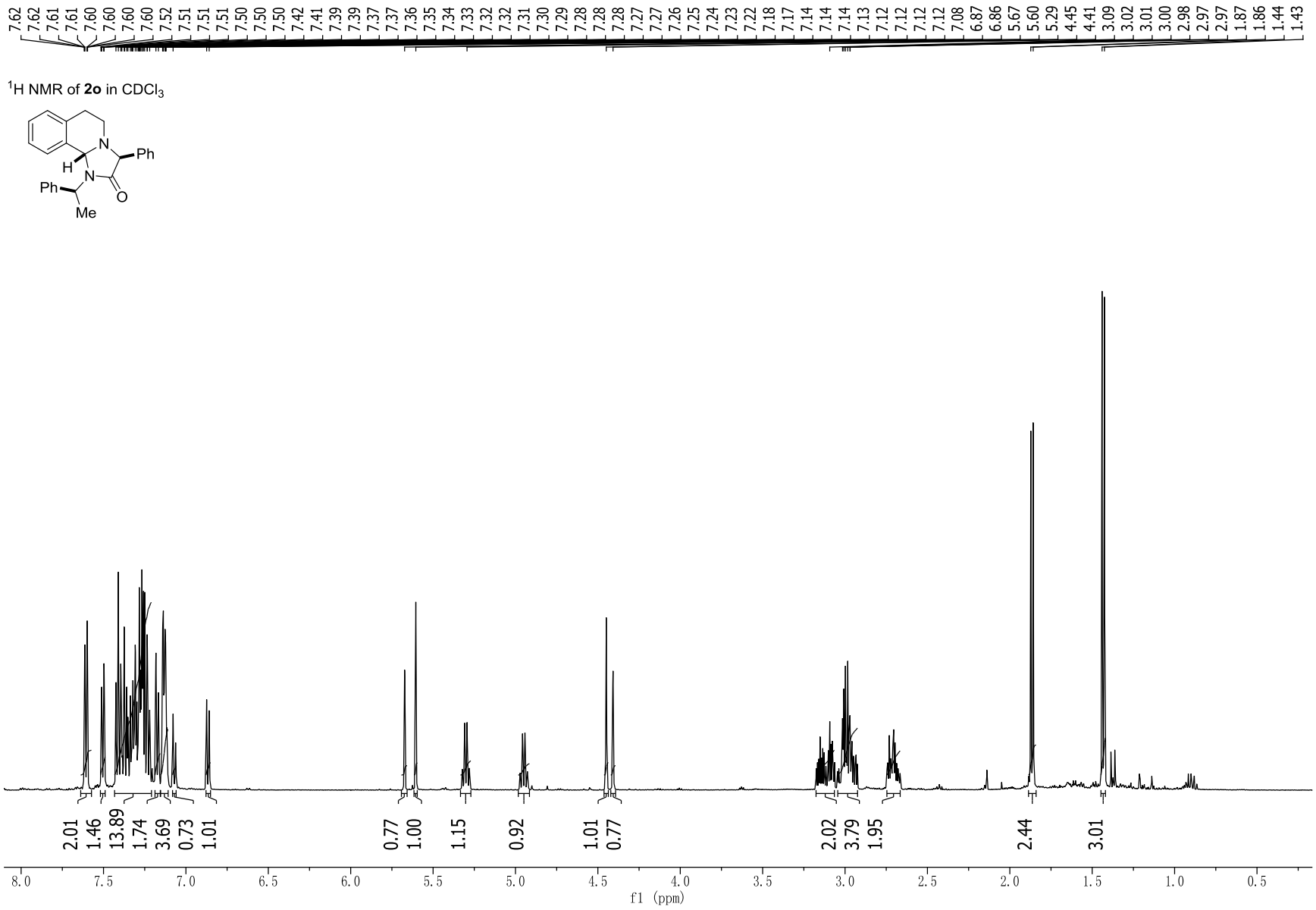


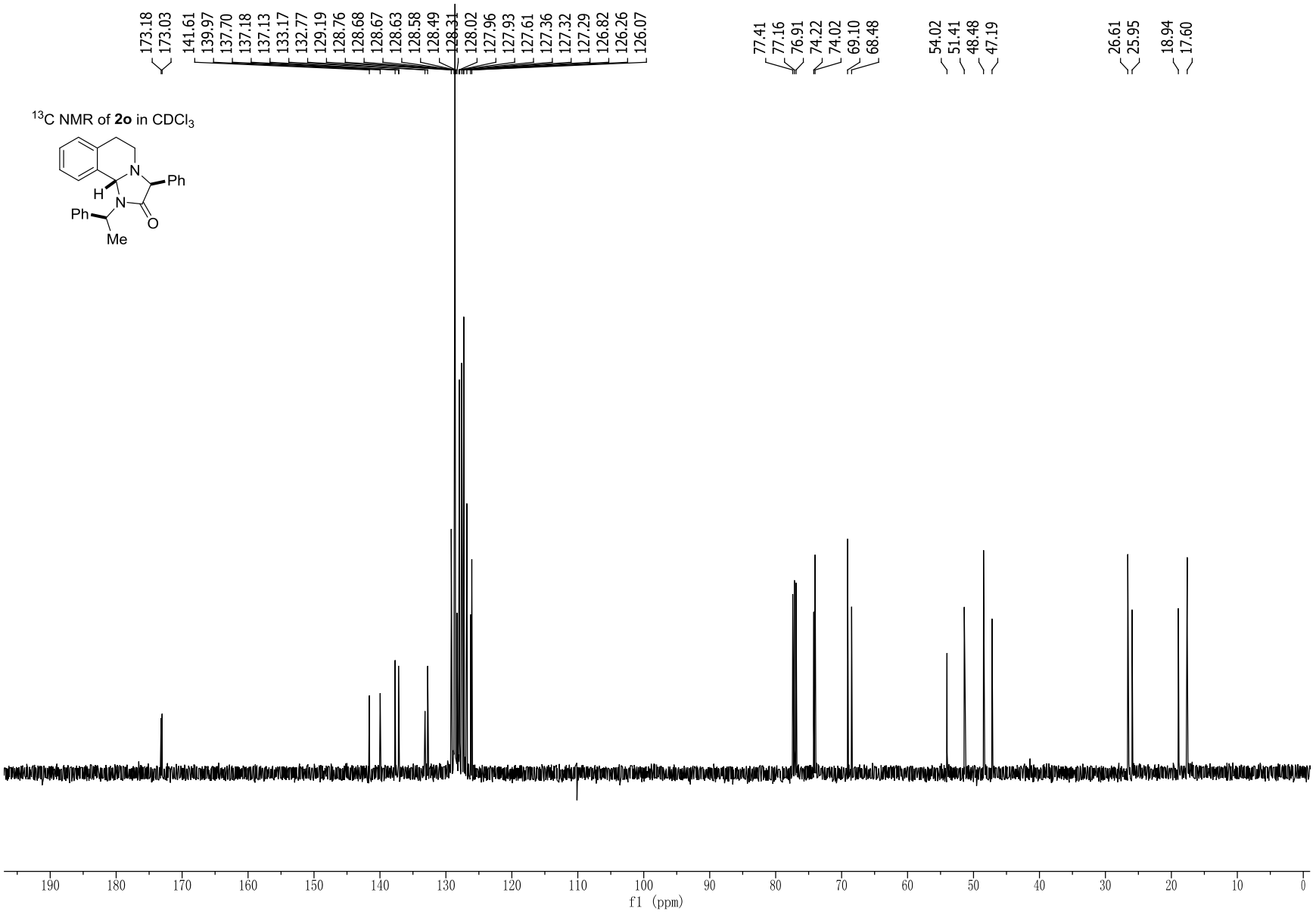

7.24 7.23 7.23 7.22 7.22 7.22 7.21 7.21 7.20 7.20 7.20 7.20 7.20 7.19 7.19 7.18 7.18 7.17 7.16 7.16 7.16 7.15 7.15 7.15 7.15 6.98 6.98 6.97 6.97 6.97 6.95 6.95 6.95 6.69 6.68 6.08 3.56 3.55 3.54 3.31 3.31 3.30 3.29 3.28 3.27 3.16 3.16 3.15 3.14 3.14 3.13 3.13 3.12 3.07 3.06 3.06 3.04 3.03 3.02 3.01 2.73 2.72 2.72 2.69 2.69 2.37 1.44 1.43
1H NMR of (±)-2m in CDCl3
N
Me
H
N
O
Me
5.61
0.99
0.95
0.96
1.00
1.07
1.05
1.13
1.08
2.85
2.91
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)

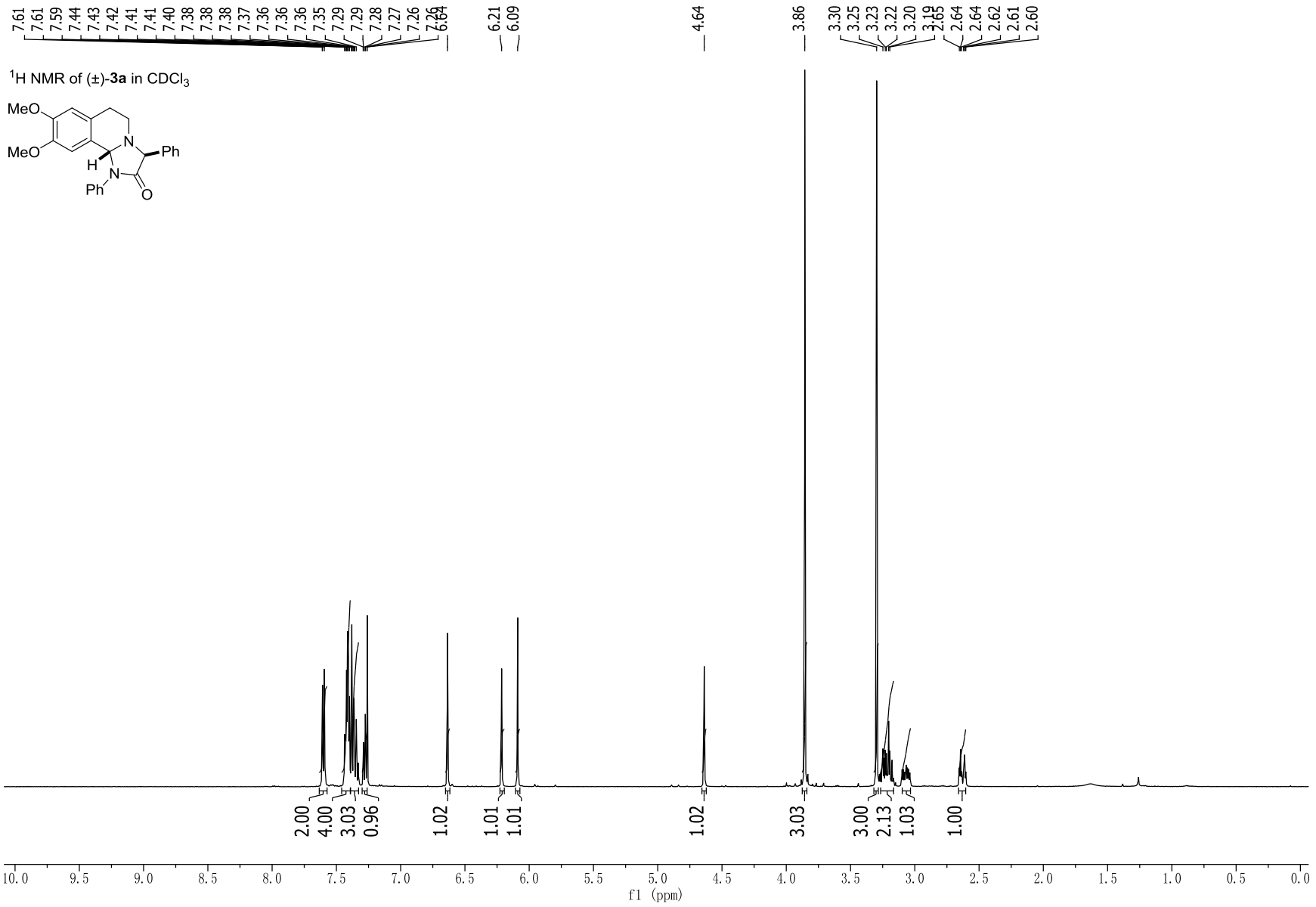

173.22
136.51
135.46
134.66
133.40
130.04
128.74
128.02
127.55
126.24
125.20
77.41
77.16
76.91
74.78
58.62
45.24
24.52
21.23
15.62
13C NMR of (±)-2m in CDCl3
N
Me
H
N
O
Me
f1 (ppm)

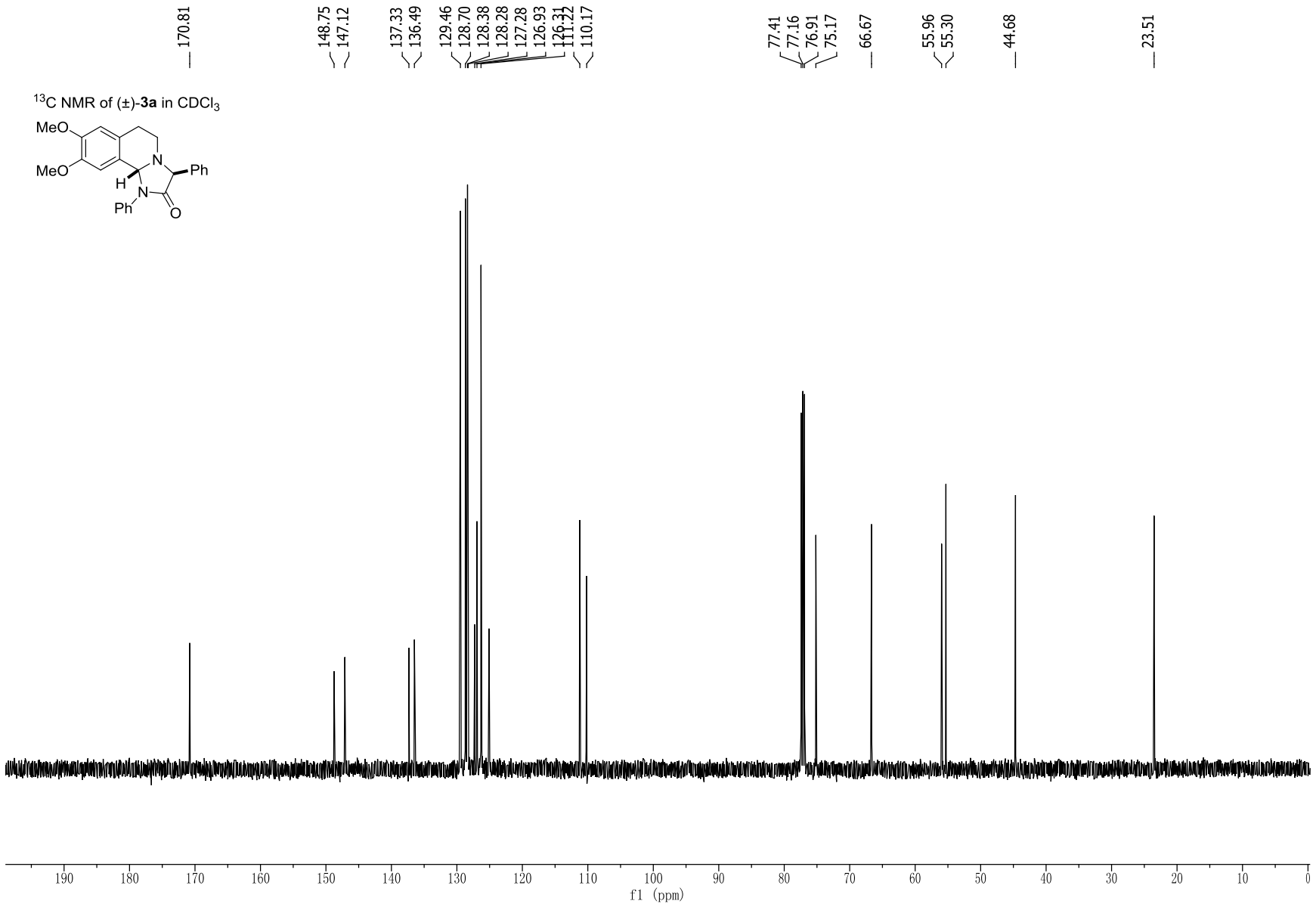

7.56 7.56 7.55 7.43 7.42 7.42 7.41 7.41 7.40 7.39 7.39 7.36 7.36 7.36 7.35 7.28 7.28 7.27 7.27 7.26 7.26 7.26 7.26 7.25 7.25 7.20 7.19 7.18 7.17 5.81
4.41
3.22 3.22 3.21 3.20 3.13 3.12 3.11 3.09 3.06 2.71 2.70 2.69 2.68 2.67
1H NMR of (±)-2n in CDCl3
Ph
H
N
H
N
O
1.91 1.98 1.12 1.78 2.22 1.20 1.03 0.99 1.03 2.40 1.11
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
f1 (ppm)

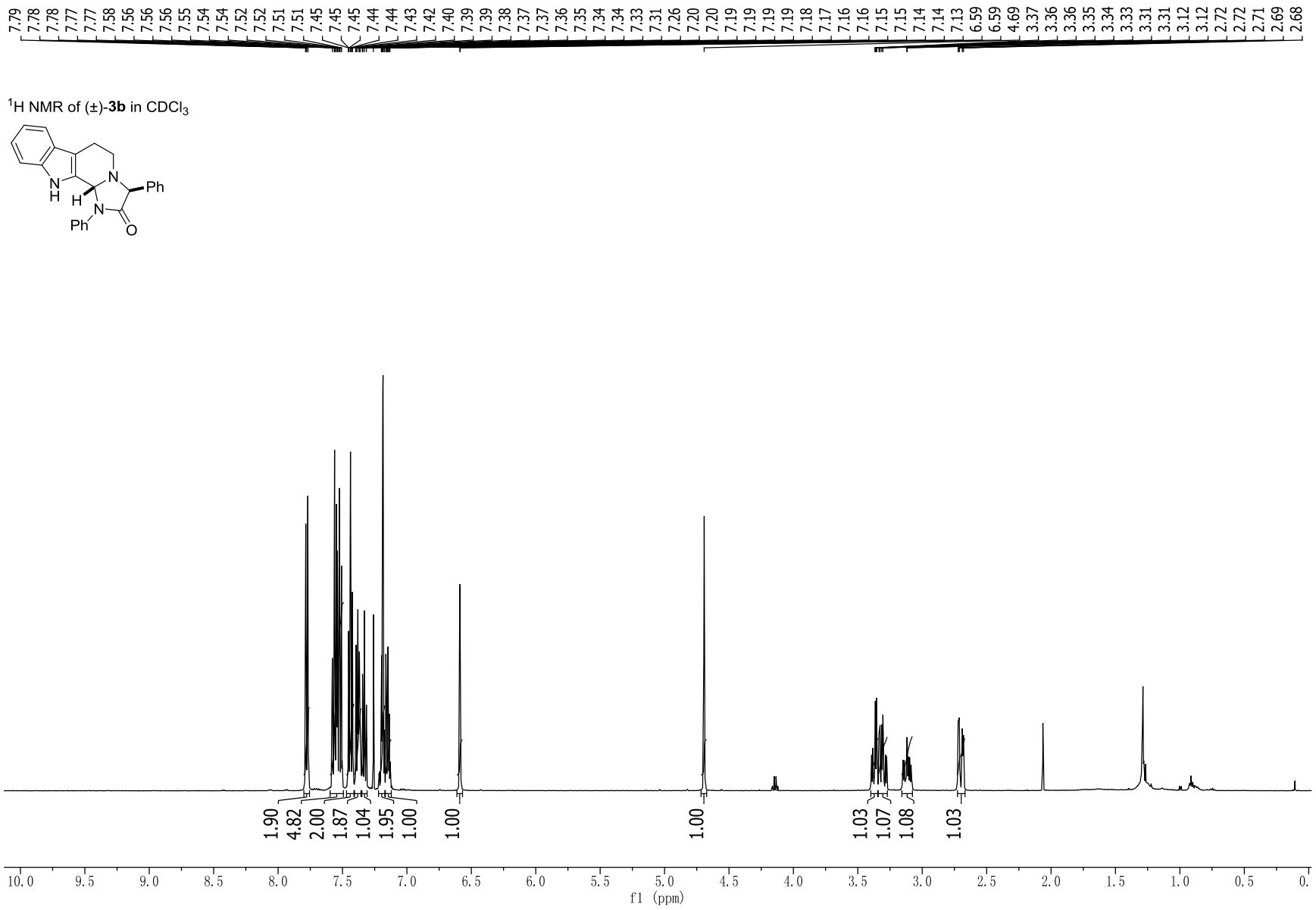

174.84
136.76
135.16
134.46
129.06
128.75
128.27
128.24
127.84
127.16
126.81
77.41
77.16
76.90
69.60
65.42
44.33
23.88
13C NMR of (±)-2n in CDCl3
Ph
H
N
H
N
O
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

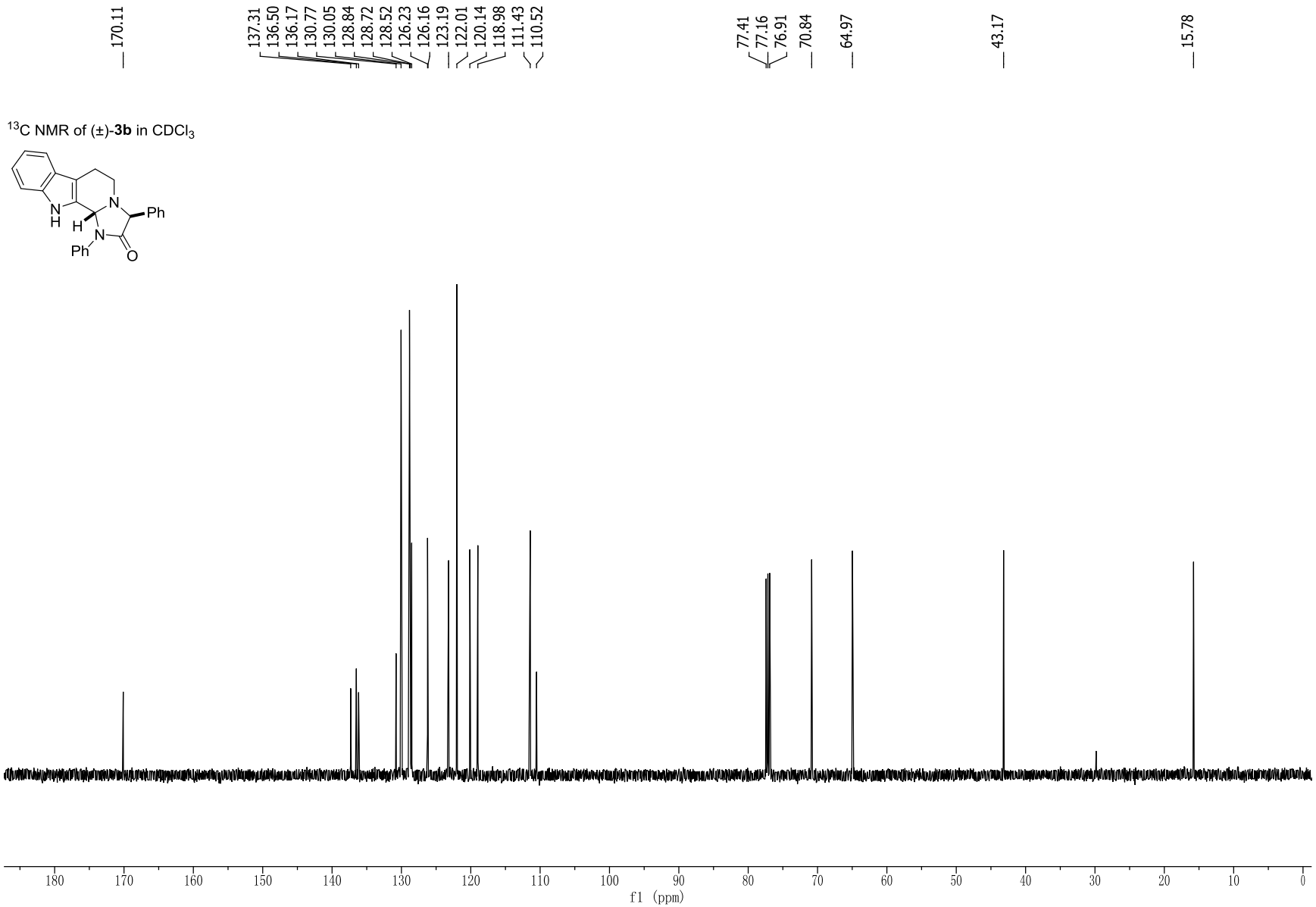

7.62 7.62 7.61 7.61 7.60 7.60 7.60 7.60 7.52 7.51 7.51 7.50 7.50 7.50 7.42 7.41 7.39 7.39 7.37 7.37 7.36 7.35 7.34 7.33 7.32 7.32 7.31 7.30 7.29 7.28 7.28 7.28 7.27 7.27 7.26 7.25 7.24 7.23 7.22 7.18 7.17 7.14 7.14 7.14 7.13 7.12 7.12 7.12 7.12 7.08 6.87 6.86 5.67 5.60 5.29 4.45 4.41 3.09 3.02 3.01 3.00 2.98 2.97 1.87 1.86 1.44 1.43
1H NMR of 2o in CDCl3
N
Ph
H
N
Ph
O
Me
2.01 1.46 13.89 1.74 3.69 0.73 1.01 0.77 1.00 1.15 0.92 1.01 0.77 2.02 3.79 1.95 2.44 3.01
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
f1 (ppm)


173.18
173.03
141.61
139.97
137.70
137.18
137.13
133.17
132.77
129.19
128.76
128.68
128.67
128.63
128.58
128.49
128.31
128.02
127.96
127.93
127.61
127.36
127.32
127.29
126.82
126.26
126.07
77.41
77.16
76.91
74.22
74.02
69.10
68.48
54.02
51.41
48.48
47.19
26.61
25.95
18.94
17.60
13C NMR of 2o in CDCl3
N
Ph
H
N
Ph
O
Me
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

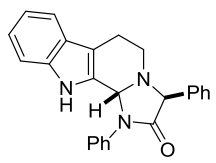

7.61
7.61
7.59
7.44
7.43
7.42
7.41
7.41
7.40
7.38
7.38
7.38
7.37
7.36
7.36
7.36
7.35
7.29
7.29
7.28
7.27
7.26
7.25
6.84
6.21
6.09
4.64
3.86
3.30
3.25
3.23
3.22
3.20
3.19
2.65
2.64
2.64
2.62
2.61
2.60
^{1}H NMR of (±)-**3a** in $CDCl_3$
MeO
MeO
N
Ph
H
N
Ph
O
2.00
4.00
3.03
0.96
1.02
1.01
1.01
1.02
3.03
3.00
2.13
1.03
1.00
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)


170.81
148.75
147.12
137.33
136.49
129.46
128.70
128.38
128.28
127.28
126.93
126.31
111.22
110.17
77.41
77.16
76.91
75.17
66.67
55.96
55.30
44.68
23.51
13C NMR of (±)-3a in CDCl3
MeO
MeO
H
N
N
Ph
Ph
O
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

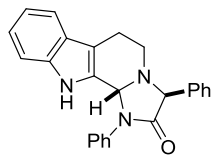

7.79 7.78 7.78 7.77 7.77 7.58 7.56 7.56 7.56 7.55 7.54 7.54 7.52 7.52 7.51 7.51 7.45 7.45 7.45 7.44 7.44 7.43 7.42 7.40 7.39 7.39 7.38 7.37 7.37 7.36 7.35 7.34 7.34 7.33 7.31 7.26 7.20 7.20 7.19 7.19 7.19 7.19 7.18 7.17 7.16 7.16 7.15 7.15 7.14 7.14 7.13 6.59 6.59 4.69 3.37 3.36 3.36 3.35 3.34 3.33 3.31 3.31 3.12 3.12 2.72 2.72 2.71 2.69 2.68
1H NMR of (±)-3b in CDCl3
N
H
H
N
Ph
N
Ph
O
1.90 4.82 2.00 1.87 1.04 1.95 1.00 1.00 1.00 1.03 1.07 1.08 1.03
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.
f1 (ppm)

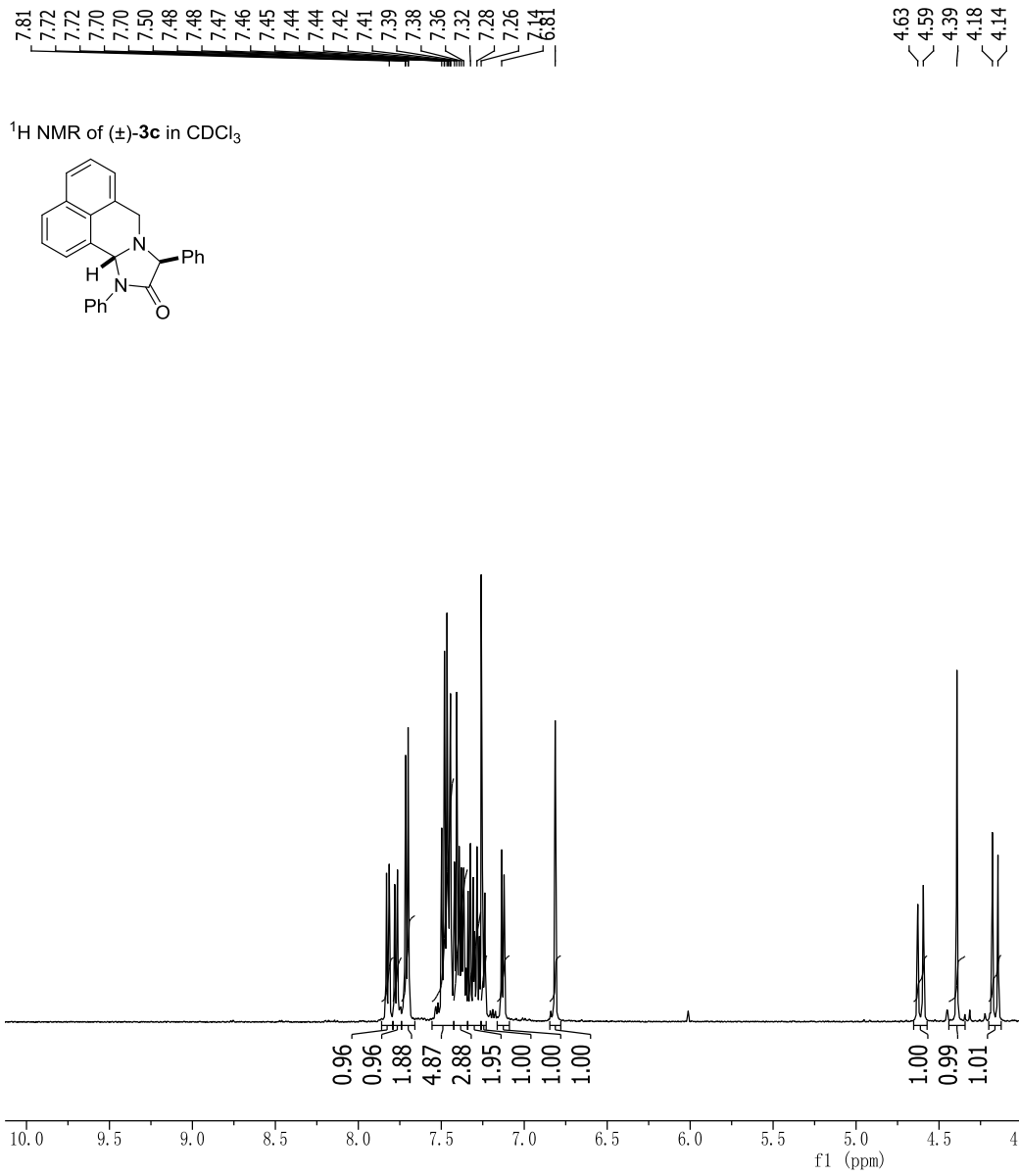

170.11
137.31
136.50
136.17
130.77
130.05
128.84
128.72
128.52
126.23
126.16
123.19
122.01
120.14
118.98
111.43
110.52
77.41
77.16
76.91
70.84
64.97
43.17
15.78
13C NMR of (±)-3b in CDCl3
N
H
H
N
N
Ph
Ph
O
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

GCOSY of (±)-**3b** in $CDCl_3$

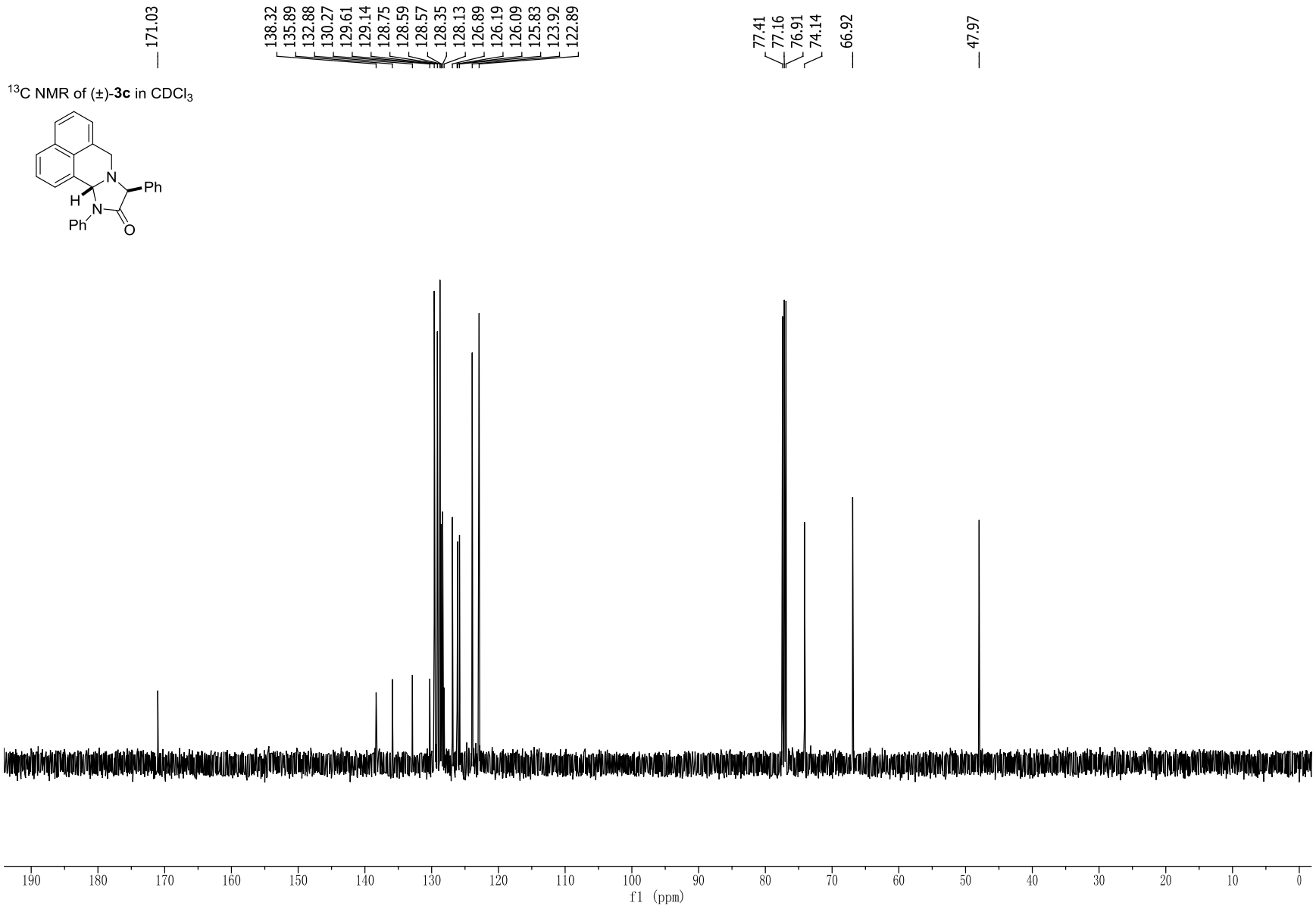


NOESY of (±)-**3b** in $CDCl_3$

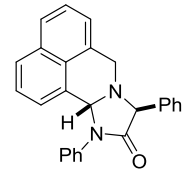




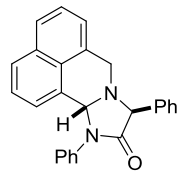
^{1}H NMR of (±)-**3c** in $CDCl_3$

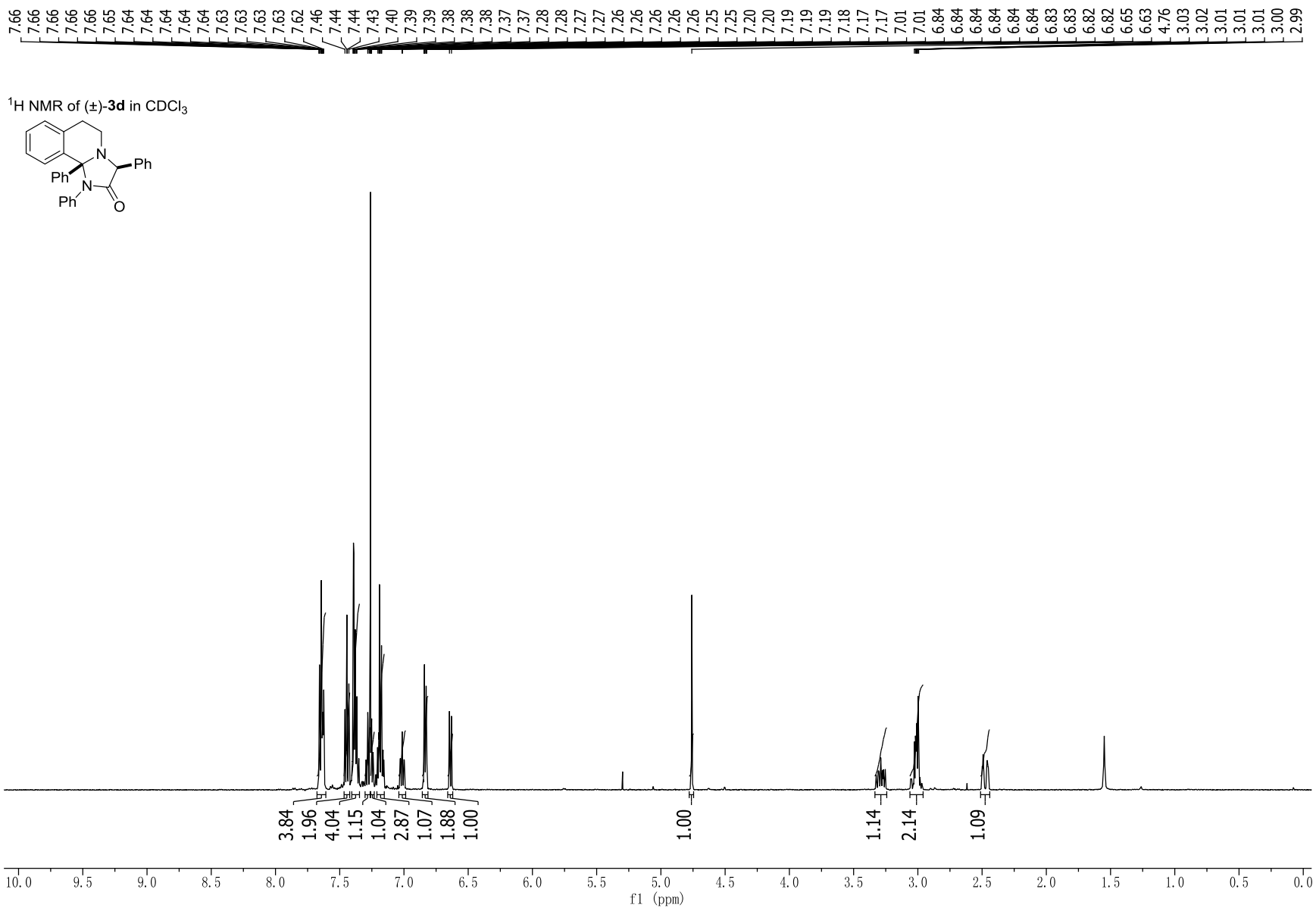

171.03
138.32
135.89
132.88
130.27
129.61
129.14
128.75
128.59
128.57
128.35
128.13
126.89
126.19
126.09
125.83
123.92
122.89
77.41
77.16
76.91
74.14
66.92
47.97
13C NMR of (±)-3c in CDCl3
Ph
H
N
N
Ph
O
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

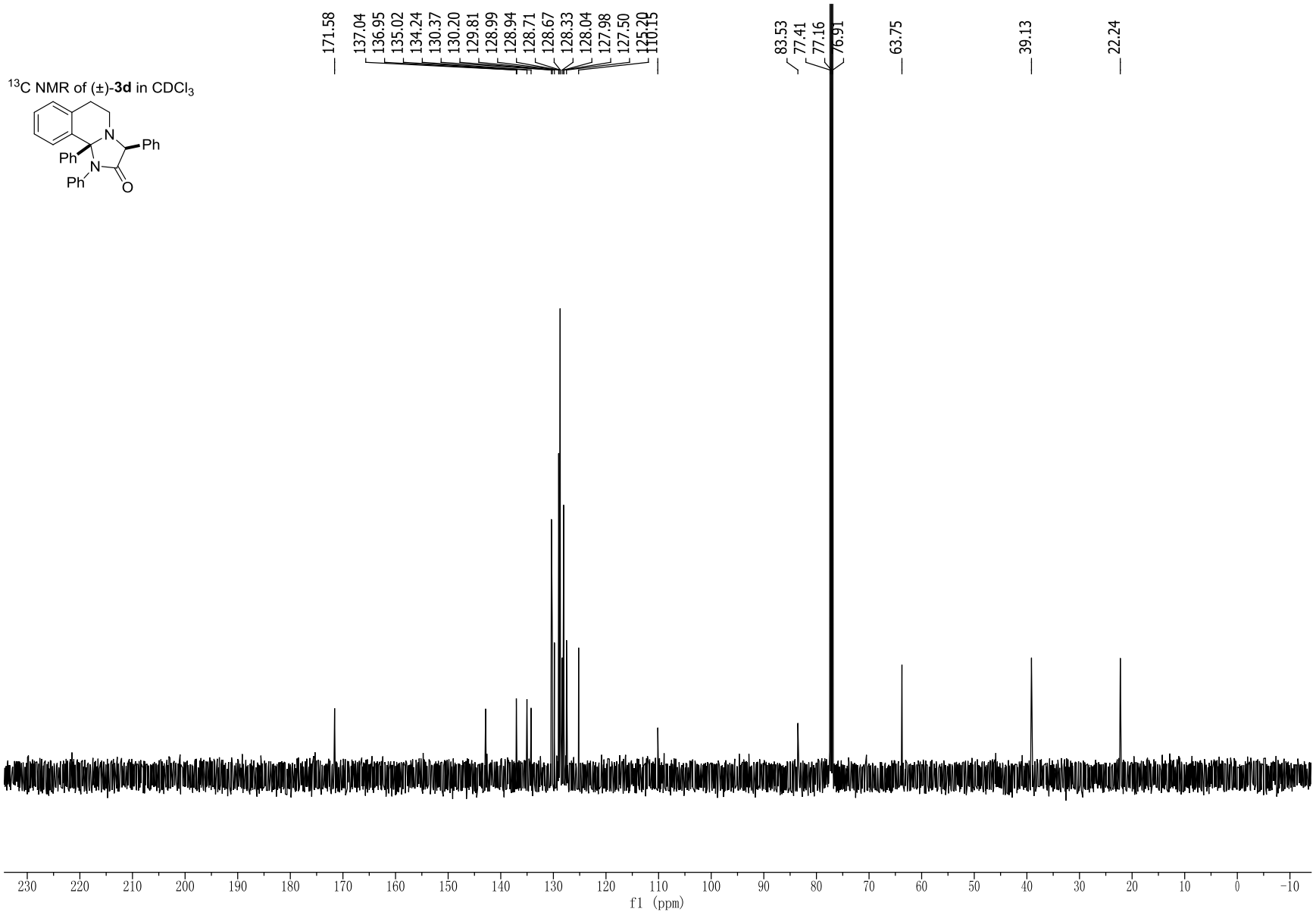
GCOSY of (±)-**3c** in $CDCl_3$

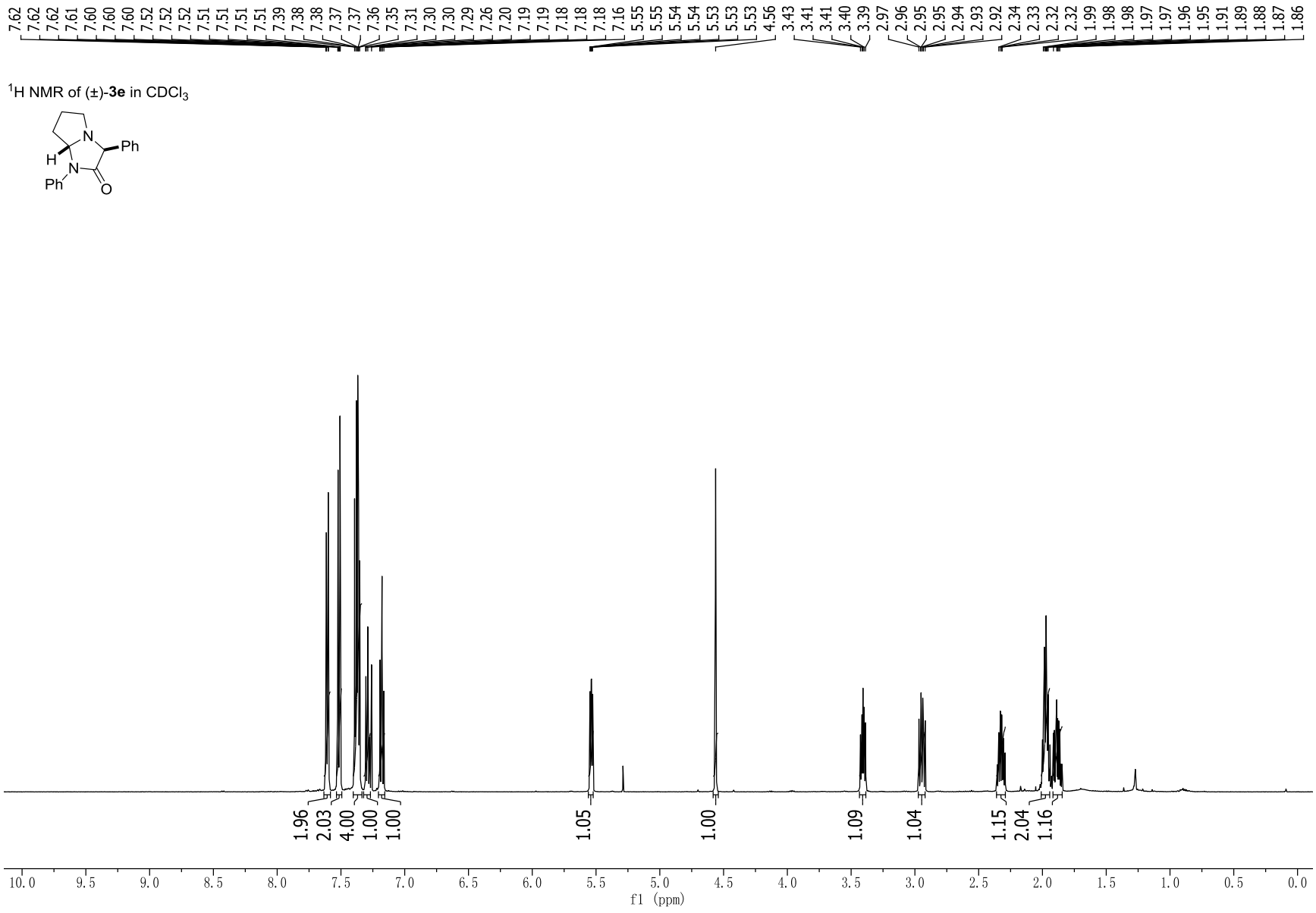


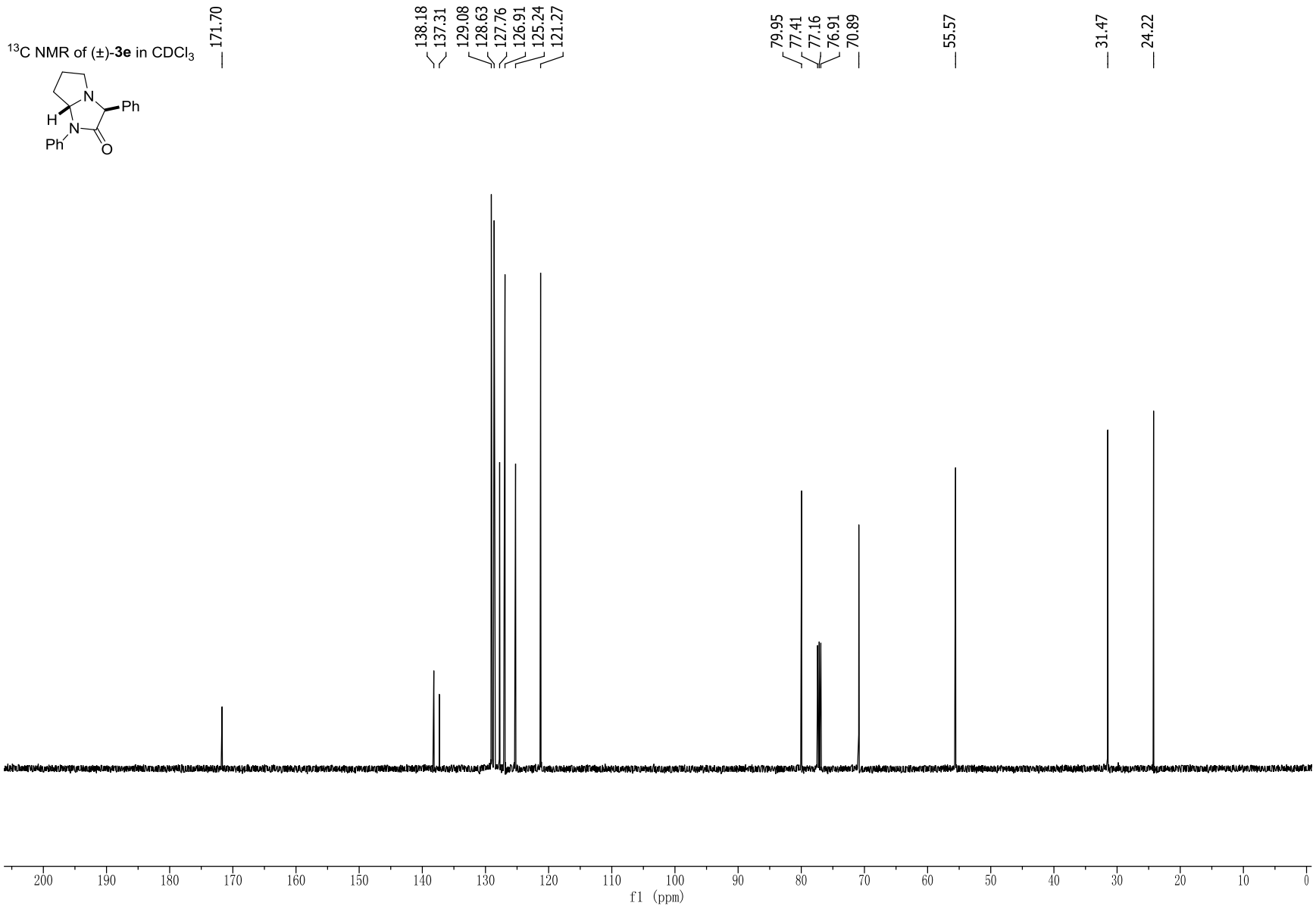
NOESY of (±)-**3c** in $CDCl_3$



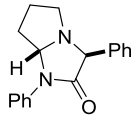

7.66 7.66 7.66 7.66 7.66 7.65 7.64 7.64 7.64 7.64 7.64 7.63 7.63 7.63 7.63 7.62 7.46 7.44 7.44 7.43 7.40 7.39 7.39 7.38 7.38 7.38 7.37 7.37 7.28 7.28 7.27 7.27 7.26 7.26 7.26 7.26 7.26 7.25 7.25 7.20 7.20 7.19 7.19 7.19 7.18 7.17 7.17 7.01 7.01 6.84 6.84 6.84 6.84 6.84 6.83 6.83 6.82 6.82 6.65 6.63 4.76 3.03 3.02 3.01 3.01 3.01 3.00 2.99
1H NMR of (±)-3d in CDCl3
Ph
N
Ph
N
Ph
O
3.84 1.96 4.04 1.15 1.04 2.87 1.07 1.88 1.00 1.00 1.14 2.14 1.09
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)


^{13}C NMR of (±)-**3d** in $CDCl_3$
171.58
137.04
136.95
135.02
134.24
130.37
130.20
129.81
128.99
128.94
128.71
128.67
128.33
128.04
127.98
127.50
125.20
110.15
83.53
77.41
77.16
76.91
63.75
39.13
22.24
Ph
Ph
N
N
Ph
O
f1 (ppm)

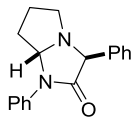

7.62 7.62 7.62 7.61 7.60 7.60 7.60 7.52 7.52 7.52 7.51 7.51 7.51 7.51 7.39 7.38 7.38 7.37 7.37 7.36 7.35 7.31 7.30 7.30 7.29 7.26 7.20 7.19 7.19 7.18 7.18 7.18 7.16 5.55 5.55 5.54 5.54 5.53 5.53 5.53 4.56 3.43 3.41 3.41 3.40 3.39 2.97 2.96 2.95 2.95 2.94 2.93 2.92 2.34 2.33 2.32 2.32 1.99 1.98 1.98 1.97 1.97 1.96 1.95 1.91 1.89 1.88 1.87 1.86
1H NMR of (±)-3e in CDCl3
N
Ph
H
N
Ph
O
1.96 2.03 4.00 1.00 1.00 1.05 1.00 1.09 1.04 1.15 2.04 1.16
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)

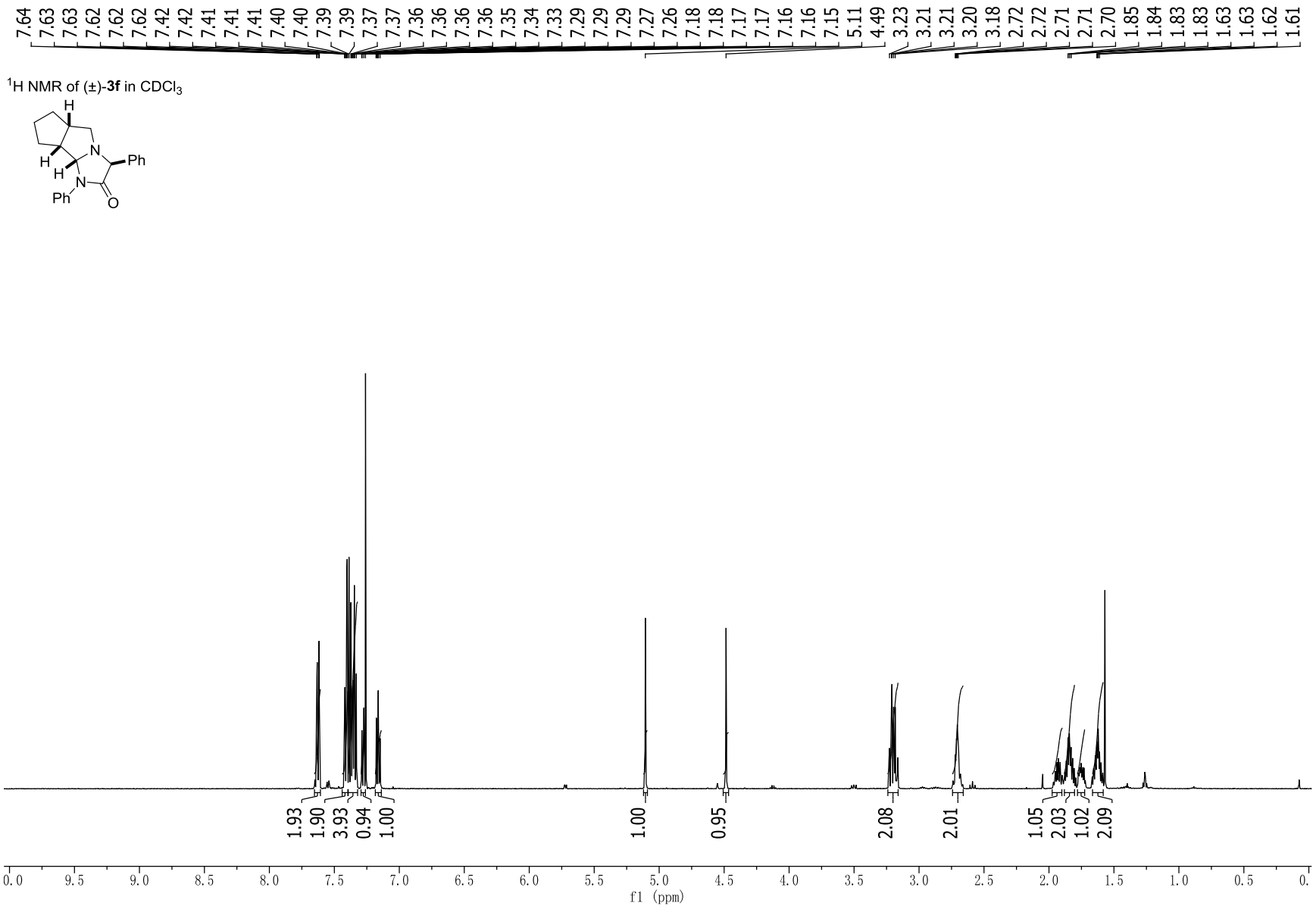

^{13}C NMR of (±)-**3e** in $CDCl_3$
171.70
138.18
137.31
129.08
128.63
127.76
126.91
125.24
121.27
79.95
77.41
77.16
76.91
70.89
55.57
31.47
24.22
Ph
H
N
Ph
N
O
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

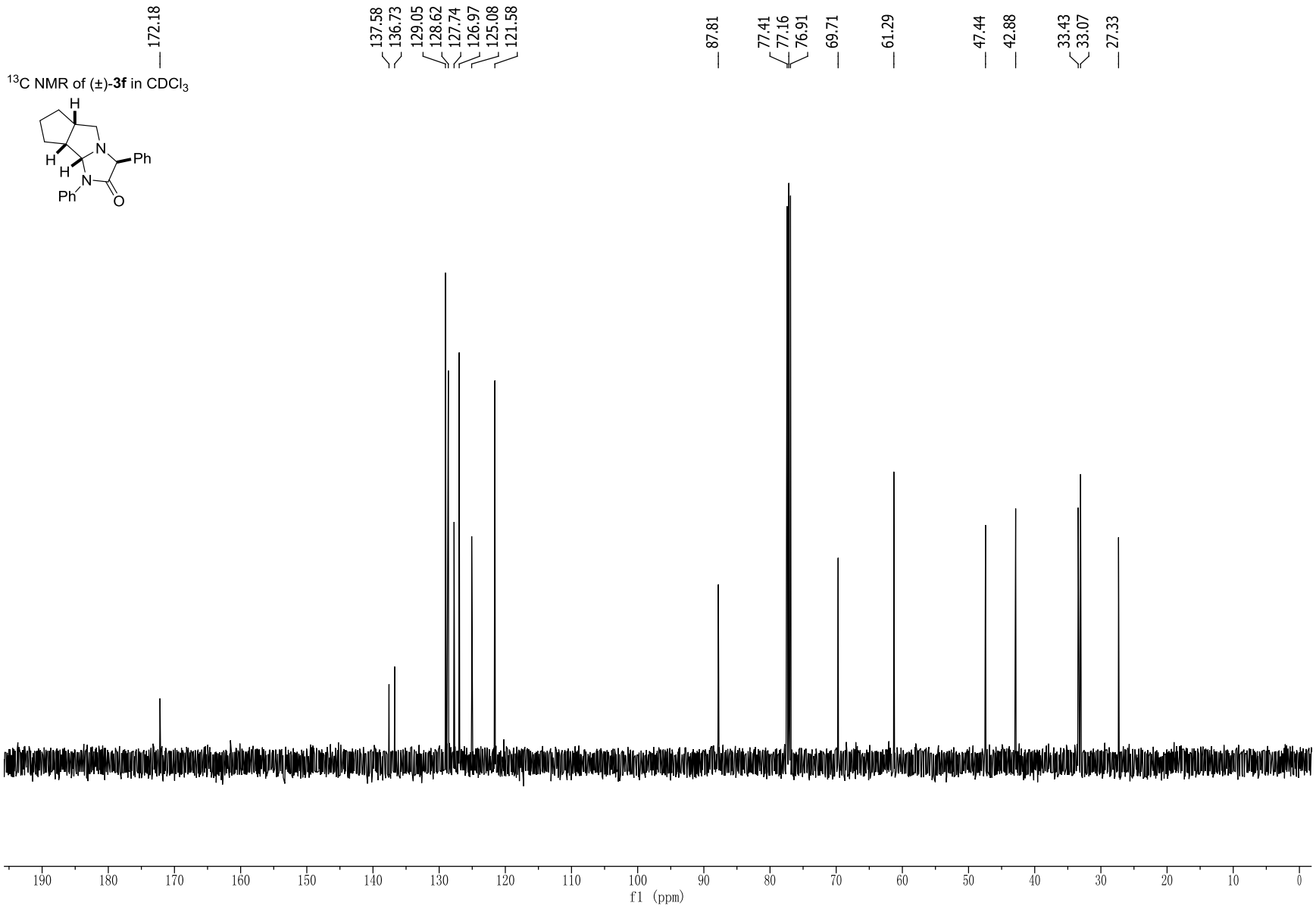
GCOSY of (±)-**3e** in $CDCl_3$



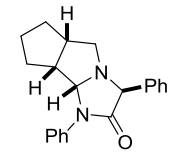
NOESY of (±)-**3e** in $CDCl_3$



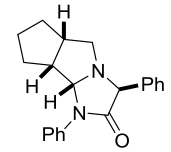

7.64 7.63 7.63 7.62 7.62 7.62 7.42 7.42 7.41 7.41 7.41 7.40 7.40 7.39 7.39 7.37 7.37 7.36 7.36 7.36 7.36 7.35 7.34 7.33 7.29 7.29 7.29 7.27 7.26 7.18 7.18 7.17 7.17 7.16 7.16 7.15 5.11 4.49 3.23 3.21 3.21 3.20 3.18 2.72 2.72 2.71 2.71 2.70 1.85 1.84 1.83 1.83 1.63 1.63 1.62 1.61
1H NMR of (±)-3f in CDCl3
H
H
H
N
Ph
N
Ph
O
1.93 1.90 3.93 0.94 1.00 1.00 0.95 2.08 2.01 1.05 2.03 1.02 2.09
f1 (ppm)

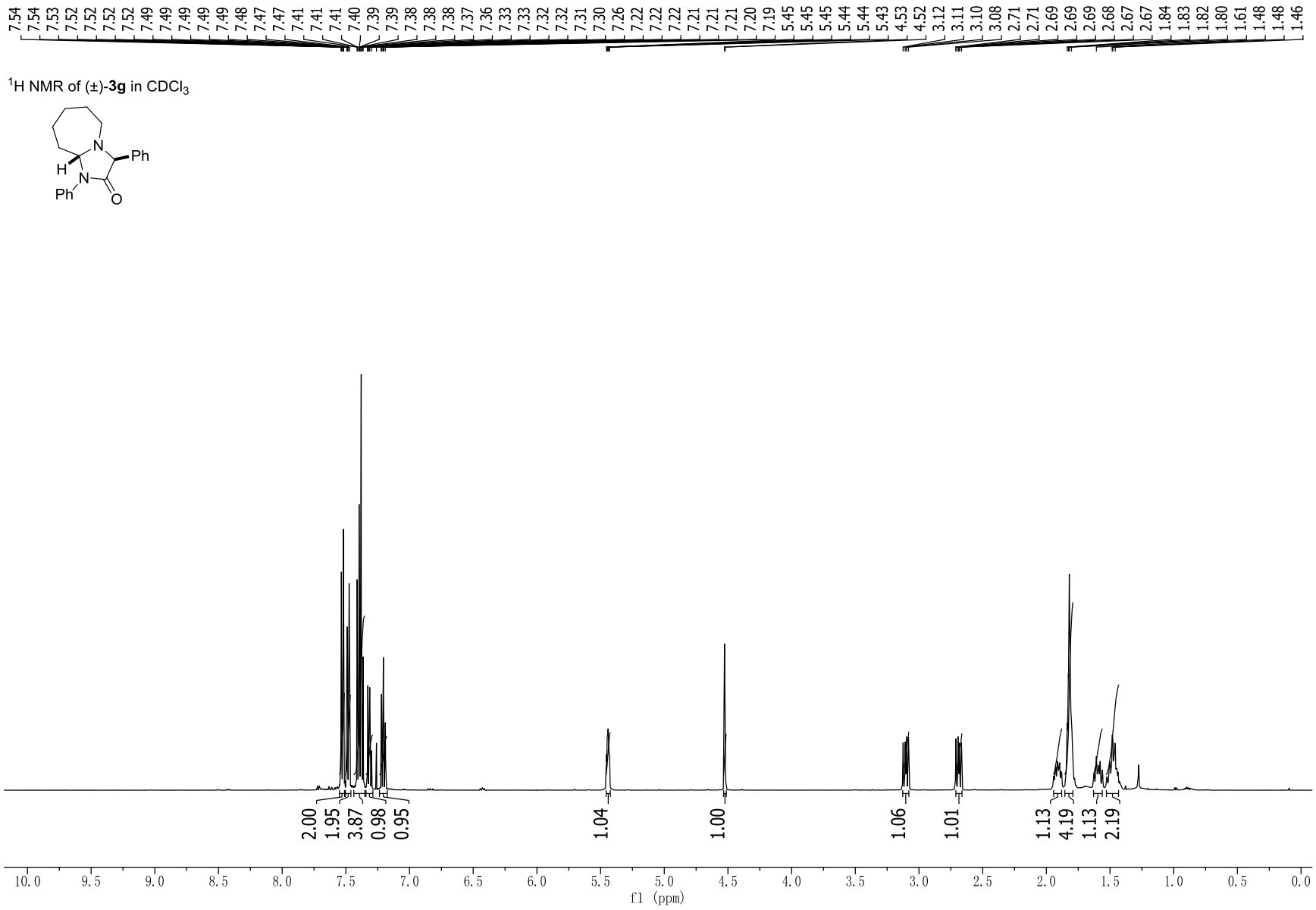

172.18
137.58
136.73
129.05
128.62
127.74
126.97
125.08
121.58
87.81
77.41
77.16
76.91
69.71
61.29
47.44
42.88
33.43
33.07
27.33
13C NMR of (±)-3f in CDCl3
H
H
H
N
Ph
N
Ph
O
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

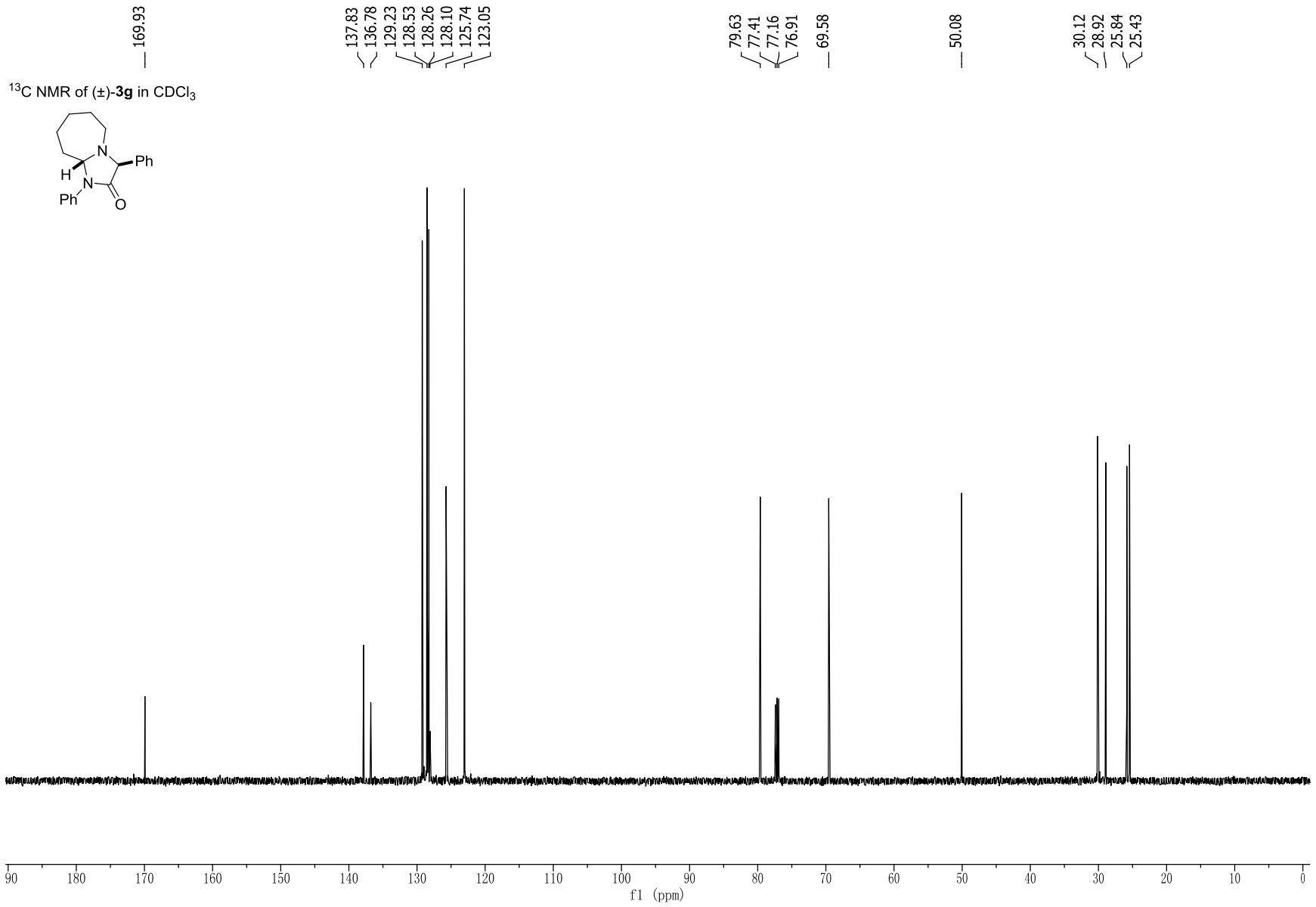
GCOSY of (±)-**3f** in $CDCl_3$



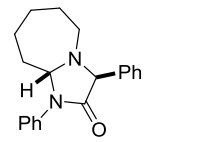
NOESY of (±)-**3f** in $CDCl_3$



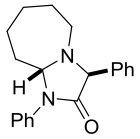

7.54 7.54 7.53 7.52 7.52 7.52 7.52 7.49 7.49 7.49 7.49 7.48 7.47 7.47 7.41 7.41 7.41 7.40 7.39 7.39 7.38 7.38 7.38 7.37 7.36 7.33 7.33 7.32 7.32 7.31 7.30 7.26 7.22 7.22 7.22 7.21 7.21 7.21 7.20 7.19 5.45 5.45 5.45 5.44 5.44 5.43 4.53 4.52 3.12 3.11 3.10 3.08 2.71 2.71 2.69 2.69 2.69 2.68 2.67 2.67 1.84 1.83 1.82 1.80 1.61 1.48 1.48 1.46
^{1}H NMR of (±)-**3g** in $CDCl_3$
N
Ph
H
N
Ph
O
2.00 1.95 3.87 0.98 0.95 1.04 1.00 1.06 1.01 1.13 4.19 1.13 2.19
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)

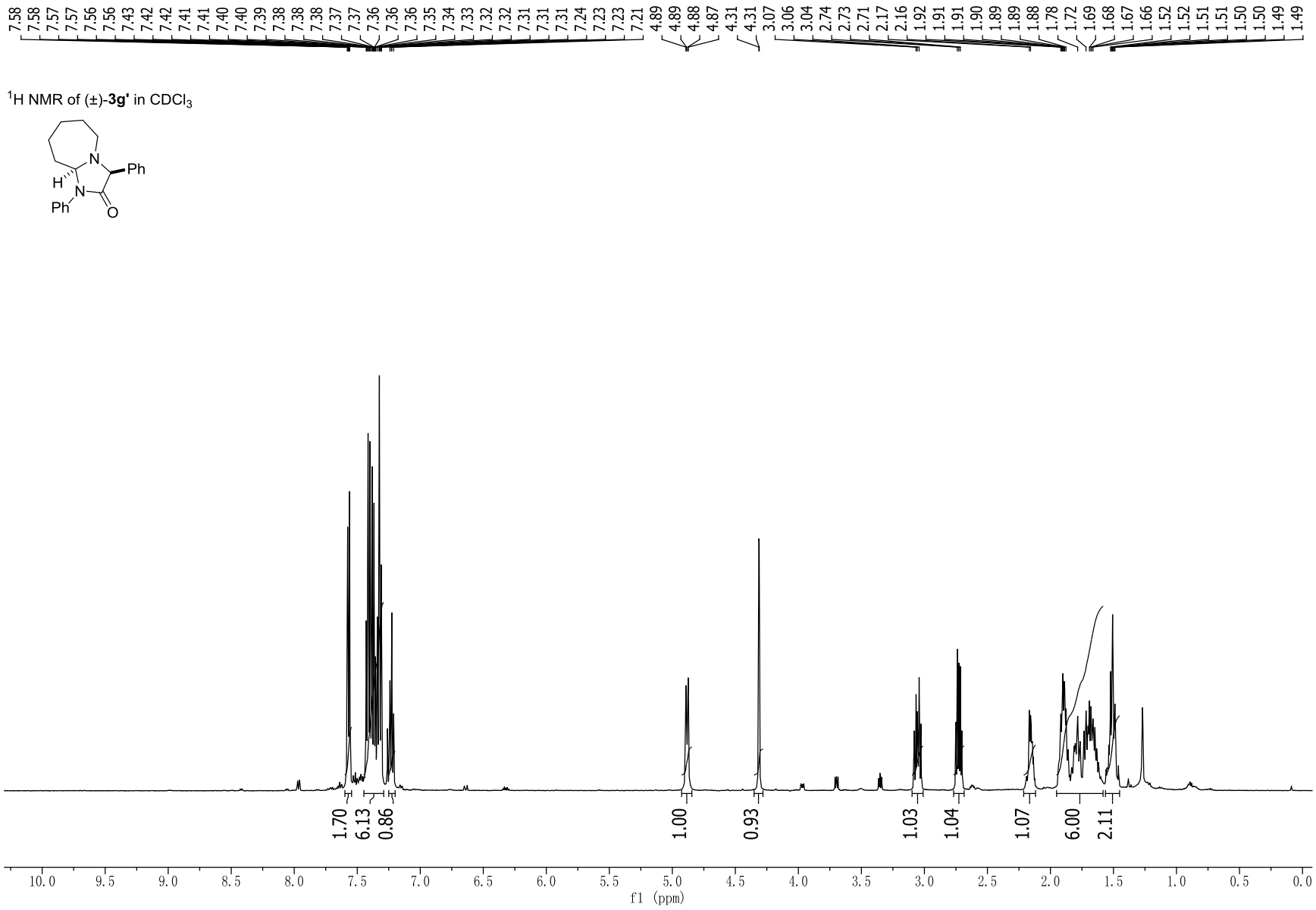

169.93
137.83
136.78
129.23
128.53
128.26
128.10
125.74
123.05
79.63
77.41
77.16
76.91
69.58
50.08
30.12
28.92
25.84
25.43
13C NMR of (±)-3g in CDCl3
Ph
H
N
Ph
O
f1 (ppm)

GCOSY of (±)-**3g** in $CDCl_3$

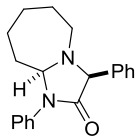


NOESY of (±)-**3g** in $CDCl_3$

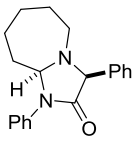



7.58 7.58 7.57 7.57 7.56 7.56 7.43 7.42 7.42 7.41 7.41 7.40 7.40 7.39 7.38 7.38 7.38 7.37 7.37 7.36 7.36 7.36 7.35 7.34 7.33 7.32 7.32 7.31 7.31 7.31 7.24 7.23 7.23 7.21 4.89 4.89 4.88 4.87 4.31 4.31 3.07 3.06 3.04 2.74 2.73 2.71 2.17 2.16 1.92 1.91 1.91 1.90 1.89 1.89 1.88 1.78 1.72 1.69 1.68 1.67 1.66 1.52 1.52 1.51 1.51 1.50 1.50 1.49 1.49
^{1}H NMR of (±)-**3g'** in $CDCl_3$
N
Ph
H
N
Ph
O
1.70
6.13
0.86
1.00
0.93
1.03
1.04
1.07
6.00
2.11
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)

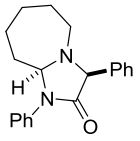
^{13}C NMR of (±)-**3g'** in $CDCl_3$

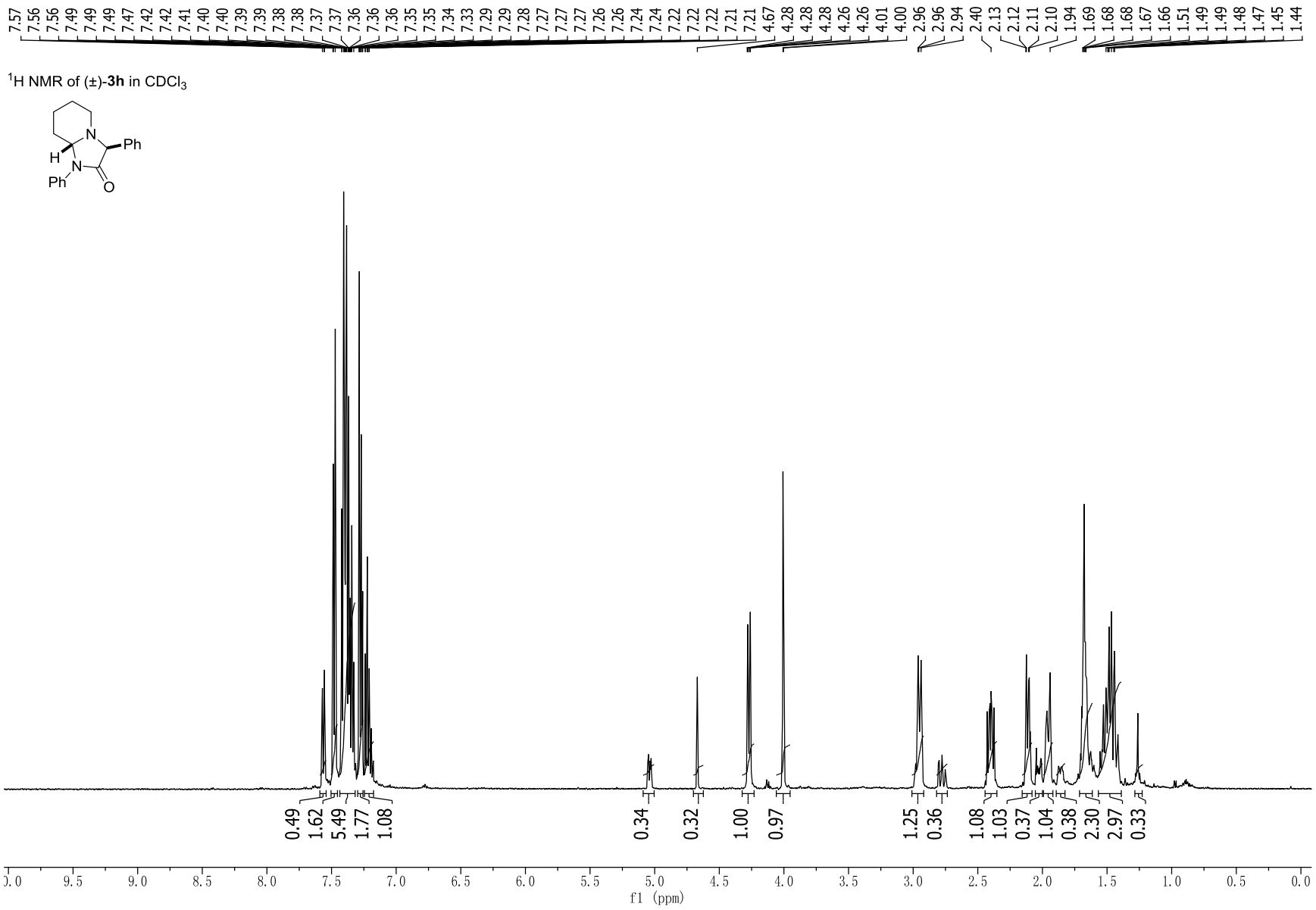


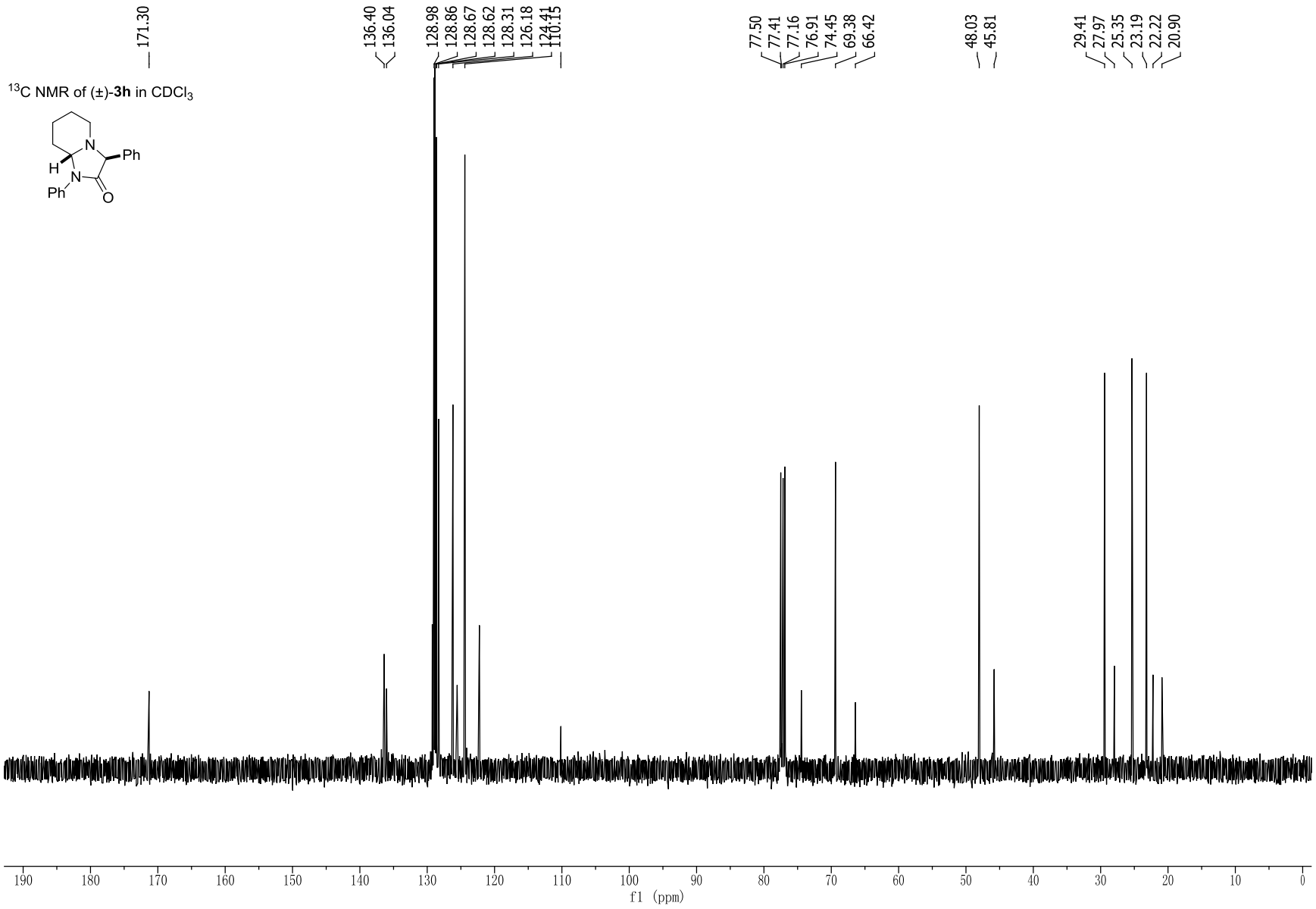
GCOSY of (±)-**3g'** in $CDCl_3$

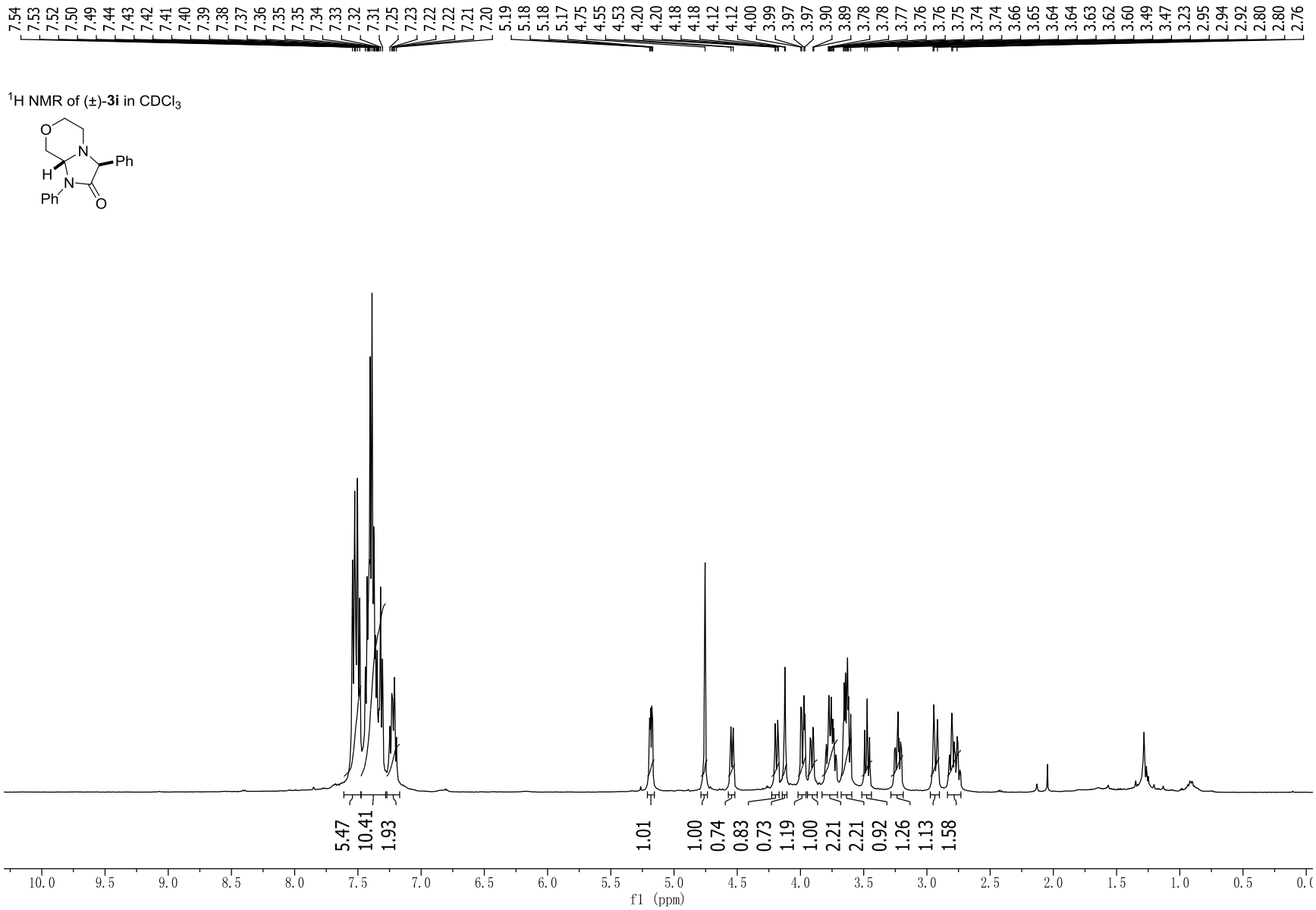


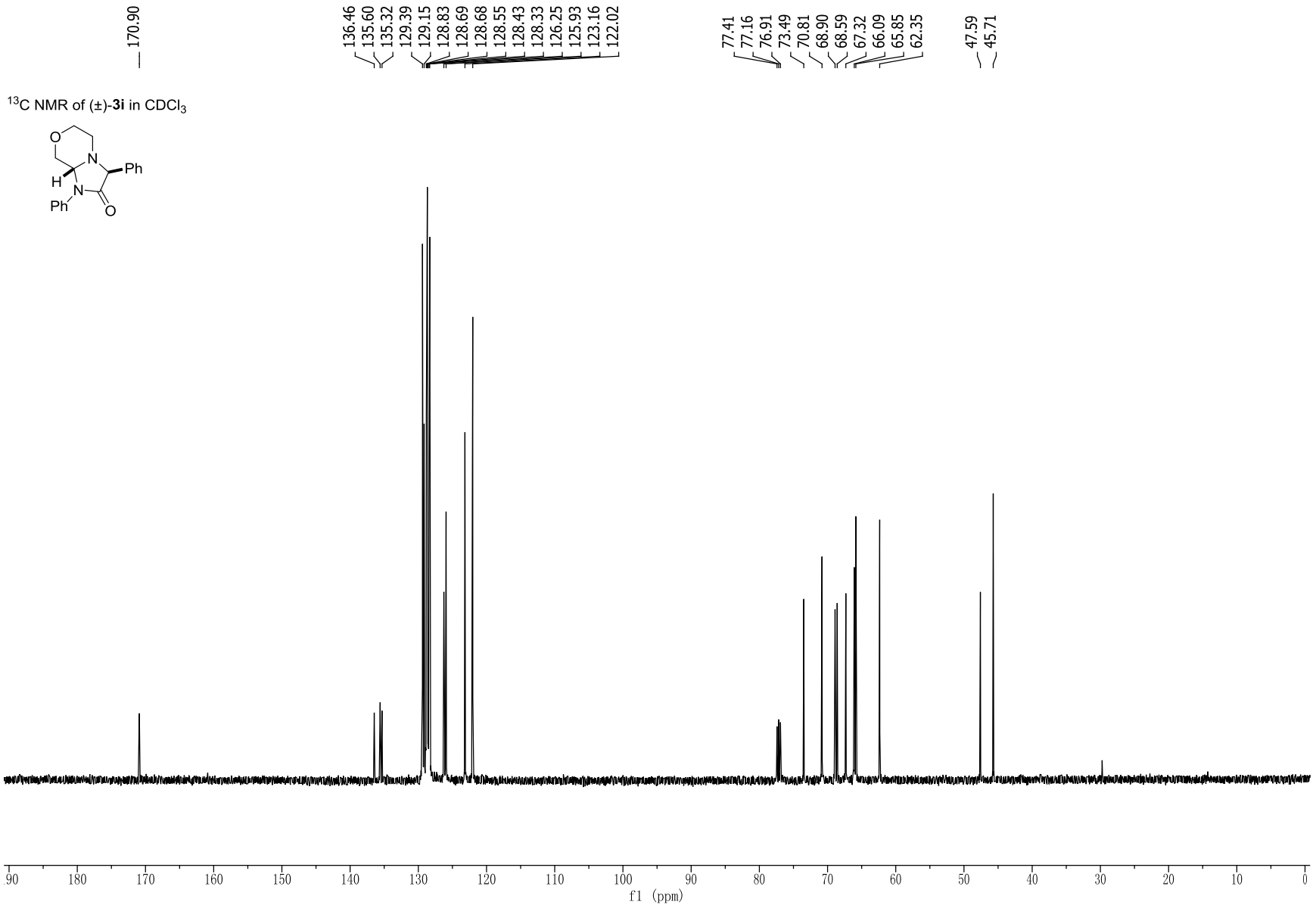
NOESY of (±)-**3g'** in $CDCl_3$



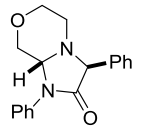

7.57 7.56 7.56 7.49 7.49 7.49 7.47 7.42 7.42 7.41 7.40 7.40 7.39 7.39 7.38 7.38 7.37 7.37 7.36 7.36 7.36 7.35 7.35 7.34 7.33 7.29 7.29 7.28 7.27 7.27 7.27 7.26 7.26 7.24 7.24 7.22 7.22 7.21 7.21 4.67 4.28 4.28 4.28 4.26 4.26 4.01 4.00 2.96 2.96 2.94 2.40 2.13 2.12 2.11 2.10 1.94 1.69 1.68 1.68 1.67 1.66 1.51 1.49 1.49 1.48 1.47 1.45 1.44
1H NMR of (±)-3h in CDCl3
Ph
H
N
N
Ph
O
0.49 1.62 5.49 1.77 1.08 0.34 0.32 1.00 0.97 1.25 0.36 1.08 1.03 0.37 1.04 0.38 2.30 2.97 0.33
f1 (ppm)


171.30
136.40
136.04
128.98
128.86
128.67
128.62
128.31
126.18
124.41
110.15
77.50
77.41
77.16
76.91
74.45
69.38
66.42
48.03
45.81
29.41
27.97
25.35
23.19
22.22
20.90
13C NMR of (±)-3h in CDCl3
N
Ph
H
N
Ph
O
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

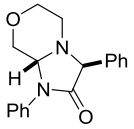

7.54 7.53 7.52 7.50 7.49 7.44 7.43 7.42 7.41 7.40 7.39 7.38 7.37 7.36 7.35 7.35 7.34 7.33 7.32 7.31 7.25 7.23 7.22 7.22 7.21 7.20
5.19 5.18 5.18 5.17 4.75 4.55 4.53 4.20 4.20 4.18 4.18 4.12 4.12 4.00 3.99 3.97 3.97 3.90 3.89 3.78 3.78 3.77 3.76 3.76 3.75 3.74 3.74 3.66 3.65 3.64 3.64 3.63 3.62 3.60 3.49 3.47 3.23 2.95 2.94 2.92 2.80 2.80 2.76
1H NMR of (±)-3i in CDCl3
O
N
Ph
H
N
Ph
O
5.47 10.41 1.93 1.01 1.00 0.74 0.83 0.73 1.19 1.00 2.21 2.21 0.92 1.26 1.13 1.58
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
f1 (ppm)

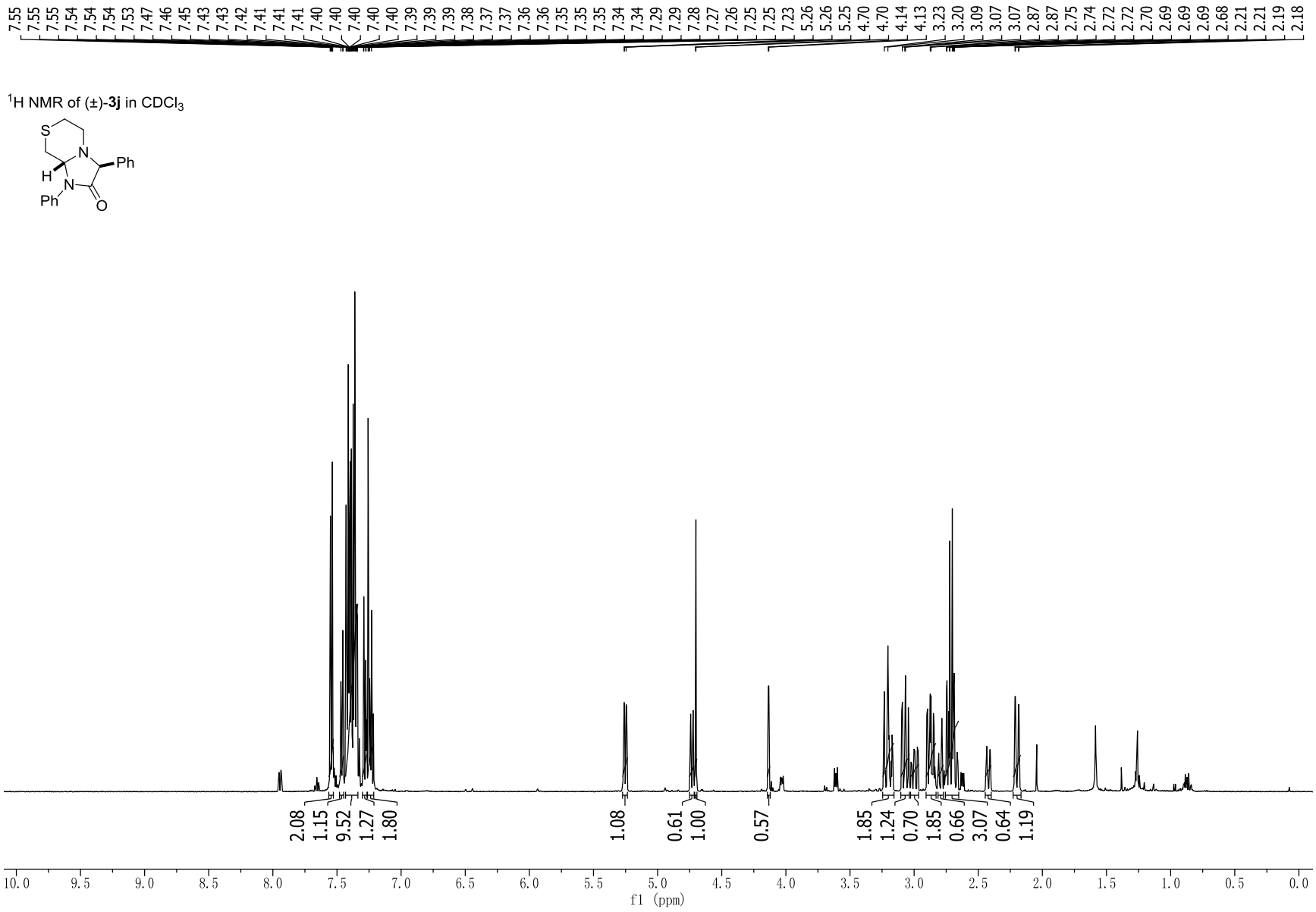

170.90
136.46
135.60
135.32
129.39
129.15
128.83
128.69
128.68
128.55
128.43
128.33
126.25
125.93
123.16
122.02
77.41
77.16
76.91
73.49
70.81
68.90
68.59
67.32
66.09
65.85
62.35
47.59
45.71
13C NMR of (±)-3i in CDCl3
O
N
Ph
H
N
Ph
O
f1 (ppm)

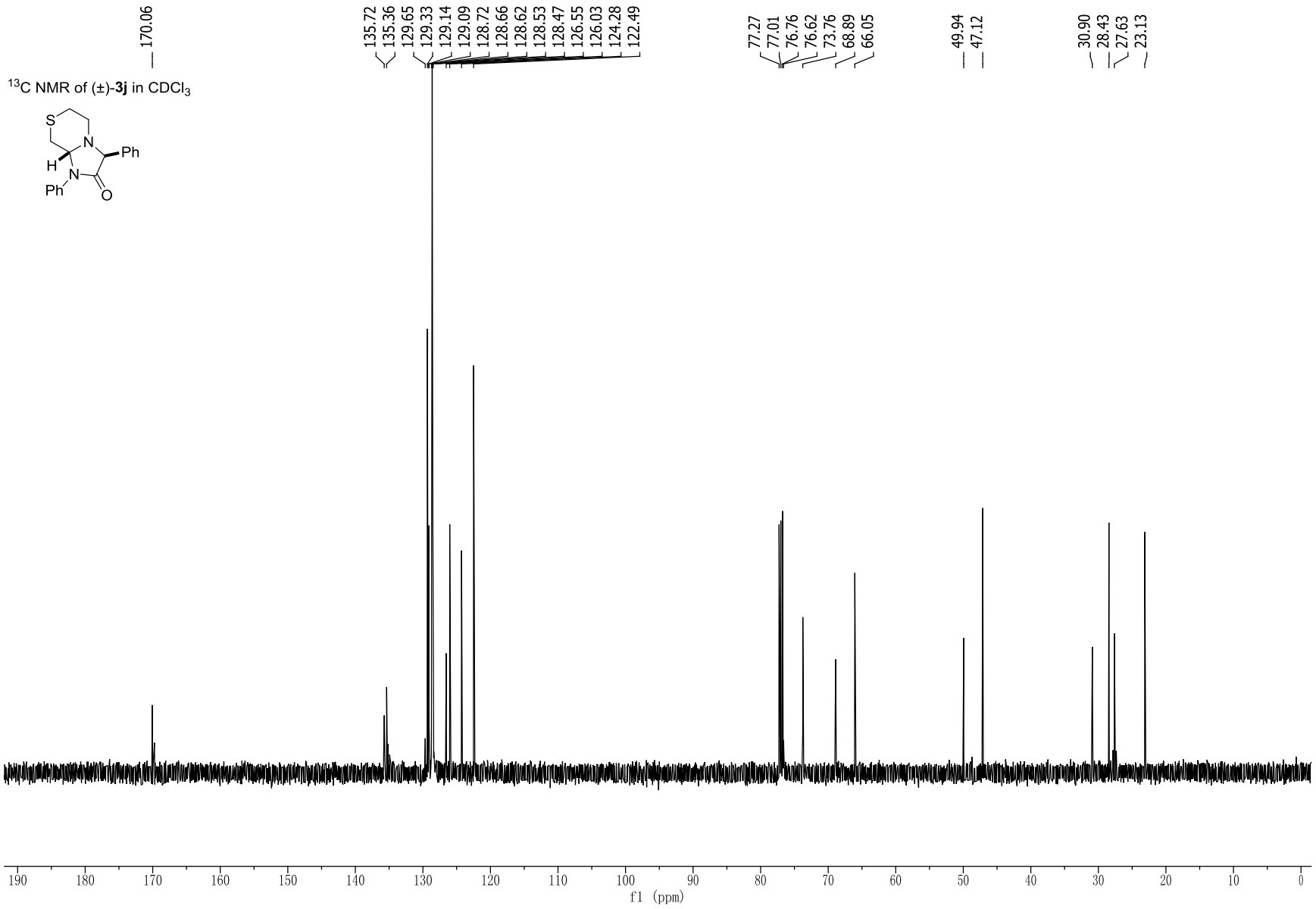
GCOSY of (±)-**3i** in $CDCl_3$



NOESY of (±)-**3i** in $CDCl_3$




7.55 7.55 7.55 7.54 7.54 7.54 7.53 7.47 7.46 7.45 7.43 7.43 7.42 7.41 7.41 7.41 7.40 7.40 7.40 7.40 7.40 7.39 7.39 7.39 7.38 7.37 7.37 7.36 7.36 7.35 7.35 7.35 7.34 7.34 7.29 7.29 7.28 7.27 7.26 7.25 7.25 7.23 5.26 5.26 5.25 4.70 4.70 4.14 4.13 3.23 3.20 3.09 3.07 3.07 2.87 2.87 2.75 2.74 2.72 2.72 2.70 2.69 2.69 2.69 2.68 2.21 2.21 2.19 2.18
1H NMR of (±)-3j in CDCl3
S
N
Ph
H
N
Ph
O
2.08 1.15 9.52 1.27 1.80 1.08 0.61 1.00 0.57 1.85 1.24 0.70 1.85 0.66 3.07 0.64 1.19
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)


170.06
135.72
135.36
129.65
129.33
129.14
129.09
128.72
128.66
128.62
128.53
128.47
126.55
126.03
124.28
122.49
77.27
77.01
76.76
76.62
73.76
68.89
66.05
49.94
47.12
30.90
28.43
27.63
23.13
13C NMR of (±)-3j in CDCl3
S
N
Ph
H
N
Ph
O
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)