Acetic Acid Promoted Redox-Annulations with Dual C–H Functionalization

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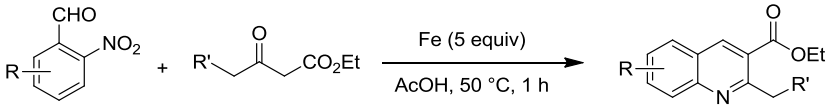
Supporting Information

General Information: Reagents and solvents were purchased from commercial sources and were 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. Melting points were recorded on a Thomas Hoover capillary melting point apparatus and are uncorrected. Infrared spectra were recorded on an ATI Mattson Genesis Series FT-Infrared spectrophotometer. 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, $(CD_3)_2SO$ at 2.50 ppm). Data are reported as app = apparent, s = singlet, d = doublet, t = triplet, q = quartet, p = pentet, br = broad, dd = doublet of doublets, dt = doublet of triplets, td = triplet of doublets, ddd = doublet of doublet of doublets, dddd = doublet of doublet of doublet of doublets, ddddd = doublet of doublet of 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, $(CD_3)_2SO$ at 39.52 ppm). Mass spectra were recorded on a Finnigan LCQ-DUO mass spectrometer or on a Finnigan 2001 Fourier Transform Ion Cyclotron Resonance Mass Spectrometer. Products **1a**,[1] **1m**,[2] **3a**,[3] and **3b**[4] were previously reported and their published characterization data matched our own in all regards. Ratios of diastereomeric products were determined by ^{1}H-NMR analysis of the crude reaction mixture.

Synthesis of ethyl 2-alkylquinoline-3-carboxylates 11 and ethyl 2-alkylnicotinates 15

General procedure A

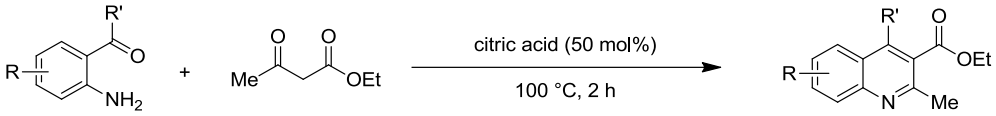
This method was adopted from a literature procedure.[5] To a stirred solution of nitrobenzaldehyde **10** (20 mmol) and β-ketoester (1.2 equiv) in acetic acid (0.3 M) was added iron (5 equiv) at room temperature. The mixture was heated to 50 °C for 1 h. The resulting mixture was allowed to cool to room temperature and filtered through a short pad of celite and washed with EtOAc. The filtrate was basified with 1 M NaOH. The aqueous layer was extracted with EtOAc (2 x 75 mL). The combined organic layers were then washed with brine and dried over anhydrous Na_2SO_4. The solvent was removed under reduced pressure and the residue purified by silica gel chromatography. Products **11a**[5] and **1l**[5] were reported previously and their published characterization data matched our own in all regards.



10 1.2 equiv **11**

General procedure B

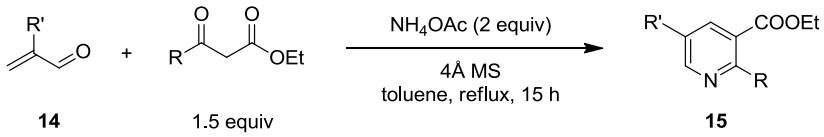
This method was adopted from a literature procedure.[6] To a mixture of **13** (20 mmol) and β-ketoester (1.2 equiv) was added citric acid (0.5 equiv) at room temperature. The mixture was heated to 100 °C for 2 h. The reaction mixture was allowed to cool to room temperature and poured into distilled water (100 mL). In case of solid products, the mixture was stirred for another hour and then filtered. The solid was recrystallized from EtOH. For liquid products, the mixture was extracted with EtOAc (2 x 75 mL). The combined organic layers were then washed with brine and dried over anhydrous Na_2SO_4. The solvent was removed under reduced pressure and the residue purified by silica gel chromatography. Products **11g**[6] and **11i**[6] were reported previously and their published characterization data matched our own in all regards.

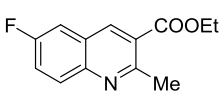


13 1.2 equiv **11**

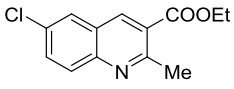
General Procedure C

This method was adopted from a literature procedure.[7] To a stirred solution of **14** (1.5 equiv) and β-ketoester (20 mmol) in toluene (0.5 M) was added ammonium acetate (2 equiv) and 4 Å molecular sieves (10 g) at room temperature. The mixture was heated under reflux for 15 h. The reaction mixture was then allowed to cool to room temperature, filtered through a short pad of celite, and washed with EtOAc. The solvent was removed under reduced pressure and the residue purified by silica gel chromatography. Compound **15a**[7] was reported previously and its published characterization data matched our own in all regards.

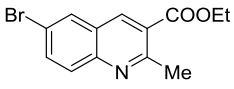


Ethyl 6-fluoro-2-methylquinoline-3-carboxylate (11b): Following the general procedure A, 

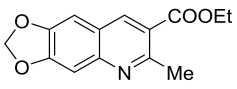
11b was obtained from 5-fluoro-2-nitrobenzaldehyde and ethyl acetoacetate as a white solid in 52% yield (R_f = 0.39 in hexane/EtOAc 80:20 v/v); mp = 85–87 °C; IR (KBr) 3055, 2989, 2938, 2364, 2346, 1720, 1603, 1561, 1493, 1480, 1403, 1391, 1378, 1271, 1246, 1210, 1148, 1122, 1066, 968, 843, 775, 758, 690, 618, 423; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.63 (s, 1H), 8.01 (dd, *J* = 9.2, 5.2 Hz, 1H), 7.52 (ddd, *J* = 9.3, 8.3, 2.9 Hz, 1H), 7.44 (dd, *J* = 8.5, 2.9 Hz, 1H), 4.43 (q, *J* = 7.1 Hz, 2H), 2.95 (s, 3H), 1.44 (t, *J* = 7.1 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 166.4, 160.4 (d, J_{C-F} = 248.5 Hz), 157.8 (d, J_{C-F} = 2.7 Hz), 145.8, 139.1 (d, J_{C-F} = 5.5 Hz), 131.2 (d, J_{C-F} = 9.0 Hz), 126.4 (d, J_{C-F} = 10.2 Hz), 124.8, 121.8 (d, J_{C-F} = 25.8 Hz), 111.4 (d, J_{C-F} = 21.7 Hz), 61.7, 25.6, 14.4; *m/z* (ESI–MS) 233.9 [M + H]$^+$.

Ethyl 6-chloro-2-methylquinoline-3-carboxylate (11c): Following the general procedure A, 

11c was obtained from 5-chloro-2-nitrobenzaldehyde and ethyl acetoacetate as a white solid in 60% yield (R_f = 0.42 in hexane/EtOAc 80:20 v/v); mp = 93–94 °C; IR (KBr) 3060, 2977, 2931, 1702, 1616, 1592, 1560, 1481, 1431, 1403, 1295, 1269, 1252, 1283, 1080, 1067, 1020, 925, 833, 803, 648, 580; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.53 (s, 1H), 7.88 (d, *J* = 8.9 Hz, 1H), 7.73 (d, *J* = 2.4 Hz, 1H), 7.62 (dd, *J* = 9.0, 2.4 Hz, 1H), 4.40 (q, *J* = 7.1 Hz, 2H), 2.91 (s, 3H), 1.41 (t, *J* = 7.1 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 166.2, 158.8, 146.9, 138.7, 132.4, 132.2, 130.2, 126.9, 126.3, 124.8, 61.6, 25.7, 14.4; *m/z* (ESI–MS) (^{35}Cl) 250.0 [M + H]$^+$, (^{37}Cl) 252.1 [M + H]$^+$.

Ethyl 6-bromo-2-methylquinoline-3-carboxylate (11d): Following the general procedure A, 

11d was obtained from 5-bromo-2-nitrobenzaldehyde and ethyl acetoacetate as a yellow solid in 50% yield (Rf = 0.44 in hexane/EtOAc 80:20 v/v); mp = 91–93 °C; IR (KBr) 2977, 1726, 1613, 1578, 1559, 1478, 1404, 1273, 1244, 1214, 1139, 1071, 918, 834, 649, 555; 1H NMR (CDCl3, 500 MHz) δ 8.63 (s, 1H), 8.02 (d, J = 2.2 Hz, 1H), 7.90 (d, J = 9.0 Hz, 1H), 7.83 (dd, J = 9.0, 2.2 Hz, 1H), 4.44 (q, J = 7.1 Hz, 2H), 2.96 (s, 3H), 1.45 (t, J = 7.1 Hz, 3H); ^{13}C NMR (125 MHz, CDCl3) δ 166.3, 159.1, 147.3, 138.8, 135.1, 130.4, 127.0, 124.9, 120.4, 61.7, 25.8, 14.4; m/z (ESI–MS) (^{79}Br) 294.1 [M + H]+ , (^{81}Br) 295.9 [M + H]$^+$.

Ethyl 6-methyl-[1,3]dioxolo[4,5-*g*]quinoline-7-carboxylate (11e): Following the general 

procedure A, **11e** was obtained from 6-Nitropiperonal and ethyl acetoacetate as a yellow solid in 54% yield (R_f = 0.21 in hexane/EtOAc 80:20 v/v); mp = 159–161 °C; IR (KBr) 3032, 2910, 1687, 1612, 1590, 1461, 1421, 1395, 1276, 1245, 1214, 1105, 1070, 1039, 1018, 943, 888, 863, 583; ^{1}H NMR (DMSO, 500 MHz) δ 8.64 (s, 1H), 7.47 (s, 1H), 7.37–7.23 (m, 1H), 6.23 (s, 2H), 4.35 (q, *J* = 7.1 Hz, 2H), 2.78 (s, 3H), 1.36 (t, *J* = 7.1 Hz, 3H); ^{13}C NMR (125 MHz, DMSO) δ 166.1, 155.3, 152.4, 147.6, 146.9, 137.9, 122.2, 121.4, 104.3, 103.3, 102.3, 60.9, 24.8, 14.1; *m/z* (ESI–MS) 260.0 [M + H]$^+$.

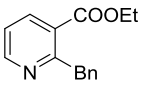
Ethyl 2-methyl-7-(trifluoromethyl)quinoline-3-carboxylate (11f): Following a modified literature procedure,[8] to a stirred solution of (2-amino-4-(trifluoromethyl)phenyl)methanol (20 mmol) and ethyl acetoacetate (1.2 equiv) in EtOH (0.3 M) was added MnO_2 (2 equiv) at room temperature. The mixture was heated under reflux for 15 h. The reaction mixture was then allowed to cool to room temperature, filtered through a short pad of celite, and washed with EtOAc. The solvent was removed under reduced pressure and the residue purified by silica gel chromatography. **11f** was obtained as a yellow solid in 89% yield (R_f = 0.52 in hexane/EtOAc 80:20 v/v); mp = 42–43 °C; IR (KBr) 2987, 1730, 1728, 1603, 1563, 1501, 1368, 1326, 1285, 1233, 1147, 1110, 1058, 933, 900, 818, 775, 720, 683, 538, 478 ; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.86–8.57 (m, 1H), 8.25 (dq, *J* = 1.7, 0.9 Hz, 1H), 7.87 (app dt, *J* = 8.6, 0.9 Hz, 1H), 7.61 (dd, *J* = 8.5, 1.8 Hz, 1H), 4.41 (q, *J* = 7.2 Hz, 2H), 2.94 (s, 3H), 1.42 (t, *J* = 7.1 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 166.0, 160.0, 147.6, 139.30 133.0 (q, J_{C-F} = 32.7 Hz), 129.6, 127.2, 126.5 (q, J_{C-F} = 4.5 Hz), 125.8, 123.8 (q, J_{C-F} = 272.7 Hz), 122.1 (q, J_{C-F} = 3.2 Hz), 61.8, 25.7, 14.3; *m/z* (ESI–MS) 284.3 [M + H]$^+$.

Ethyl 4-ethyl-2-methylquinoline-3-carboxylate (11h): Following the general procedure B, **11h** was obtained from 1-(2-aminophenyl)propan-1-one and ethyl acetoacetate as a yellow liquid in 85% yield (R_f = 0.30 in hexane/EtOAc 80:20 v/v); IR (KBr) 3054, 2986, 2684, 2305, 1723, 1583, 1489, 1422, 1266, 1224, 1166, 1131, 1078, 1060, 896, 749, 705; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.10–7.95 (comp, 2H), 7.69 (dddd, *J* = 8.3, 6.9, 1.3, 0.6 Hz, 1H), 7.60–7.45 (m, 1H), 4.48 (q, *J* = 7.1 Hz, 2H), 3.03 (q, *J* = 7.6 Hz, 2H), 2.70 (s, 3H), 1.43 (td, *J* = 7.2, 0.6 Hz, 3H). 1.35 (t, *J* = 7.6 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 169.3, 154.6, 147.8, 147.1, 130.0, 129.7, 127.4, 126.4, 124.8, 124.0, 61.7, 23.9, 23.5, 15.4, 14.4; *m/z* (ESI–MS) 244.0 [M + H]$^+$.

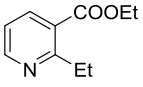
Ethyl 2-benzylquinoline-3-carboxylate (11j): Following the general procedure A, **11j** was obtained from 2-nitrobenzaldehyde and ethyl 3-oxo-4-phenylbutanoate as a yellow solid in 62% yield (R_f = 0.43 in hexane/EtOAc 80:20 v/v); mp = 60–63 °C; IR (KBr) 3060, 2977, 2923, 1719, 1620, 1565, 1489, 1425, 1278, 1250, 1203, 1128, 1061, 1025, 868, 833, 785, 760, 748, 718, 695, 505; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.67 (s, 1H), 8.13 (d, *J* = 8.4 Hz, 1H), 7.82 (d, *J* = 8.6 Hz, 1H), 7.77 (ddd, *J* = 8.4, 6.9, 1.5 Hz, 1H), 7.52 (ddd, *J* = 8.1, 7.0, 1.2 Hz, 1H), 7.40–7.21 (comp, 4H), 7.18–7.07 (m, 1H), 4.81 (s, 2H), 4.31 (q, *J* = 7.2 Hz, 2H), 1.31 (t, *J* = 7.1 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 166.4, 159.7, 148.6, 140.0, 139.6, 131.5, 129.02, 129.00, 128.4, 128.2, 126.8, 126.0, 125.9, 124.3, 61.4, 43.3, 14.2; *m/z* (ESI–MS) 292.2 [M + H]$^+$.

Ethyl 2-ethylquinoline-3-carboxylate (11k): Following the general procedure A, **11k** was obtained from 2-nitrobenzaldehyde and ethyl 3-oxopentanoate as a yellow solid in 60% yield (R_f = 0.47 in hexane/EtOAc 80:20 v/v); mp = 61–63 °C; IR (KBr) 3043, 2973, 2936, 1711, 1619, 1560, 1478, 1421, 1275, 1254, 1204, 1132, 1071, 1052, 1015, 960, 878, 798, 755, 618, 590, 478; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.68 (s, 1H), 8.05 (dd, *J* = 8.6, 0.9 Hz, 1H), 7.85 (ddd, *J* = 8.1, 1.4, 0.6 Hz, 1H), 7.80–7.66 (m, 1H), 7.52 (ddd, *J* = 8.1, 6.9, 1.1 Hz, 1H), 4.44 (q, *J* = 7.1 Hz, 2H), 3.34 (q, *J* = 7.5 Hz, 2H), 1.45 (t, *J* = 7.1 Hz, 3H), 1.38 (t, *J* = 7.5 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 166.9, 163.1, 148.8,

140.0, 131.6, 128.9, 128.5, 126.6, 125.8, 124.0, 61.6, 31.2, 14.4, 14.2; *m/z* (ESI–MS) 230.1 [M + H]$^+$.

Ethyl 2-benzylnicotinate (15b): Following the general procedure C, **15b** was obtained from 

ethyl 3-oxo-4-phenylbutanoate, acrolein and ammonium acetate as a yellow liquid in 23% yield (R_f = 0.28 in hexane/EtOAc 80:20 v/v); IR (KBr) 3049, 2984. 2684, 2359, 2304, 1721, 1421, 1261, 1156, 895, 750, 700; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.73 (dd, *J* = 4.8, 1.8 Hz, 1H), 8.21 (dd, *J* = 7.9, 1.9 Hz, 1H), 7.34–7.24 (comp, 5H), 7.24–7.15 (m, 1H), 4.63 (s, 2H), 4.36 (q, *J* = 7.1 Hz, 2H), 1.36 (t, *J* = 7.1 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 166.6, 161.2, 151.9, 139.7, 138.8, 129.0, 128.3, 126.3, 126.2, 121.4, 61.5, 42.4, 14.3; *m/z* (ESI–MS) 242.1 [M + H]$^+$.

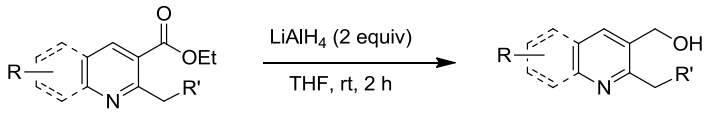
Ethyl 2-ethylnicotinate (15c): Following the general procedure C, **15c** was obtained from ethyl 

3-oxopentanoate, acrolein and ammonium acetate as a yellow liquid in 25% yield (R_f = 0.30 in hexane/EtOAc 80:20 v/v); IR (KBr) 3054, 2987, 2687, 2411, 2305, 1721, 1581, 1566, 1422, 1266, 1143, 1093, 1023, 896, 749, 706; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.56 (dd, *J* = 4.8, 1.9 Hz, 1H), 8.06 (dd, *J* = 7.9, 1.9 Hz, 1H), 7.11 (dd, *J* = 7.9, 4.8 Hz, 1H), 4.30 (q, *J* = 7.1 Hz, 2H), 3.09 (q, *J* = 7.5 Hz, 2H), 1.32 (t, *J* = 7.1 Hz, 3H), 1.23 (t, *J* = 7.5 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 166.6, 164.3, 151.7, 138.3, 125.5, 120.7, 61.3, 30.3, 14.2, 14.0; *m/z* (ESI–MS) 180.1 [M + H]$^+$.

Synthesis of (2-alkylquinolin-3-yl)methanol 12 and (2-alkylpyridin-3-yl)methanol 16

General Procedure D

To an ice-cooled suspension of $LiAlH_4$ (531 mg, 14 mmol, 2 equiv) in dry THF (15 mL) was slowly added a solution of compound **11** or **15** (7 mmol, 1 equiv) in dry THF (5 mL). The reaction was allowed to warm to room temperature and stirred for two hours. The mixture was then cooled in an ice bath, followed by quenching with water (2 mL) and 1M NaOH (1 mL). The lithium and aluminum hydroxide salts were then filtered through a short pad of celite and washed with EtOAc (6 x 50 mL). The solvent was removed under reduced pressure. For pyridine substrates, the residue was directly used in the next step without purification. For quinoline substrates, the residue was purified by silica gel chromatography.

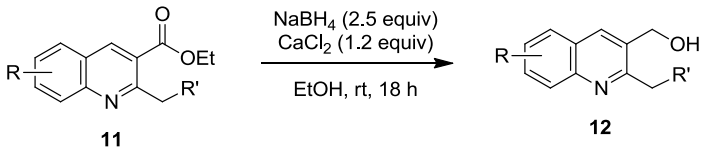


11 or 15 **12 or 16**

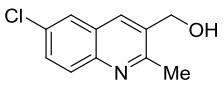
General Procedure E

This method was adopted from a literature procedure.[9] To an ice-cooled solution of $NaBH_4$ (2.5 equiv) in EtOH (10 mL) were added a solution of **11** (10 mmol) in EtOH (20 mL) and a solution of anhydrous $CaCl_2$ (1.2 equiv) in EtOH (20 mL). The reaction mixture was allowed to warm to room temperature and stirred for 18 h. After the consumption of **7** as indicated by TLC, the solvent was removed under reduced pressure. The residue was suspended in dichloromethane

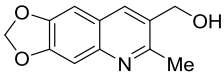
(50 mL) and water (50 mL). After separation, the organic layer was washed with brine (30 mL x 3), then dried over $NaSO_4$. The solvent was removed under reduced pressure and the residue was directly used in the next step without purification.
Following the general procedure E, **11b**, **11d**, **11f**, **11j** were reduced to the corresponding alcohols and used in the next step without purification.



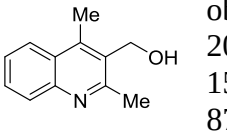
(6-chloro-2-methylquinolin-3-yl)methanol (12c): Following the general procedure D, **12c** was obtained from **11c** as a yellow solid in 60% yield (R_f = 0.30 in hexane/EtOAc 20:80 v/v); mp = 142–143 °C; IR (KBr) 3301, 2933, 2361, 2339, 1598, 1561, 1483, 1382, 1345, 1224, 1115, 1076, 1052, 987, 933, 926, 824, 720, 558, 487; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.02 (s, 1H), 7.92 (d, *J* = 8.9 Hz, 1H), 7.71 (d, *J* = 2.3 Hz, 1H), 7.57 (dd, *J* = 8.9, 2.3 Hz, 1H), 4.85 (s, 2H), 2.76 (br s, 1H), 2.66 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 157.8, 145.3, 133.8, 132.7, 131.7, 130.2, 129.9, 127.8, 126.2, 62.3, 22.6; *m/z* (ESI–MS) (^{35}Cl) 208.1 [M + H]$^+$, (^{37}Cl) 210.1 [M + H]$^+$.



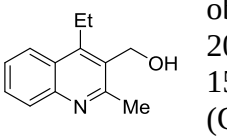
(6-methyl-[1,3]dioxolo[4,5-*g*]quinolin-7-yl)methanol (12e): Following the general procedure D, **12e** was obtained from **11e** as a yellow solid in 60% yield (R_f = 0.24 in hexane/EtOAc 20:80 v/v); mp = 198–200 °C; IR (KBr) 3097, 2912, 2823, 2364, 1624, 1499, 1468, 1370, 1241, 1204, 1099, 1043, 946, 918, 849, 799, 614; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.02 (d, *J* = 1.1 Hz, 1H), 7.31 (s, 1H), 7.25 (s, 1H), 6.16 (s, 2H), 5.31 (t, *J* = 5.3 Hz, 1H) 4.60 (dd, *J* = 5.3, 1.0 Hz, 2H), 2.51 (s, 3H); ^{13}C NMR (125 MHz, DMSO) δ 154.4, 149.7, 146.7, 144.0, 132.1, 132.0, 123.2, 104.4, 102.7, 101.6, 60.4, 21.8; *m/z* (ESI–MS) 218.1 [M + H]$^+$.



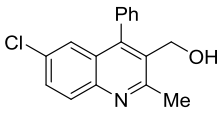
(2,4-dimethylquinolin-3-yl)methanol (12g): Following the general procedure D, **12g** was obtained from **11g** as a yellow solid in 78% yield (R_f = 0.18 in hexane/EtOAc 20:80 v/v); mp = 119–121 °C; IR (KBr) 3119, 2922, 2847, 2739, 2617, 1616, 1586, 1572, 1501, 1449, 1405, 1369, 1346, 1315, 1218, 1181, 1108, 1038, 1017, 873, 831, 765, 721, 675, 675, 648, 508; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.00–7.85 (comp, 2H), 7.59 (ddd, *J* = 8.3, 6.8, 1.3 Hz, 1H), 7.44 (ddd, *J* = 8.3, 6.8, 1.2 Hz, 1H), 4.87 (s, 2H), 3.13 (br s, 1H), 2.70 (s, 3H), 2.65 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 158.6, 146.6, 143.5, 129.9, 129.2, 128.9, 127.1, 125.8, 124.1, 59.0, 23.7, 14.2; *m/z* (ESI–MS) 188.2 [M + H]$^+$.



(4-ethyl-2-methylquinolin-3-yl)methanol (12h): Following the general procedure D, **12h** was obtained from **11h** as a yellow solid in 83% yield (R_f = 0.24 in hexane/EtOAc 20:80 v/v); mp = 106–108 °C; IR (KBr) 3186, 2979, 2934, 2874, 1611, 1585, 1571, 1502, 1438, 1404, 1373, 1181, 1108, 1031, 1015, 865, 774, 650; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.92 (app dt, *J* = 8.5, 1.7 Hz, 2H), 7.65–7.55 (m, 1H), 7.45 (app td, *J* = 7.8, 6.9, 1.9 Hz, 1H), 4.86 (s, 2H), 3.30 (br s, 1H), 3.13 (q, *J* = 7.5 Hz, 3H), 2.72 (s, 3H), 1.28 (t, *J* = 7.6 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 159.1, 149.1, 147.2, 129.2, 129.1, 126.0, 125.9, 124.0, 58.6, 23.6, 21.3, 15.8; *m/z* (ESI–MS) 202.1 [M + H]$^+$.



(6-chloro-2-methyl-4-phenylquinolin-3-yl)methanol (12i): Following the general procedure D, **12i** was obtained from **11i** as a yellow solid in 55% yield (R_f = 0.42 in hexane/EtOAc 20:80 v/v); mp = 137–139 °C; IR (KBr) 3060, 2910, 2826, 2724, 2361, 1584, 1570, 1484, 1444, 1375, 1348, 1301, 1078, 1040, 950, 831, 707, 603, 516; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.96 (dd, *J* = 8.9, 1.6 Hz, 1H), 7.70–7.44 (comp, 4H), 7.38–7.22 (comp, 3H), 4.60 (s, 2H), 2.90 (s, 3H), 1.86 (br s, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 159.9, 147.3, 145.6, 135.7, 131.8, 130.4, 130.3, 130.2, 129.5, 128.8, 128.6, 127.5, 125.6, 60.1, 23.7; *m/z* (ESI–MS) (^{35}Cl) 284.0 [M + H]$^+$, (^{37}Cl) 286.1 [M + H]$^+$.

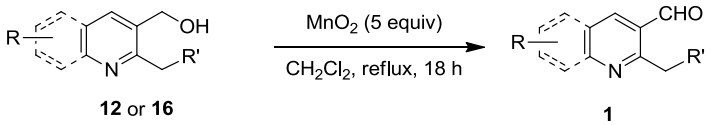


(2-ethylquinolin-3-yl)methanol (12k): Following the general procedure D, **12k** was obtained from **11k** as a yellow solid in 56% yield (R_f = 0.56 in hexane/EtOAc 20:80 v/v); mp = 87–90 °C; IR (KBr) 3134, 2970, 2832, 1619, 1603, 1566, 1495, 1458, 1420, 1353, 1217, 1187, 1136, 1100, 1048, 919, 845, 786, 743, 653, 621, 481; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.13 (s, 1H), 8.05 (d, *J* = 8.5 Hz, 1H), 7.82–7.73 (m, 1H), 7.66 (ddd, *J* = 8.4, 6.9, 1.5 Hz, 1H), 7.48 (ddd, *J* = 8.2, 6.9, 1.2 Hz, 1H), 4.92 (s, 2H), 3.00 (q, *J* = 7.5 Hz, 2H), 1.39 (t, *J* = 7.5 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 162.1, 134.3, 132.2, 129.3, 128.6, 127.6, 127.1, 126.1, 110.2, 62.4, 28.8, 13.5; *m/z* (ESI–MS) 188.1 [M + H]$^+$.

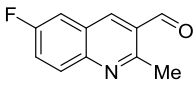
Synthesis of 2-alkylquinoline-3-carbaldehydes and 2-alkylnicotinaldehydes 1

General Procedure F

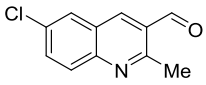
To a stirred solution of compounds **12** or **16** (2 mmol, 1 equiv) in CH_2Cl_2 (15 mL) was added MnO_2 (869 mg, 10 mmol, 5 equiv) at room temperature. The resulting mixture was heated under reflux until **12** or **16** was consumed as indicated by TLC. The reaction mixture was then allowed to cool to room temperature, filtered through a short pad of celite and washed with CH_2Cl_2. The solvent was removed under reduced pressure and the residue purified by silica gel chromatography.



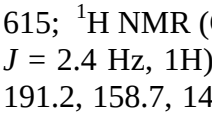
6-fluoro-2-methylquinoline-3-carbaldehyde (1b): Following the general procedure F, **1b** was obtained from **12b** as a yellow solid in 75% yield (over 2 steps), (R_f = 0.20 in hexane/EtOAc 80:20 v/v); mp = 104–105 °C; IR (KBr) 1714, 1685, 1592, 1564, 1496, 1373, 1349, 1210, 1154, 1105, 965, 833, 750, 585, 488; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 10.39 (s, 1H), 8.54 (s, 1H), 8.07 (dd, *J* = 9.2, 5.1 Hz, 1H), 7.60 (dddd, *J* = 9.1, 8.2, 2.8, 0.8 Hz, 1H), 7.55 (dd, *J* = 8.3, 2.8 Hz, 1H), 3.02 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 191.3, 160.5 (d, $J_{C\text{-}F}$ = 249.6 Hz), 157.7 (d, $J_{C\text{-}F}$ = 2.8 Hz), 146.4, 141.5 (d, $J_{C\text{-}F}$ = 5.4 Hz), 131.4 (d, $J_{C\text{-}F}$ = 9.0 Hz), 128.6, 126.7 (d, $J_{C\text{-}F}$ = 10.1 Hz), 122.8 (d, $J_{C\text{-}F}$ = 25.7 Hz), 111.9 (d, $J_{C\text{-}F}$ = 21.7 Hz), 23.9; *m/z* (ESI–MS) 222.0 [M + MeOH +H]$^+$.



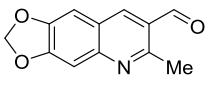
6-chloro-2-methylquinoline-3-carbaldehyde (1c): Following the general procedure F, **1c** was obtained from **12c** as a yellow solid in 70% yield (R_f = 0.24 in hexane/EtOAc 80:20 v/v); mp = 130–132 °C; IR (KBr) 3033, 3002, 1709, 1691, 1616, 1592, 1560, 1482, 1397, 1338, 1226, 1155, 1121, 1075, 928, 830, 769, 728, 667, 615; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 10.37 (s, 1H), 8.48 (s, 1H), 7.99 (d, *J* = 9.0 Hz, 1H), 7.90 (d, *J* = 2.4 Hz, 1H), 7.75 (dd, *J* = 8.9, 2.3 Hz, 1H), 3.01 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 191.2, 158.7, 147.7, 141.2, 133.5, 132.8, 130.6, 128.7, 127.6, 126.8, 24.1; *m/z* (ESI–MS) (^{35}Cl) 238.0 [M + MeOH +H]$^+$, (^{37}Cl) 240.0 [M + MeOH + H]$^+$.



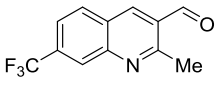
6-bromo-2-methylquinoline-3-carbaldehyde (1d): Following the general procedure F, **1d** was obtained from **12d** as a yellow solid in 60% yield (over 2 steps), (R_f = 0.23 in hexane/EtOAc 80:20 v/v); mp = 136–138 °C; IR (KBr) 2975, 1708, 1692, 1612, 1588, 1559, 1481, 1391, 1375, 1226, 1156, 1118, 1058, 923, 830, 765, 648; ^{1}H NMR ($CDCl_3$, 500 MHz) δ10.37 (s, 1H), 8.48 (s, 1H), 8.08 (d, *J* = 2.1 Hz, 1H), 7.92 (ddd, *J* = 9.2, 2.4, 1.0 Hz, 1H), 7.88 (dt, *J* = 9.0, 1.8 Hz, 1H), 3.01 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 191.12, 158.9, 147.9, 141.1, 136.1, 131.0, 130.6, 128.7, 127.3, 120.8, 24.1; *m/z* (ESI–MS) (^{79}Br) 282.0 [M + MeOH + H]$^+$, (^{81}Br) 283.9 [M + MeOH + H]$^+$.



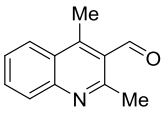
6-methyl-[1,3]dioxolo[4,5-*g*]quinoline-7-carbaldehyde (1e): Following the general procedure C, **1e** was obtained from **12e** as a yellow solid in 99% yield (R_f = 0.11 in hexane/EtOAc 80:20 v/v); mp = 171–173 °C; IR (KBr) 3043, 2927, 2718, 1692, 1590, 1501, 1465, 1389, 1245, 1205, 1170, 1136, 1035, 936, 801, 765, 595, 433; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 10.30 (s, 1H), 8.33 (s, 1H), 7.32 (s, 1H), 7.10 (s, 1H), 6.13 (s, 2H), 2.93 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 191.4, 156.8, 153.5, 148.5, 148.2, 140.0, 126.5, 123.1, 105.6, 103.7, 102.3, 23.4; *m/z* (ESI–MS) 248.0 [M + MeOH + H]$^+$.



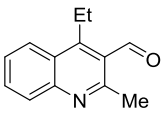
2-methyl-7-(trifluoromethyl)quinoline-3-carbaldehyde (1f): Following the general procedure F, **1f** was obtained from **12f** as a yellow solid in 90% yield (over 2 steps), (R_f = 0.29 in hexane/EtOAc 80:20 v/v); mp = 64–66 °C; IR (KBr) 2896, 1697, 1685, 1599, 1558, 1351, 1323, 1284, 1162, 1125, 1055, 935, 895, 870, 825, 763, 690; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 10.42 (s, 1H), 8.64 (s, 1H), 8.38 (dd, *J* = 1.7, 0.9 Hz, 1H), 8.07 (d, *J* = 8.4 Hz, 1H), 7.75 (dd, *J* = 8.5, 1.7 Hz, 1H), 3.06 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 191.1, 159.9, 148.4, 141.8, 134.1 (q, *J* = 33.0 Hz), 130.3, 129.5, 126.8 (q, *J* = 4.3 Hz), 123.74 (q, *J* = 272.8 Hz), 122.74 (q, *J* = 3.2 Hz), 24.2; *m/z* (ESI–MS) 271.9 [M + MeOH + H]$^+$.



2,4-dimethylquinoline-3-carbaldehyde (1g): Following the general procedure F, **1g** was obtained from **12g** as a yellow solid in 80% yield, (R_f = 0.29 in hexane/EtOAc 70:30 v/v); mp = 111–113 °C; IR (KBr) 2924, 2356, 2344, 1689, 1608, 1570, 1496, 1441, 1398, 1374, 1188, 1138, 1110, 1030, 754, 645; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 10.77 (s, 1H), 8.12 (dd, *J* = 8.3, 1.5 Hz, 1H), 8.06–7.95 (m, 1H), 7.77 (ddd, *J* = 8.4, 6.9, 1.4 Hz, 1H), 7.56 (ddd, *J* = 8.3, 6.8, 1.3 Hz, 1H), 2.93 (s, 3H), 2.92 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 193.3, 158.0, 148.8, 148.3, 131.8, 129.6, 126.9, 126.8, 126.7, 124.8, 24.9, 14.2.; *m/z* (ESI–MS) 218.1 [M + MeOH + H]$^+$.



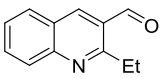
4-ethyl-2-methylquinoline-3-carbaldehyde (1h): Following the general procedure F, **1h** was obtained from **12h** as a yellow solid in 75% yield (R_f = 0.35 in hexane/EtOAc 70:30 v/v); mp = 78–80 °C; IR (KBr) 3057, 2967, 2925, 2864, 2762, 1689, 1611, 1561, 1498, 1406, 1238, 1181, 1133, 1103, 1063, 905, 853, 770, 743, 643, 508; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 10.76 (s, 1H), 8.18–8.11 (m, 1H), 8.08–7.99 (m, 1H), 7.85–7.72 (m, 1H), 7.64–7.47 (m, 1H), 3.41 (q, *J* = 7.5 Hz, 2H), 2.92 (s, 3H), 1.39 (t, *J* = 7.6 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 193.0, 158.2, 154.8, 148.8, 131.7, 129.8, 126.8, 125.8, 125.7, 124.5, 25.1, 20.9, 16.0; *m/z* (ESI–MS) 232.0 [M + MeOH + H]$^+$.



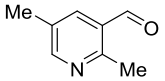
6-chloro-2-methyl-4-phenylquinoline-3-carbaldehyde (1i): Following the general procedure F, **1i** was obtained from **12i** as a yellow solid in 55% yield (R_f = 0.54 in hexane/EtOAc 70:30 v/v); mp = 220–222 °C; IR (KBr) 3052, 2999, 2869, 1685, 1559, 1478, 1406, 1379, 1338, 1296, 1148, 1123, 1073, 953, 833, 750, 707, 655, 618, 465; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 9.96 (s, 1H), 8.03 (dd, *J* = 8.9, 0.5 Hz, 1H), 7.73 (dd, *J* = 8.9, 2.3 Hz, 1H), 7.65–7.55 (comp, 3H), 7.50 (dd, *J* = 2.3, 0.5 Hz, 1H), 7.41–7.34 (comp, 2H), 2.99 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 193.3, 158.3, 154.0, 147.0, 133.1, 133.0, 132.8, 130.8, 130.2, 129.5, 129.0, 126.8, 126.3, 125.7, 25.8; *m/z* (ESI–MS) (^{35}Cl) 314.0 [M + MeOH + H]$^+$, (^{37}Cl) 316.0 [M + MeOH + H]$^+$.

2-benzylquinoline-3-carbaldehyde (1j): Following the general procedure F, **1j** was obtained from **12j** as a yellow solid in 70% yield (over 2 steps), (R_f = 0.52 in hexane/EtOAc 70:30 v/v); mp = 114–116 °C; IR (KBr) 2918, 2363, 2345, 1698, 1619, 1561, 1419, 1430, 1171, 1108, 923, 786, 759, 723, 696, 463; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 10.31 (s, 1H), 8.63 (d, *J* = 0.8 Hz, 1H), 8.16 (dd, *J* = 8.5, 0.9 Hz, 1H), 7.95 (dd, *J* = 8.1, 1.4 Hz, 1H), 7.87 (ddd, *J* = 8.5, 6.9, 1.5 Hz, 1H), 7.61 (ddd, *J* = 8.1, 6.9, 1.1 Hz, 1H), 7.32–7.22 (comp, 4H), 7.17 (ddt, *J* = 8.5, 6.1, 1.9 Hz, 1H), 4.77 (s, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 190.8, 160.0, 149.4, 142.4, 139.0, 132.7, 129.3, 129.2, 129.0, 128.6, 127.9, 127.3, 126.6, 126.4, 42.3; *m/z* (ESI–MS) 280.1 [M + MeOH + H]$^+$.

2-ethylquinoline-3-carbaldehyde (1k): Following the general procedure F, **1k** was obtained from **12k** as a yellow solid in 70% yield (R_f = 0.46 in hexane/EtOAc 70:30 v/v); mp = 57–59 °C; IR (KBr) 2975, 2934, 2874, 2360, 2342, 1698, 1685, 1619, 1595, 1561, 1490, 1452, 1421, 1373, 1343, 1213, 1186, 1151, 1104, 1013, 955, 921, 803, 750, 598, 482; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 10.39 (s, 1H), 8.61 (s, 1H), 8.09 (d, *J* = 8.5 Hz, 1H), 7.93 (d, *J* = 8.1 Hz, 1H), 7.87–7.72 (m, 1H), 7.57 (t, *J* = 7.5 Hz, 1H), 3.39 (q, *J* = 7.5 Hz, 2H), 1.40 (t, *J* = 7.5 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 191.3, 163.4, 149.6, 142.5, 132.7, 129.2, 129.1, 127.6, 127.0, 126.2, 29.8, 14.2; *m/z* (ESI–MS) 218.2 [M + MeOH + H]$^+$.



2,5-dimethylnicotinaldehyde (1n): Following the general procedure F, **1n** was obtained from **16a** as a white solid in 70% yield (over 2 steps), (R_f = 0.18 in hexane/EtOAc 70:30 v/v); mp = 37–40 °C; IR (KBr) 2364, 2341, 1683, 1598, 1558, 1468, 1291, 1226, 1198, 1118, 995, 950, 890, 793, 720, 665, 623, 508; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 10.31 (s, 1H), 8.50 (d, *J* = 2.4 Hz, 1H), 8.09–7.77 (m, 1H), 2.83 (s, 3H), 2.38 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 191.7, 157.5, 153.9, 138.4, 131.5, 129.0, 21.9, 18.0; *m/z* (ESI–MS) 168.1 [M + MeOH + H]$^+$.



2-benzylnicotinaldehyde (1o): Following the general procedure F, **1o** was obtained from **16b** as a yellow liquid in 80% yield (over 2 steps), (R_f = 0.29 in hexane/EtOAc 70:30 v/v); IR (KBr) 3410, 3054, 2922, 2852, 2356, 2341, 1696, 1633, 1581, 1493, 1453, 1266, 743; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 10.34 (s, 1H), 8.77 (dd, *J* = 4.8, 1.9 Hz, 1H), 8.14 (dd, *J* = 7.8, 1.9 Hz, 1H), 7.37 (dd, *J* = 7.8, 4.8 Hz, 1H), 7.31–7.22 (comp, 4H), 7.21–7.15 (m, 1H), 4.61 (s, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 191.1, 162.4, 153.8, 139.1, 138.4, 129.6, 128.9, 128.8, 126.7, 122, 41.2; *m/z* (ESI–MS) 230.0 [M + MeOH + H]$^+$.

2-ethylnicotinaldehyde (1p): Following the general procedure F, **1p** was obtained from **16c** as a yellow liquid in 80% yield (over 2 steps), (R_f = 0.24 in hexane/EtOAc 70:30 v/v); IR (KBr) 2969, 2934, 2874, 2371, 1701, 1696, 1681, 1581, 1453, 1261, 1218, 1098, 1063, 1033, 845, 758, 588; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 10.34 (d, *J* = 0.5 Hz, 1H), 8.72 (dd, *J* = 4.8, 1.9 Hz, 1H), 8.11 (dd, *J* = 7.8, 1.9 Hz, 1H), 7.32 (ddd, *J* = 7.8, 4.8, 0.6 Hz, 1H), 3.23 (q, *J* = 7.6 Hz, 2H), 1.34 (t, *J* = 7.6 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 191.1, 165.6, 153.5, 138.2, 129.0, 121.9, 28.3, 14.6; *m/z* (ESI–MS) 168.2 [M + MeOH + H]$^+$.

General Procedure G for the redox-annulation of 2-alkylquinoline-3-carbaldehydes with secondary amines

To a solution of the aldehyde (0.5 mmol, 1 equiv) in toluene (2.5 mL) and acetic acid (2.5 mL) was added the amine (0.75 mmol, 1.5 equiv). The resulting mixture was heated under reflux until the aldehyde was consumed as indicated by TLC. The reaction mixture was then allowed to cool to room temperature, diluted with EtOAc (15 mL), and washed with 1N NaOH (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.

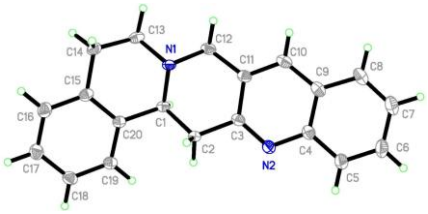
General Procedure H for the redox-annulation of 2-methylpyridine-3-carbaldehyde with secondary amines

To a solution of the aldehyde (0.5 mmol, 1 equiv) in acetic acid (0.1 M, 5 mL) was added the amine (0.75 mmol, 1.5 equiv). The resulting mixture was heated under reflux until the aldehyde was consumed as indicated by TLC. The reaction mixture was then allowed to cool to room temperature, diluted with EtOAc (15 mL), and washed with 1N NaOH (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-methylquinoline-3-carbaldehyde (342 mg, 2 mmol, 1 equiv) in toluene (10 mL) and acetic acid (10 mL) was added 1,2,3,4-tetrahydroisoquinoline (0.377 mL, 3 mmol, 1.5 equiv). The resulting mixture was heated under reflux for 1.5 h. The reaction mixture was then allowed to cool to room temperature, diluted with EtOAc (40 mL), and washed with 1N NaOH (2 x 50 mL). The combined aqueous layers were extracted with EtOAc (2 x 40 mL), and the combined organic layers washed with brine (50 mL) and dried over anhydrous Na_2SO_4. Solvent was then removed under reduced pressure and the residue purified

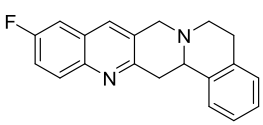
by silica gel chromatography. Product (±)-**2a** was obtained as a yellow solid in 85% yield (486 mg). (R_f = 0.19 in hexane/EtOAc 70:30 v/v); mp = 173–175 °C; IR (KBr) 3016, 2923, 2816, 2748, 2360, 1671, 1584, 1493, 1466, 1427, 1315, 1270, 1250, 1163, 1144, 1099, 1045, 815, 738, 520, 478; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.01 (app d, *J* = 8.4 Hz, 1H), 7.84 (s, 1H), 7.74 (app d, *J* = 8.1, 1H), 7.65 (ddd, *J* = 8.4, 6.8, 1.5 Hz, 1H), 7.47 (ddd, *J* = 8.2, 6.7, 1.2 Hz, 1H), 7.35 (app d, *J* = 7.7 Hz, 1H), 7.25 (app td, *J* = 7.6, 7.2, 1.7 Hz, 1H), 7.22–7.11 (comp, 2H), 4.24 (d, *J* = 14.9 Hz, 1H), 3.99–3.73 (comp, 3H), 3.38–3.14 (comp, 3H), 2.81 (dd, *J* = 14.7, 3.7 Hz, 1H), 2.75–2.61 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 156.7, 147.2, 137.4, 134.3, 132.6, 129.1, 129.0, 128.6, 128.3, 127.3, 127.1, 126.4, 126.0, 125.6, 60.2, 58.1, 51.4, 40.8, 29.6; *m/z* (ESI–MS) 287.1 $[M + H]^+$.

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

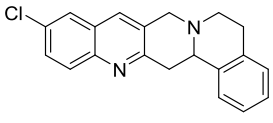


The requisite CIF has been submitted to the journal and deposited with the CCDC (deposition # 1541944).

(±)-**2b**: Following general procedure G, product (±)-**2b** was obtained from **1b** and 1,2,3,4-tetrahydroisoquinoline (reflux for 1 h) as a yellow solid in 85% yield (130 mg), (R_f = 0.23 in hexane/EtOAc 70:30 v/v); mp = 171–172 °C; IR (KBr) 2924, 2813, 2760, 1627, 1612, 1497, 1410, 1361, 1329, 1300, 1235, 1213, 1163, 1140, 1108, 1046, 984, 961, 901, 875, 829, 760, 736, 470; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.00 (dd, *J* = 9.2, 5.3 Hz, 1H), 7.80 (s, 1H), 7.42 (ddd, *J* = 9.2, 8.3, 2.8 Hz, 1H), 7.38–7.32 (comp, 2H), 7.28–7.23 (m, 1H), 7.21 (app tdd, *J* = 7.1, 1.5, 0.7 Hz, 1H), 7.18–7.15 (m, 1H), 4.24 (d, *J* = 14.9, 1H), 3.94–3.84 (comp, 2H), 3.80 (dd, *J* = 17.1, 3.9 Hz, 1H), 3.33–3.15 (comp, 3H), 2.90–2.78 (m, 1H), 2.75–2.63 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 160.3 (d, $J_{C\text{-}F}$ = 246.9 Hz), 156.1 (d, $J_{C\text{-}F}$ = 2.8 Hz), 144.3, 137.3, 134.3, 132.0 (d, $J_{C\text{-}F}$ = 5.5 Hz), 131.0 (d, $J_{C\text{-}F}$ = 9.2 Hz), 129.2, 129.0, 127.6 (d, $J_{C\text{-}F}$ = 9.9 Hz), 126.5 (d, $J_{C\text{-}F}$ = 3.4 Hz), 125.7, 119.4 (d, $J_{C\text{-}F}$ = 25.8 Hz), 110.2 (d, $J_{C\text{-}F}$ = 21.7 Hz), 60.1, 58.0, 51.3, 40.7, 29.6; *m/z* (ESI–MS) 305.1 $[M + H]^+$.

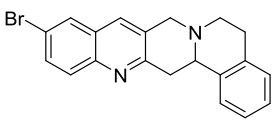


(±)-**2c**: Following general procedure G, product (±)-**2c** was obtained from **1c** and 1,2,3,4-tetrahydroisoquinoline (reflux for 1 h) as a yellow solid in 86% yield (137 mg), (R_f = 0.31 in hexane/EtOAc 70:30 v/v); mp = 179–181 °C; IR (KBr) 2926, 2810, 2757, 1601, 1484, 1407, 1372, 1325, 1299, 1160, 1142, 1104, 1075, 932, 919, 827, 757, 734, 670, 485; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.93 (d, *J* = 8.9 Hz, 1H), 7.74 (s, 1H), 7.71 (d, *J* = 2.3 Hz, 1H), 7.57 (dd, *J* = 8.9, 2.3 Hz, 1H), 7.33 (d, *J* = 7.7 Hz, 1H), 7.28–7.23 (m, 1H), 7.23–7.18 (m, 1H), 7.16 (d, *J* = 7.2 Hz, 1H), 4.22 (d, *J* = 15.1 Hz, 1H), 3.93–3.83 (comp, 2H), 3.79 (dd, *J* = 17.2, 3.9 Hz, 1H), 3.33–3.14 (comp, 3H), 2.88–2.76 (m, 1H), 2.74–2.62 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 157.1, 145.6, 137.2, 134.2, 131.67, 131.66, 130.2, 130.0, 129.4, 129.0, 127.6, 126.52, 126.48, 125.9,

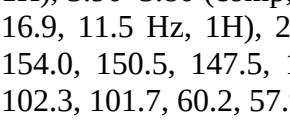


125.6, 60.0, 58.0, 51.3, 40.8, 29.6; *m/z* (ESI–MS) (^{35}Cl) 321.0 [M + H]$^+$, (^{37}Cl) 323.0 [M + H]$^+$.

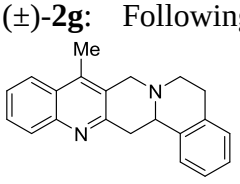
(±)-**2d**: Following general procedure G, product (±)-**2d** was obtained from **1d** and 1,2,3,4-tetrahydroisoquinoline (reflux for 1.5 h) as a yellow solid in 85% yield (156 mg), (R_f = 0.33 in hexane/EtOAc 70:30 v/v); mp = 180–183 °C; IR (KBr) 2924, 2756, 2360, 1596, 1480, 1406, 1324, 1298, 1141, 1103, 913, 900, 827, 737; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.90 (d, *J* = 2.2 Hz, 1H), 7.87 (d, *J* = 8.9 Hz, 1H), 7.75 (s, 1H), 7.71 (dd, *J* = 9.0, 2.2 Hz, 1H), 7.33 (d, *J* = 7.7 Hz, 1H), 7.28–7.23 (m, 1H), 7.23–7.19 (m, 1H), 7.17 (d, *J* = 7.2, 1H), 4.23 (d, *J* = 15.1 Hz, 1H), 3.93–3.83 (comp, 2H), 3.79 (dd, *J* = 17.3, 3.9 Hz, 1H), 3.35–3.13 (comp, 3H), 2.90–2.78 (m, 1H), 2.75–2.63 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 157.3, 145.8, 137.2, 134.2, 132.6, 131.6, 130.4, 129.4, 129.3, 129.0, 128.2, 126.54, 126.50, 125.6, 119.8, 60.0, 58.0, 51.3, 40.8, 29.6; *m/z* (ESI–MS) (^{79}Br) 365.0 [M + H]$^+$, (^{81}Br) 366.9 [M + H]$^+$.



(±)-**2e**: Following general procedure G, product (±)-**2e** was obtained from **1e** and 1,2,3,4-tetrahydroisoquinoline (reflux for 1.5 h) as a yellow solid in 83% yield (136 mg), (R_f = 0.12 in hexane/EtOAc 70:30 v/v); mp = 201–203 °C; IR (KBr) 2893, 2813, 1621, 1495, 1461, 1395, 1238, 1214, 1164, 1098, 1035, 947, 877, 800, 732, 585, 458; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.67 (s, 1H), 7.34 (d, *J* = 7.7 Hz, 1H), 7.30 (s, 1H), 7.25–7.22 (m, 1H), 7.22–7.18 (m, 1H), 7.17–7.13 (m, 1H), 6.99 (s, 1H), 6.08 (d, *J* = 1.1 Hz, 1H), 6.07 (d, *J* = 1.2 Hz, 1H), 4.17 (d, *J* = 14.7 Hz, 1H), 3.90–3.80 (comp, 2H), 3.73 (dd, *J* = 17.0, 3.9 Hz, 1H), 3.34–3.20 (comp, 2H), 3.16 (dd, *J* = 16.9, 11.5 Hz, 1H), 2.88–2.77 (m, 1H), 2.74–2.63 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 154.0, 150.5, 147.5, 145.4, 137.6, 134.3, 131.8, 129.0, 126.43, 126.40, 125.7, 123.9, 105.2, 102.3, 101.7, 60.2, 57.9, 51.4, 40.4, 29.6; *m/z* (ESI–MS) 331.1 [M + H]$^+$.



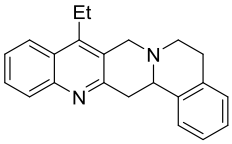
(±)-**2f**: Following general procedure G, product (±)-**2f** was obtained from **1f** and 1,2,3,4-tetrahydroisoquinoline (reflux for 1 h) as a light yellow solid in 80% yield (142 mg), (R_f = 0.26 in hexane/EtOAc 70:30 v/v); mp = 159–160 °C; IR (KBr) 3027, 2930, 2815, 2748, 2361, 2344, 1673, 1616, 1561, 1493, 1436, 1334, 1319, 1297, 1224, 1197, 1171, 1141, 1123, 1104, 1059, 940, 905, 823, 738, 688, 483; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.33 (dd, *J* = 2.0, 1.1 Hz, 1H), 7.92 (s, 1H), 7.87 (d, *J* = 8.5 Hz, 1H), 7.65 (dd, *J* = 8.5, 1.8 Hz, 1H), 7.35 (d, *J* = 7.7 Hz, 1H), 7.30–7.25 (m, 1H), 7.25–7.20 (m, 1H), 7.18 (dd, *J* = 7.7, 1.5 Hz, 1H), 4.28 (d, *J* = 15.2 Hz, 1H), 3.97–3.88 (comp, 2H), 3.84 (dd, *J* = 17.3, 3.9 Hz, 1H), 3.36–3.21 (comp, 3H), 2.90–2.80 (m, 1H), 2.78–2.67 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 158.6, 146.1, 137.1, 134.2, 132.4, 130.8 (q, $J_{C\text{-}F}$ = 32.5 Hz), 130.6, 129.0, 128.5, 126.59, 126.57, 126.54, 126.50, 125.6, 124.2 (q, $J_{C\text{-}F}$ = 272.4 Hz), 121.7 (q, $J_{C\text{-}F}$ = 3.2 Hz), 60.0, 58.0, 51.3, 40.8, 29.6; *m/z* (ESI–MS) 355.1 [M + H]$^+$.



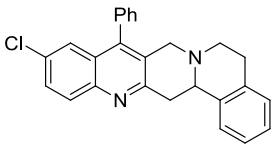
(±)-**2g**: Following general procedure G, product (±)-**2g** was obtained from **1g** and 1,2,3,4-tetrahydroisoquinoline (reflux for 1 h) as a white solid in 87% yield (131 mg), (R_f = 0.11 in hexane/EtOAc 50:50 v/v); mp = 171–173 °C; IR (KBr) 3061, 2945, 2897, 2792, 1671, 1600, 1497, 1466, 1387, 1269, 1252, 1111, 1051, 813, 756, 744, 518; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.01 (app d, *J* =

8.3 Hz, 1H), 7.92 (app d, *J* = 8.1 Hz, 1H), 7.64 (ddd, *J* = 8.4, 6.8, 1.4 Hz, 1H), 7.51 (ddd, *J* = 8.2, 6.8, 1.3 Hz, 1H), 7.40 (app d, *J* = 7.6 Hz, 1H), 7.31–7.27 (m, 1H), 7.26–7.22 (m, 1H), 7.21–7.17 (m, 1H), 4.16 (d, *J* = 15.6 Hz, 1H), 3.89–3.76 (comp, 2H), 3.72 (app dt, *J* = 15.6, 2.0 Hz, 1H), 3.37–3.21 (comp, 2H), 3.19–3.07 (m, 1H), 2.90–2.74 (comp, 2H), 2.65 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 156.2, 146.1, 139.2, 137.5, 134.7, 129.25, 129.17, 128.6, 126.8, 126.6, 126.4, 126.3, 125.9, 125.6, 122.5, 58.7, 56.4, 51.4, 33.6, 29.6, 22.8; *m/z* (ESI–MS) 300.9 $[M + H]^+$.

(±)-**2h**: Following general procedure G, product (±)-**2h** was obtained from **1h** and 1,2,3,4-tetrahydroisoquinoline (reflux for 1 h) as a yellow solid in 87% yield (137 mg), (R_f = 0.24 in hexane/EtOAc 70:30 v/v); mp = 116–119 °C; IR (KBr) 3059, 2970, 2939, 2742, 1588, 1496, 1458, 1408, 1371, 1291, 1156, 1101, 1036, 1013, 742, 477; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.06–7.96 (comp, 2H), 7.64 (ddd, *J* = 8.4, 6.7, 1.4 Hz, 1H), 7.50 (ddd, *J* = 8.4, 6.6, 1.3 Hz, 1H), 7.35 (app d, *J* = 7.7 Hz, 1H), 7.29–7.13 (comp, 3H), 4.35 (d, *J* = 15.1 Hz, 1H), 3.94–3.67 (comp, 3H), 3.38–3.17 (comp, 3H) 3.15–2.95 (comp, 2H), 2.93–2.79 (m, 1H), 2.78–2.69 (m, 1H), 1.32 (t, *J* = 7.6 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 156.3, 147.1, 145.2, 137.4, 134.3, 129.4, 128.9, 128.6, 126.43, 126.40, 125.84, 125.79, 125.7, 125.3, 123.3, 59.5, 56.2, 51.6, 41.5, 29.6, 20.6, 13.9; *m/z* (ESI–MS) 315.2 $[M + H]^+$.

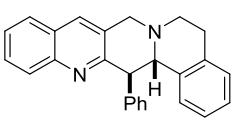


(±)-**2i**: Following general procedure G, product (±)-**2i** was obtained from **1i** and 1,2,3,4-tetrahydroisoquinoline (reflux for 1 h) as a yellow solid in 93% yield (184 mg), (R_f = 0.46 in hexane/EtOAc 70:30 v/v); mp = 205–207 °C; IR (KBr) 3026, 2893, 2746, 1671, 1581, 1483, 1366, 1333, 1270, 1172, 1103, 953, 841, 758, 724, 704, 678, 613; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.99 (d, *J* = 9.0 Hz, 1H), 7.64–7.48 (comp, 4H), 7.40–7.33 (comp, 2H), 7.32–7.28 (m, 1H), 7.28–7.23 (comp, 2H), 7.22–7.17 (m, 1H), 7.14 (app d, *J* = 7.5 Hz, 1H), 3.91–3.81 (comp, 3H), 3.55 (d, *J* = 15.8 Hz, 1H), 3.28 (dd, *J* = 17.7, 12.2 Hz, 1H), 3.17 (ddd, *J* = 16.7, 12.0, 5.2 Hz, 1H), 3.05 (ddd, *J* = 11.2, 5.3, 1.9 Hz, 1H), 2.79–2.70 (m, 1H), 2.57 (app td, *J* = 11.6, 3.4 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 156.9, 145.5, 144.2, 137.2, 135.4, 134.3, 131.8, 130.4, 129.9, 129.2, 129.1, 129.0, 128.9, 128.6, 127.3, 126.9, 126.51, 126.50, 125.6, 124.7, 59.8, 57.2, 51.4, 41.4, 29.6; *m/z* (ESI–MS) (^{35}Cl) 397.0 $[M + H]^+$, (^{37}Cl) 399.0 $[M + H]^+$.



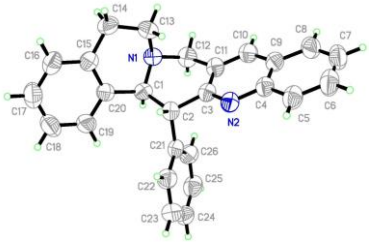
(±)-**2j and** (±)-**2j'**: Following the general procedure G, products (±)-**2j** and (±)-**2j'** were obtained from **1j** and 1,2,3,4-tetrahydroisoquinoline (reflux for 18 h) in a 4:1 ratio (153 mg, 84% combined yield).

Characterization data for major diastereomer (±)-**2j**: Yellow liquid (68% yield, 123 mg), (R_f = 0.20 in hexane/EtOAc 70:30 v/v); mp = 113–115 °C; IR (KBr) 3025, 2916, 1736, 1619, 1599, 1493, 1452, 1421, 1308, 1240, 1155, 1041, 783, 746, 699; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.91 (s, 1H), 7.85 (d, *J* = 8.5 Hz, 1H), 7.74 (dd, *J* = 8.4, 1.5 Hz, 1H), 7.54 (ddd, J = 8.5, 6.8, 1.5 Hz, 1H), 7.44 (ddd, *J* = 8.2, 6.8, 1.2 Hz, 1H), 3.35–3.10 (comp, 3H), 7.27–7.20 (m, 1H), 7.20–7.09 (comp, 4H), 6.89 (ddd, *J* = 8.3, 5.8, 2.9 Hz, 1H), 6.35 (app d, *J* = 7.8 Hz, 1H), 4.66 (d, *J* = 15.9 Hz, 1H), 4.52 (comp, 2H), 4.19 (d, *J* = 15.9 Hz, 1H), 3.19 (ddd, *J* = 11.4, 8.0, 5.4 Hz, 1H), 3.13–2.98 (comp, 2H), 2.90 (ddd, *J* = 11.1, 6.3, 4.6 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 159.8, 147.9, 144.6,



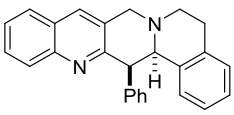
137.2, 133.5, 133.2, 130.4, 129.3, 129.1, 128.8, 128.5, 127.7, 127.2, 127.01, 126.98, 126.8, 126.4, 126.2, 125.0, 66.1, 56.8, 52.9, 46.7, 29.1; *m/z* (ESI–MS) 363.0 $[M + H]^+$.

X-ray quality crystals of (±)-**2j** were obtained from hexane/ethyl acetate through slow diffusion at room temperature. The relative stereochemistry was assigned by X-ray crystallography



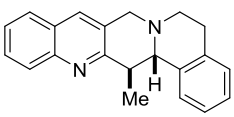
The requisite CIF has been submitted to the journal and deposited with the CCDC (deposition # 1541946).

Characterization data for minor diastereomer (±)-**2j'**: Yellow solid (17% yield, 30 mg), (R_f = 0.51 in hexane/EtOAc 70:30 v/v); mp = 162–165 °C; IR (KBr) 3059, 3024, 2912, 2804, 2759, 2361, 2345, 1654, 1600, 1560, 1493, 1459, 1426, 1377, 1314, 1226, 1147, 1095, 913, 746, 733, 700, 575, 474; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.03–7.93 (comp, 2H), 7.77 (dd, *J* = 8.4, 1.3 Hz, 1H), 7.60 (ddd, *J* = 8.5, 6.8, 1.5 Hz, 1H), 7.53–7.41 (comp, 2H), 7.24–7.19 (m, 1H), 7.17–7.09 (comp, 2H), 7.04 (app tt, *J* = 7.5, 1.0 Hz, 1H), 7.00–6.89 (comp, 4H), 4.92 (d, *J* = 3.4 Hz, 1H), 4.43 (dd, *J* = 15.0, 1.0 Hz, 1H), 4.34 (d, *J* = 2.9 Hz, 1H), 4.02 (d, *J* = 15.1 Hz, 1H), 3.21 (ddd, *J* = 10.5, 4.4, 2.0 Hz, 1H), 2.77 (dddd, *J* = 14.8, 11.7, 4.5, 1.4 Hz, 1H), 2.68 (ddd, *J* = 12.2, 10.3, 2.2 Hz, 1H), 2.52 (app dt, *J* = 15.0, 2.2 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$). δ 159.8, 147.5, 141.4, 136.0, 135.4, 133.0, 131.0, 129.2, 128.9, 128.64, 128.55, 127.18, 127.16, 126.8, 126.7, 126.2, 125.85, 125.82, 64.6, 57.6, 54.4, 50.6, 29.7; *m/z* (ESI–MS) 363.1 $[M + H]^+$.

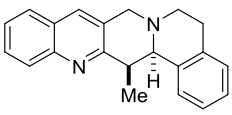


(±)-**2k and** (±)-**2k'**: Following the general procedure G, products (±)-**2k** and (±)-**2k'** were obtained from **1k** and 1,2,3,4-tetrahydroisoquinoline (reflux for 2 h) in a 2:1 ratio (143 mg, 95% combined yield).

Characterization data for major diastereomer (±)-**2k**: Yellow oil (63% yield, 95 mg), (R_f = 0.14 in hexane/EtOAc 70:30 v/v); IR (KBr) 2967, 2908, 2878, 2849, 2363, 1601, 1561, 1492, 1420, 1380, 1306, 1218, 1159, 1140, 1123, 1102, 1013, 966, 941, 908, 758, 742, 592, 450; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.02 (app d, *J* = 8.5 Hz, 1H), 7.81 (s, 1H), 7.73 (app d, *J* = 7.6 Hz, 1H), 7.63 (ddd, *J* = 8.5, 6.8, 1.5 Hz, 1H), 7.46 (ddd, *J* = 8.1, 6.8, 1.2 Hz, 1H), 7.36 (dd, *J* = 7.0, 2.2 Hz, 1H), 7.23–7.08 (comp, 3H), 4.26 (d, *J* = 15.4 Hz, 1H), 4.03 (d, *J* = 15.7 Hz, 1H), 3.92 (d, *J* = 7.8 Hz, 1H), 3.51 (app p, *J* = 7.2 Hz, 1H), 3.21–3.11 (m, 1H), 3.08–2.96 (comp, 2H), 2.93–2.83 (m, 1H), 1.73 (d, *J* = 7.1 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 161.1, 147.6, 138.1, 133.8, 132.9, 129.2, 128.9, 128.8, 127.4, 127.1, 127.0, 126.80, 126.78, 126.0, 125.6, 65.7, 56.1, 48.0, 40.1, 28.5, 22.0; *m/z* (ESI–MS) 301.1 $[M + H]^+$.

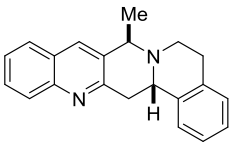


Characterization data for minor diastereomer (±)-**2k'**: Yellow oil (32% yield, 48 mg), (R_f = 0.54 in hexane/EtOAc 70:30 v/v); IR (KBr) 3024, 2983, 2935, 2796, 2756, 1619, 1494, 1450, 1424, 1359, 1318, 1234, 1191, 1111, 1060, 1019, 894, 859, 762, 743, 615, 481; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.07–8.03 (m, 1H), 7.86 (s, 1H), 7.79–7.73 (m, 1H), 7.66 (ddd, *J* = 8.4, 6.8, 1.5 Hz, 1H), 7.48 (ddd, *J* = 8.1, 6.8, 1.2 Hz, 1H), 7.31 (app d, *J* = 7.8 Hz, 1H), 7.28–7.22 (m, 1H), 7.22–7.13 (comp, 2H), 4.25 (d, *J* = 15.0 Hz, 1H), 4.03 (d, *J* = 2.6 Hz, 1H), 3.85 (d, *J* = 15.0 Hz, 1H), 3.74 (qd, *J* = 7.0, 3.3 Hz, 1H), 3.28–3.14 (m, 2H), 2.80–2.71 (m, 1H), 2.70–2.62 (m, 1H) 1.13 (d, *J* = 7.0 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 162.3, 147.3, 136.1, 135.9, 132.8, 129.0, 128.9, 128.7, 128.0, 127.3, 127.1, 126.5, 126.03, 125.98, 125.8, 63.7, 58.1, 51.1, 42.8, 29.8, 17.2; *m/z* (ESI–MS) 301.1 $[M + H]^+$.

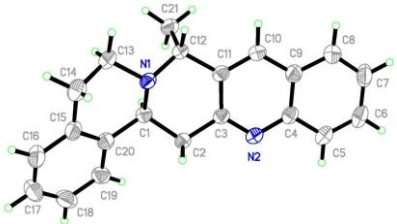


(±)-**2l and** (±)-**2l'**: To a solution of 1-(2-methylquinolin-3-yl)ethan-1-one (93 mg, 0.5 mmol, 1 equiv) in toluene (0.1 M, 5 mL) were added 1,2,3,4-tetrahydroisoquinoline (94 µL, 0.75 mmol, 1.5 equiv) and acetic acid (0.572 mL, 10 mmol, 20 equiv). The mixture was heated under reflux for 24 h. The reaction was then allowed to cool to room temperature, diluted with EtOAc (15 mL), and washed with 1N NaOH (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 was purified by silica gel chromatography. Products (±)-**2l** and (±)-**2l'** were obtained in a 3:1 ratio (126 mg, 83% combined yield).

Characterization data for major diastereomer (±)-**2l**: Yellow solid (63% yield, 95 mg), (R_f = 0.21 in hexane/EtOAc 70:30 v/v); mp = 154–156 °C; IR (KBr) 2969, 2941, 2904, 2821, 2364, 1622, 1604, 1496, 1450, 1381, 1381, 1297, 1171, 1143, 1104, 991, 908, 855, 761, 746, 479; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.01 (d, *J* = 8.5Hz, 1H), 7.88 (s, 1H), 7.76 (dd, *J* = 8.1, 1.4 Hz, 1H), 7.65 (ddd, *J* = 8.4, 6.9, 1.5 Hz, 1H), 7.47 (ddd, *J* = 8.0, 6.9, 1.2 Hz, 1H), 7.31 (d, *J* = 7.6 Hz, 1H), 7.24 (app td, *J* = 7.3, 1.7 Hz, 1H), 7.19 (app td, *J* = 7.3, 1.4 Hz, 1H), 7.16 (dd, *J* = 7.6, 1.7 Hz, 1H), 4.53 (dd, *J* = 11.4, 4.3 Hz, 1H), 4.46 (q, *J* = 6.9 Hz, 1H), 3.64 (dd, *J* = 17.5, 4.3 Hz, 1H), 3.18 (dd, *J* = 17.5, 11.4 Hz, 1H), 3.14–3.03 (comp, 2H), 2.97 (ddd, *J* = 10.7, 9.7, 3.9 Hz, 1H), 2.91 (app dt, *J* = 16.2, 4.2 Hz, 1H), 1.48 (d, *J* = 6.8 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 156.2, 147.2, 138.4, 134.4, 133.9, 133.5, 129.10, 129.08, 128.5, 127.3, 127.2, 126.5, 126.2, 125.9, 59.1, 51.0, 47.3, 40.5, 30.2, 17.8; *m/z* (ESI–MS) 301.1 $[M + H]^+$.

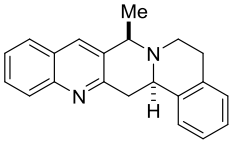


X-ray quality crystals of (±)-**2l** were obtained from hexane/ethyl acetate through slow diffusion at room temperature. The relative stereochemistry was assigned by X-ray crystallography

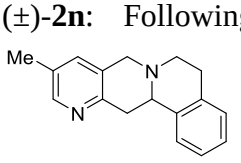


The requisite CIF has been submitted to the journal and deposited with the CCDC (deposition # 1541945).

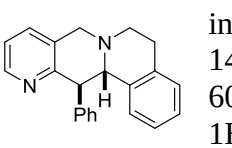
Characterization data for minor diastereomer (±)-**2l'**: Yellow oil (20% yield, 31 mg), (R_f = 0.29 in hexane/EtOAc 70:30 v/v); IR (KBr) 3024, 2983, 2935, 2796, 2756, 1619, 1494, 1450, 1424, 1359, 1318, 1234, 1191, 1111, 1060, 1019, 894, 859, 762, 743, 615, 481; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.02 (app d, *J* = 8.5, 1H), 7.99 (s, 1H), 7.78 (dd, *J* = 8.1, 1.4 Hz, 1H), 7.67 (ddd, *J* = 8.4, 6.8, 1.4 Hz, 1H), 7.48 (ddd, *J* = 8.1, 6.8, 1.2 Hz, 1H), 7.34 (app d, *J* = 7.6 Hz, 1H), 7.23 (app td, *J* = 7.3, 1.9 Hz, 1H), 7.21–7.15 (comp, 2H), 4.03 (dd, *J* = 11.5, 2.3 Hz, 1H), 3.98 (q, *J* = 6.4 Hz, 1H), 3.72 (dd, *J* = 16.8, 3.3 Hz, 1H), 3.44 (ddd, *J* = 11.3, 5.2, 3.1 Hz, 1H), 3.30 (dd, *J* = 16.8, 11.6 Hz, 1H), 3.17 (ddd, *J* = 15.8, 10.9, 4.9 Hz, 1H), 2.86 (app dt, *J* = 16.2, 3.4 Hz, 1H), 2.55 (app td, *J* = 11.1, 3.6 Hz, 1H), 1.69 (d, *J* = 6.4 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$). δ 156.8, 146.7, 138.0, 134.7, 133.5, 133.4, 129.3, 128.9, 128.4, 127.5, 127.4, 126.4, 126.3, 126.0, 59.5, 59.3, 46.6, 41.2, 30.2, 21.8; *m/z* (ESI–MS) 301.0 $[M + H]^+$.



(±)-**2m**: Following general procedure H, product (±)-**2m** was obtained from **1m** and 1,2,3,4-tetrahydroisoquinoline (reflux for 2 h) as a white solid in 89% yield (105 mg), (R_f = 0.15 in hexane/EtOAc 20:80 v/v); mp = 98–100 °C; IR (KBr) 2957, 2920, 2816, 2755, 1668, 1578, 1493, 1446, 1338, 1293, 1140, 1104, 976, 778, 740, 722, 490; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.43 (app d, *J* = 4.8 Hz, 1H), 7.39 (app d, *J* = 7.7 Hz, 1H), 7.30 (app d, *J* = 7.8 Hz, 1H), 7.27–7.13 (comp, 3H), 7.09 (dd, *J* = 7.8, 4.8 Hz, 1H), 4.02 (d, *J* = 14.9 Hz, 1H), 3.79 (dd, *J* = 11.3, 3.1 Hz, 1H), 3.72 (d, *J* = 14.9 Hz, 1H), 3.59 (dd, *J* = 17.0, 4.0 Hz, 1H), 3.37–3.16 (comp, 2H), 3.06 (dd, *J* = 17.0, 11.4 Hz, 1H), 2.87–2.74 (m, 1H), 2.71–2.57 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 155.0, 147.9, 137.5, 134.3, 134.0, 129.9, 128.9, 126.39, 126.37, 125.6, 121.2, 59.9, 57.6, 51.2, 39.8, 29.6; *m/z* (ESI–MS) 237.2 $[M + H]^+$.



(±)-**2n**: Following general procedure H, product (±)-**2n** was obtained from **1n** and 1,2,3,4-tetrahydroisoquinoline (reflux for 2 h) as a white solid in 74% yield (93 mg), (R_f = 0.20 in hexane/EtOAc 20:80 v/v); mp = 152–155 °C; IR (KBr) 3009, 2938, 2892, 2795, 2750, 1601, 1576, 1496, 1468, 1360, 1287, 1248, 1141, 1108, 1015, 973, 873, 788, 751, 723, 560, 515, 478; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.26 (s, 1H), 7.29 (app d, *J* = 7.7 Hz, 1H), 7.23 (app td, *J* = 7.4, 1.7 Hz, 1H), 7.20–7.16 (comp, 2H), 7.14 (app d, *J* = 7.3 Hz, 1H), 3.98 (d, *J* = 14.9 Hz, 1H), 3.76 (dd, *J* = 11.4, 4.0 Hz, 1H), 3.68 (d, *J* = 14.9 Hz, 1H), 3.54 (dd, *J* = 16.9, 3.9 Hz, 1H), 3.27–3.12 (comp, 2H), 3.00 (dd, *J* = 16.8, 11.4 Hz, 1H), 2.83–2.73 (m, 1H), 2.65 (ddd, *J* = 12.0, 11.2, 3.5 Hz, 1H), 2.29 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 151.9, 148.3, 137.6, 134.4, 134.3, 130.5, 129.2, 128.9, 126.4, 126.3, 125.6, 60.0, 57.5, 51.2, 39.4, 29.6, 18.2; *m/z* (ESI–MS) 251.1 $[M + H]^+$.

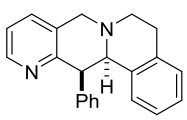


(±)-**2o and** (±)-**2o'**: Following the general procedure H, products (±)-**2o** and (±)-**2o'** were obtained from **1o** and 1,2,3,4-tetrahydroisoquinoline (reflux for 1.5 h) in a 3:1 ratio (125 mg, 81% combined yield).

Characterization data for major diastereomer (±)-**2o**: Yellow oil (60% yield, 160 mg), (R_f = 0.34 in hexane/EtOAc 50:50 v/v; IR (KBr) 3027, 2922, 2891, 2781, 1576, 1490, 1437, 1388, 1356, 1287, 1206, 1143, 1105, 1078, 1036, 948, 789, 755, 732, 703, 604, 564, 465; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.42–8.37 (m, 1H), 7.50–7.42 (m, 1H), 7.37–7.28 (comp, 2H), 7.28–7.19 (m, 1H), 7.17–7.09 (comp, 2H), 7.08–

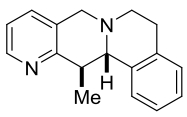
7.00 (comp, 3H), 6.89–6.76 (m, 1H), 6.16 (app d, *J* = 7.8 Hz, 1H), 4.49 (d, *J* = 16.4 Hz, 1H), 4.37 (d, *J* = 9.6 Hz, 1H), 4.29 (d, *J* = 9.6 Hz, 1H), 3.97 (d, *J* = 16.0 Hz, 1H), 3.22 (ddd, *J* = 11.4, 7.7, 5.8 Hz, 1H), 3.13–2.99 (comp, 2H), 2.92–2.82 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 158.0, 148.7, 144.0, 136.8, 134.3, 133.6, 130.3, 129.0, 128.9, 128.6, 127.8, 126.6, 126.5, 124.7, 121.2, 65.7, 56.5, 52.1, 46.7, 29.3; *m/z* (ESI–MS) 313.1 $[M + H]^+$.

Characterization data for minor diastereomer (±)-**2o'**: Yellow oil (21% yield, 32 mg), (R_f = 0.19 in hexane/EtOAc 50:50 v/v); mp = 142–144 °C; IR (KBr) 3026, 2890, 2837, 1575, 1490, 1437, 1388, 1356, 1211, 1143, 1105, 788, 755, 731, 703, 604, 564, 473; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.48–8.39 (m, 1H), 7.49 (app d, *J* = 7.7 Hz, 1H), 7.37 (app d, *J* = 7.9 Hz, 1H), 7.24–7.14 (comp, 3H), 7.11 (app dd, *J* = 7.7, 4.7 Hz, 1H), 7.06–6.92 (comp, 4H), 6.89 (app d, *J* = 7.6 Hz, 1H), 4.68 (d, *J* = 3.6 Hz, 1H), 4.23 (d, *J* = 3.7 Hz, 1H), 4.19 (d, *J* = 15.0 Hz, 1H), 3.83 (d, *J* = 15.0 Hz, 1H), 3.19 (ddd, *J* = 10.7, 4.8, 2.1 Hz, 1H), 2.82 (app tdd, *J* = 13.2, 5.1, 2.7 Hz, 1H), 2.65 (ddd, *J* = 11.9, 10.6, 2.7 Hz, 1H), 2.52 (app dt, *J* = 15.3, 2.3 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 158.2, 148.4, 141.2, 135.8, 135.4, 134.2, 130.9, 130.1, 128.6, 127.0, 126.7, 126.0, 125.74, 125.72, 121.4, 64.3, 57.2, 53.5, 50.6, 29.7; *m/z* (ESI–MS) 313.0 $[M + H]^+$.

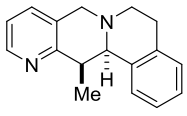


(±)-**2p and** (±)-**2p'**: Following the general procedure H, products (±)-**2p** and (±)-**2p'** were obtained from **1p** and 1,2,3,4-tetrahydroisoquinoline (reflux for 1.5 h) in a 2:1 ratio (120 mg, 96% combined yield).

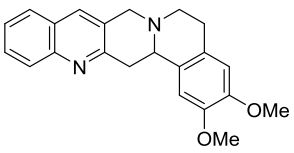
Characterization data for major diastereomer (±)-**2p**: Yellow oil (63% yield, 79 mg), (R_f = 0.10 in hexane/EtOAc 50:50 v/v; IR (KBr) 3052, 2987, 2687, 2411, 2359, 2301, 1421, 1266, 893, 748, 689, ; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.50–8.44 (m, 1H), 7.35 (ddd, *J* = 7.6, 1.7, 0.8 Hz, 1H), 7.29 (dd, *J* = 7.3, 1.5 Hz, 1H), 7.23–7.12 (comp, 3H), 7.07 (dd, *J* = 7.6, 4.8 Hz, 1H), 4.16 (d, *J* = 15.7 Hz, 1H), 3.85–3.78 (comp, 2H), 3.26 (app p, *J* = 7.2 Hz, 1H), 3.17 (ddd, *J* = 11.6, 7.1, 6.0 Hz, 1H), 3.09–2.99 (comp, 2H), 2.86 (app dt, *J* = 12.0, 6.1 Hz, 1H), 1.61 (d, *J* = 6.9 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 159.0, 148.0, 137.8, 134.3, 133.9, 129.3, 128.8, 127.1, 126.8, 125.4, 121.1, 65.1, 55.6, 47.5, 38.4, 28.4, 20.4; *m/z* (ESI–MS) 250.9 $[M + H]^+$.



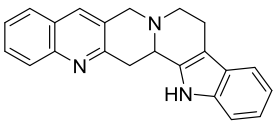
Characterization data for minor diastereomer (±)-**2p'**: Yellow oil (33% yield, 41 mg), (R_f = 0.29 in hexane/EtOAc 50:50 v/v); IR (KBr) 3057, 2969, 2919, 1586, 1572, 1492, 1449, 1439, 1380, 1307, 1142, 1123, 1100, 1010, 941, 845, 795, 780, 762, 732, 455; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.50–8.43 (m, 1H), 7.43–7.38 (m, 1H), 7.26–7.20 (comp, 2H), 7.19–7.12 (comp, 2H), 7.10 (dd, *J* = 7.7, 4.7 Hz, 1H), 4.01 (d, *J* = 15.0 Hz, 1H), 3.92 (d, *J* = 3.4, 1H), 3.68 (dd, *J* = 14.9, 1.0 Hz, 1H), 3.50 (qd, *J* = 7.0, 3.4 Hz, 1H), 3.28–3.10 (comp, 2H), 2.82–2.68 (m, 1H), 2.68–2.57 (m, 1H), 1.04 (d, *J* = 7.0 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$). δ 160.5, 147.8, 136.1, 135.8, 134.0, 129.5, 128.8, 126.4, 125.90, 125.86, 121.1, 63.4, 57.6, 50.9, 41.7, 29.8, 16.6; *m/z* (ESI–MS) 251.0 $[M + H]^+$..



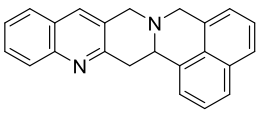
(±)-**2q**: Following general procedure G, product (±)-**2q** was obtained from **1a** and 6,7-dimethoxy-1,2,3,4-tetrahydroisoquinoline (reflux for 1.5 h) as a yellow solid in 93% yield (162 mg), (R_f = 0.12 in hexane/EtOAc 50:50 v/v); mp = 67–69 °C; IR (KBr) 2935, 2827, 1609, 1516 1498, 1464, 1430, 1375, 1363, 1260, 1230, 1209, 1134, 1110, 1021, 855, 786, 752; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 7.95 (app d, *J* = 8.5 Hz, 1H), 7.74 (s, 1H), 7.66 (app d, *J* = 7.9 Hz, 1H), 7.58 (ddd, *J* = 8.4, 6.8, 1.5 Hz, 1H), 7.40 (ddd, *J* = 8.1, 6.9, 1.2 Hz, 1H), 6.76 (s, 1H), 6.57 (s, 1H), 4.13 (d, *J* = 14.9 Hz, 1H), 3.82 (s, 3H), 3.80 (s, 3H), 3.77–3.62 (comp, 3H), 3.24–3.04 (comp, 3H), 2.72–2.53 (comp, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 13C NMR (126 MHz, cdcl3) δ 156.4, 147.52, 147.48, 146.9, 132.5, 129.03, 128.95, 128.3, 128.1, 127.2, 126.9, 126.2, 125.8, 111.2, 108.2, 77.4, 59.6, 57.8, 55.82, 55.78, 51.4, 40.8, 28.9; *m/z* (ESI–MS) 347.0 $[M + H]^+$.



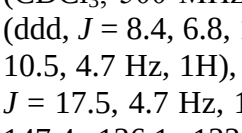
(±)-**2r**: Following general procedure G, product (±)-**2r** was obtained from **1a** and 2,3,4,9-tetrahydro-1*H*-pyrido[3,4-*b*]indole (reflux for 1.5 h) as a yellow solid in 74% yield (120 mg), (R_f = 0.29 in hexane/EtOAc 50:50 v/v); mp (decomposition at 230 °C); IR (KBr) 3268, 2942, 2896, 2824, 1604, 1492, 1452, 1431, 1314, 1276, 1164, 1128, 1063, 983, 906, 858, 743, 617, 479; ^{1}H NMR (DMSO, 500 MHz) δ 10.99 (s, 1H), 8.07 (s, 1H), 7.94 (app d, *J* = 8.4 Hz, 1H), 7.88 (app d, *J* = 8.1 Hz, 1H), 7.71–7.65 (m, 1H), 7.52 (app t, *J* = 7.5 Hz, 1H), 7.43 (app d, *J* = 7.8 Hz, 1H), 7.37 (app d, *J* = 8.0 Hz, 1H), 7.08 (app t, *J* = 7.5 Hz, 1H), 6.99 (app t, *J* = 7.4 Hz, 1H), 4.27 (d, *J* = 15.3 Hz, 1H), 4.00–3.75 (comp, 3H), 3.28 (dd, *J* = 11.2, 5.2 Hz, 1H), 3.01 (dd, *J* = 16.5, 11.4 Hz, 1H), 2.89 (dddd, *J* = 14.3, 11.7, 5.5, 2.3 Hz, 1H), 2.80–2.72 (m, 1H), 2.65 (app td, *J* = 11.3, 4.0 Hz, 1H); ^{13}C NMR (125 MHz, DMSO) δ 155.8, 146.4, 136.3, 134.9, 132.5, 128.9, 128.6, 128.1, 127.4, 126.6, 126.5, 125.9, 120.7, 118.4, 117.7, 111.1, 106.6, 56.4, 56.1, 51.6, 38.4, 21.2; *m/z* (ESI–MS) 326.0 $[M + H]^+$.



(±)-**2s**: Following general procedure G, product (±)-**2s** was obtained from **1a** and 2,3-dihydro-1*H*-benzo[*de*]isoquinoline (reflux for 1 h) as a white solid in 50% yield (80 mg), (R_f = 0.24 in hexane/EtOAc 50:50 v/v); mp (decomposition at 150 °C); IR (KBr) 3047, 2928, 2743, 2361, 1624, 1600, 1497, 1430, 1415, 1320, 1306, 1164, 1129, 1086, 1006, 976, 822, 776, 745, 479; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.04 (app d, *J* = 8.5 Hz, 1H), 7.86 (s, 1H), 7.83–7.72 (comp, 3H), 7.67 (ddd, *J* = 8.4, 6.8, 1.5 Hz, 1H), 7.59–7.40 (comp, 4H), 7.28 (app d, *J* = 7.0 Hz, 1H), 4.41 (dd, *J* = 10.5, 4.7 Hz, 1H), 4.35 (app d, *J* = 14.9 Hz, 2H), 4.07 (app dd, *J* = 20.6, 14.9 Hz, 2H), 3.94 (dd, *J* = 17.5, 4.7 Hz, 1H), 3.45 (dd, *J* = 17.5, 10.4 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 155.8, 147.4, 136.1, 133.2, 132.8, 132.1, 129.2, 128.6, 127.5, 127.4, 127.34, 127.27, 126.7, 126.6, 126.11, 126.09, 125.7, 122.1, 121.8, 59.1, 56.7, 56.0, 38.6; *m/z* (ESI–MS) 323.2 $[M + H]^+$.

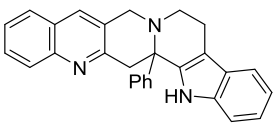


(±)-**2t**: Following general procedure G, product (±)-**2t** was obtained from **1a** and 1-phenyl-1,2,3,4-tetrahydroisoquinoline (reflux for 12 h) as a white solid in 63% yield (114 mg), (R_f = 0.69 in hexane/EtOAc 50:50 v/v); mp = 176–179 °C; IR (KBr) 3050, 2918, 2853, 2361, 2209, 1621, 1492, 1470, 1428, 1316, 1159, 1015, 916, 905, 771, 751, 727, 704, 479 ; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.10–8.06 (m, 1H), 7.76–7.70 (comp, 2H), 7.68 (ddd, *J* = 8.4, 6.9, 1.5 Hz, 1H), 7.48 (ddd, *J* = 8.1, 6.9, 1.2 Hz, 1H), 7.25–7.12 (comp, 7H), 7.08 (ddd, *J* = 7.8, 6.9, 1.8 Hz, 1H), 6.87 (dd, *J* =

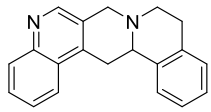


7.9, 1.2 Hz, 1H), 4.07 (d, *J* = 18.1 Hz, 1H), 3.94–3.77 (comp, 2H), 3.71 (d, *J* = 18.1 Hz, 1H), 3.35–3.22 (m, 1H), 3.22–3.11 (comp, 2H), 3.07 (app dt, *J* = 15.3, 5.0 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 156.6, 147.4, 143.9, 143.4, 133.1, 132.5, 129.0, 128.8, 128.6, 128.3, 127.9, 127.31, 127.29, 127.0, 126.3, 126.2, 126.0, 63.1, 52.8, 46.4, 40.9, 29.9; *m/z* (ESI–MS) 363.1 [M + H]$^+$.

(±)-**2u**: Following general procedure G, product (±)-**2u** was obtained from **1a** and 1-phenyl-2,3,4,9-tetrahydro-1*H*-pyrido[3,4-*b*]indole (reflux for 14 h) as a white solid in 80% yield (160 mg), (R_f = 0.60 in hexane/EtOAc 50:50 v/v); mp = 240–242 °C; IR (KBr) 3053, 2910, 2834, 1736, 1618, 1598, 1561, 1491, 1464, 1446, 1413, 1312, 1303, 1236, 1178, 1110, 1098, 1045, 988, 908, 744, 704, 435; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.03 (app d, *J* = 8.6 Hz, 1H), 7.84 (br s, 1H), 7.80 (s, 1H), 7.77–7.72 (m, 1H), 7.67 (ddd, *J* = 8.4, 6.9, 1.4 Hz, 1H), 7.61–7.56 (m, 1H), 7.50 (ddd, *J* = 8.1, 6.8, 1.2 Hz, 1H), 7.29–7.23 (m, 1H), 7.21–7.11 (comp, 7H), 4.06 (d, *J* = 17.6 Hz, 1H), 3.98 (dd, *J* = 16.8 Hz, 1H), 3.85 (d, *J* = 7.1 Hz, 1H), 3.81 (d, *J* = 5.9 Hz, 1H), 3.26–3.17 (comp, 2H), 3.14–3.07 (comp, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 156.0, 147.3, 140.6, 138.7, 136.6, 132.8, 129.2, 128.8, 128.6, 128.5, 128.3, 127.9, 127.4, 127.3, 127.1, 126.2, 121.9, 119.6, 118.6, 111.3, 108.1, 60.4, 51.7, 47.1, 40.8, 21.8; *m/z* (ESI–MS) 402.0 [M + H]$^+$.

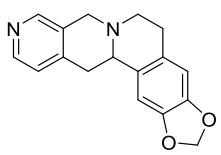


(±)-**4a**: Following general procedure H, product (±)-**4a** was obtained from **3a** and 1,2,3,4-tetrahydroisoquinoline (reflux for 2 h) as a yellow solid in 57% yield (82 mg), (R_f = 0.10 in hexane/EtOAc 50:50 v/v); mp = 70–72 °C; IR (KBr) 3027, 2917, 2752, 2364, 2344, 1736, 1689, 1653, 1621, 1506, 1501, 1456, 1388, 1143, 1108, 753, 733; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.67 (s, 1H), 8.09 (dd, *J* = 8.5, 1.2 Hz, 1H), 7.95 (dd, *J* = 8.4, 1.3 Hz, 1H), 7.67 (ddd, *J* = 8.3, 6.8, 1.4 Hz, 1H), 7.56 (ddd, *J* = 8.2, 6.8, 1.3 Hz, 1H), 7.39 (app d, *J* = 7.5 Hz, 1H), 7.28 (app td, *J* = 7.5, 1.6 Hz, 1H), 7.25–7.21 (m, 1H), 7.19 (app d, *J* = 7.4 Hz, 1H), 4.19 (d, *J* = 15.4 Hz, 1H), 3.98–3.75 (comp, 3H), 3.32–3.19 (comp, 2H), 3.18–3.06 (m, 1H), 2.93–2.56 (comp, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 149.4, 146.8, 139.4, 137.5, 134.6, 130.2, 129.2, 128.6, 127.6, 127.2, 126.8, 126.6, 126.4, 125.6, 122.6, 59.2, 56.4, 51.2, 33.1, 29.6; *m/z* (ESI–MS) 287.0 [M + H]$^+$.

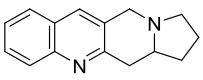


(±)-**4b**: Following general procedure G, product (±)-**4b** was obtained from **3b** and 1,2,3,4-tetrahydroisoquinoline (reflux for 1 h) as a yellow oil in 91% yield (107 mg), (R_f = 0.22 in hexane/EtOAc 20:80 v/v); IR (KBr) 3054, 2987, 2684, 2411, 2304, 1596, 1418, 1263, 1148, 1100, 888, 745, 698; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.43–8.32 (comp, 2H), 7.26–7.17 (comp, 3H), 7.17–7.12 (m, 1H), 7.09 (app d, *J* = 5.0 Hz, 1H), 4.07 (d, *J* = 15.0 Hz, 1H), 3.78–3.66 (comp, 2H), 3.37 (dd, *J* = 17.0, 4.0 Hz, 1H), 3.27–3.14 (comp, 2H), 2.90 (app dddt, *J* = 17.0, 11.1, 1.9, 1.0 Hz, 1H), 2.84–2.75 (m, 1H), 2.73–2.60 (m, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 147.8, 147.4, 143.8, 137.2, 134.6, 130.8, 129.1, 126.5, 126.3, 125.5, 123.6, 59.4, 56.0, 51.4, 36.2, 29.5; *m/z* (ESI–MS) 237.1 [M + H]$^+$.

(±)-**4c**: Following general procedure G, product (±)-**4c** was obtained from **3b** and 5,6,7,8-tetrahydro-[1,3]dioxolo[4,5-g]isoquinoline (reflux for 1.5 h) as a yellow solid in 85% yield (119 mg), (R_f = 0.24 in MeOH/EtOAc 10:90 v/v); mp = 140–142 °C; IR (KBr) 2945, 1915, 2794, 2734, 1597, 1501, 1486, 1420, 1393, 1361, 1339, 1303, 1246, 1225, 1127, 1036, 932, 861, 823, 760, 543, 453; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.36 (app s, 2H), 7.06 (d, *J* = 4.8 Hz, 1H), 6.70 (app s, 1H), 6.59 (s, 1H), 5.93–5.91 (comp, 2H), 4.04 (d, *J* = 15.0 Hz, 1H), 3.68 (d, *J* = 15.0 Hz, 1H), 3.60 (dd, *J* = 11.1, 3.7 Hz, 1H), 3.26 (dd, *J* = 16.9, 3.9 Hz, 1H), 3.19–3.02 (comp, 2H), 2.85 (dd, *J* = 16.9, 11.2 Hz, 1H), 2.70–2.56 (comp, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 147.8, 147.4, 146.4, 146.3, 143.6, 130.1, 127.8, 123.6, 108.6, 105.5, 101.0, 59.4, 55.9, 51.5, 36.4, 29.6; *m/z* (ESI–MS) 280.9 $[M + H]^+$.



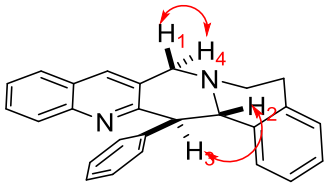
(±)-**5**: A 10 mL microwave reaction tube was charged with a 10 x 8 mm SiC passive heating element, 2-methylquinoline-3-carbaldehyde (34.2 mg, 0.2 mmol, 1 equiv), acetic acid (2 mL), and pyrrolidine (81 µL, 1 mmol, 5 equiv). The reaction tube was sealed with a Teflon-lined snap cap, and heated in a microwave reactor at 200 °C (200 W, 70–100 psi) for 15 minutes. After cooling with compressed air flow, the reaction mixture was diluted with EtOAc (15 mL) and then washed with 1N NaOH (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. (±)-**5** was obtained as a yellow solid in 59% yield (27 mg), (R_f = 0.24 in MeOH/EtOAc 10:90 v/v); mp = 115–118 °C; IR (KBr) 2964, 2827, 2799, 2364, 1621, 1558, 1491, 1456, 1413, 1303, 1161, 1126, 1023, 998, 955, 900, 780, 750, 615, 478; ^{1}H NMR ($CDCl_3$, 500 MHz) δ 8.01–7.95 (m, 1H), 7.81 (s, 1H), 7.75–7.69 (m, 1H), 7.63 (ddd, *J* = 8.4, 6.9, 1.5 Hz, 1H), 7.45 (ddd, *J* = 8.1, 6.8, 1.2 Hz, 1H), 4.32 (d, *J* = 14.7 Hz, 1H), 3.56 (d, *J* = 14.6 Hz, 1H), 3.39 (dd, *J* = 16.9, 3.8 Hz, 1H), 3.32 (app td, *J* = 8.6, 2.3 Hz, 1H), 3.00 (dd, *J* = 16.8, 11.1 Hz, 1H), 2.54 (dddd, *J* = 10.9, 9.7, 6.8, 3.8 Hz, 1H), 2.32 (app q, *J* = 8.9 Hz, 1H), 2.19 (dddd, *J* = 12.3, 9.8, 6.8, 4.2 Hz, 1H), 1.98 (app ddtd, *J* = 12.8, 11.2, 8.7, 4.2 Hz, 1H), 1.87 (ddddd, *J* = 12.4, 9.7, 8.8, 6.4, 2.3 Hz, 1H), 1.65 (dddd, *J* = 12.4, 11.3, 9.5, 6.4 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 157.2, 147.1, 133.1, 129.0, 128.63, 128.59, 127.3, 126.9, 126.0, 61.4, 55.2, 54.8, 40.3, 31.5, 22.0; *m/z* (ESI–MS) 225.2 $[M + H]^+$.



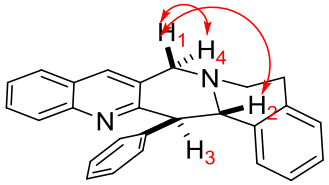
Alternate procedure for the preparation of (±)-**5:** Following general procedure H, **1a** and proline were heated under reflux for 1h. Product (±)-**5** was obtained as a yellow solid in 70% yield (78 mg). The characterization data matched those shown above.

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

GCOSY



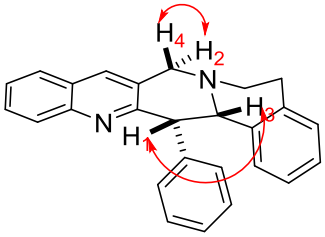
NOESY



Protons	Chemical Shifts (ppm)
H_1,	4.66,
H_2, H_3,	4.56–4.49
H_4	4.19

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

GCOSY



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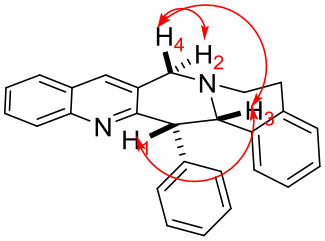
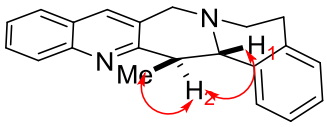


Protons	Chemical Shifts (ppm)
H_1	4.91
H_2	4.43
H_3	4.34
H_4	4.02

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

GCOSY



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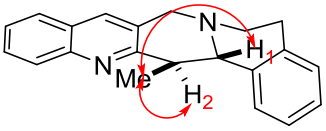
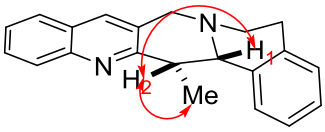


Protons	Chemical Shifts (ppm)
H_1	3.92
H_2	3.51
Me	1.73

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

GCOSY



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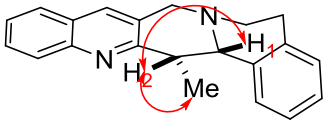
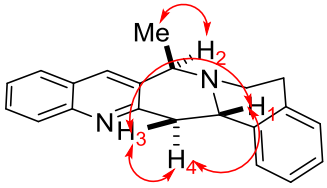


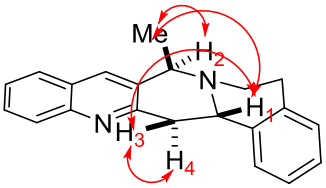
Protons	Chemical Shifts (ppm)
H_1	4.02
H_2	3.74
Me	1.13

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

GCOSY



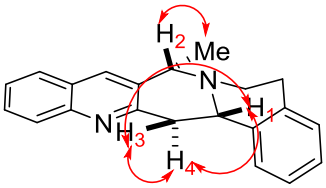
NOESY



Protons	Chemical Shifts (ppm)
H_1	4.53
H_2	4.45
H_3	3.65
H_4	3.18
Me	1.48

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

GCOSY



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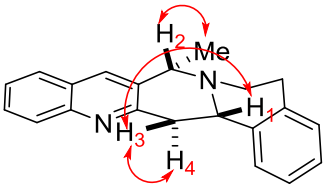
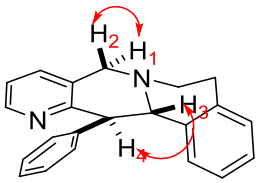


Protons	Chemical Shifts (ppm)
H_1	4.03
H_2	3.98
H_3	3.72
H_4	3.30
Me	1.69

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

GCOSY



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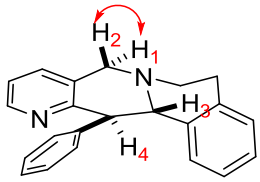
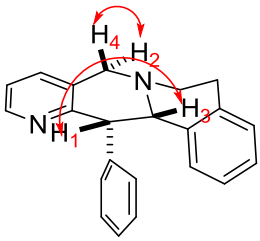


Protons	Chemical Shifts (ppm)
H_1, H_2	4.50, 3.97
H_3, H_4	4.38, 4.31

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

GCOSY



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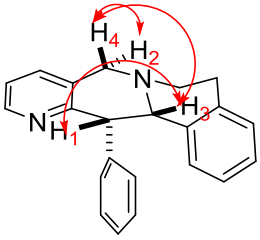
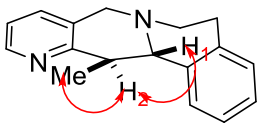


Protons	Chemical Shifts (ppm)
H_1	4.67
H_2	4.23
H_3	4.19
H_4	3.83

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

GCOSY



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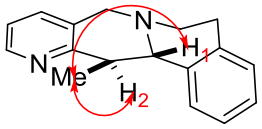
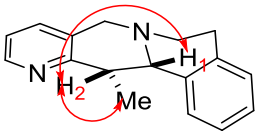


Protons	Chemical Shifts (ppm)
H_1	3.85–3.79
H_2	3.26
Me	1.61

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

GCOSY



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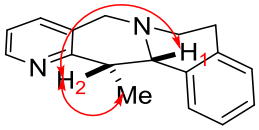

Protons	Chemical Shifts (ppm)
H_1	3.90
H_2	3.49
Me	1.04

References:

(1) Huo, M.; Kuang, Y.; Chen, F. *Org. Prep. Proced. Int*. **2004**, *36*, 331.
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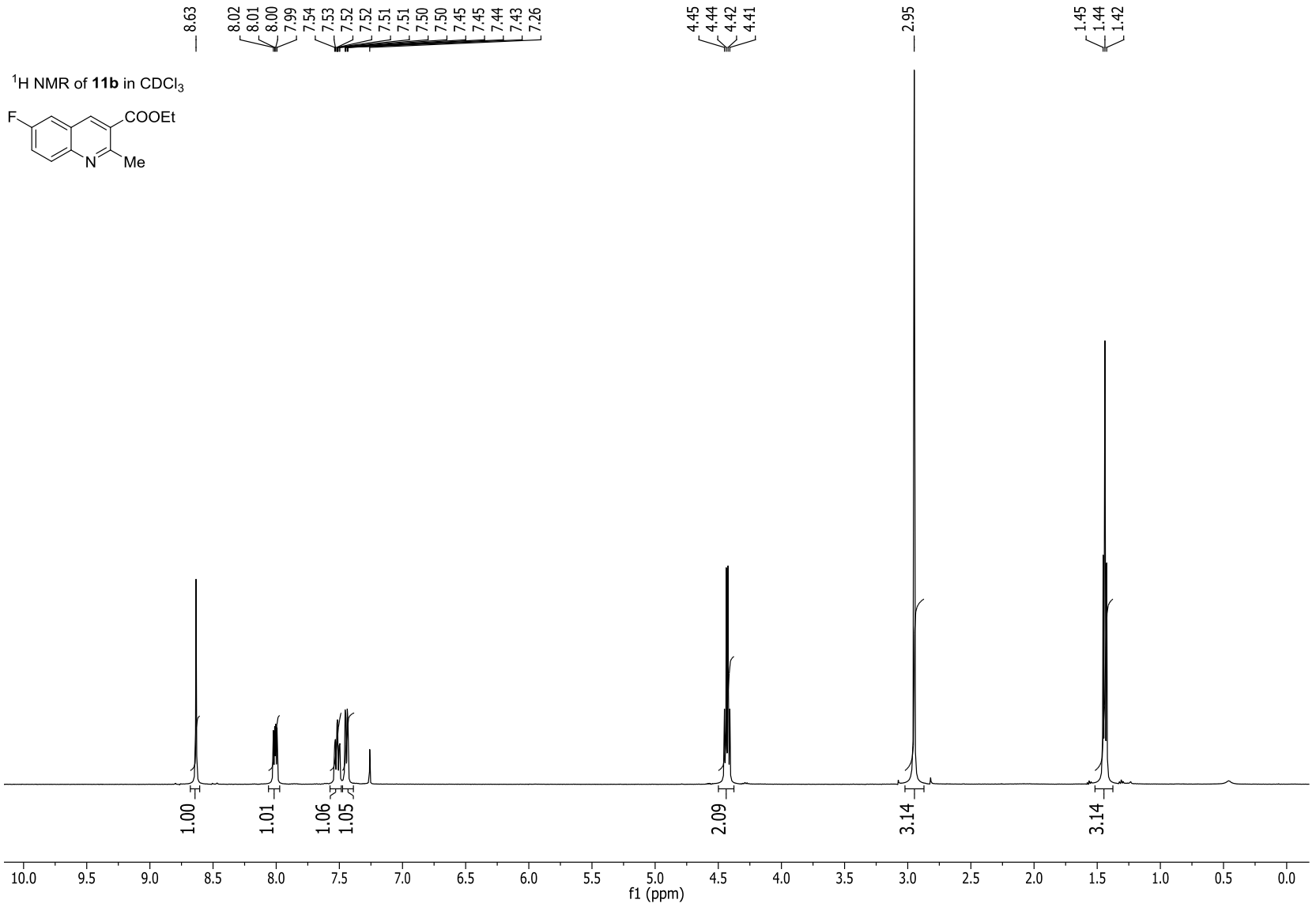
(9) Huo, M.; Kuang, Y.; Chen, F. *Org. Prep. Proced. Int*. **2004**, *36*, 331.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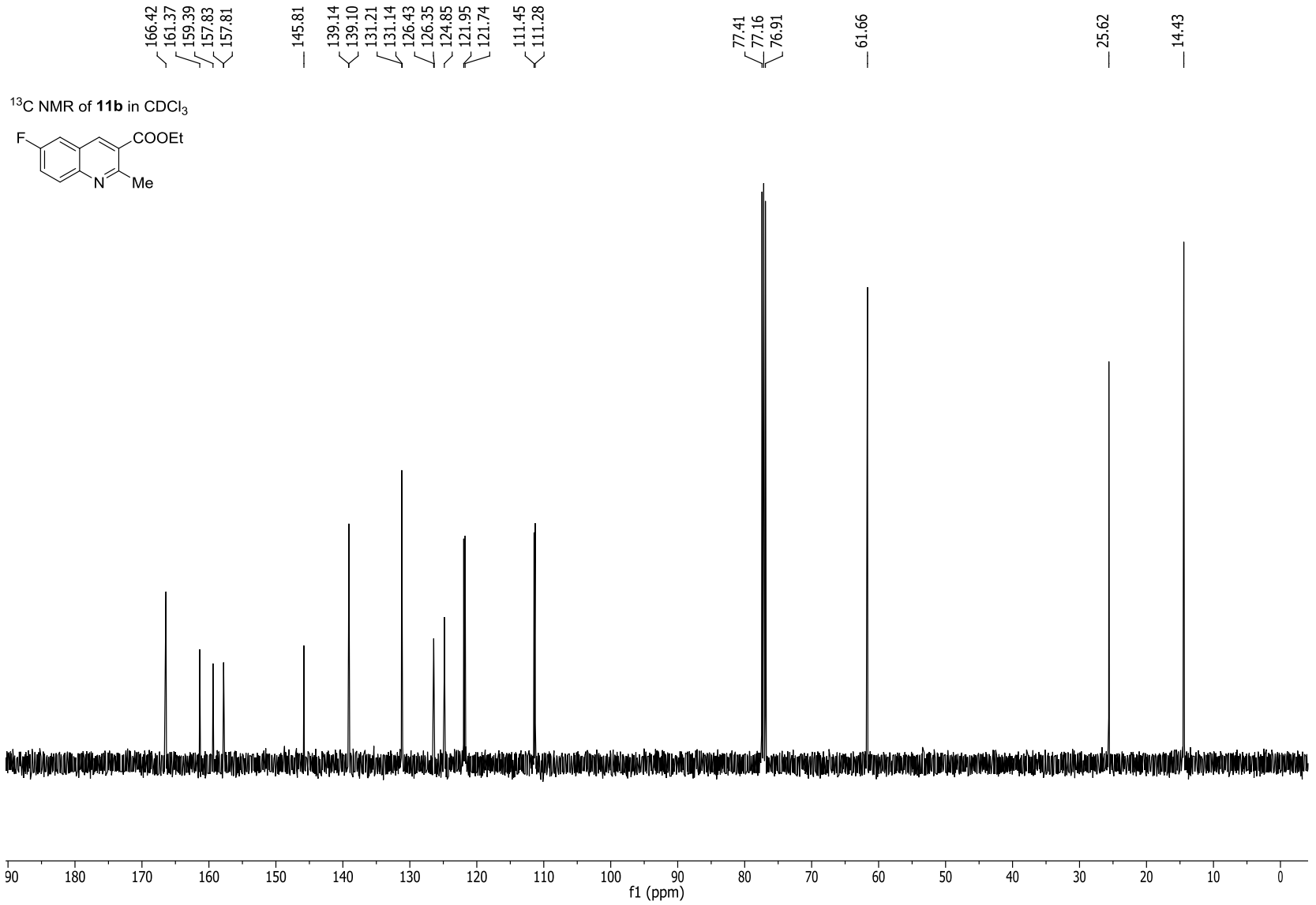
8.02
8.01
8.00
7.99
7.54
7.53
7.52
7.52
7.51
7.51
7.50
7.50
7.45
7.45
7.44
7.43
7.26
4.45
4.44
4.42
4.41
2.95
1.45
1.44
1.42
1H NMR of 11b in CDCl3
F
COOEt
N
Me
1.00
1.01
1.06
1.05
2.09
3.14
3.14
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)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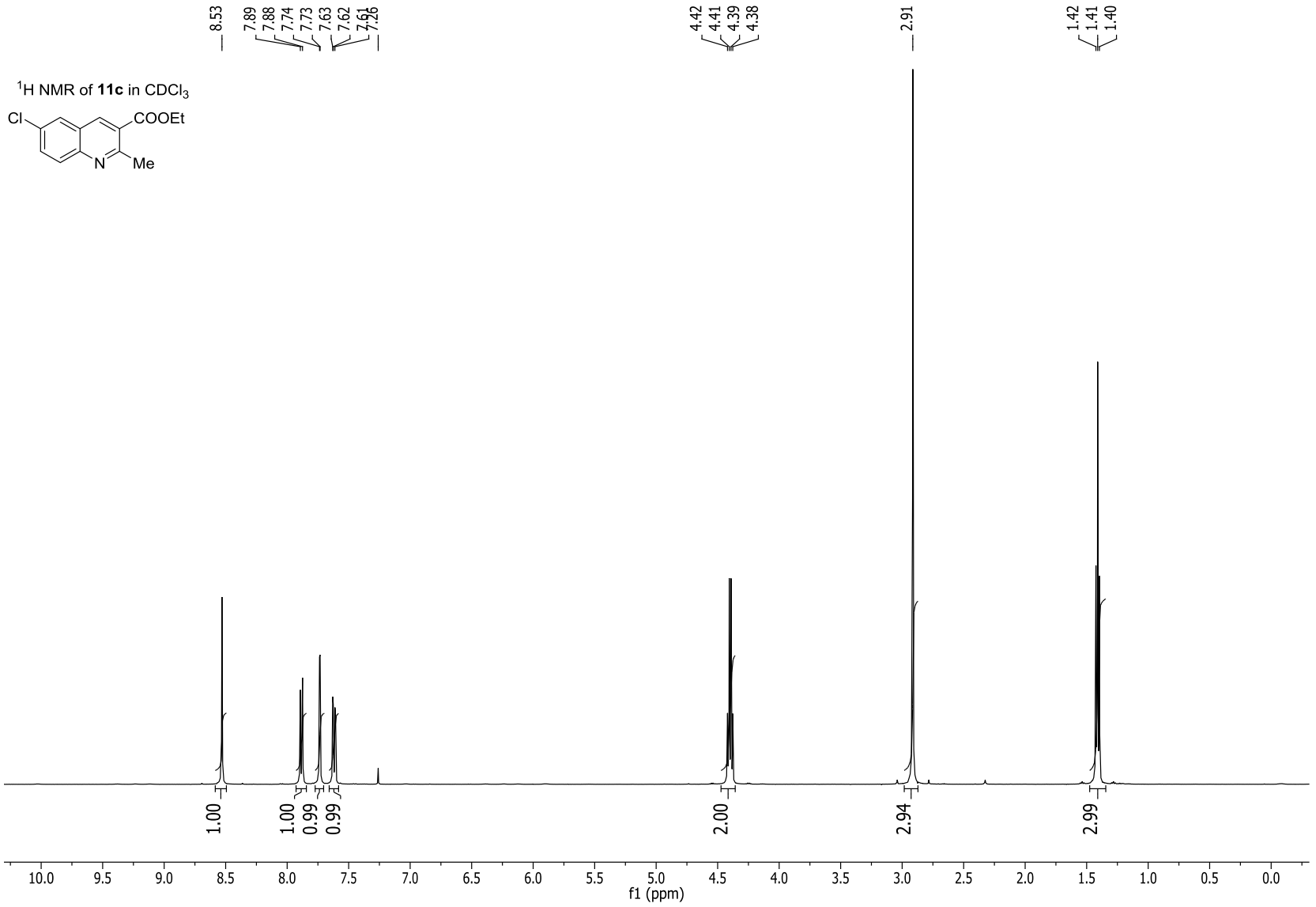
161.37
159.39
157.83
157.81
145.81
139.14
139.10
131.21
131.14
126.43
126.35
124.85
121.95
121.74
111.45
111.28
77.41
77.16
76.91
61.66
25.62
14.43
13C NMR of 11b in CDCl3
F
COOEt
N
Me
90
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)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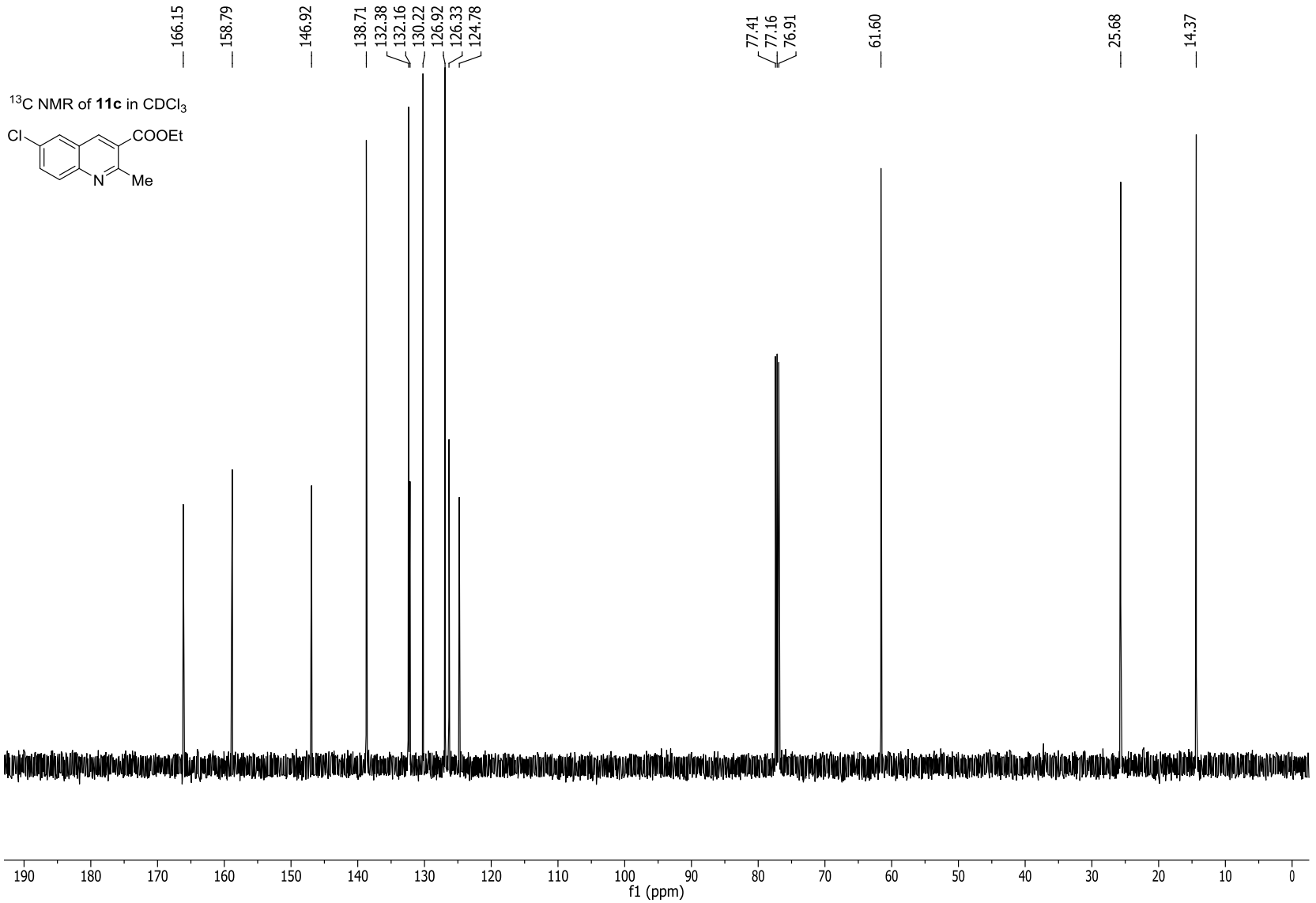
7.89
7.88
7.74
7.73
7.63
7.62
7.61
7.26
4.42
4.41
4.39
4.38
2.91
1.42
1.41
1.40
1H NMR of 11c in CDCl3
Cl
COOEt
N
Me
1.00
1.00
0.99
0.99
2.00
2.94
2.99
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)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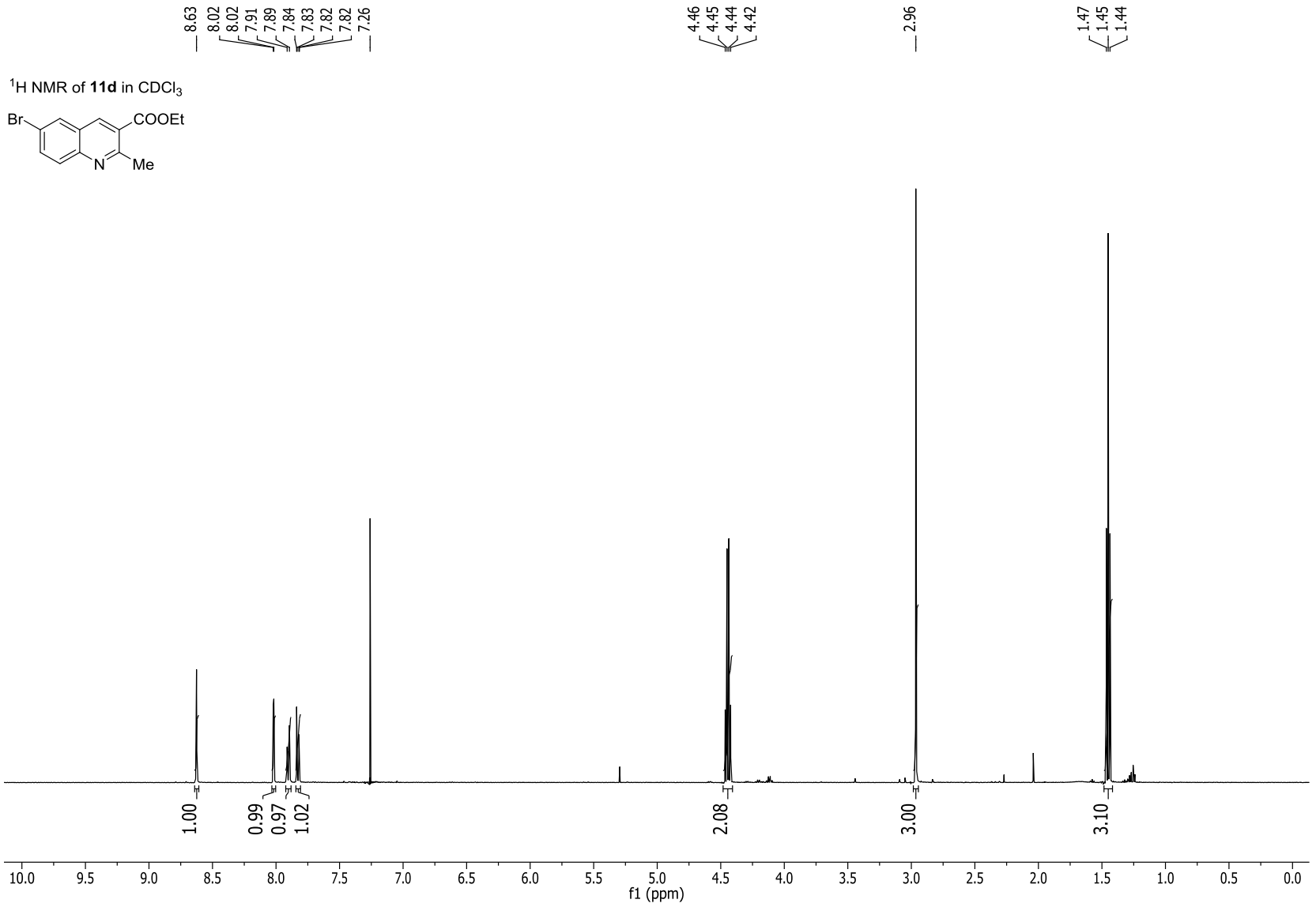
146.92
138.71
132.38
132.16
130.22
126.92
126.33
124.78
77.41
77.16
76.91
61.60
25.68
14.37
13C NMR of 11c in CDCl3
Cl
COOEt
N
Me
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)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8.02
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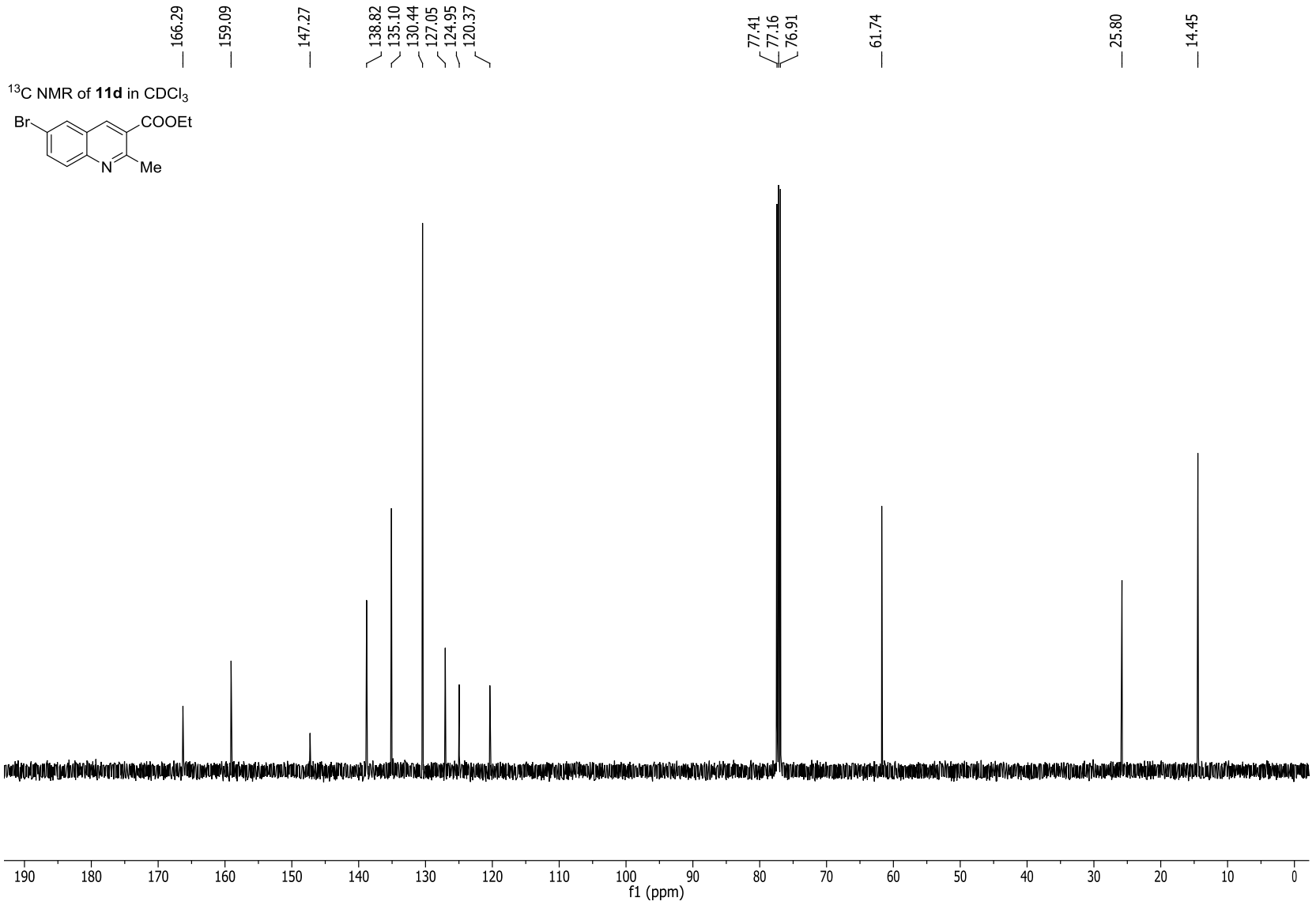
7.89
7.84
7.83
7.82
7.82
7.26
4.46
4.45
4.44
4.42
2.96
1.47
1.45
1.44
1H NMR of 11d in CDCl3
Br
COOEt
N
Me
1.00
0.99
0.97
1.02
2.08
3.00
3.10
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)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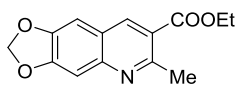
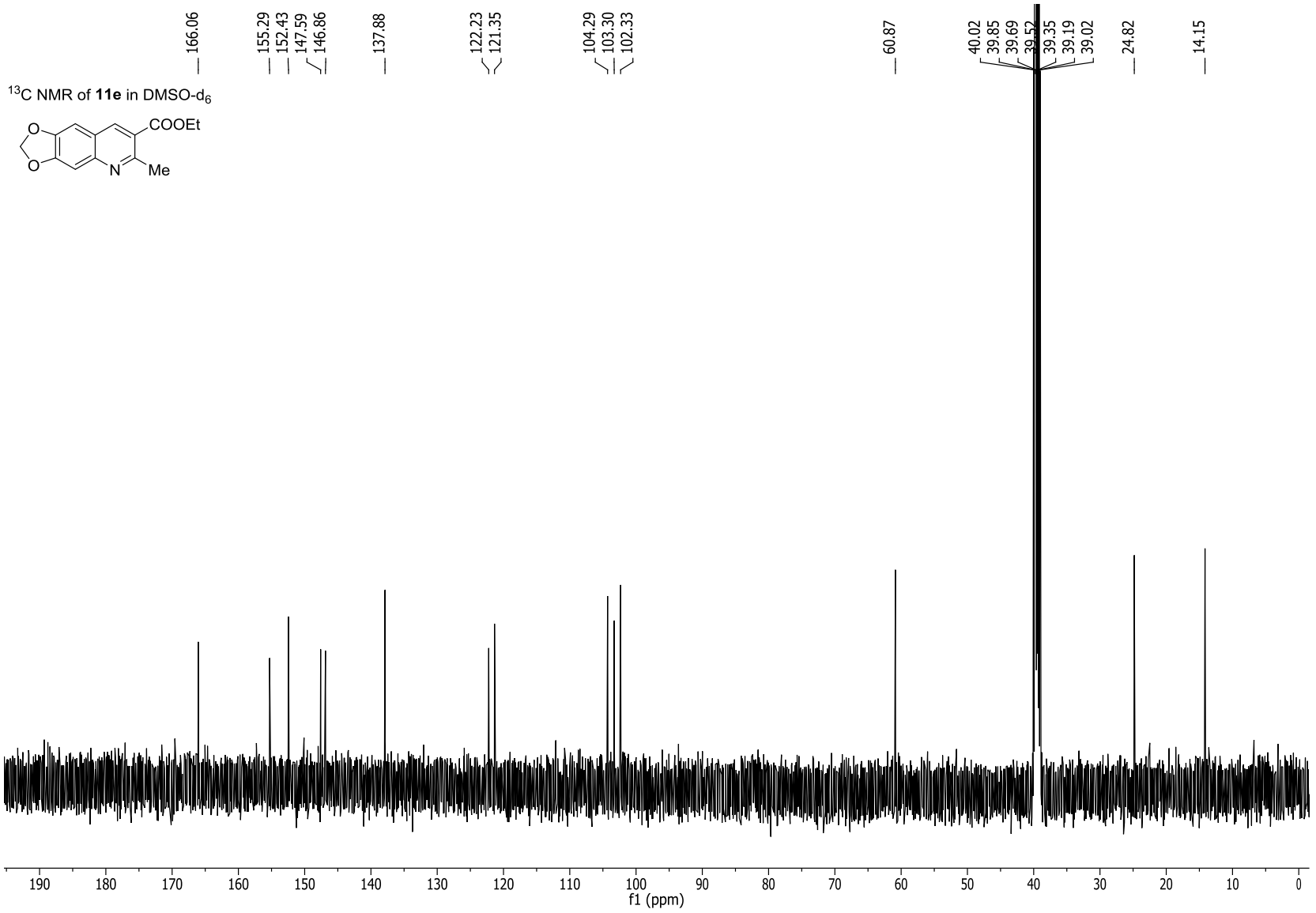
159.09
147.27
138.82
135.10
130.44
127.05
124.95
120.37
77.41
77.16
76.91
61.74
25.80
14.45
^{13}C NMR of **11d** in $CDCl_3$
Br
COOEt
N
Me
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

^{1}H NMR of **11e** in DMSO-d_6

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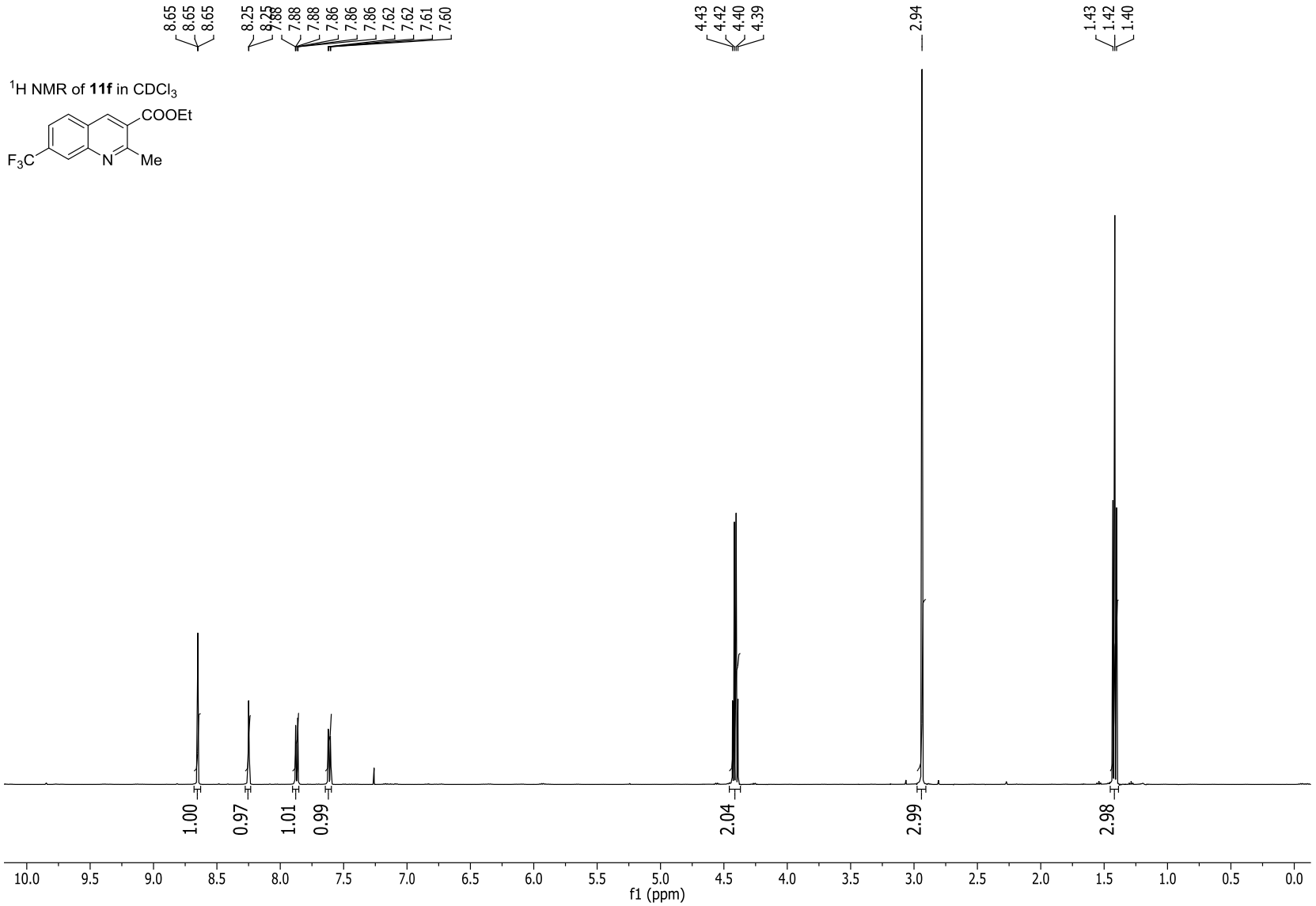
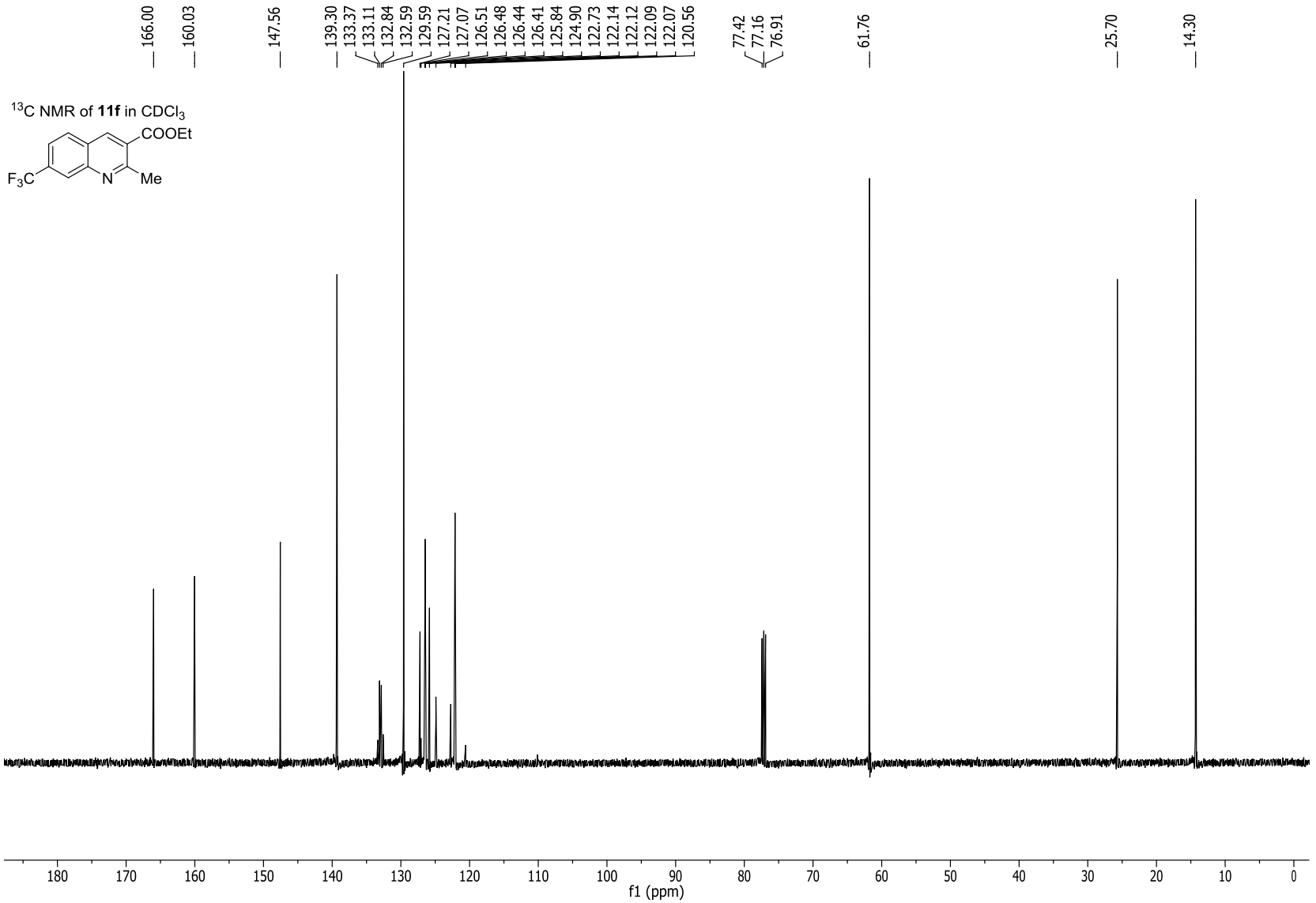
155.29
152.43
147.59
146.86
137.88
122.23
121.35
104.29
103.30
102.33
60.87
40.02
39.85
39.69
39.52
39.35
39.19
39.02
24.82
14.15
13C NMR of 11e in DMSO-d6
COOEt
O
O
N
Me
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


8.65
8.65
8.65
8.25
7.88
7.88
7.88
7.86
7.86
7.86
7.62
7.61
7.60
4.43
4.42
4.40
4.39
2.94
1.43
1.42
1.40
1H NMR of 11f in CDCl3
COOEt
F3C
N
Me
1.00
0.97
1.01
0.99
2.04
2.99
2.98
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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133.37
133.11
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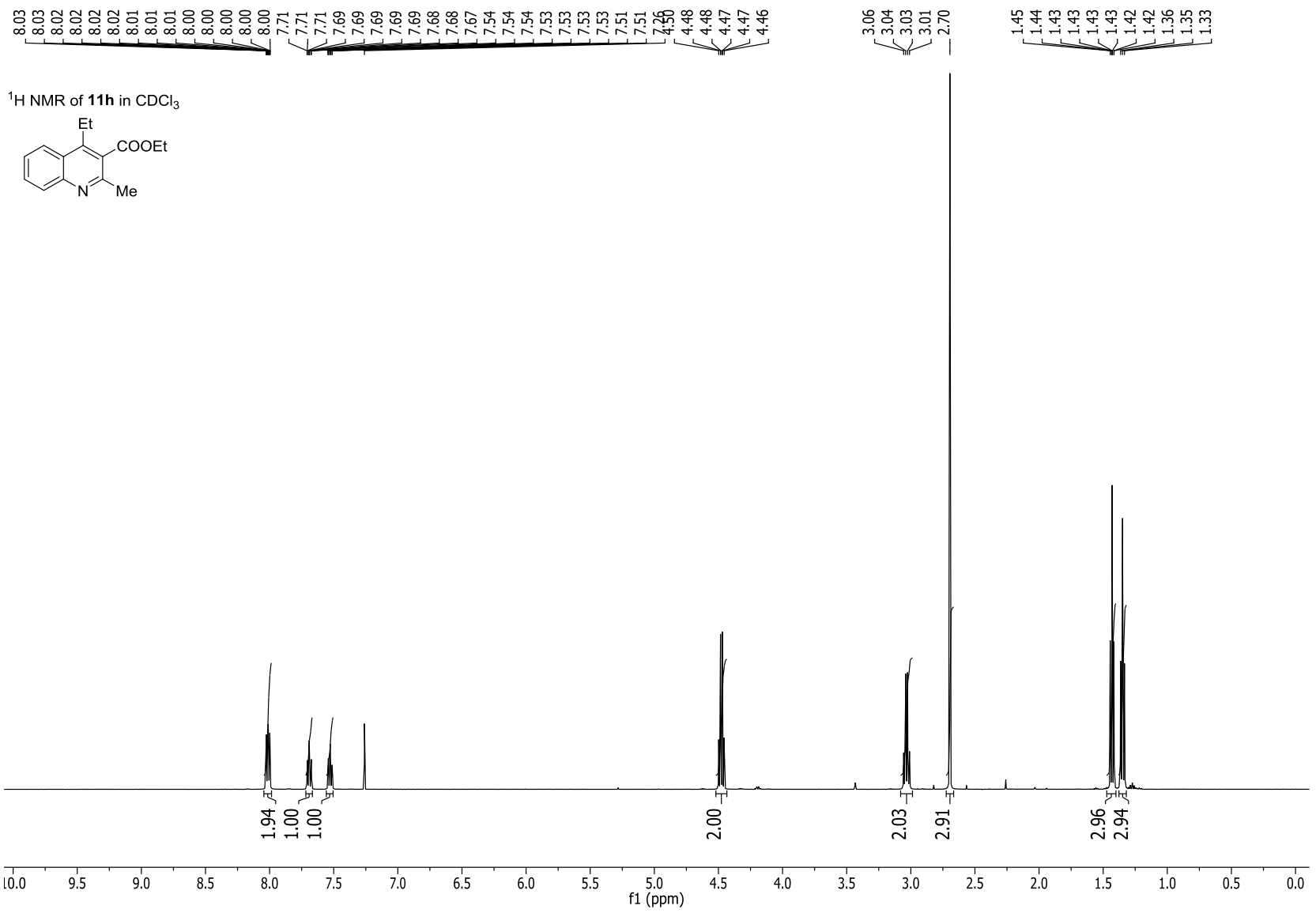
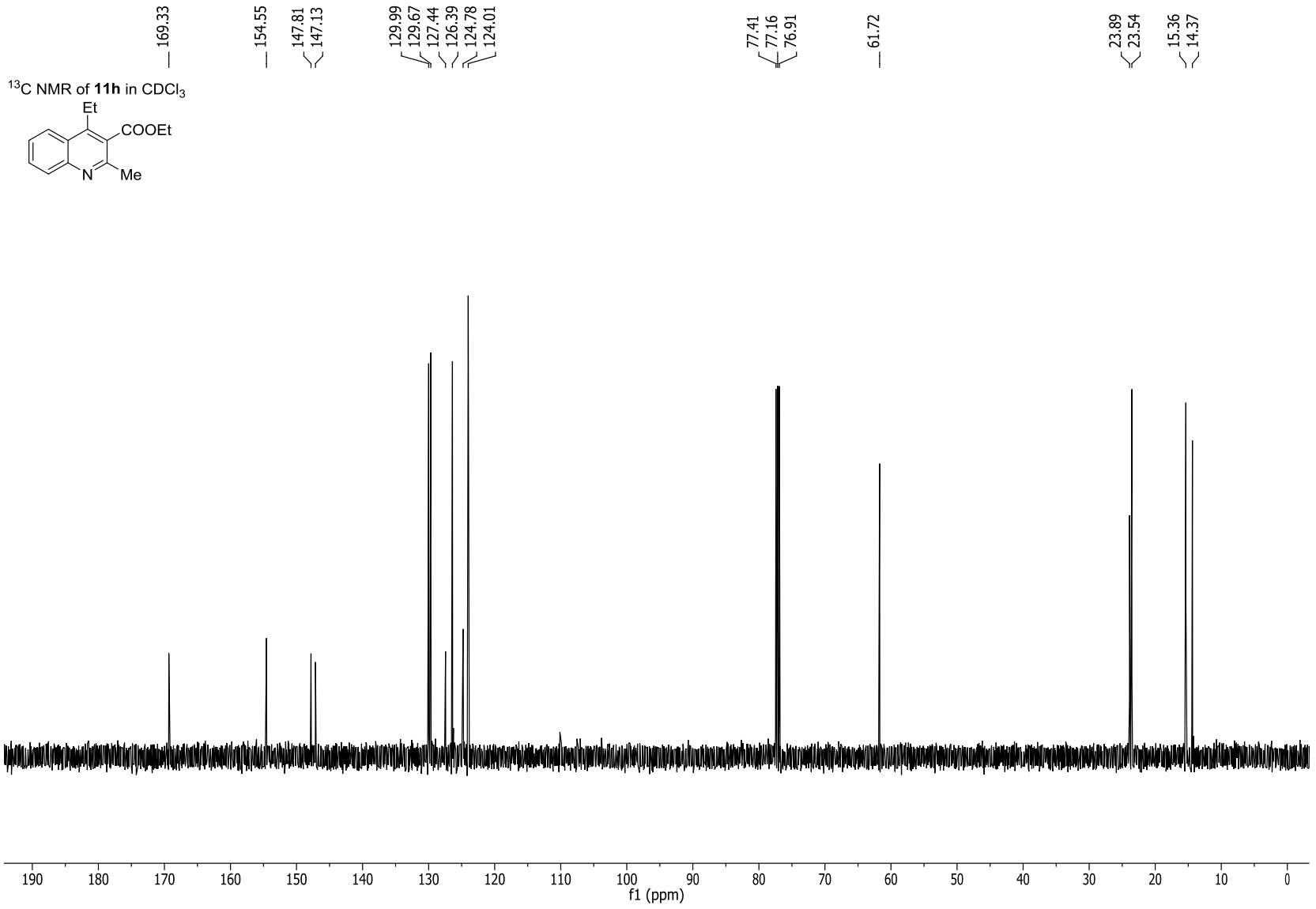
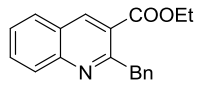
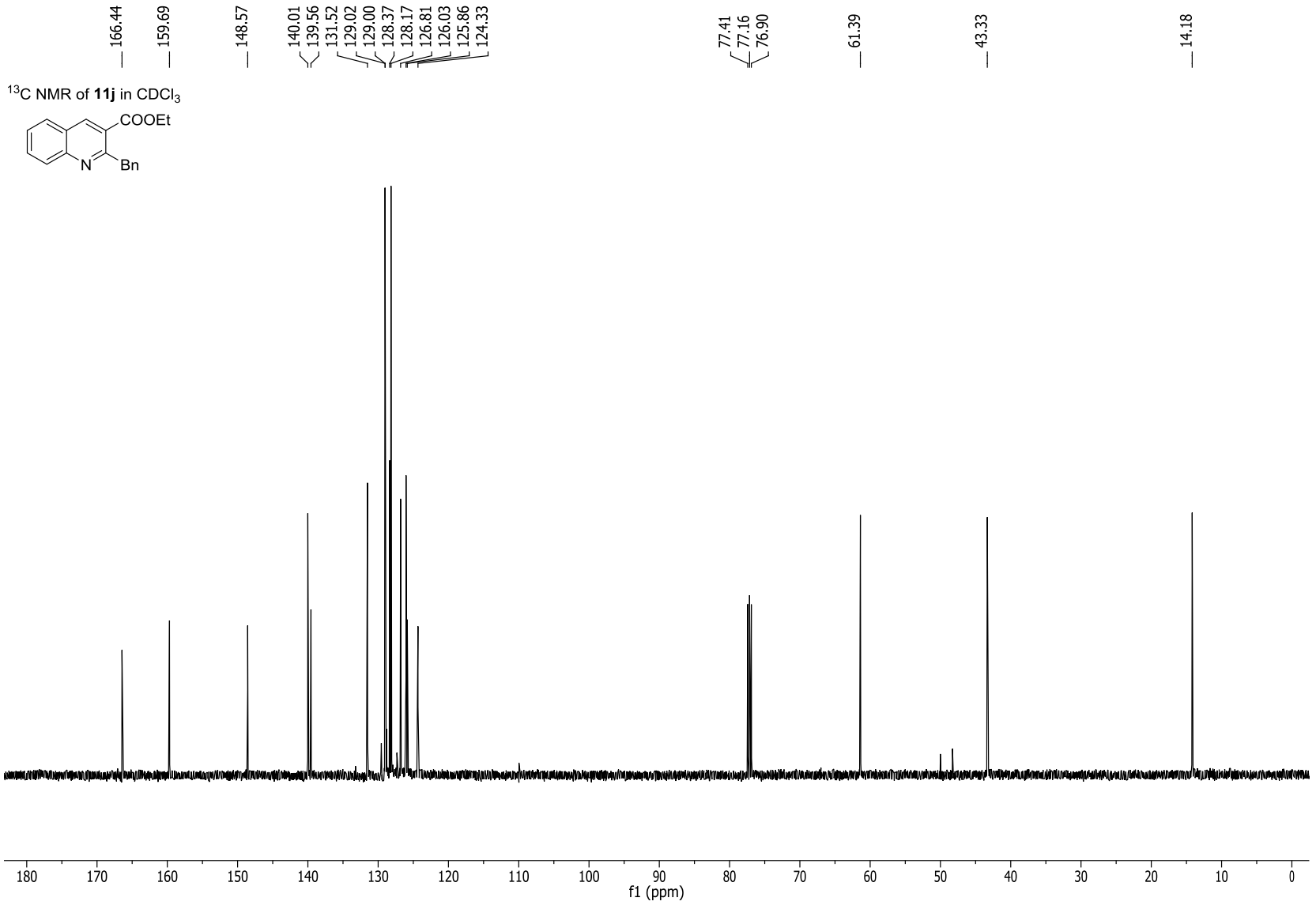
132.59
129.59
127.21
127.07
126.51
126.48
126.44
126.41
125.84
124.90
122.73
122.14
122.12
122.09
122.07
120.56
77.42
77.16
76.91
61.76
25.70
14.30
13C NMR of 11f in CDCl3
COOEt
F3C
N
Me
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


8.03 8.03 8.02 8.02 8.02 8.02 8.01 8.01 8.01 8.00 8.00 8.00 8.00 8.00 7.71 7.71 7.71 7.69 7.69 7.69 7.69 7.69 7.68 7.68 7.67 7.54 7.54 7.54 7.53 7.53 7.53 7.51 7.51 7.26 4.50 4.48 4.48 4.47 4.47 4.46 3.06 3.04 3.03 3.01 2.70 1.45 1.44 1.43 1.43 1.43 1.43 1.42 1.42 1.36 1.35 1.33
1H NMR of 11h in CDCl3
Et
COOEt
N
Me
1.94 1.00 1.00 2.00 2.03 2.91 2.96 2.94
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)


169.33
154.55
147.81
147.13
129.99
129.67
127.44
126.39
124.78
124.01
77.41
77.16
76.91
61.72
23.89
23.54
15.36
14.37
13C NMR of 11h in CDCl3
Et
COOEt
N
Me
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 **11j** in $CDCl_3$

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148.57
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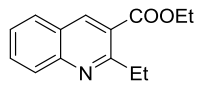
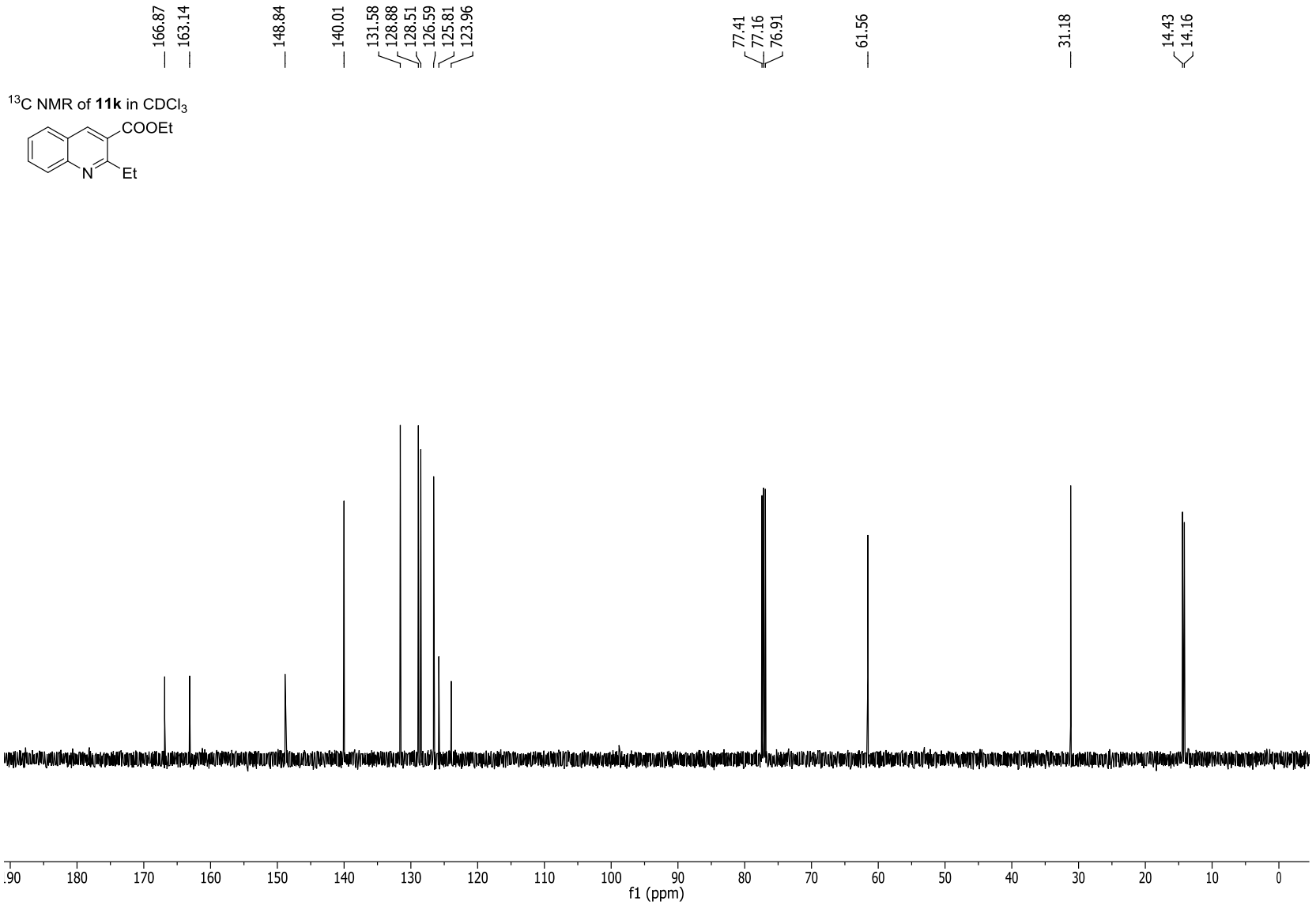
139.56
131.52
129.02
129.00
128.37
128.17
126.81
126.03
125.86
124.33
77.41
77.16
76.90
61.39
43.33
14.18
13C NMR of 11j in CDCl3
COOEt
N
Bn
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

^{1}H NMR of **11k** in $CDCl_3$

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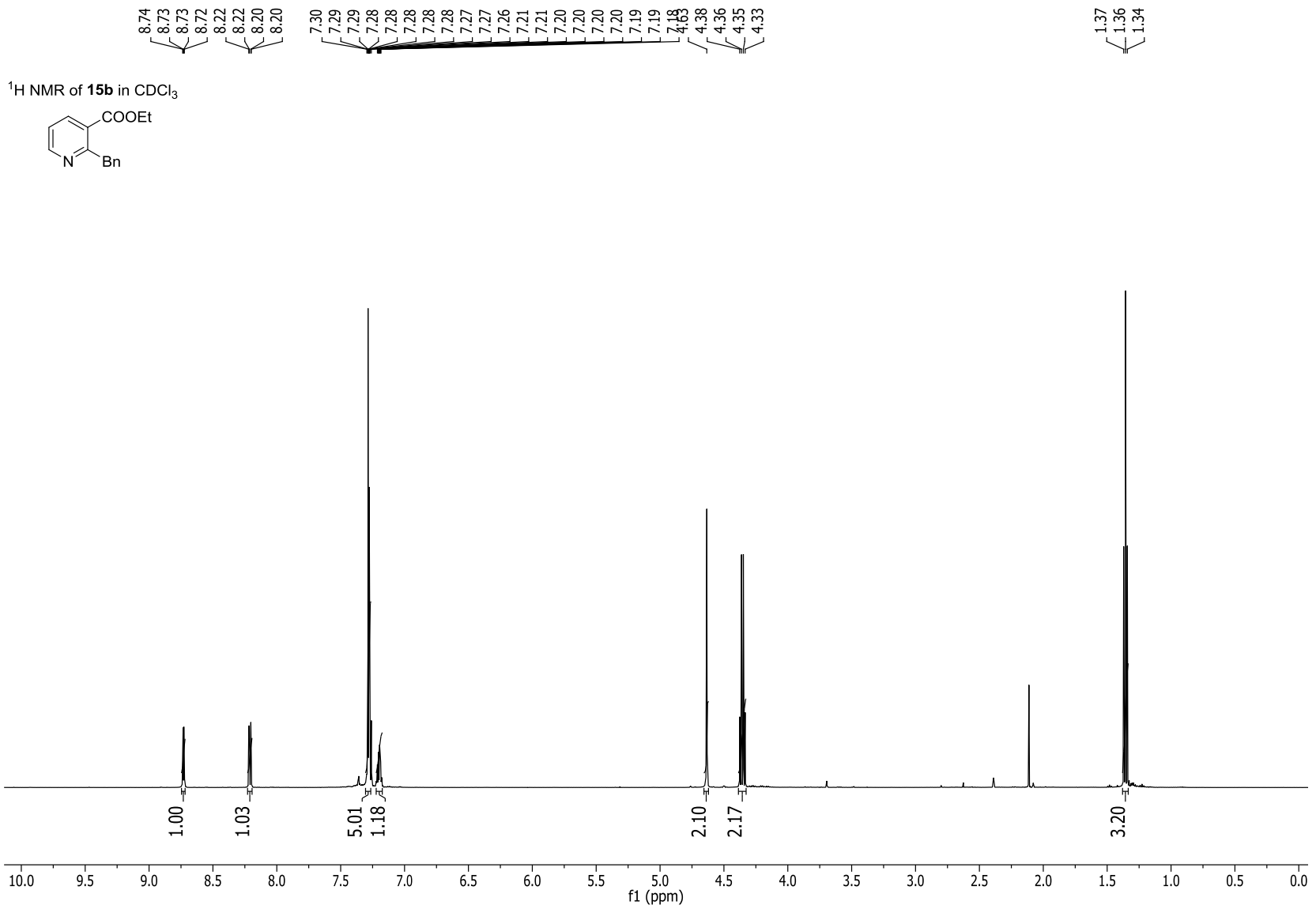
126.59
125.81
123.96
77.41
77.16
76.91
61.56
31.18
14.43
14.16
^{13}C NMR of **11k** in $CDCl_3$
COOEt
N
Et
f1 (ppm)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8.20
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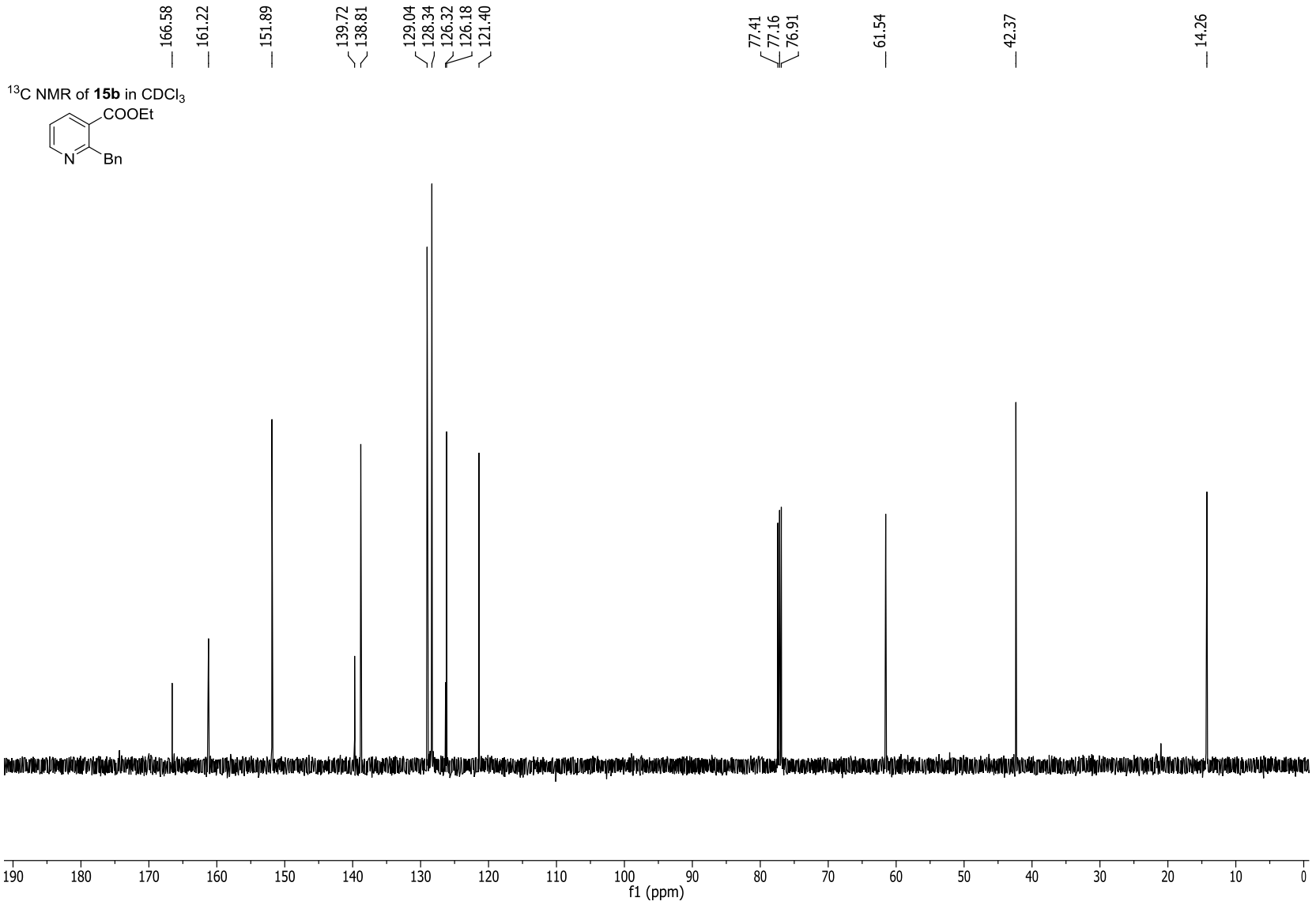
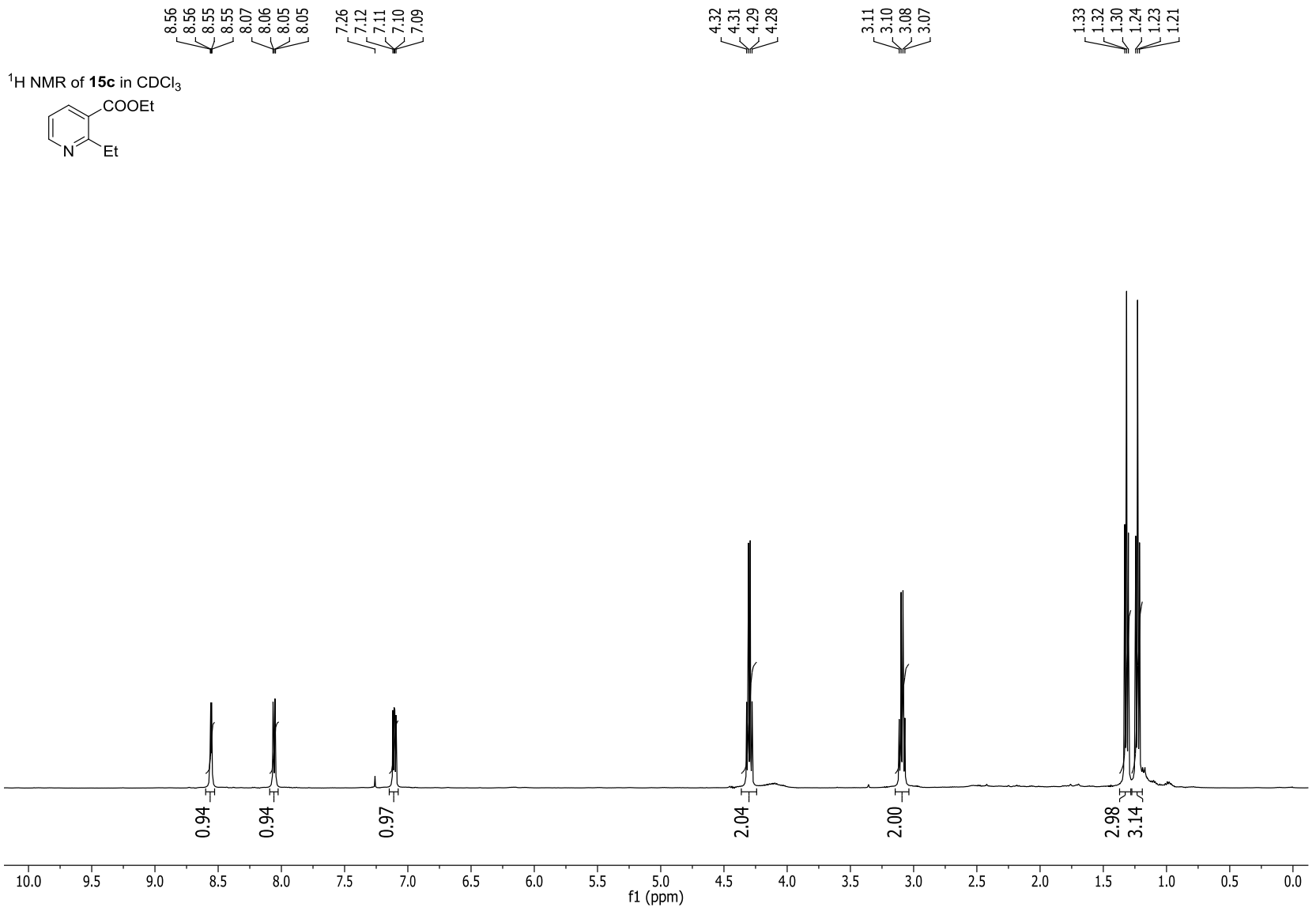
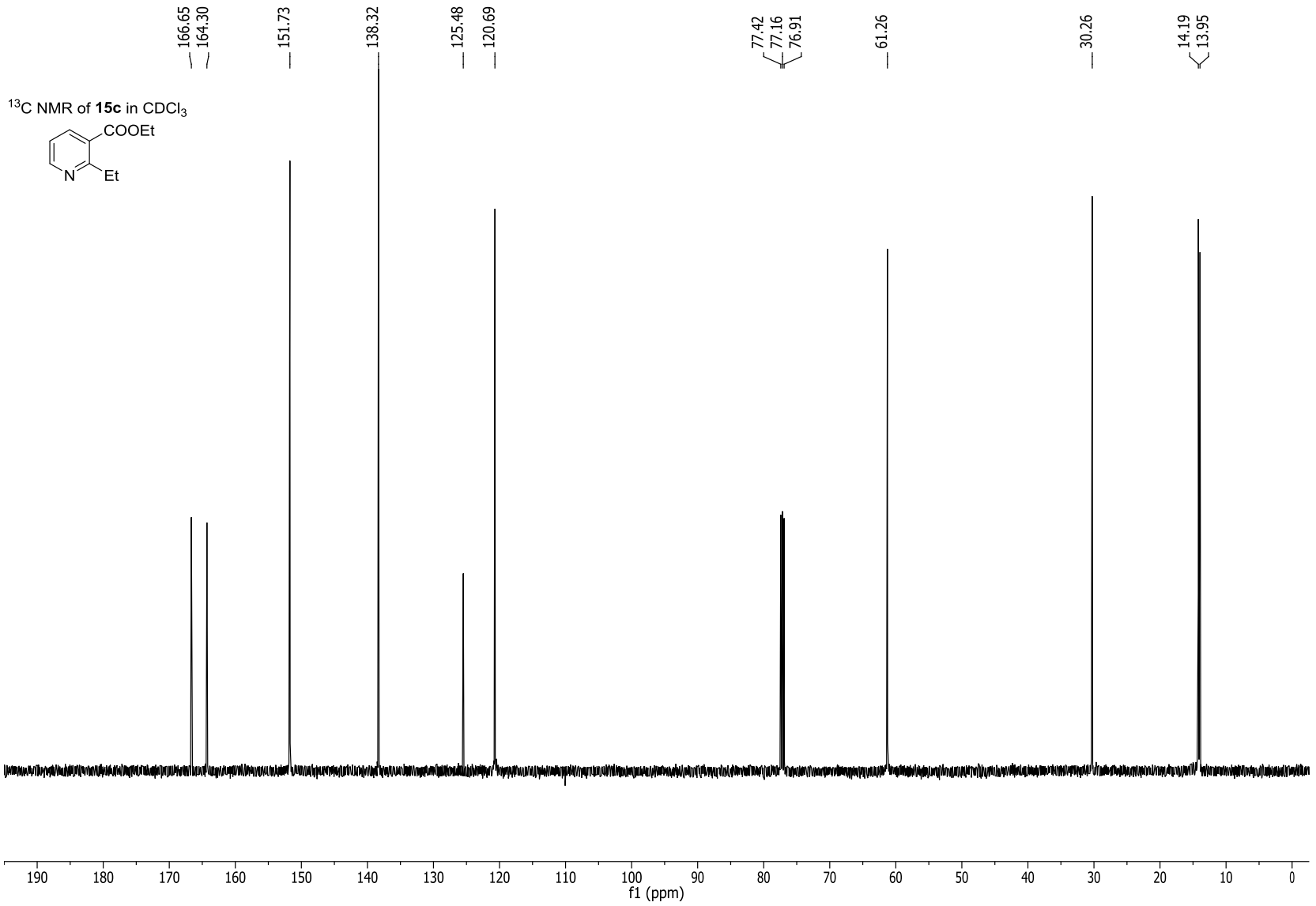
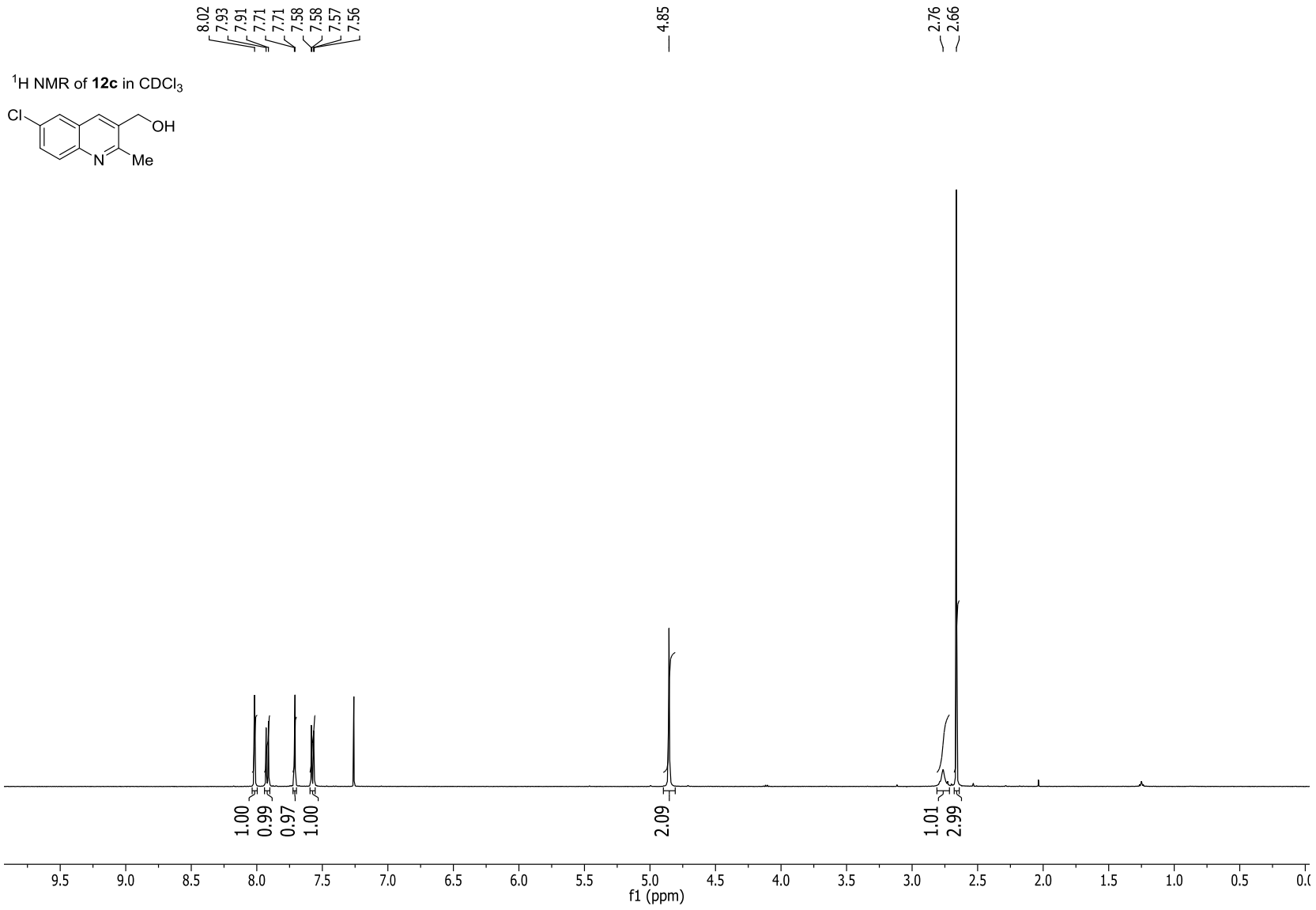
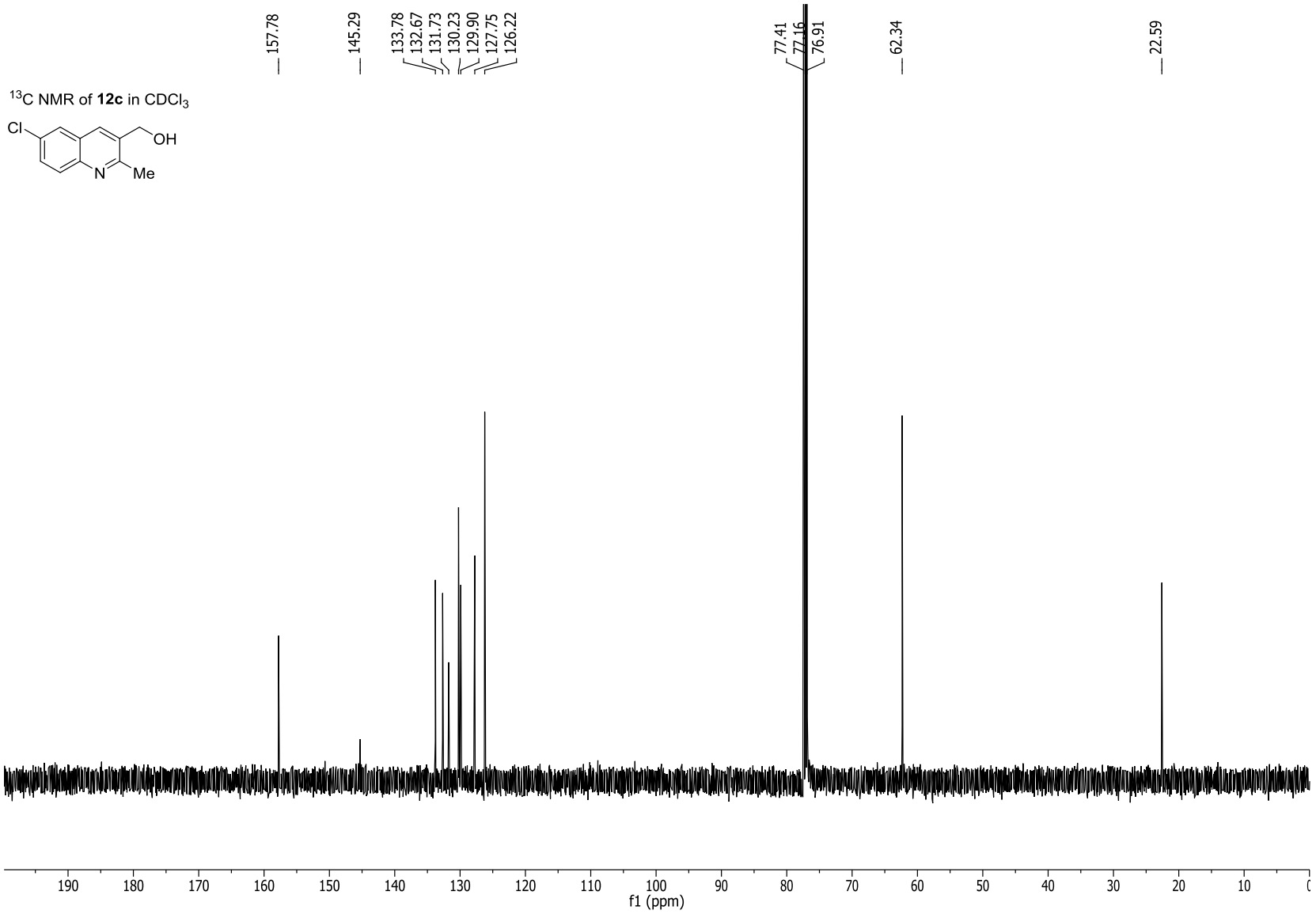
7.29
7.29
7.28
7.28
7.28
7.28
7.28
7.27
7.27
7.26
7.21
7.21
7.20
7.20
7.20
7.20
7.19
7.19
7.18
4.63
4.38
4.36
4.35
4.33
1.37
1.36
1.34
^{1}H NMR of **15b** in $CDCl_3$
COOEt
N
Bn
1.00
1.03
5.01
1.18
2.10
2.17
3.20
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

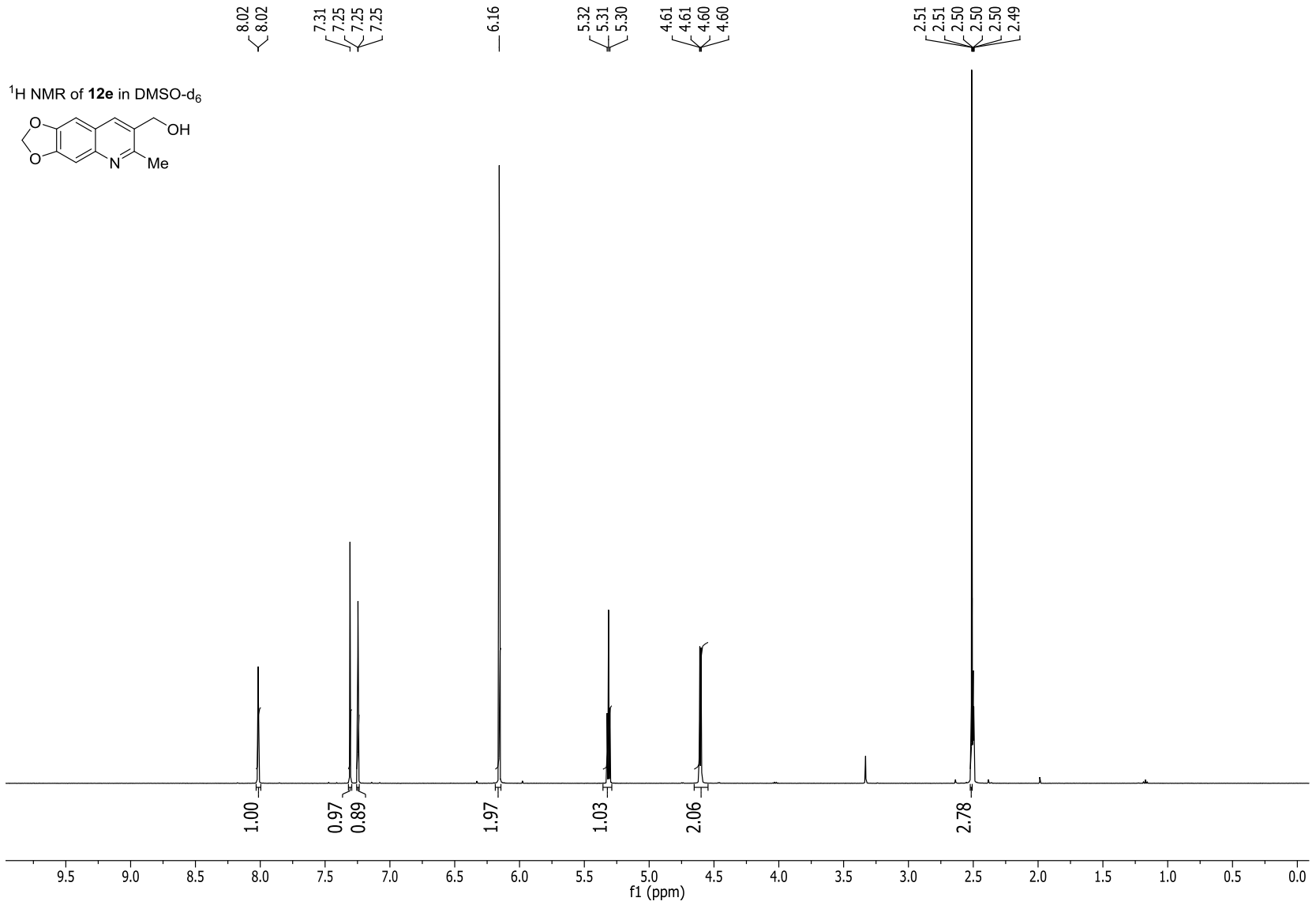

166.58
161.22
151.89
139.72
138.81
129.04
128.34
126.32
126.18
121.40
77.41
77.16
76.91
61.54
42.37
14.26
^{13}C NMR of **15b** in $CDCl_3$
COOEt
N
Bn
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

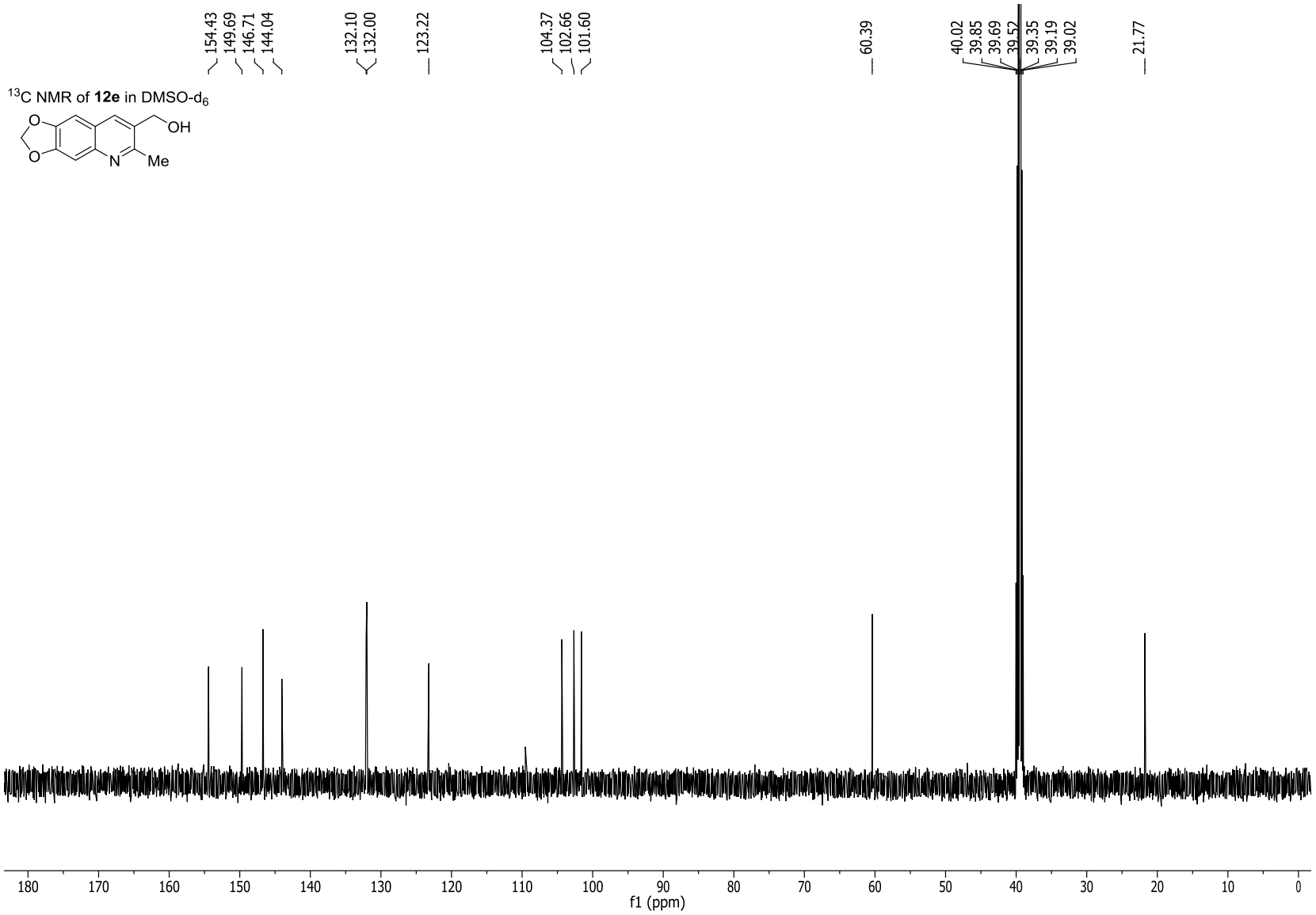

8.56
8.56
8.55
8.55
8.07
8.06
8.05
8.05
7.26
7.12
7.11
7.10
7.09
4.32
4.31
4.29
4.28
3.11
3.10
3.08
3.07
1.33
1.32
1.30
1.24
1.23
1.21
1H NMR of 15c in CDCl3
COOEt
N
Et
0.94
0.94
0.97
2.04
2.00
2.98
3.14
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


166.65
164.30
151.73
138.32
125.48
120.69
77.42
77.16
76.91
61.26
30.26
14.19
13.95
^{13}C NMR of **15c** in $CDCl_3$
COOEt
N
Et
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

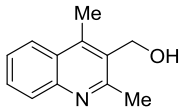

8.02
7.93
7.91
7.71
7.71
7.58
7.58
7.57
7.56
4.85
2.76
2.66
^{1}H NMR of **12c** in $CDCl_3$
Cl
OH
N
Me
1.00
0.99
0.97
1.00
2.09
1.01
2.99
f1 (ppm)

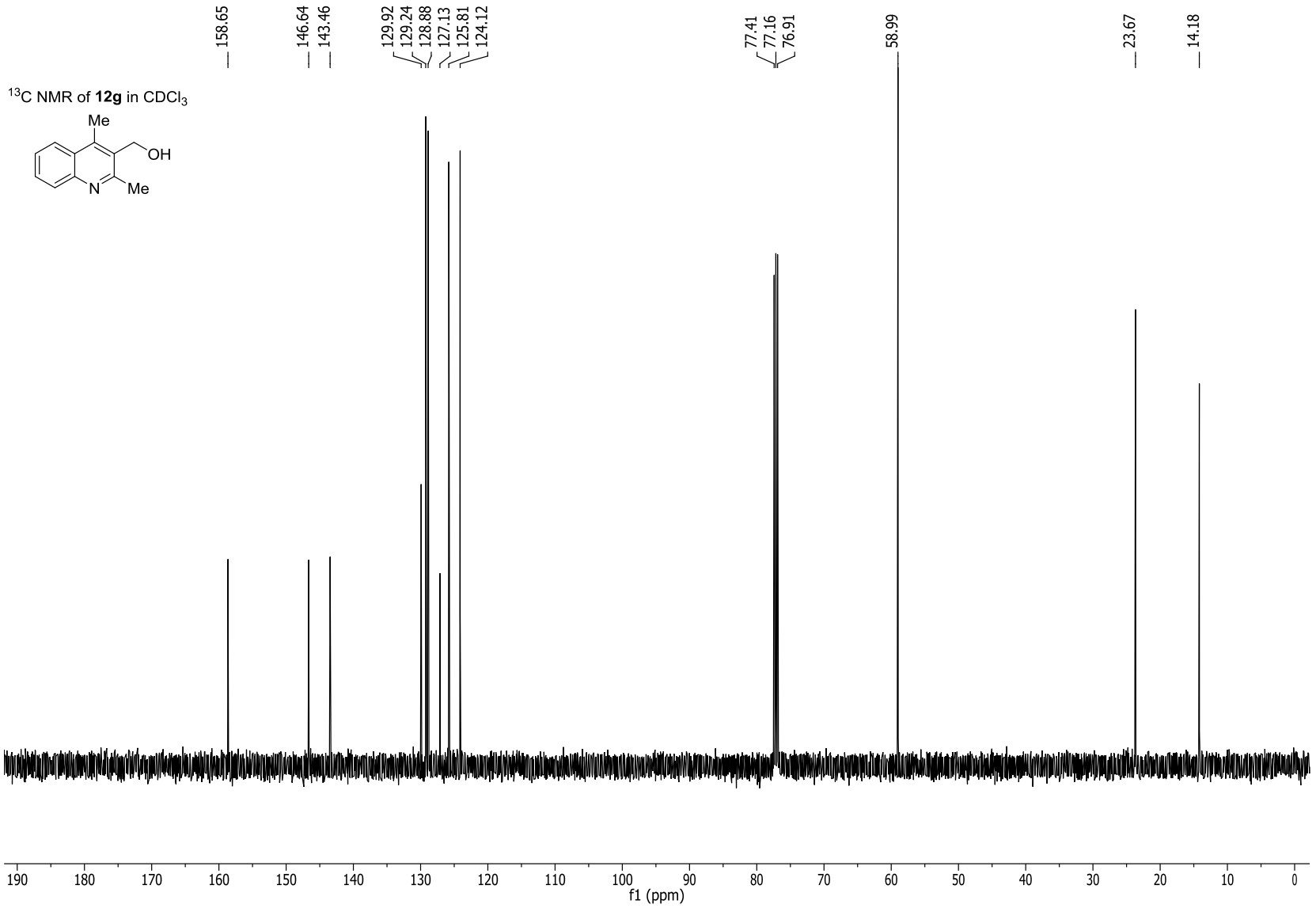

157.78
145.29
133.78
132.67
131.73
130.23
129.90
127.75
126.22
77.41
77.16
76.91
62.34
22.59
^{13}C NMR of **12c** in $CDCl_3$
Cl
OH
N
Me
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
f1 (ppm)

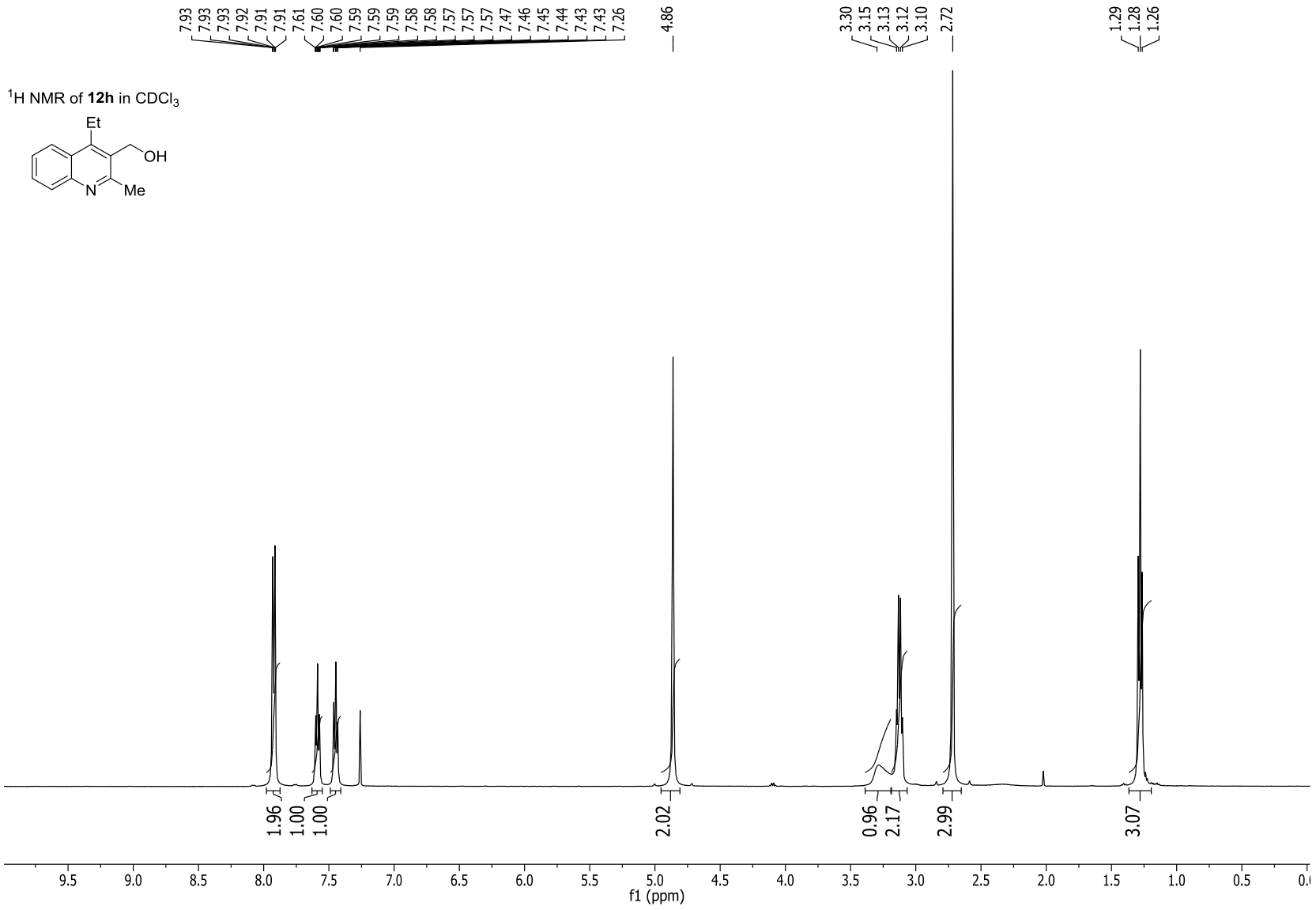

8.02
8.02
7.31
7.25
7.25
7.25
6.16
5.32
5.31
5.30
4.61
4.61
4.60
4.60
2.51
2.51
2.50
2.50
2.50
2.49
1H NMR of 12e in DMSO-d6
O
O
N
OH
Me
1.00
0.97
0.89
1.97
1.03
2.06
2.78
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

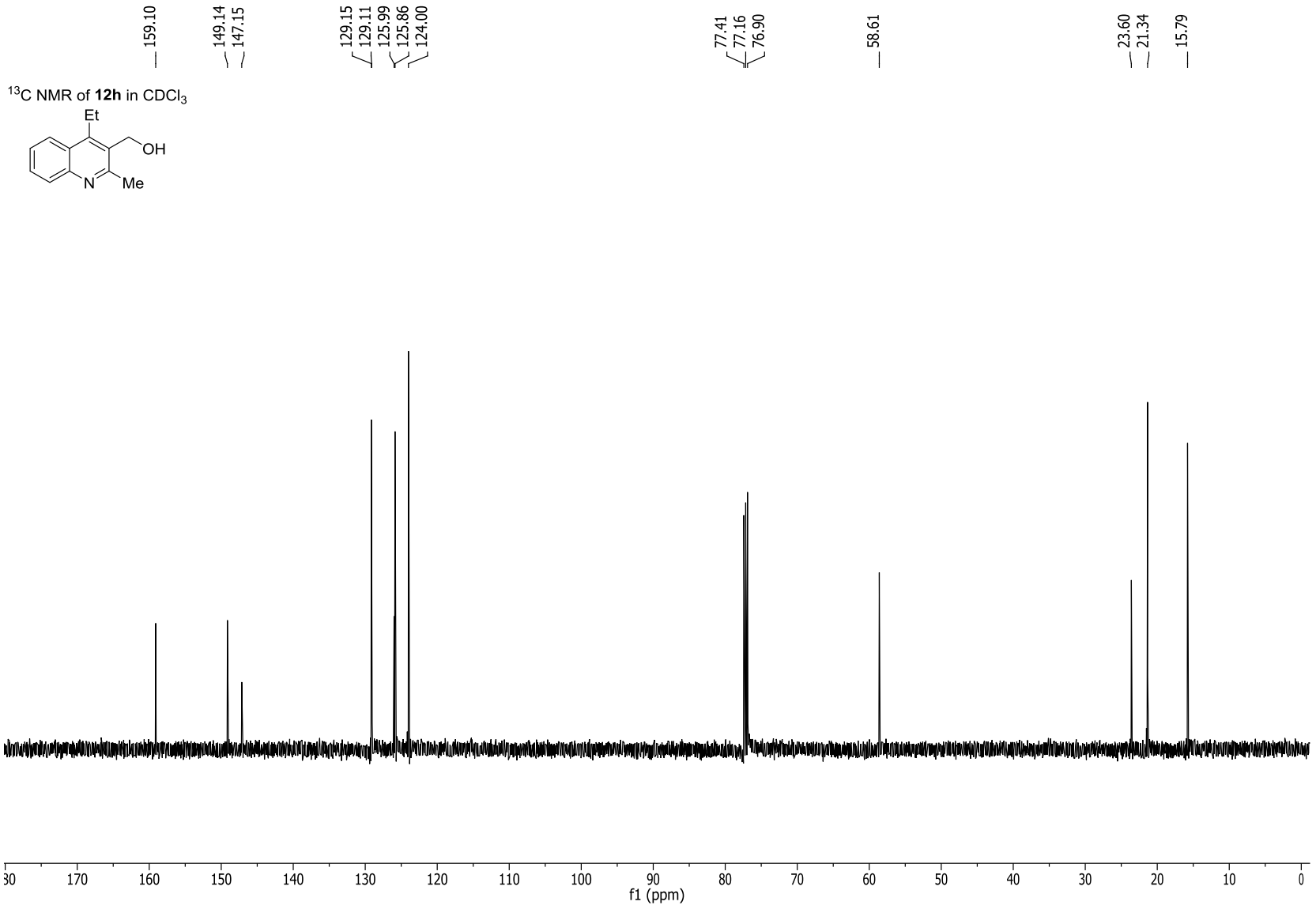

154.43
149.69
146.71
144.04
132.10
132.00
123.22
104.37
102.66
101.60
60.39
40.02
39.85
39.69
39.52
39.35
39.19
39.02
21.77
13C NMR of 12e in DMSO-d6
O
O
N
OH
Me
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

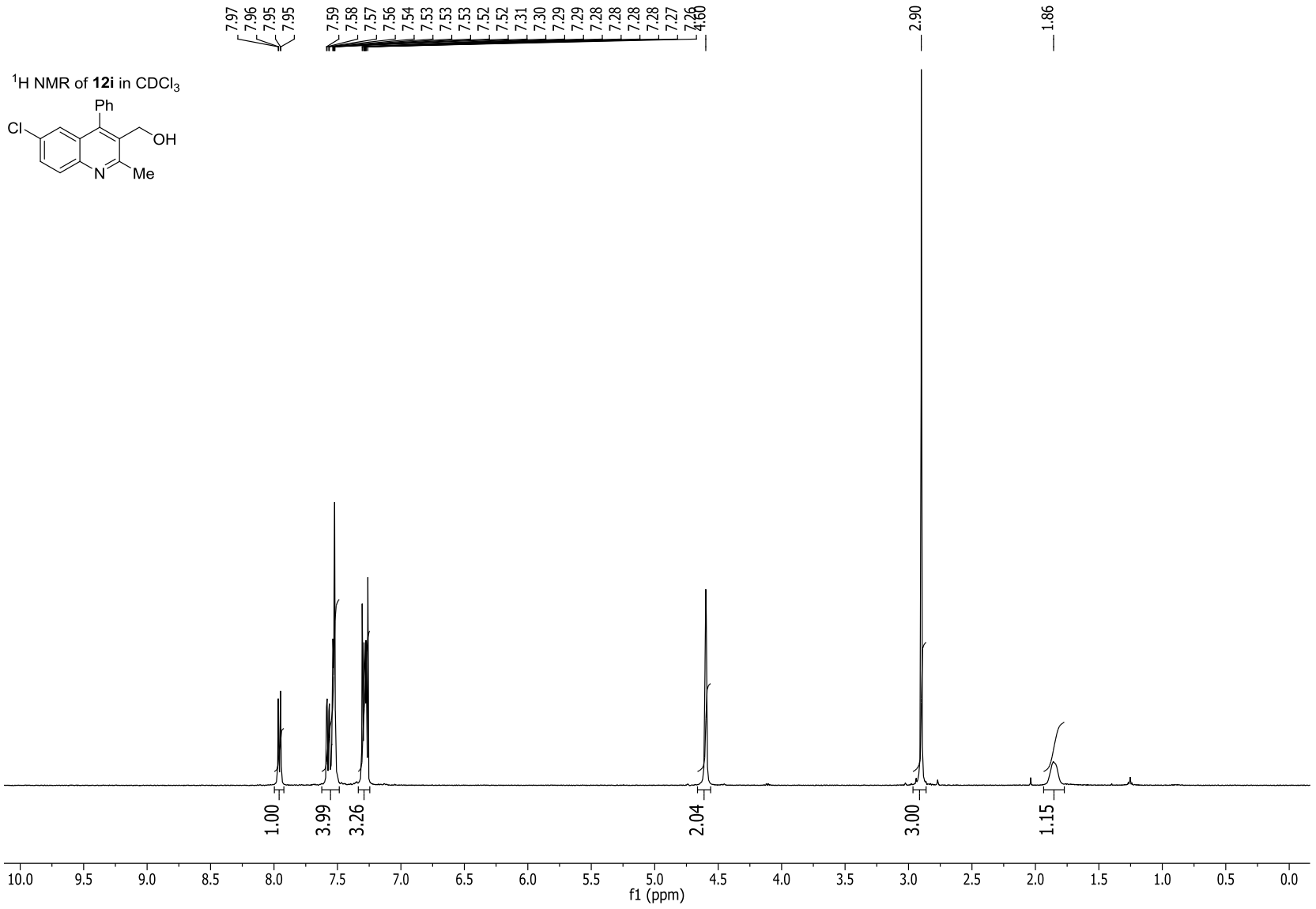
^{1}H NMR of **12g** in $CDCl_3$

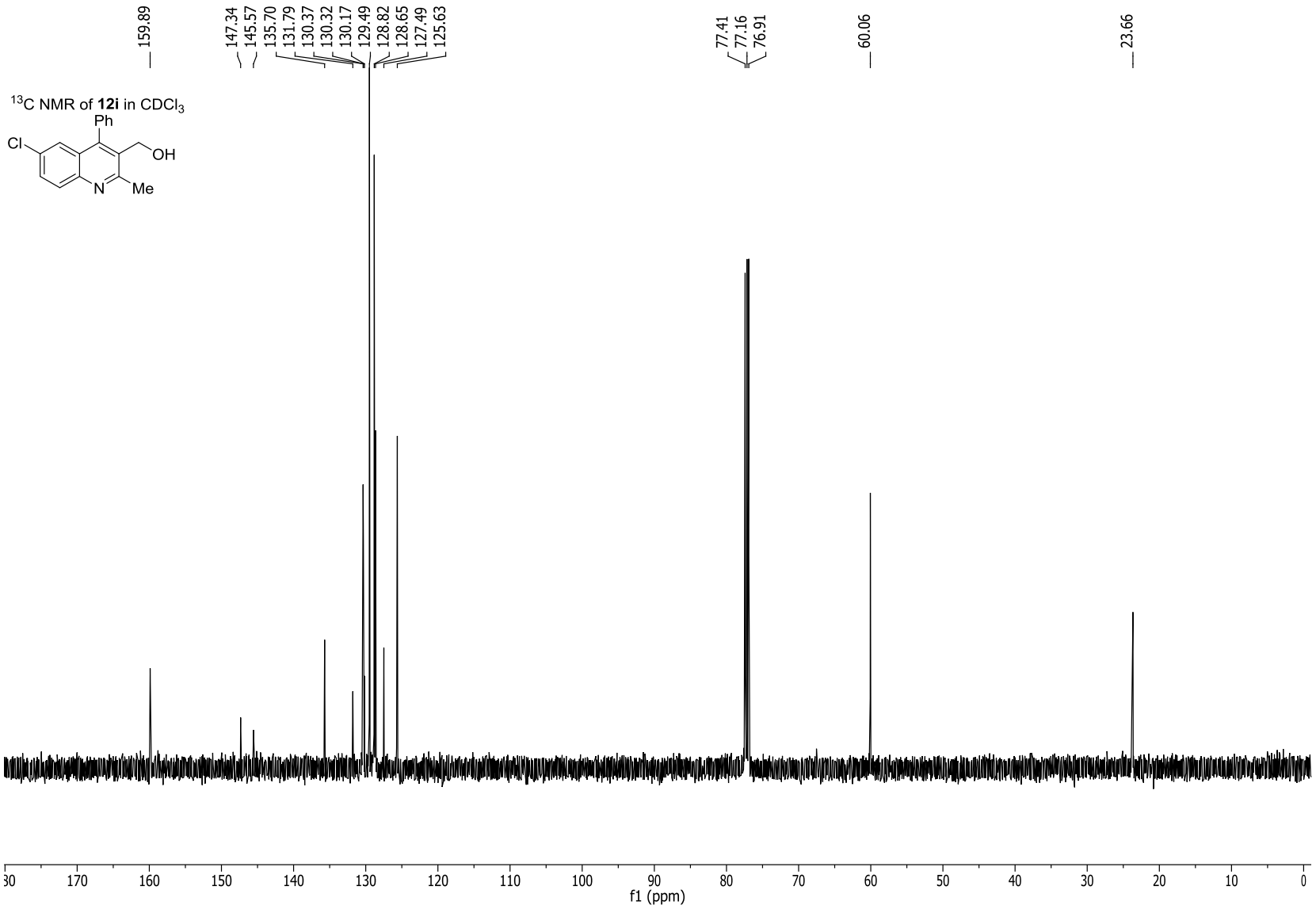


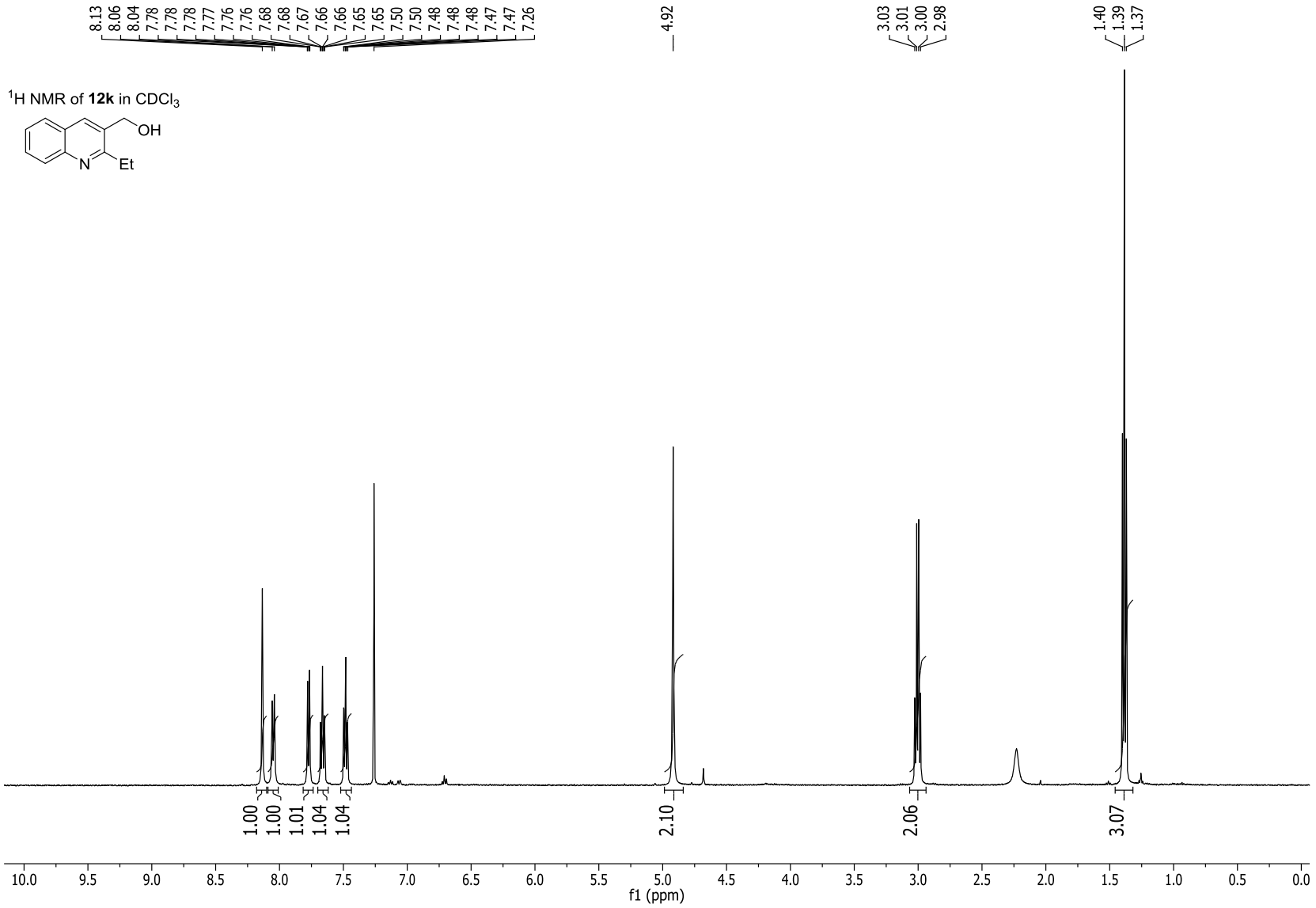

158.65
146.64
143.46
129.92
129.24
128.88
127.13
125.81
124.12
77.41
77.16
76.91
58.99
23.67
14.18
13C NMR of 12g in CDCl3
Me
OH
N
Me
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

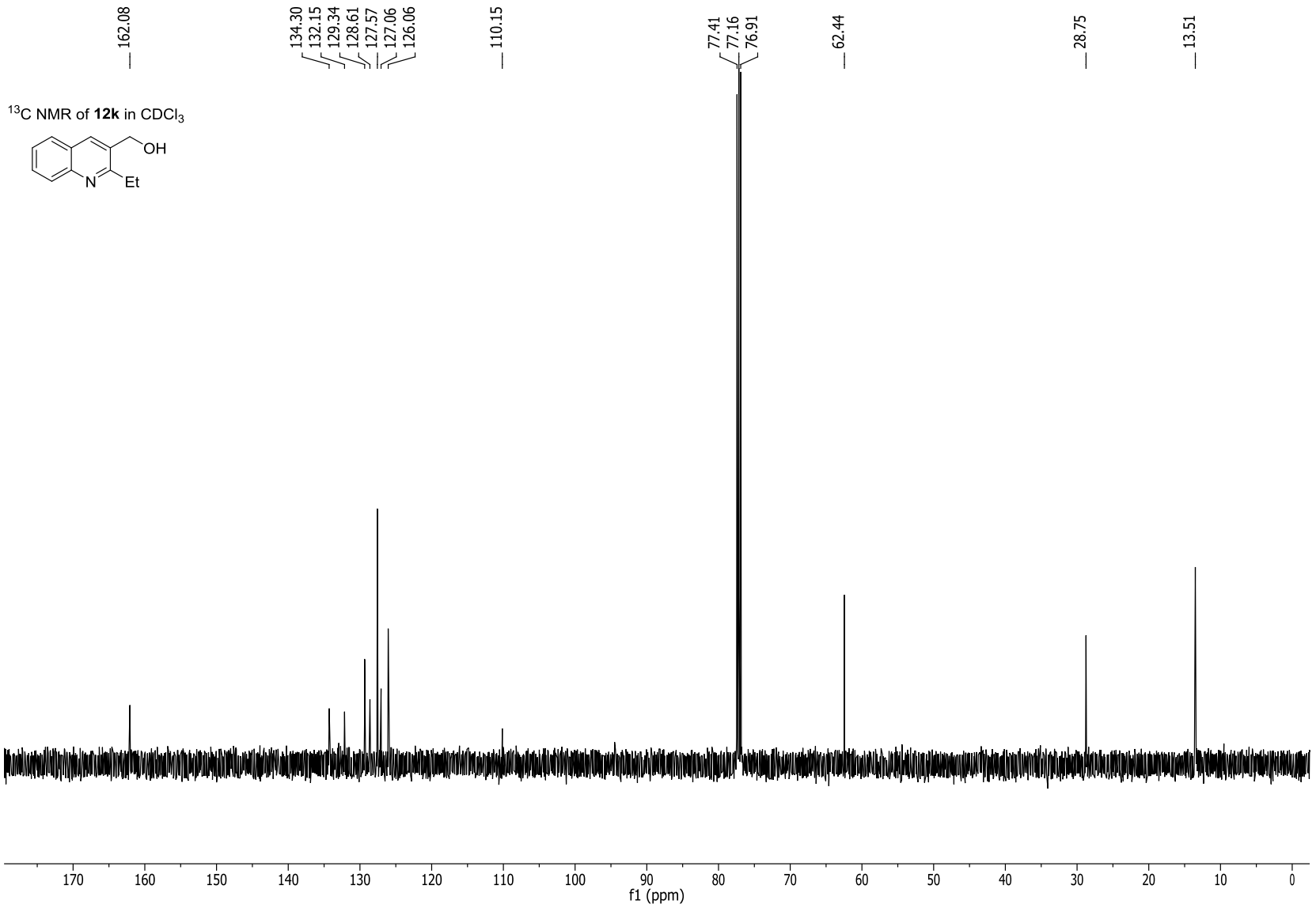

7.93
7.93
7.93
7.92
7.91
7.91
7.61
7.60
7.60
7.59
7.59
7.59
7.58
7.58
7.57
7.57
7.57
7.47
7.46
7.45
7.44
7.43
7.43
7.26
4.86
3.30
3.15
3.13
3.12
3.10
2.72
1.29
1.28
1.26
1H NMR of 12h in CDCl3
Et
OH
N
Me
1.96
1.00
1.00
2.02
0.96
2.17
2.99
3.07
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
f1 (ppm)

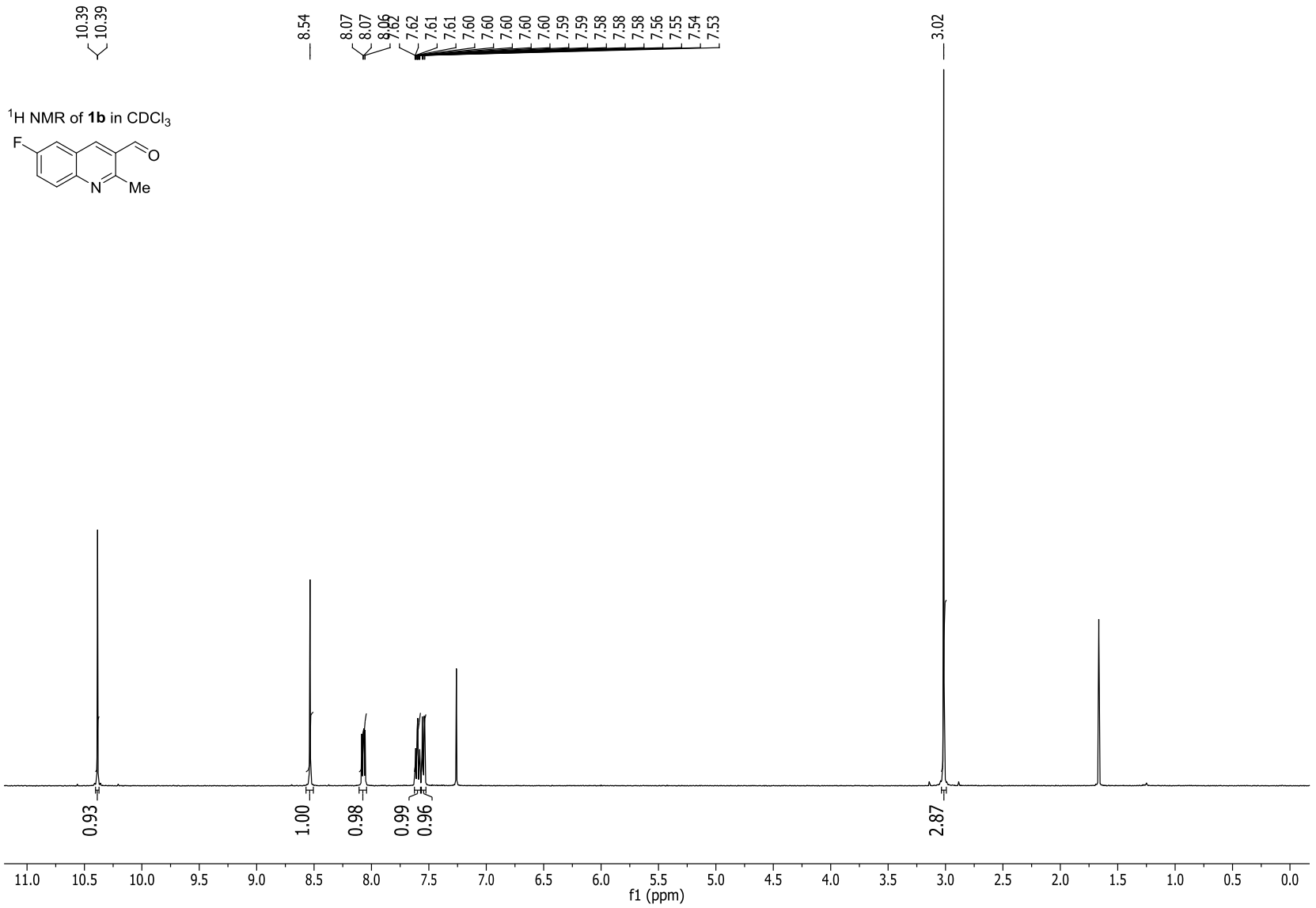

159.10
149.14
147.15
129.15
129.11
125.99
125.86
124.00
77.41
77.16
76.90
58.61
23.60
21.34
15.79
^{13}C NMR of **12h** in $CDCl_3$
Et
OH
N
Me
f1 (ppm)

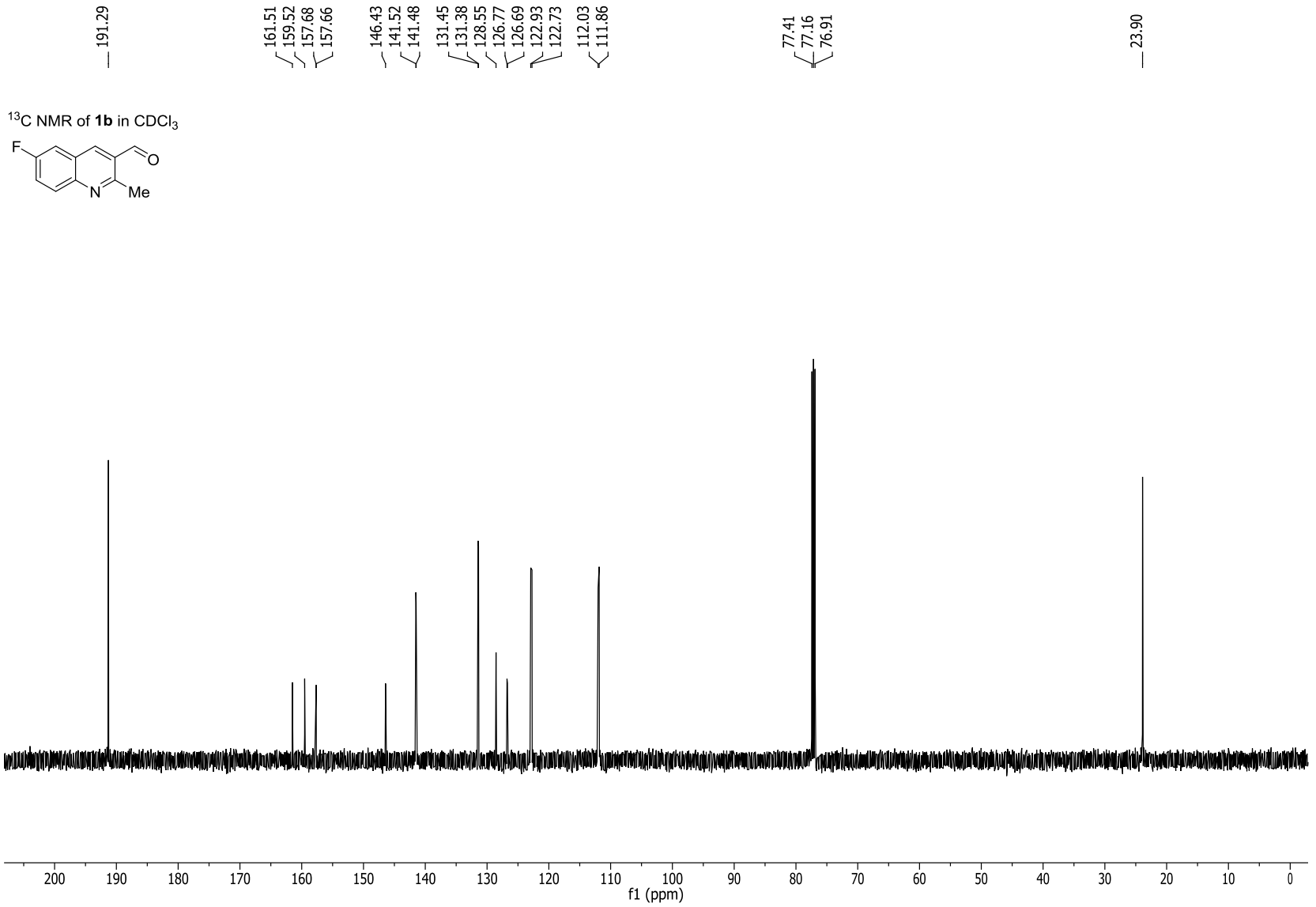

^{1}H NMR of **12i** in $CDCl_3$
Ph
Cl
OH
N
Me
7.97
7.96
7.95
7.95
7.59
7.58
7.57
7.56
7.54
7.53
7.53
7.53
7.52
7.52
7.31
7.30
7.29
7.29
7.28
7.28
7.28
7.28
7.27
7.26
4.60
2.90
1.86
1.00
3.99
3.26
2.04
3.00
1.15
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

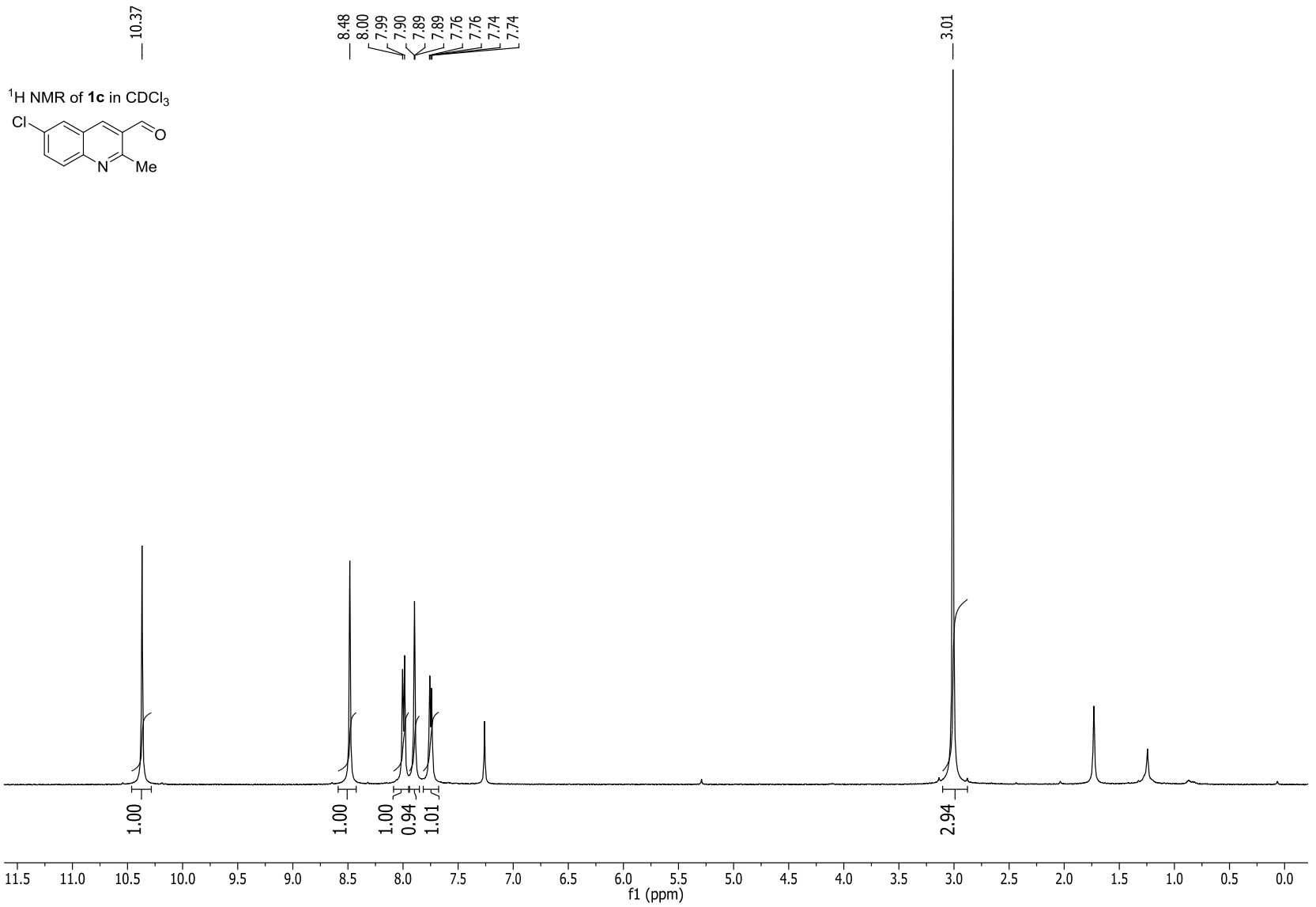

159.89
147.34
145.57
135.70
131.79
130.37
130.32
130.17
129.49
128.82
128.65
127.49
125.63
77.41
77.16
76.91
60.06
23.66
13C NMR of 12i in CDCl3
Ph
Cl
OH
N
Me
f1 (ppm)

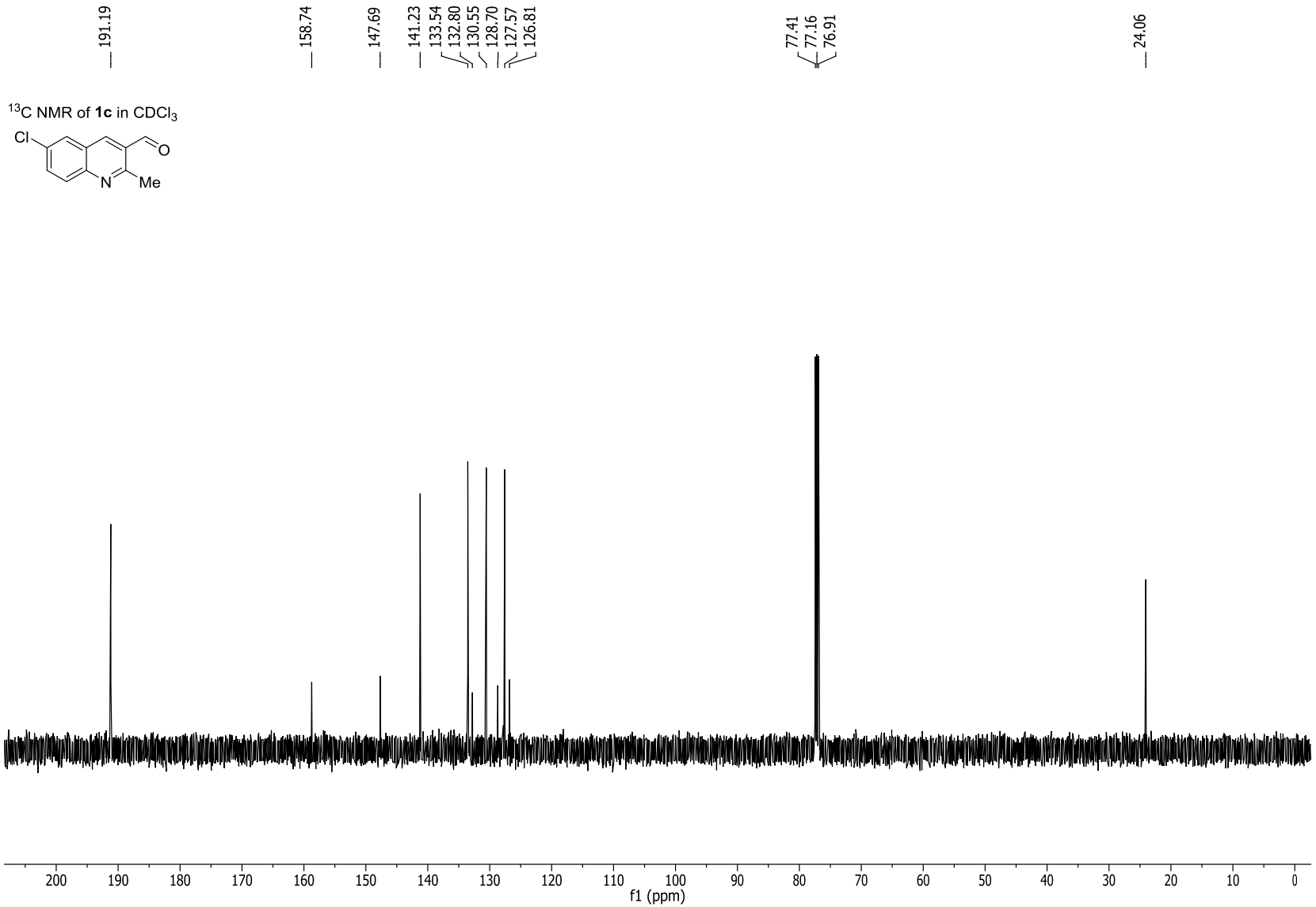

8.13
8.06
8.04
7.78
7.78
7.78
7.77
7.76
7.76
7.68
7.68
7.67
7.66
7.66
7.65
7.65
7.50
7.50
7.48
7.48
7.48
7.47
7.47
7.26
4.92
3.03
3.01
3.00
2.98
1.40
1.39
1.37
1H NMR of 12k in CDCl3
OH
N
Et
1.00
1.00
1.01
1.04
1.04
2.10
2.06
3.07
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

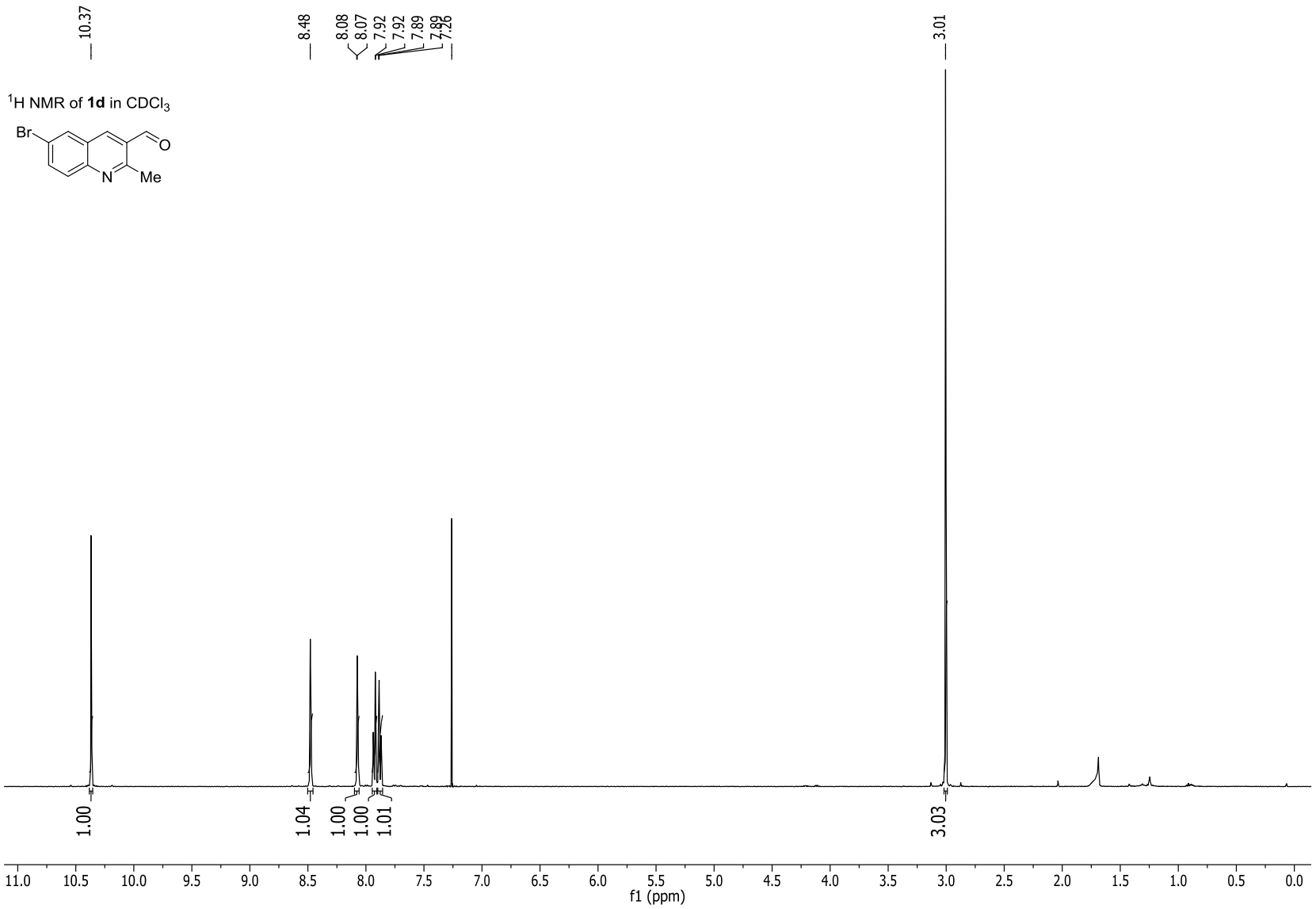

162.08
134.30
132.15
129.34
128.61
127.57
127.06
126.06
110.15
77.41
77.16
76.91
62.44
28.75
13.51
^{13}C NMR of **12k** in $CDCl_3$
OH
N
Et
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

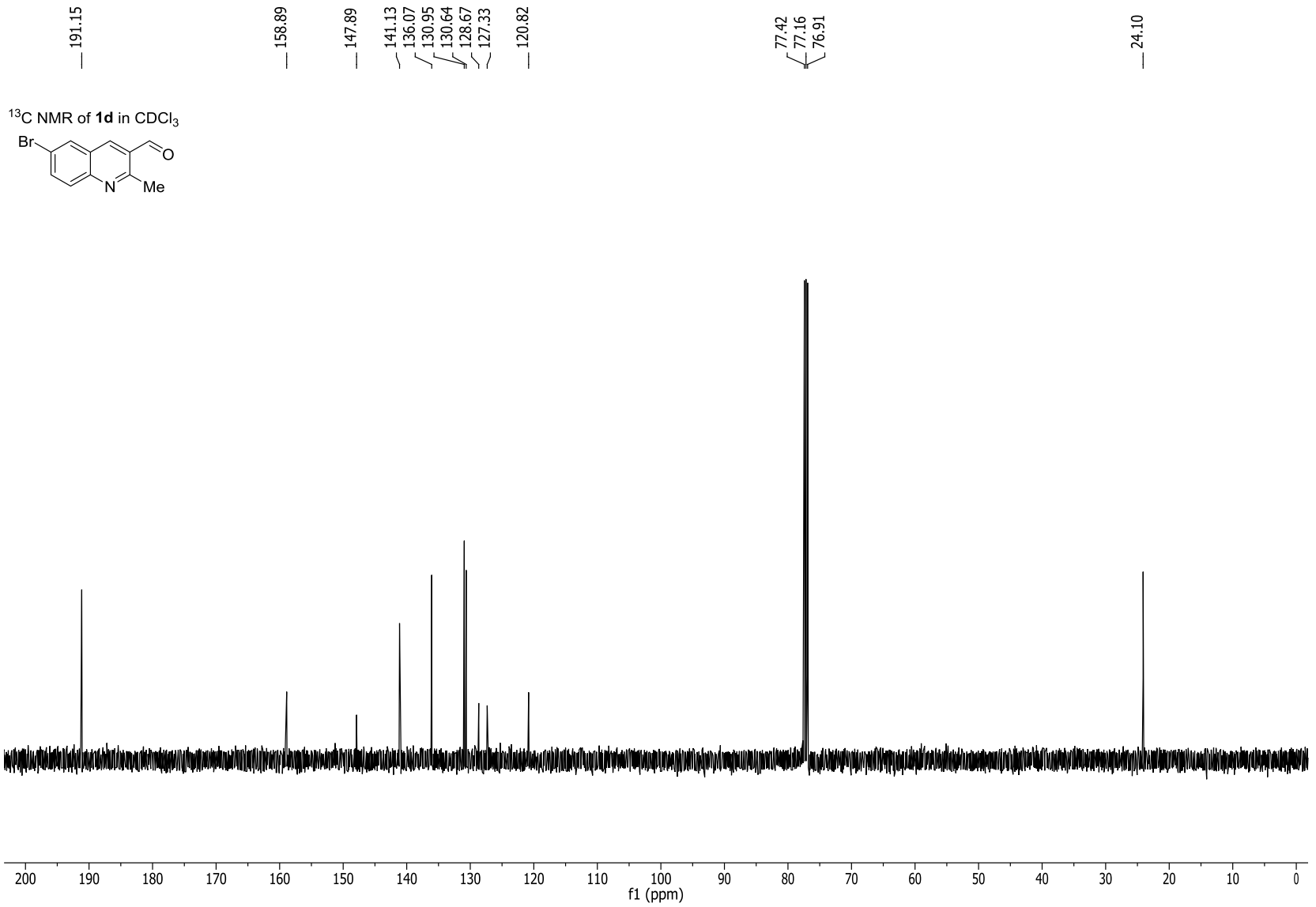

10.39
10.39
8.54
8.07
8.07
8.06
7.62
7.62
7.61
7.61
7.60
7.60
7.60
7.60
7.60
7.59
7.59
7.58
7.58
7.58
7.56
7.55
7.54
7.53
3.02
1H NMR of 1b in CDCl3
F
O
N
Me
0.93
1.00
0.98
0.99
0.96
2.87
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


191.29
161.51
159.52
157.68
157.66
146.43
141.52
141.48
131.45
131.38
128.55
126.77
126.69
122.93
122.73
112.03
111.86
77.41
77.16
76.91
23.90
13C NMR of 1b in CDCl3
F
O
N
Me
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

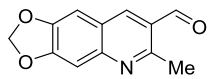

10.37
8.48
8.00
7.99
7.90
7.89
7.89
7.76
7.76
7.74
7.74
3.01
^{1}H NMR of **1c** in $CDCl_3$
Cl
O
N
Me
1.00
1.00
1.00
0.94
1.01
2.94
11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

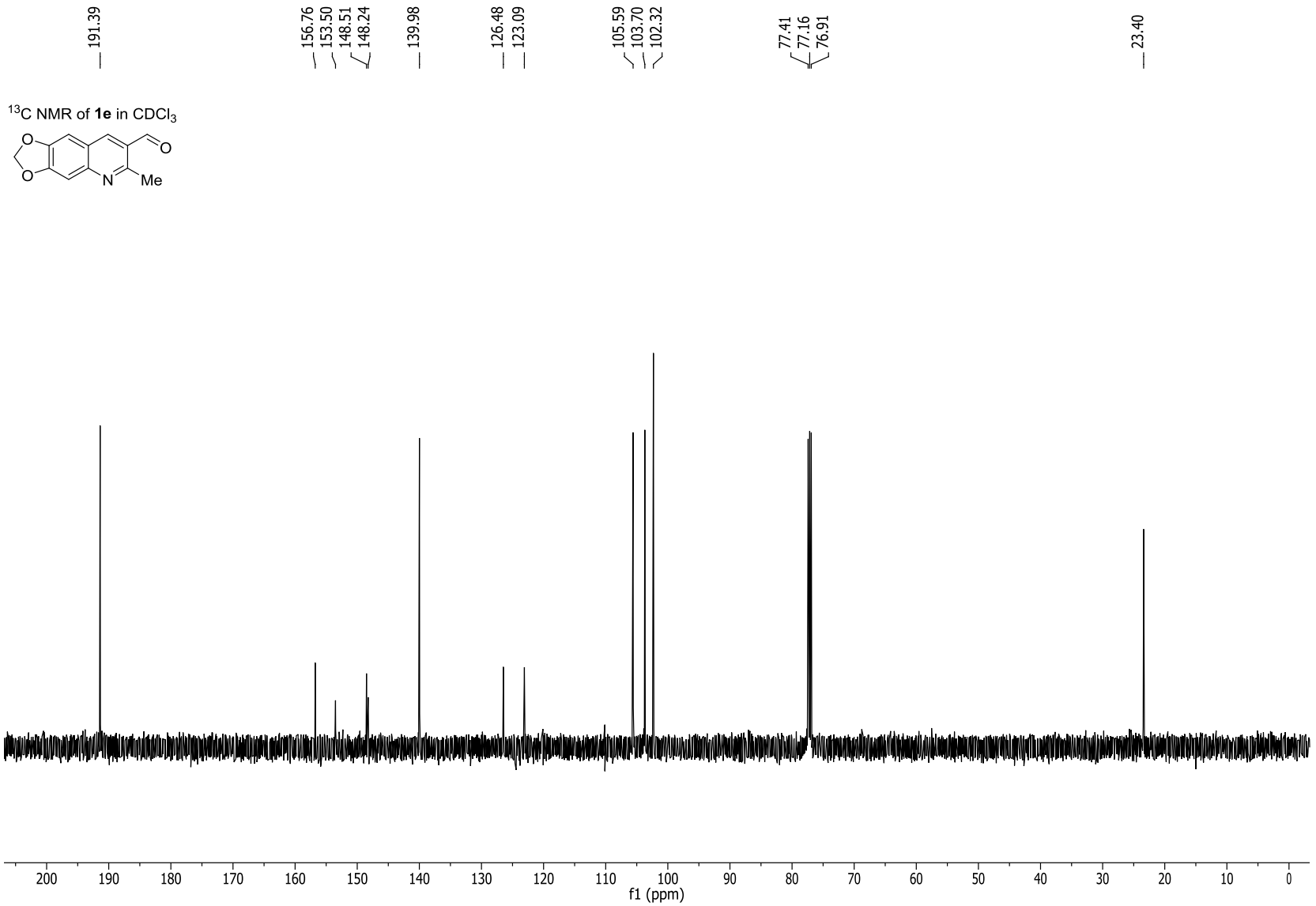

191.19
158.74
147.69
141.23
133.54
132.80
130.55
128.70
127.57
126.81
77.41
77.16
76.91
24.06
13C NMR of 1c in CDCl3
Cl
O
N
Me
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

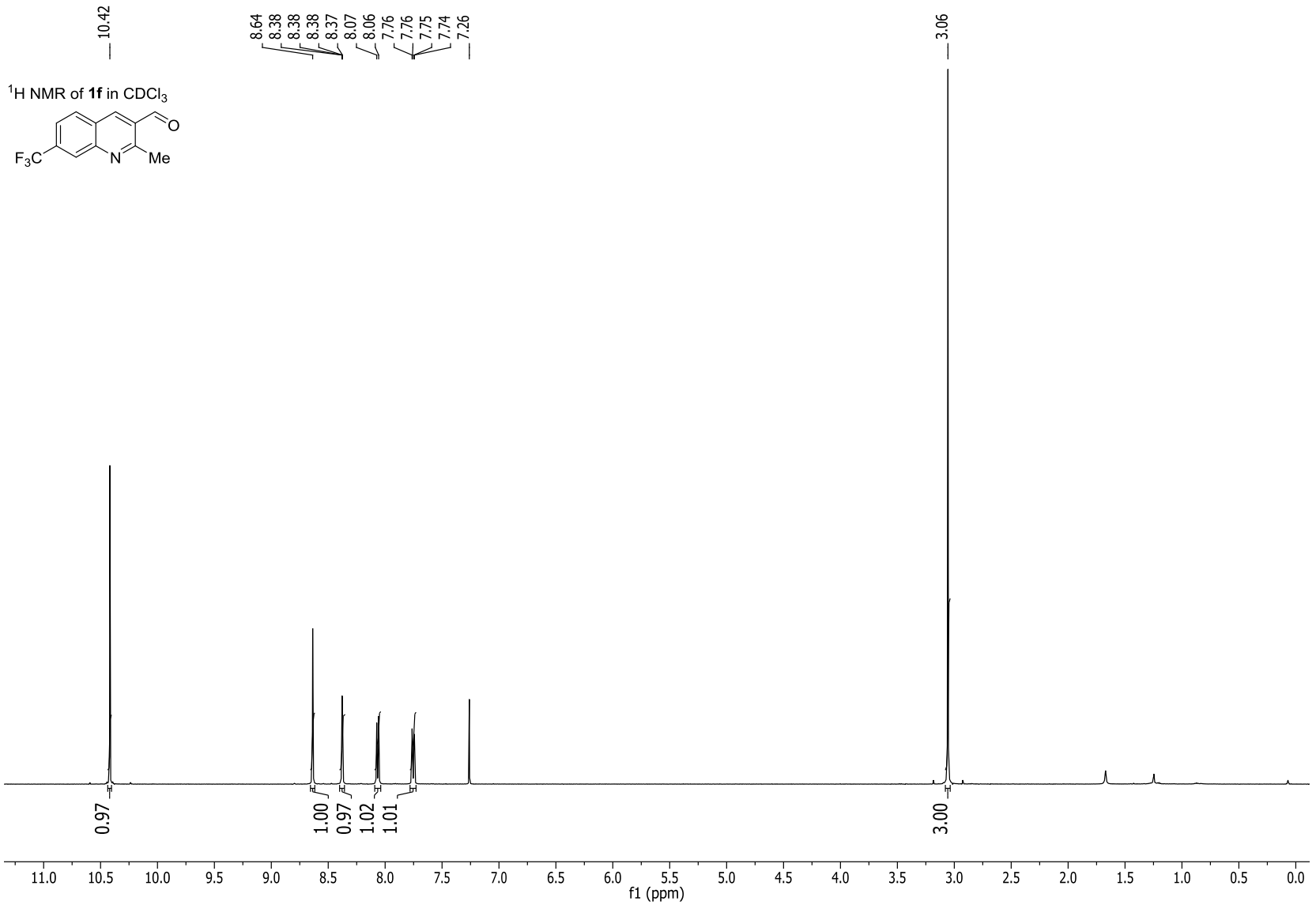

10.37
8.48
8.08
8.07
7.92
7.92
7.89
7.89
7.26
3.01
^{1}H NMR of **1d** in $CDCl_3$
Br
O
N
Me
1.00
1.04
1.00
1.00
1.01
3.03
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

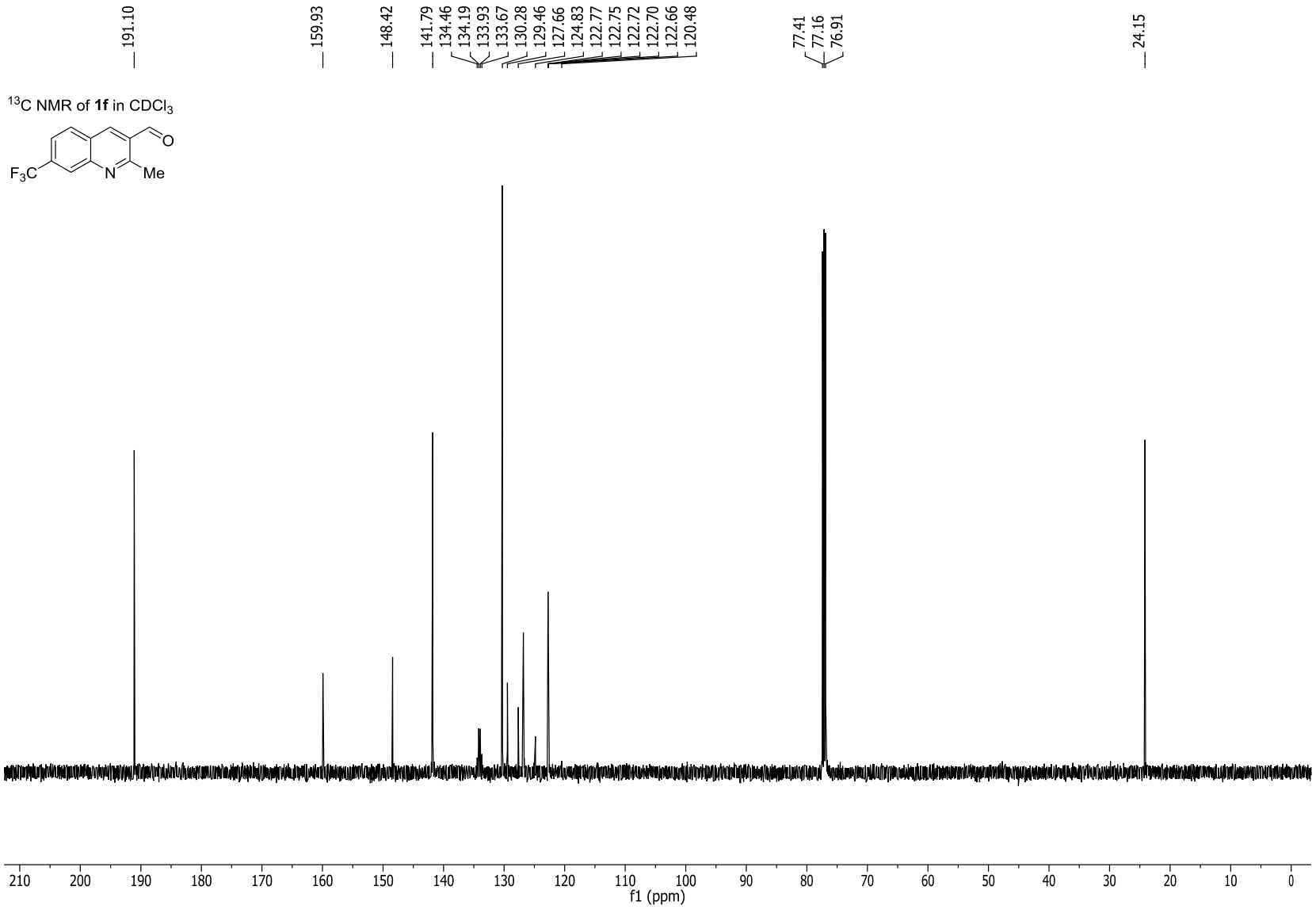

191.15
158.89
147.89
141.13
136.07
130.95
130.64
128.67
127.33
120.82
77.42
77.16
76.91
24.10
^{13}C NMR of **1d** in $CDCl_3$
Br
O
N
Me
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

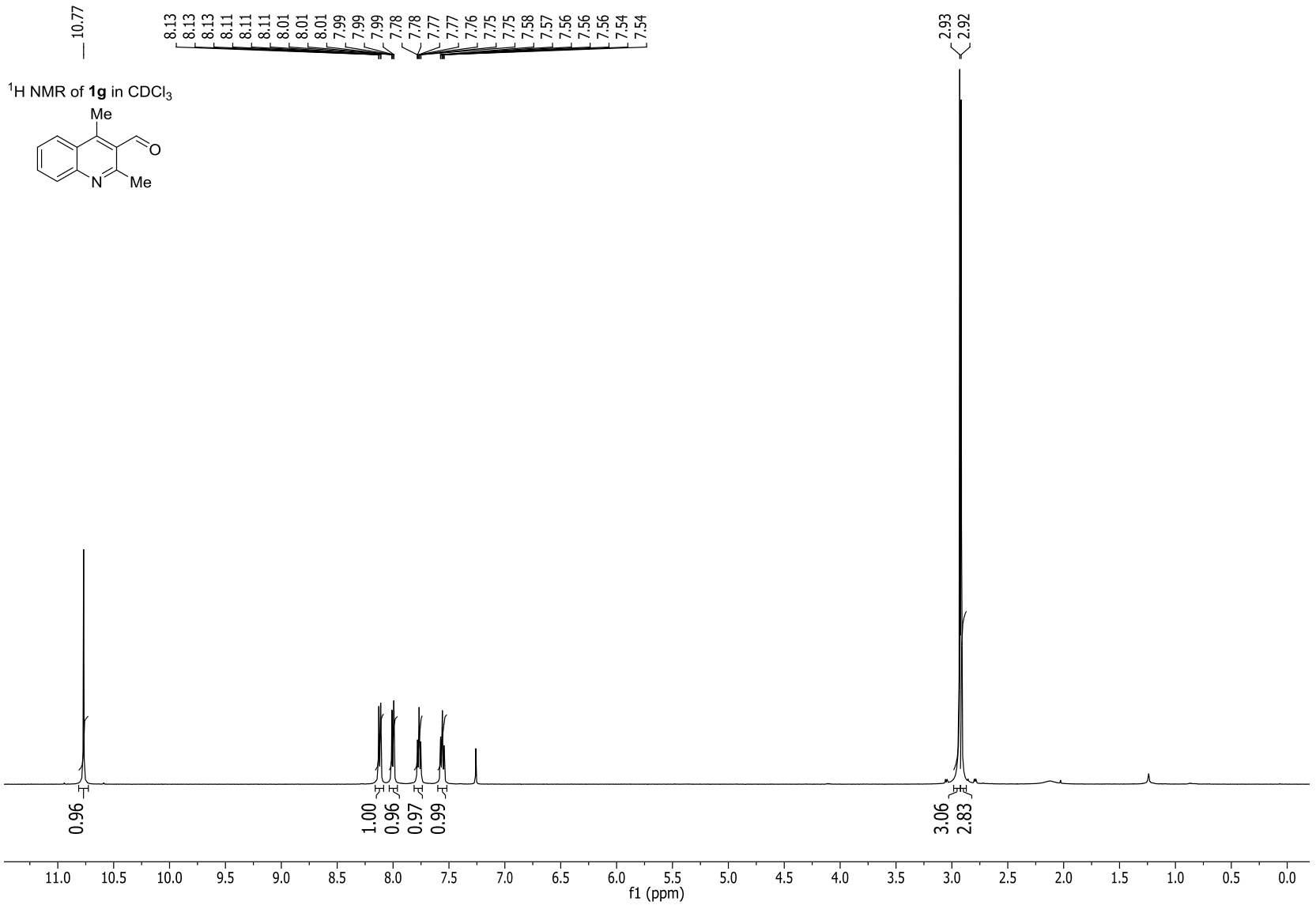
^{1}H NMR of **1e** in $CDCl_3$

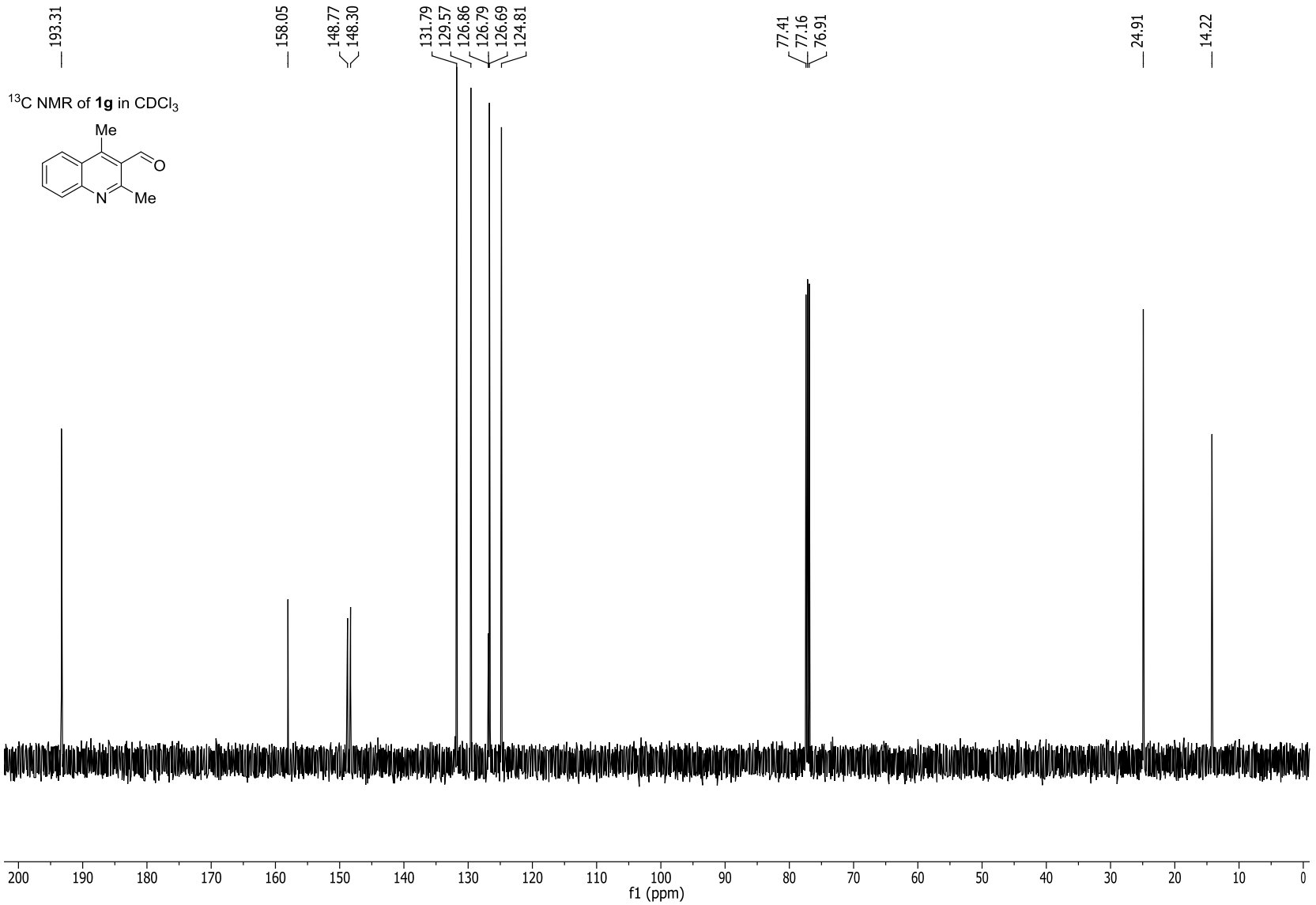


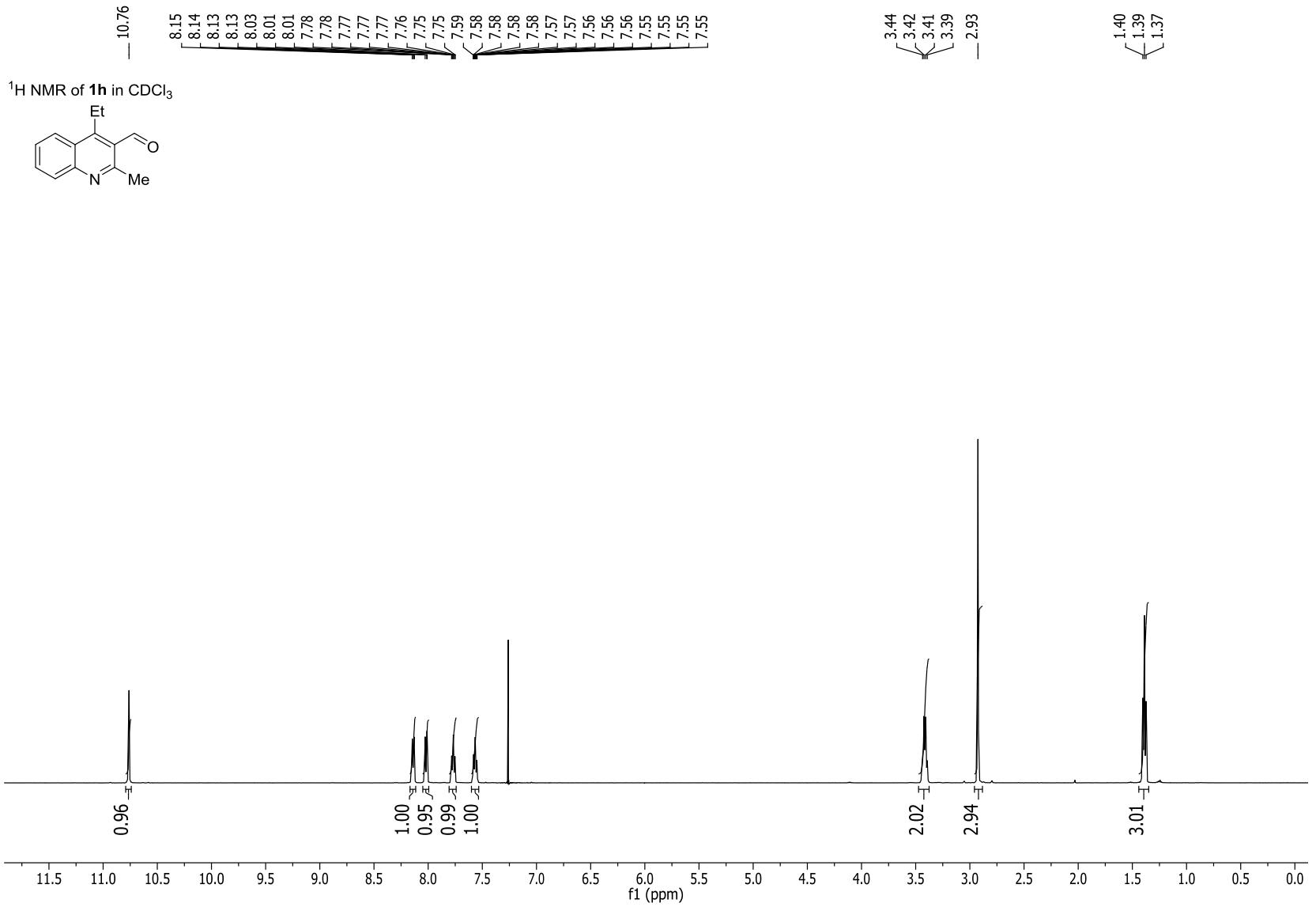

191.39
156.76
153.50
148.51
148.24
139.98
126.48
123.09
105.59
103.70
102.32
77.41
77.16
76.91
23.40
^{13}C NMR of **1e** in $CDCl_3$
O
O
O
N
Me
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

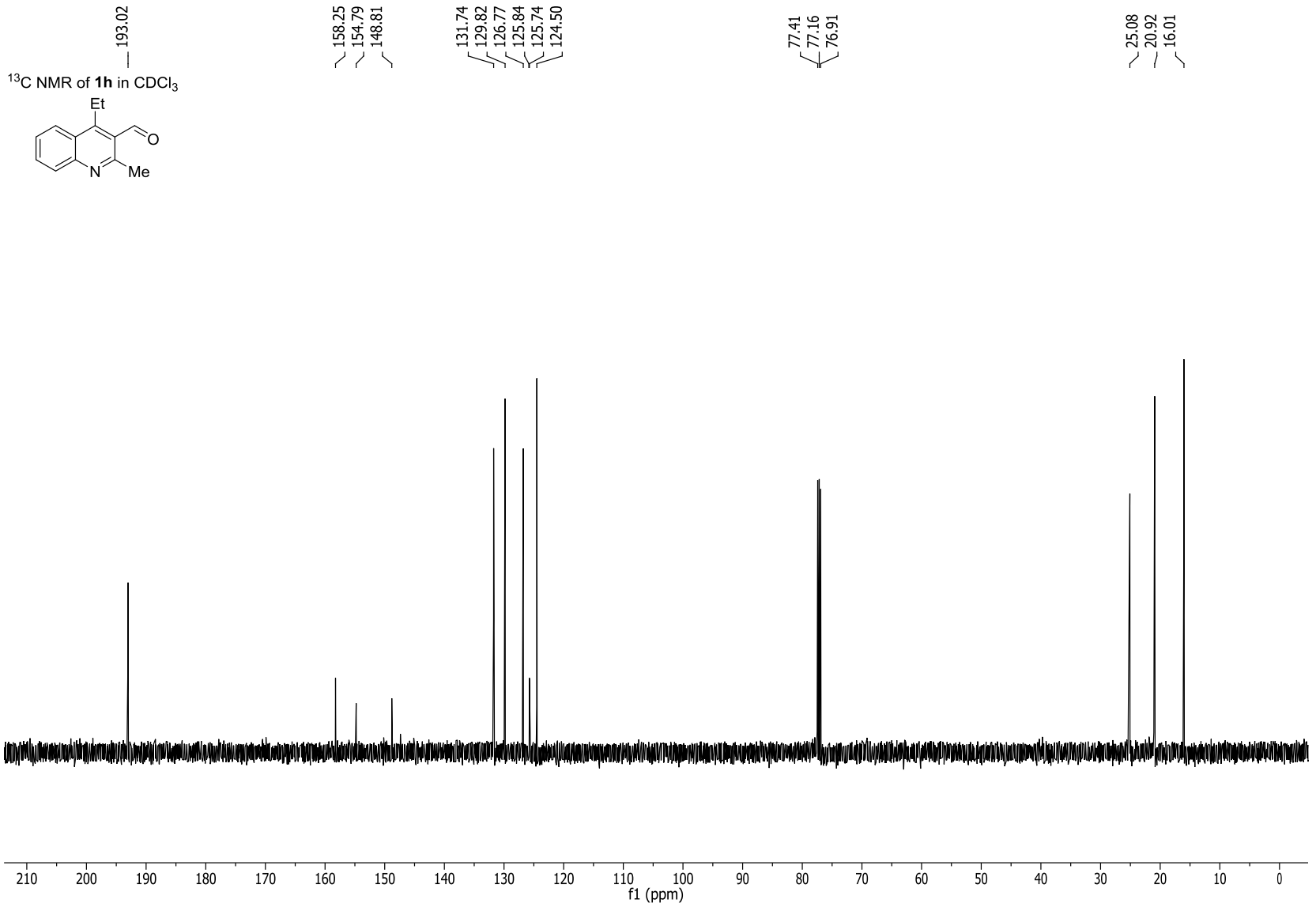

10.42
8.64
8.38
8.38
8.38
8.37
8.07
8.06
7.76
7.76
7.75
7.74
7.26
3.06
^{1}H NMR of **1f** in $CDCl_3$
F_3C
N
Me
O
0.97
1.00
0.97
1.02
1.01
3.00
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

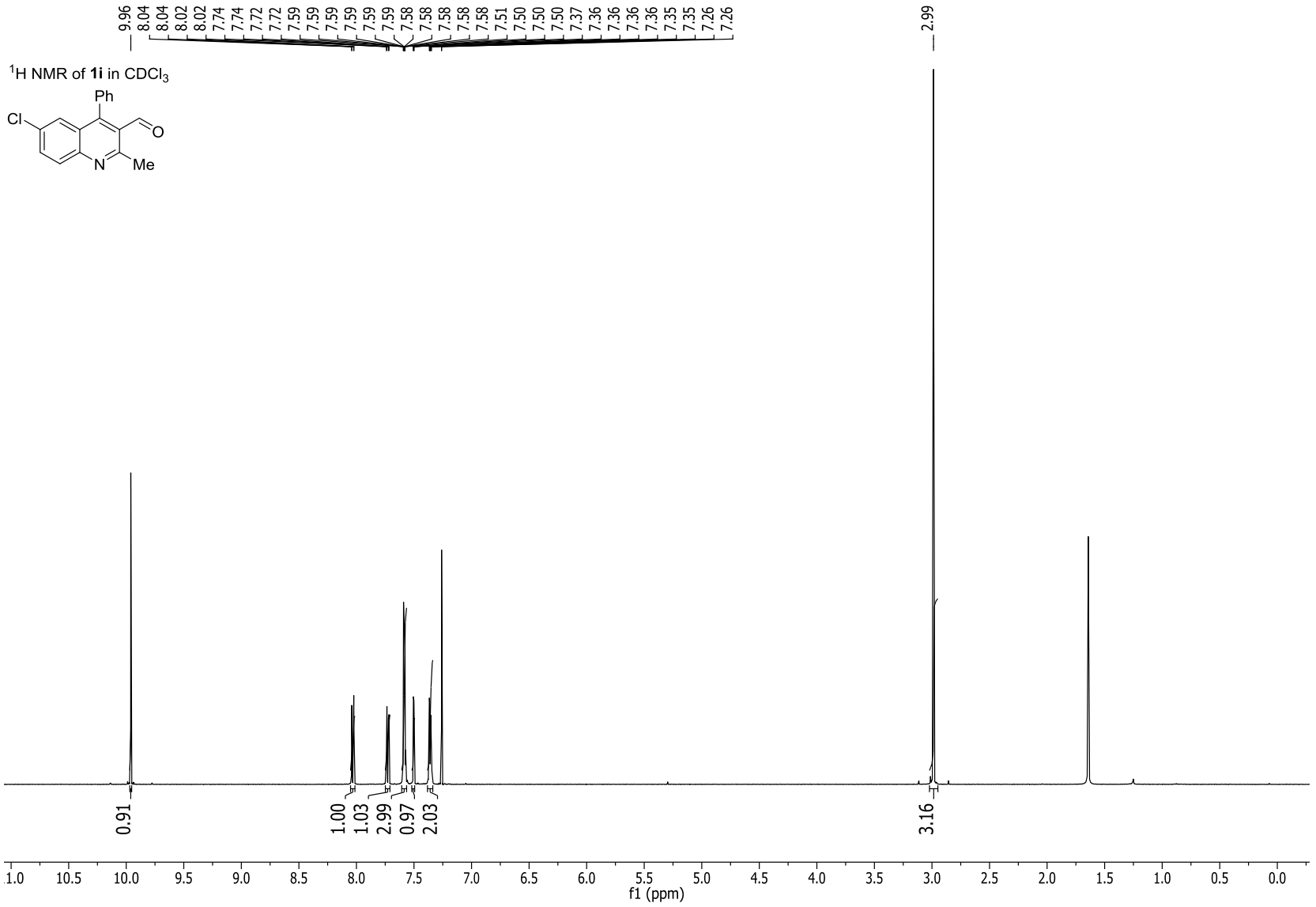

191.10
159.93
148.42
141.79
134.46
134.19
133.93
133.67
130.28
129.46
127.66
124.83
122.77
122.75
122.72
122.70
122.66
120.48
77.41
77.16
76.91
24.15
13C NMR of 1f in CDCl3
F3C
N
Me
O
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

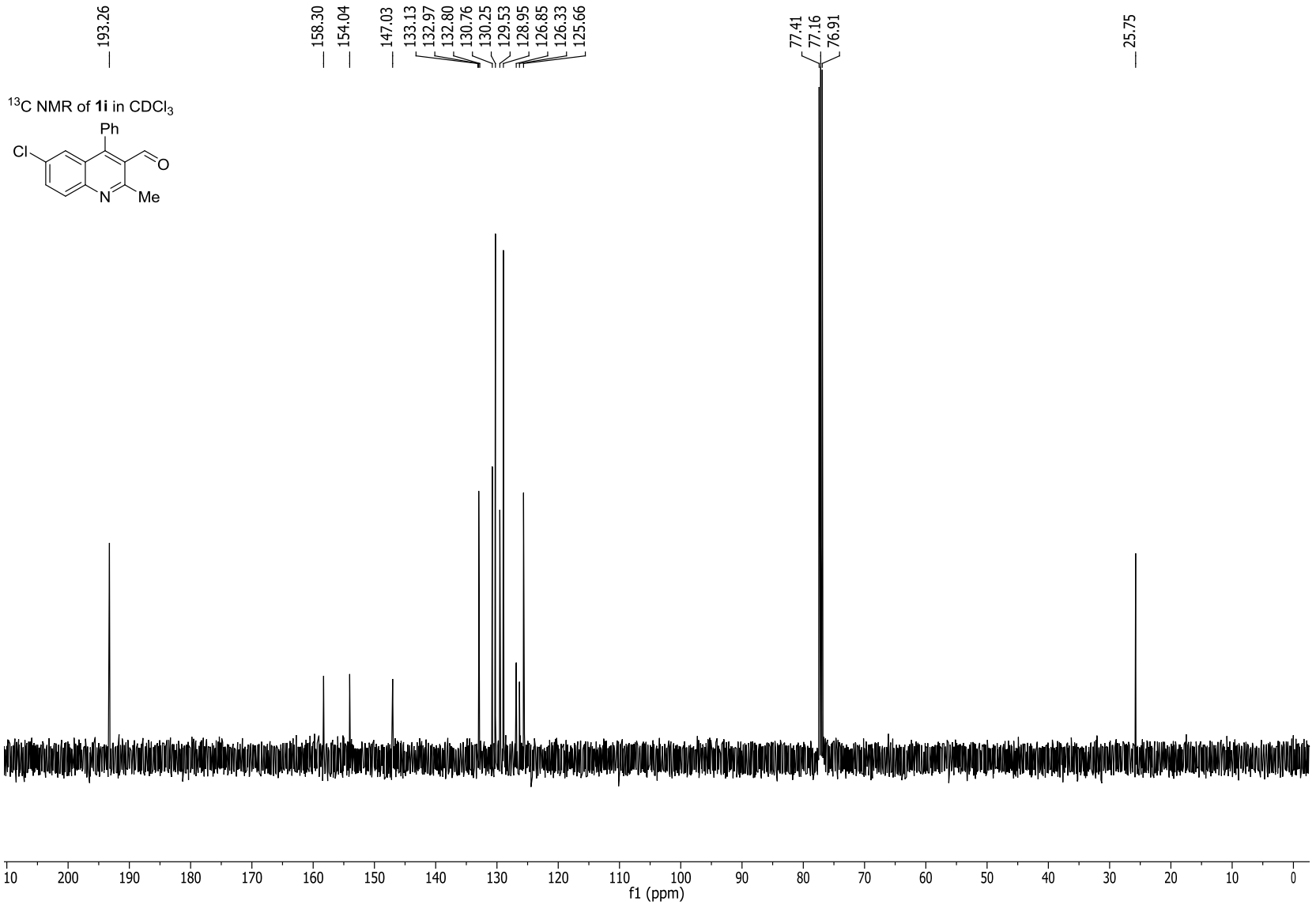

10.77
8.13
8.13
8.13
8.11
8.11
8.11
8.01
8.01
8.01
7.99
7.99
7.99
7.78
7.78
7.77
7.77
7.76
7.75
7.75
7.58
7.57
7.56
7.56
7.56
7.54
7.54
2.93
2.92
^{1}H NMR of **1g** in $CDCl_3$
Me
O
N
Me
0.96
1.00
0.96
0.97
0.99
3.06
2.83
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


193.31
158.05
148.77
148.30
131.79
129.57
126.86
126.79
126.69
124.81
77.41
77.16
76.91
24.91
14.22
^{13}C NMR of **1g** in $CDCl_3$
Me
O
N
Me
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

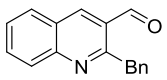

10.76
8.15
8.14
8.13
8.13
8.03
8.01
8.01
7.78
7.78
7.77
7.77
7.77
7.76
7.75
7.75
7.59
7.58
7.58
7.58
7.58
7.57
7.57
7.56
7.56
7.56
7.55
7.55
7.55
7.55
3.44
3.42
3.41
3.39
2.93
1.40
1.39
1.37
^{1}H NMR of **1h** in $CDCl_3$
Et
O
N
Me
0.96
1.00
0.95
0.99
1.00
2.02
2.94
3.01
11.5
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

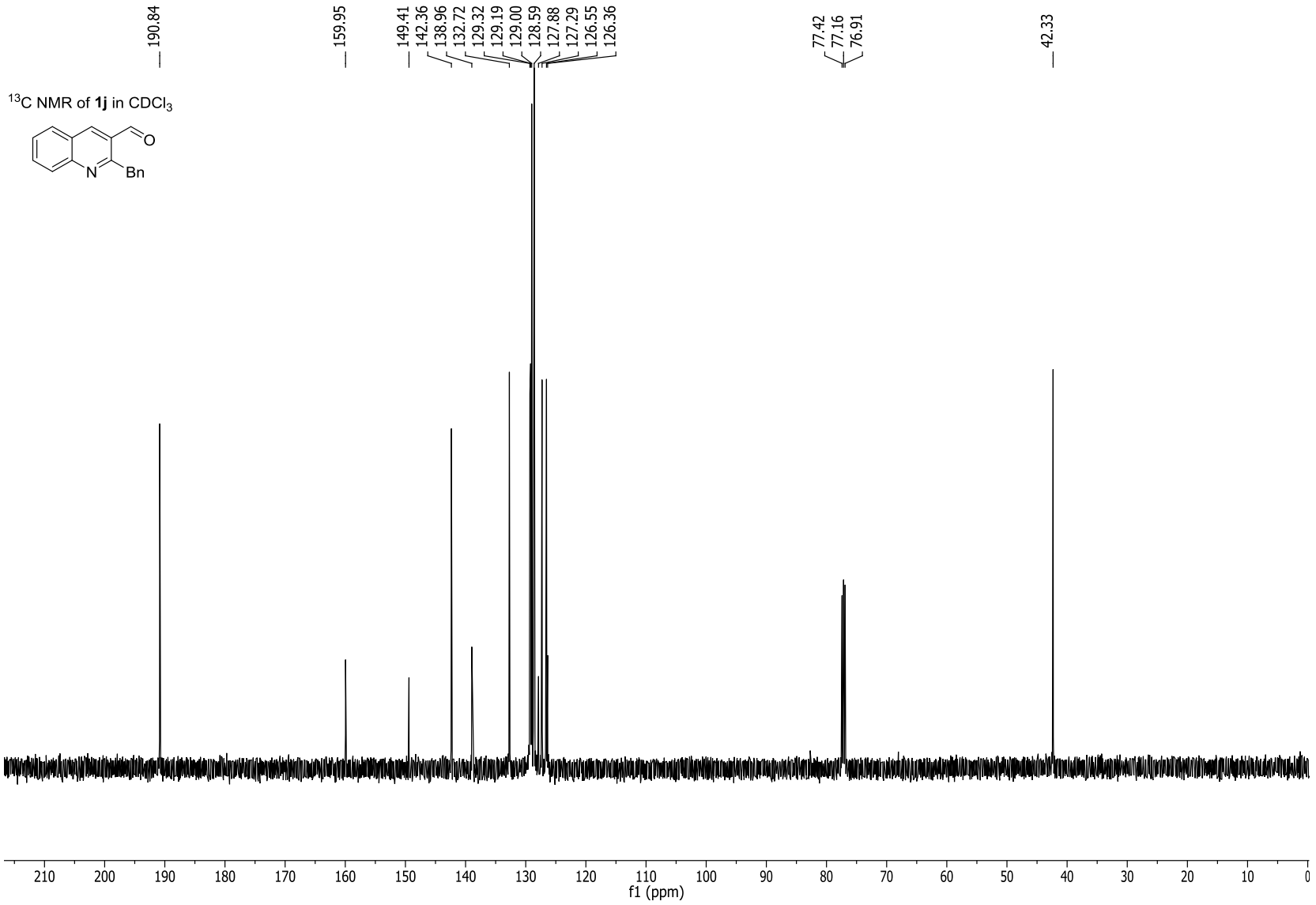

193.02
158.25
154.79
148.81
131.74
129.82
126.77
125.84
125.74
124.50
77.41
77.16
76.91
25.08
20.92
16.01
^{13}C NMR of **1h** in $CDCl_3$
Et
O
N
Me
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

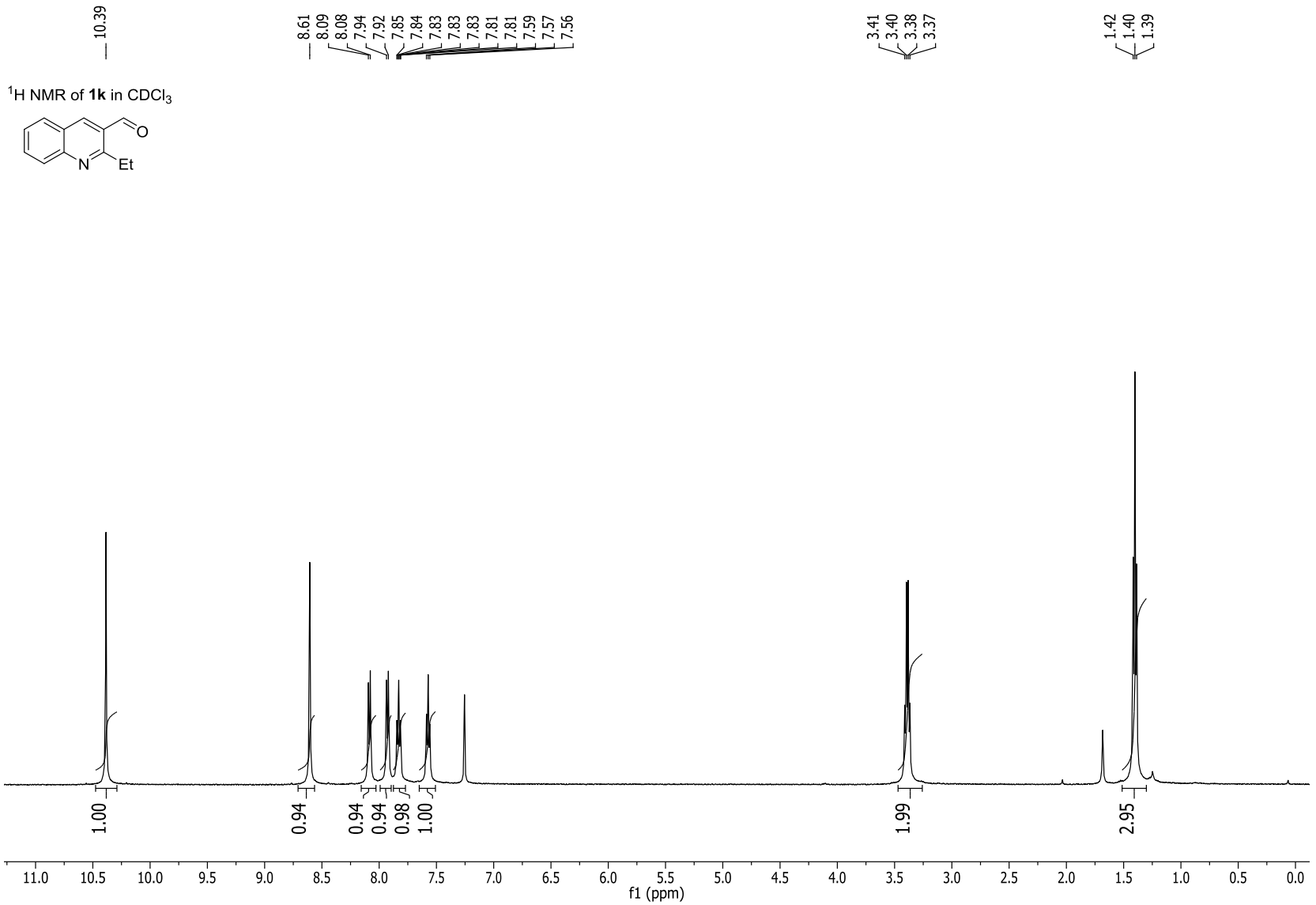

9.96
8.04
8.04
8.02
8.02
7.74
7.74
7.72
7.72
7.59
7.59
7.59
7.59
7.59
7.59
7.58
7.58
7.58
7.58
7.58
7.51
7.50
7.50
7.50
7.37
7.36
7.36
7.36
7.36
7.35
7.35
7.26
7.26
2.99
1H NMR of 1i in CDCl3
Ph
Cl
O
N
Me
0.91
1.00
1.03
2.99
0.97
2.03
3.16
f1 (ppm)

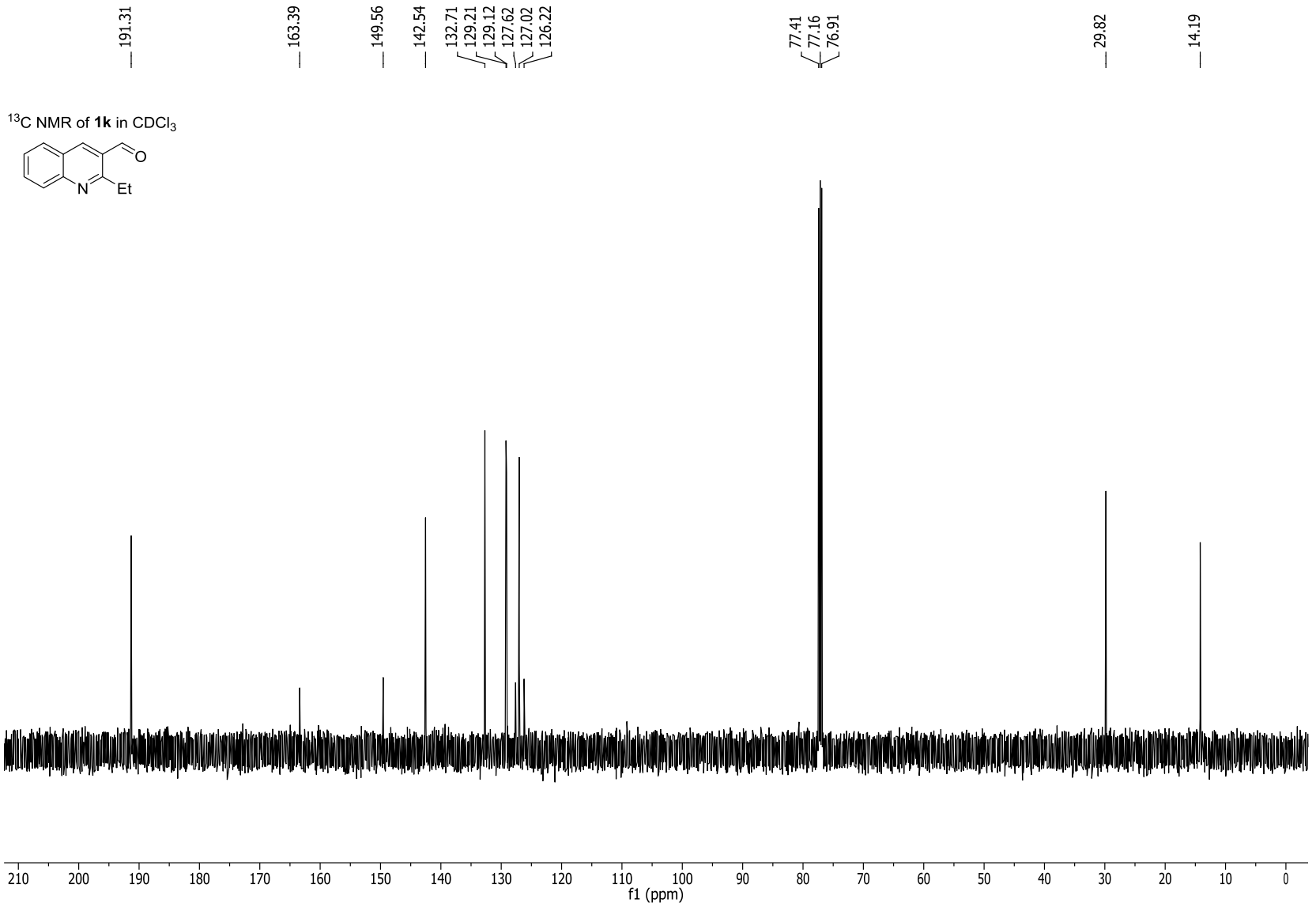

193.26
158.30
154.04
147.03
133.13
132.97
132.80
130.76
130.25
129.53
128.95
126.85
126.33
125.66
77.41
77.16
76.91
25.75
13C NMR of 1i in CDCl3
Ph
Cl
O
N
Me
f1 (ppm)

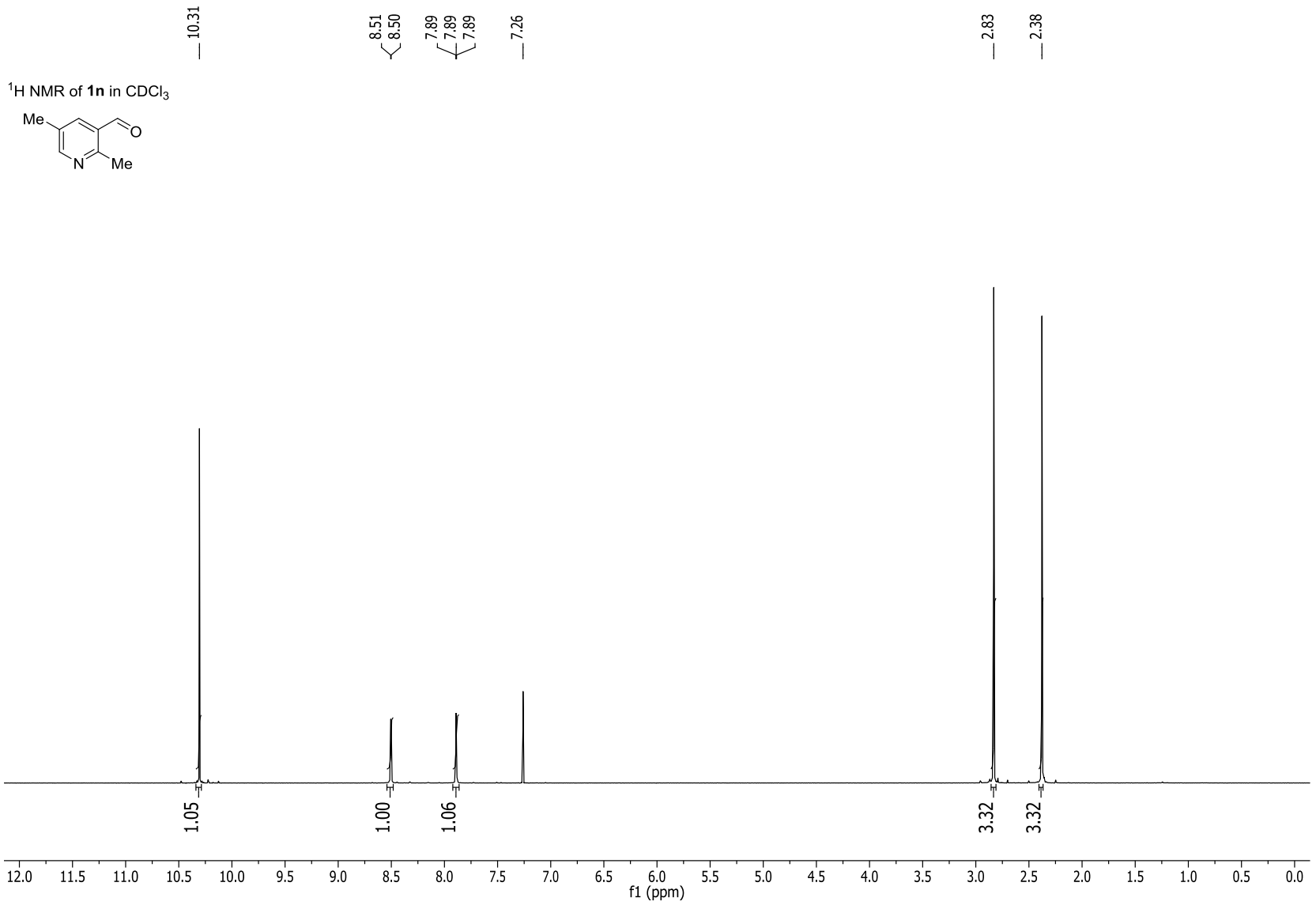
^{1}H NMR of **1j** in $CDCl_3$

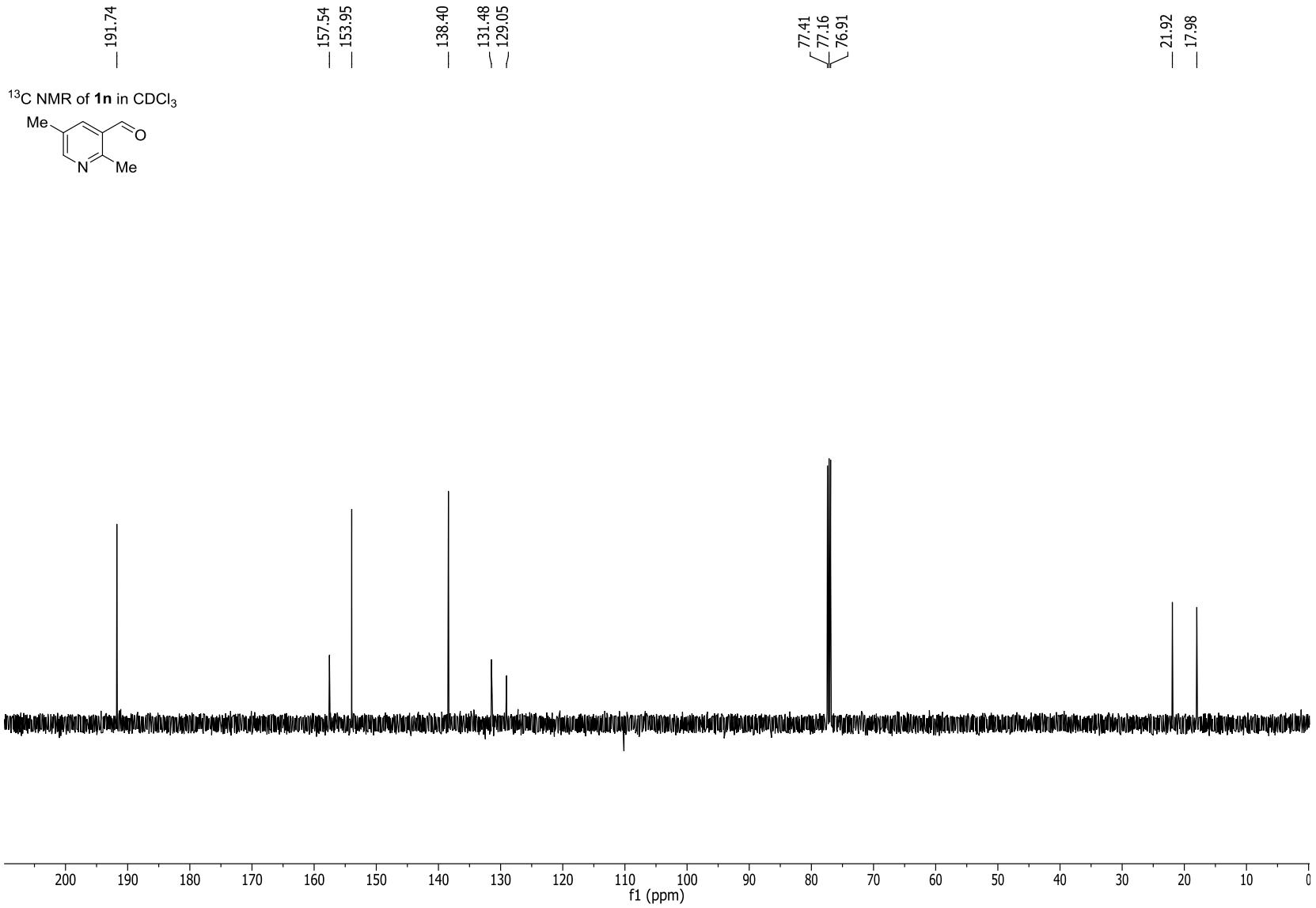


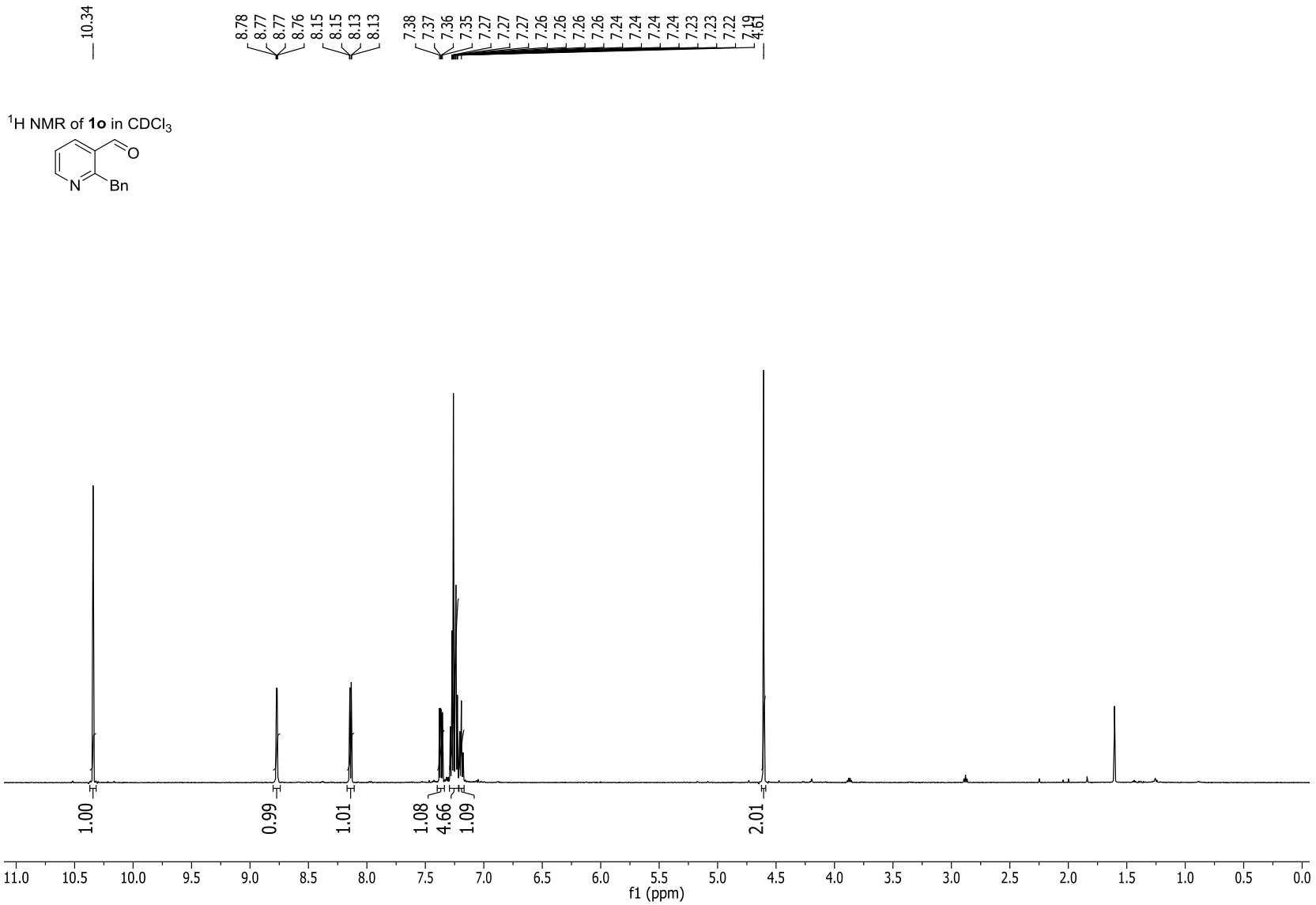

190.84
159.95
149.41
142.36
138.96
132.72
129.32
129.19
129.00
128.59
127.88
127.29
126.55
126.36
77.42
77.16
76.91
42.33
13C NMR of 1j in CDCl3
O
N
Bn
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

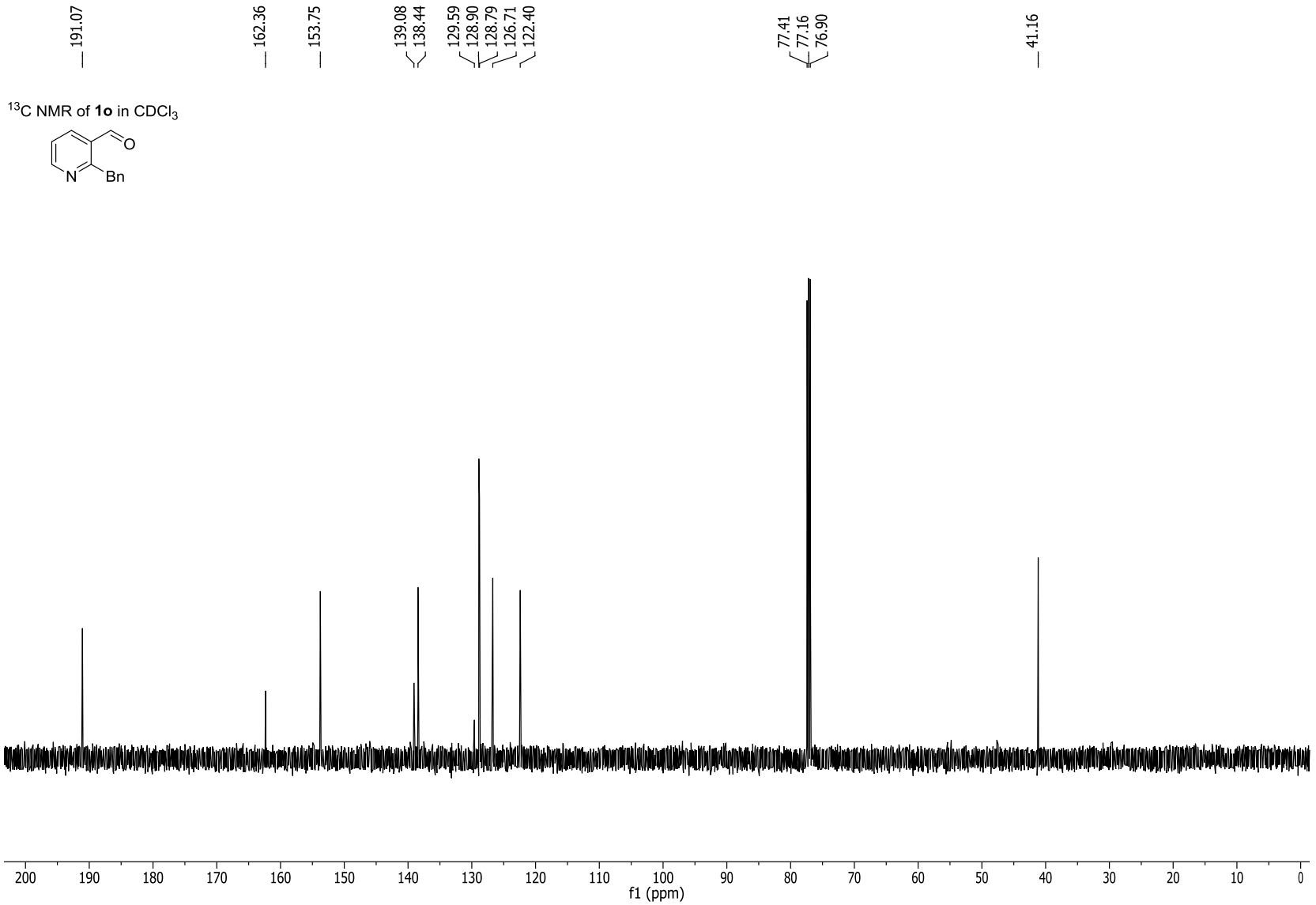

10.39
8.61
8.09
8.08
7.94
7.92
7.85
7.84
7.83
7.83
7.83
7.81
7.81
7.59
7.57
7.56
3.41
3.40
3.38
3.37
1.42
1.40
1.39
1H NMR of 1k in CDCl3
O
N
Et
1.00
0.94
0.94
0.94
0.98
1.00
1.99
2.95
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

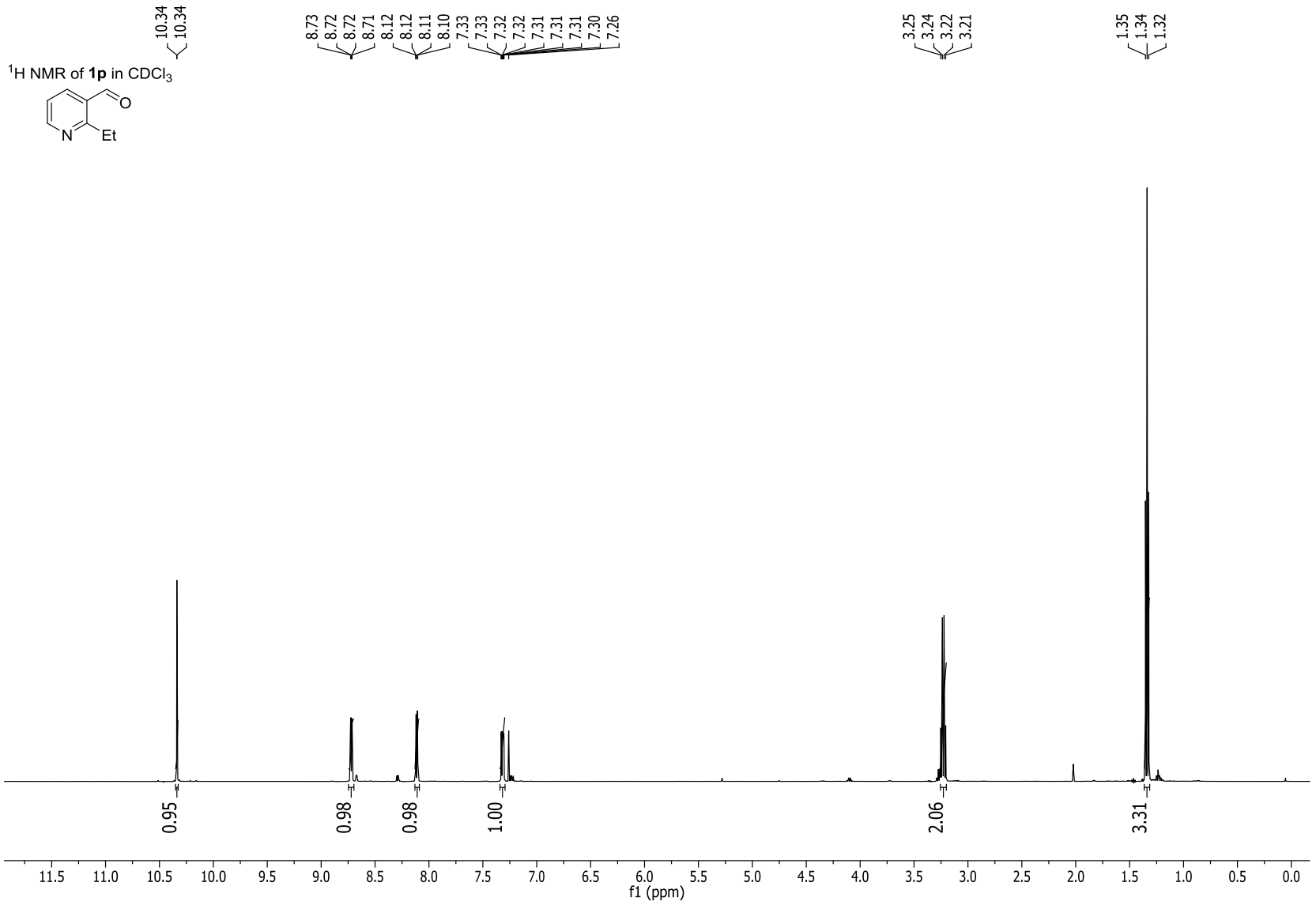

191.31
163.39
149.56
142.54
132.71
129.21
129.12
127.62
127.02
126.22
77.41
77.16
76.91
29.82
14.19
^{13}C NMR of **1k** in $CDCl_3$
O
N
Et
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

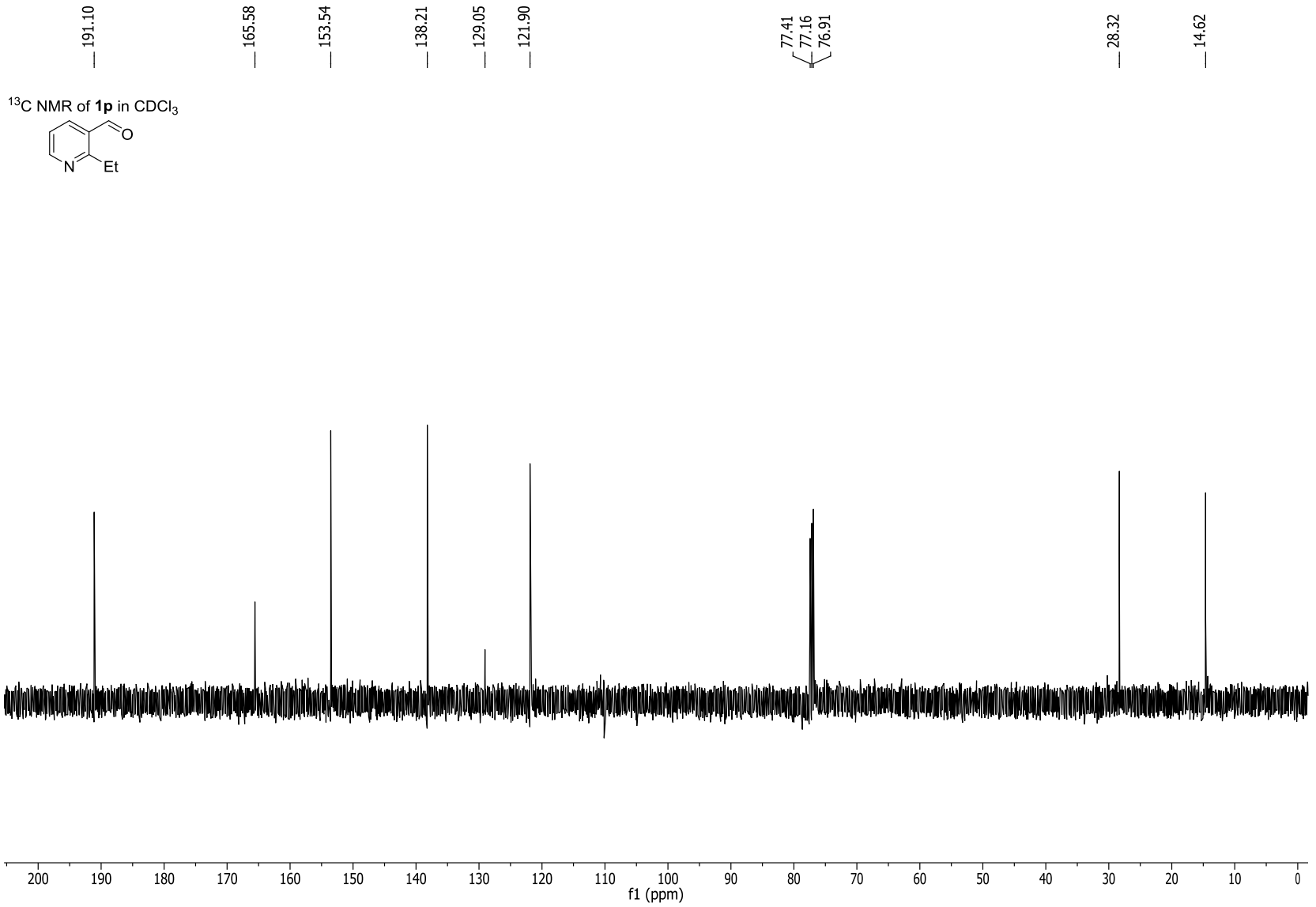

10.31
8.51
8.50
7.89
7.89
7.89
7.26
2.83
2.38
^{1}H NMR of **1n** in $CDCl_3$
Me
O
N
Me
1.05
1.00
1.06
3.32
3.32
12.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

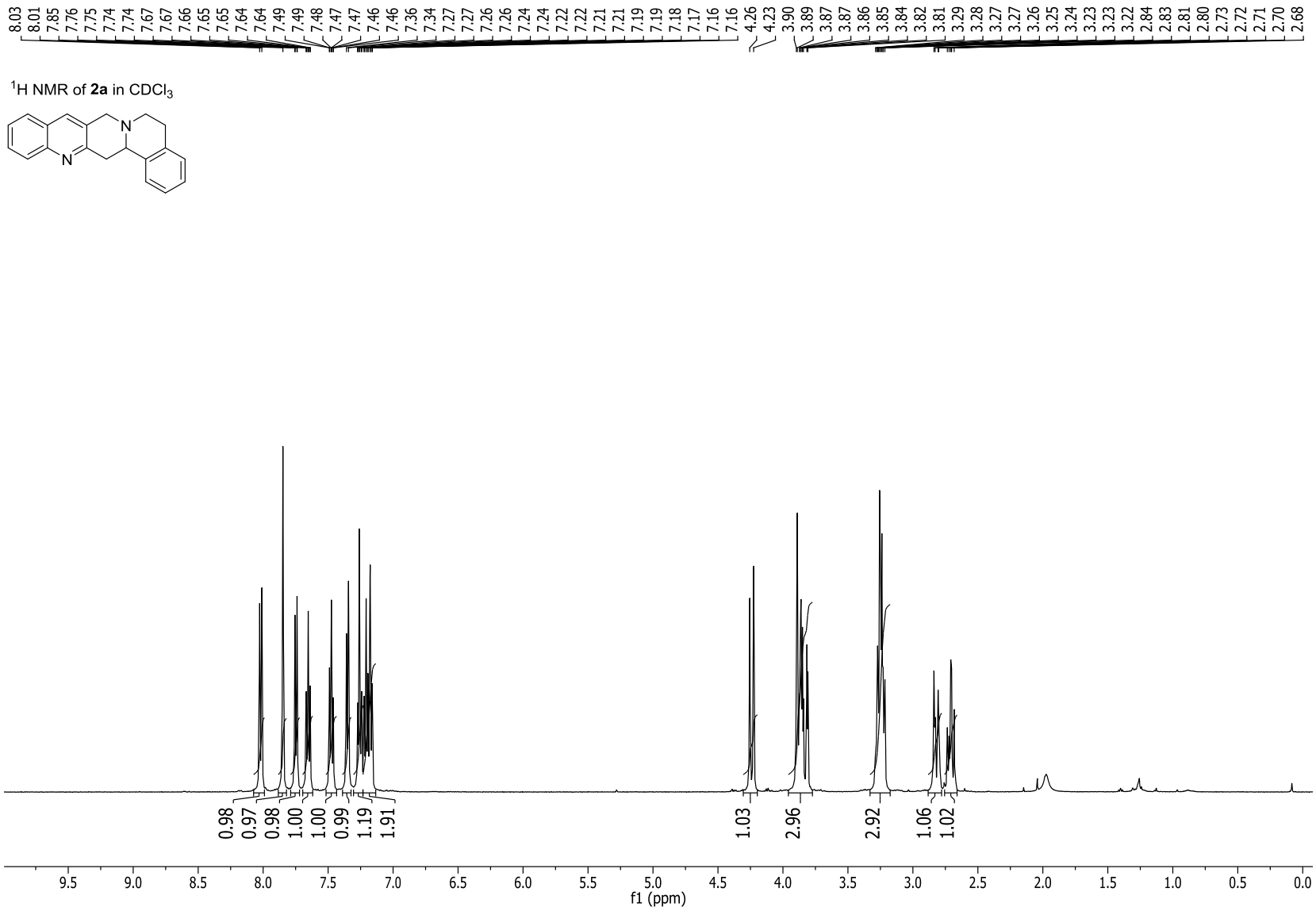

191.74
157.54
153.95
138.40
131.48
129.05
77.41
77.16
76.91
21.92
17.98
^{13}C NMR of **1n** in $CDCl_3$
Me
O
N
Me
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

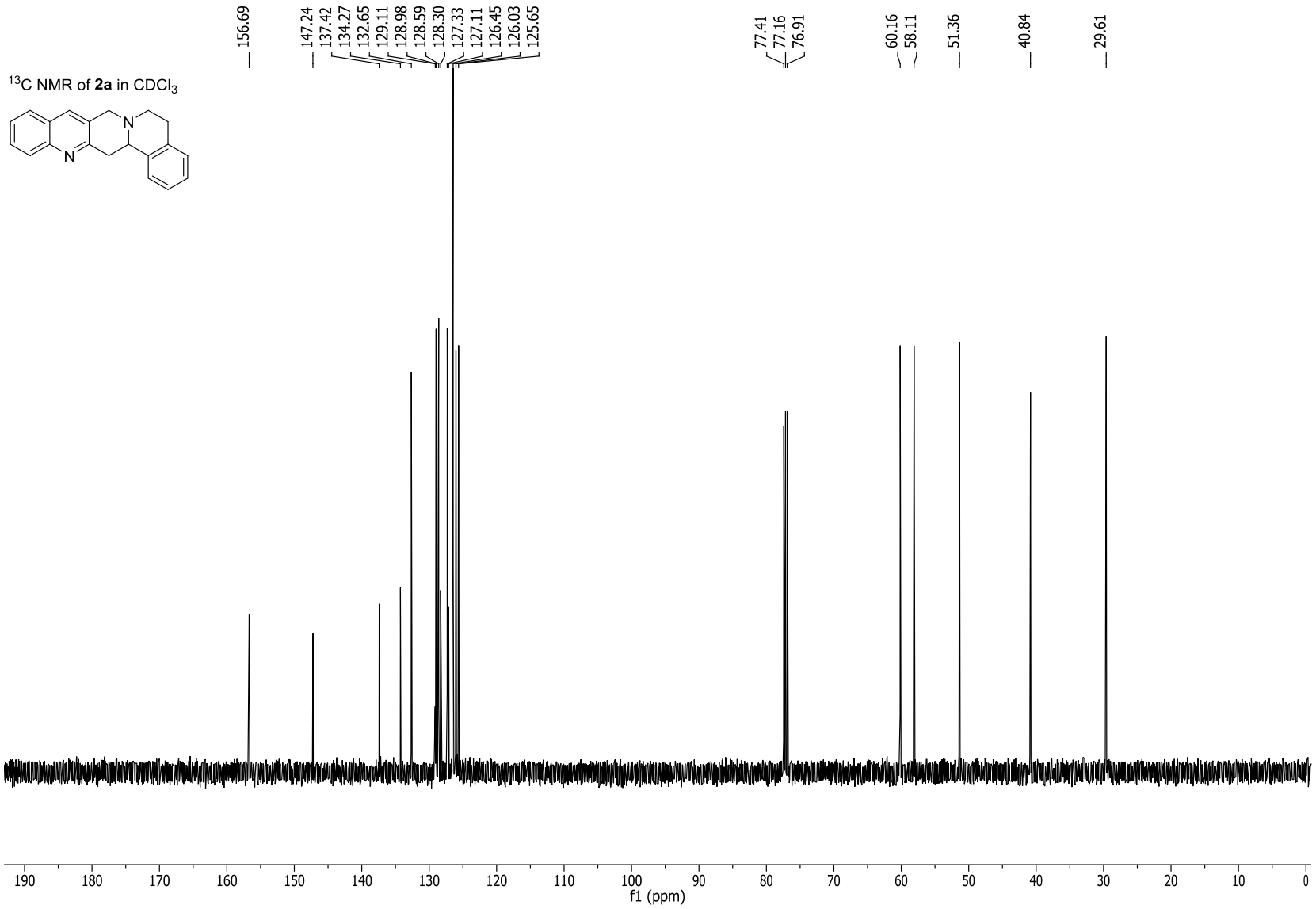

10.34
8.78
8.77
8.77
8.76
8.15
8.15
8.13
8.13
7.38
7.37
7.36
7.35
7.27
7.27
7.27
7.26
7.26
7.26
7.24
7.24
7.24
7.24
7.23
7.23
7.22
7.19
4.61
1H NMR of 1o in CDCl3
O
N
Bn
1.00
0.99
1.01
1.08
4.66
1.09
2.01
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

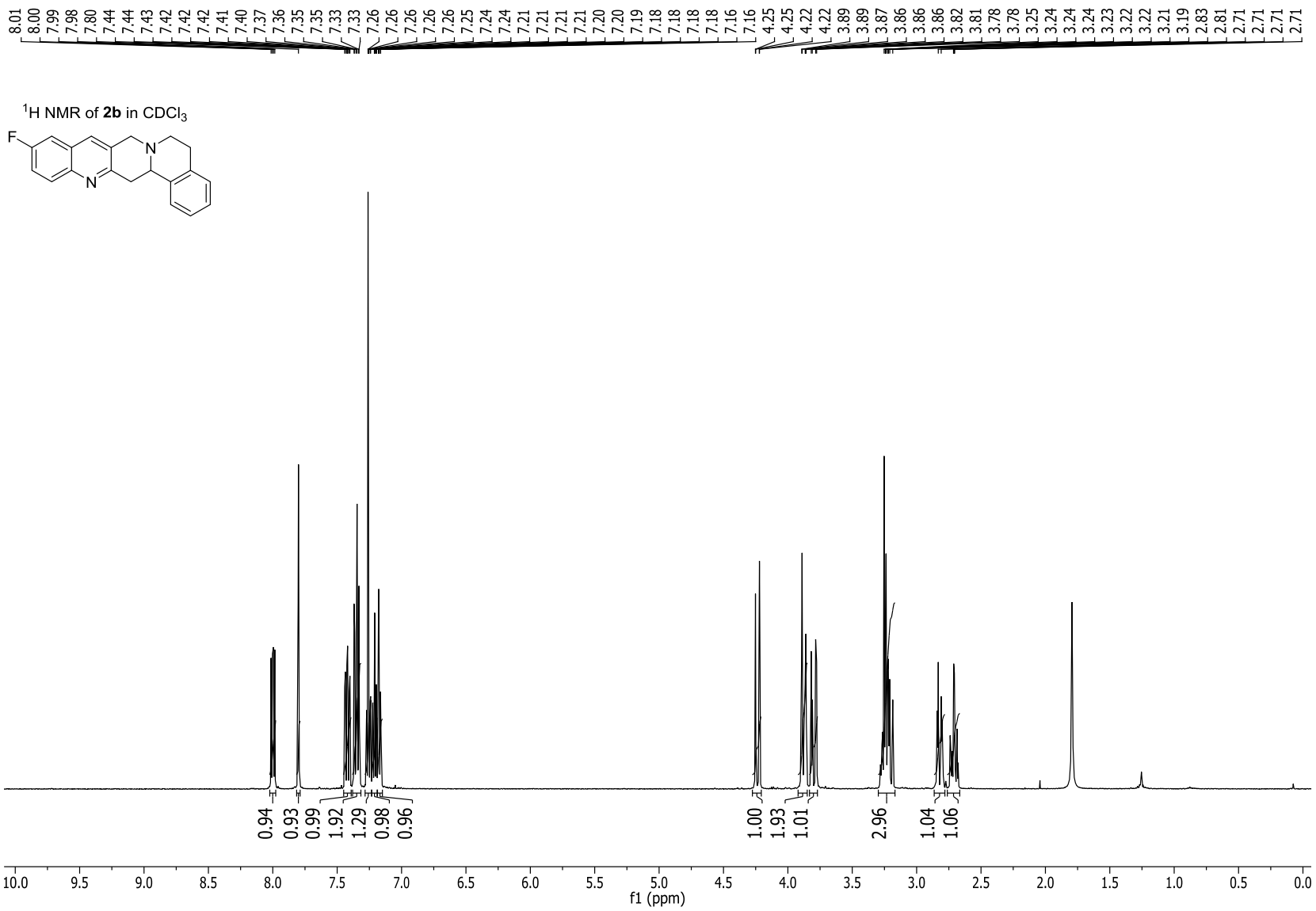

191.07
162.36
153.75
139.08
138.44
129.59
128.90
128.79
126.71
122.40
77.41
77.16
76.90
41.16
^{13}C NMR of **1o** in $CDCl_3$
O
N
Bn
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

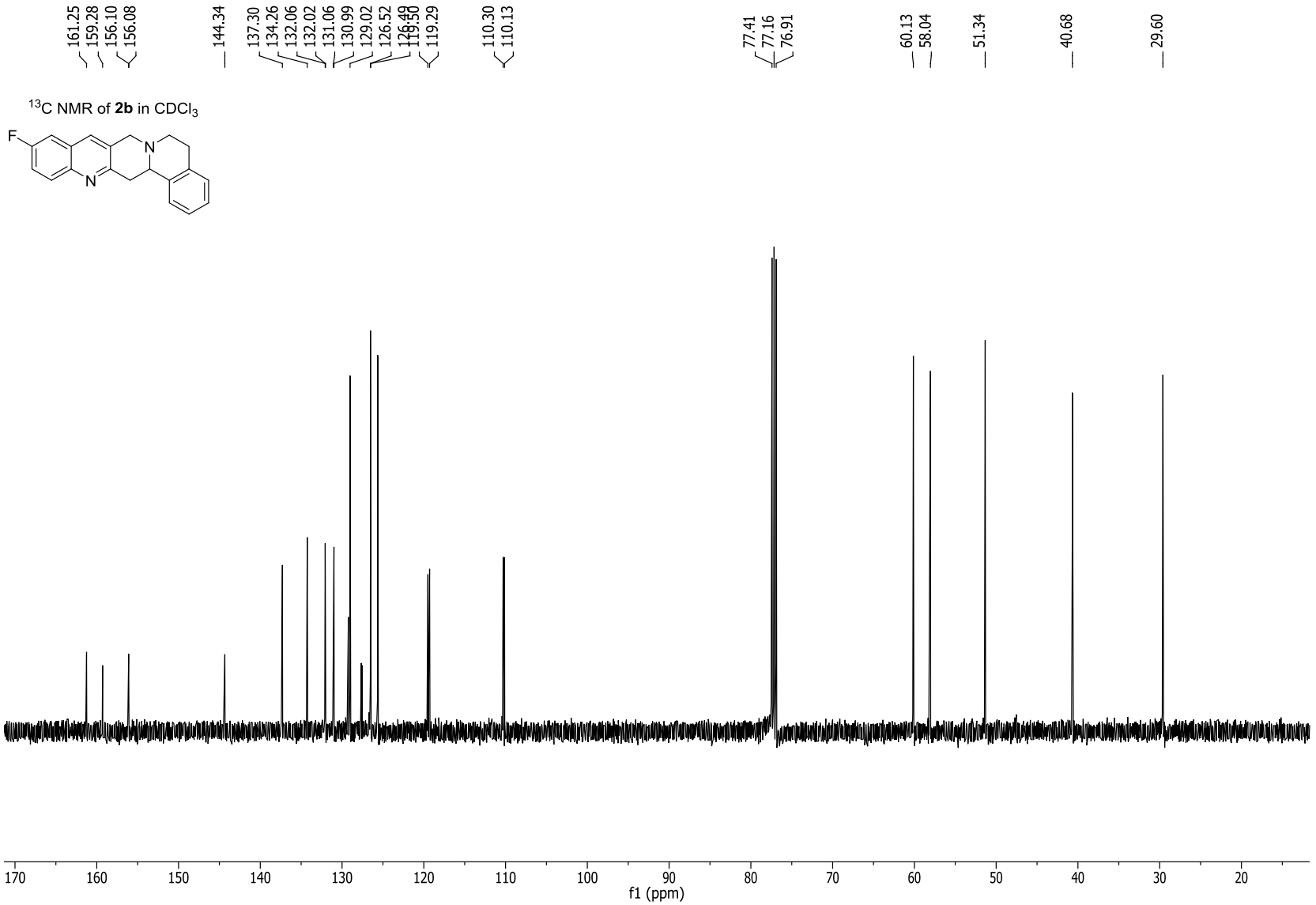

10.34
10.34
8.73
8.72
8.72
8.71
8.12
8.12
8.11
8.10
7.33
7.33
7.32
7.32
7.31
7.31
7.31
7.30
7.26
3.25
3.24
3.22
3.21
1.35
1.34
1.32
^{1}H NMR of **1p** in $CDCl_3$
O
N
Et
0.95
0.98
0.98
1.00
2.06
3.31
11.5
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

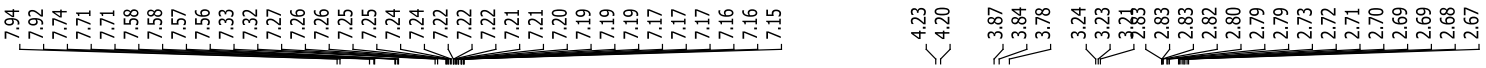

191.10
165.58
153.54
138.21
129.05
121.90
77.41
77.16
76.91
28.32
14.62
^{13}C NMR of **1p** in $CDCl_3$
O
N
Et
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


8.03 8.01 7.85 7.76 7.75 7.74 7.74 7.67 7.67 7.66 7.65 7.65 7.64 7.64 7.49 7.49 7.48 7.47 7.47 7.46 7.46 7.36 7.34 7.27 7.27 7.26 7.26 7.24 7.24 7.22 7.22 7.21 7.21 7.19 7.19 7.18 7.17 7.16 7.16 4.26 4.23 3.90 3.89 3.87 3.87 3.86 3.85 3.84 3.82 3.81 3.29 3.28 3.27 3.27 3.26 3.25 3.24 3.23 3.23 3.22 2.84 2.83 2.81 2.80 2.73 2.72 2.71 2.70 2.68
^{1}H NMR of **2a** in $CDCl_3$
N
N
0.98 0.97 0.98 1.00 1.00 0.99 1.19 1.91
1.03 2.96 2.92 1.06 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 0.0
f1 (ppm)

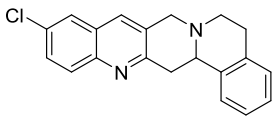

156.69
147.24
137.42
134.27
132.65
129.11
128.98
128.59
128.30
127.33
127.11
126.45
126.03
125.65
77.41
77.16
76.91
60.16
58.11
51.36
40.84
29.61
13C NMR of 2a in CDCl3
N
N
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

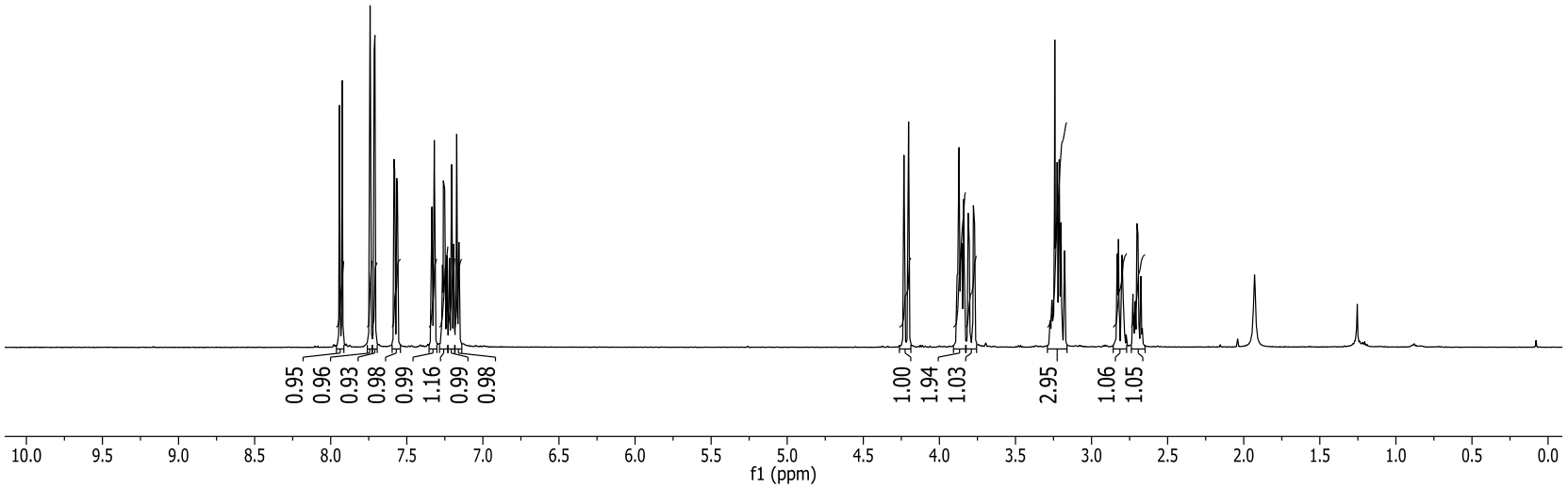

8.01 8.00 7.99 7.98 7.80 7.44 7.44 7.43 7.42 7.42 7.42 7.41 7.40 7.37 7.36 7.35 7.35 7.33 7.33 7.26 7.26 7.26 7.26 7.25 7.24 7.24 7.21 7.21 7.21 7.21 7.20 7.20 7.19 7.18 7.18 7.18 7.16 7.16 4.25 4.25 4.22 4.22 3.89 3.89 3.87 3.86 3.86 3.86 3.82 3.81 3.78 3.78 3.25 3.24 3.24 3.24 3.23 3.22 3.22 3.21 3.19 2.83 2.81 2.71 2.71 2.71
^{1}H NMR of **2b** in $CDCl_3$
F
N
N
0.94 0.93 0.99 1.92 1.29 0.98 0.96
1.00 1.93 1.01 2.96 1.04 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)

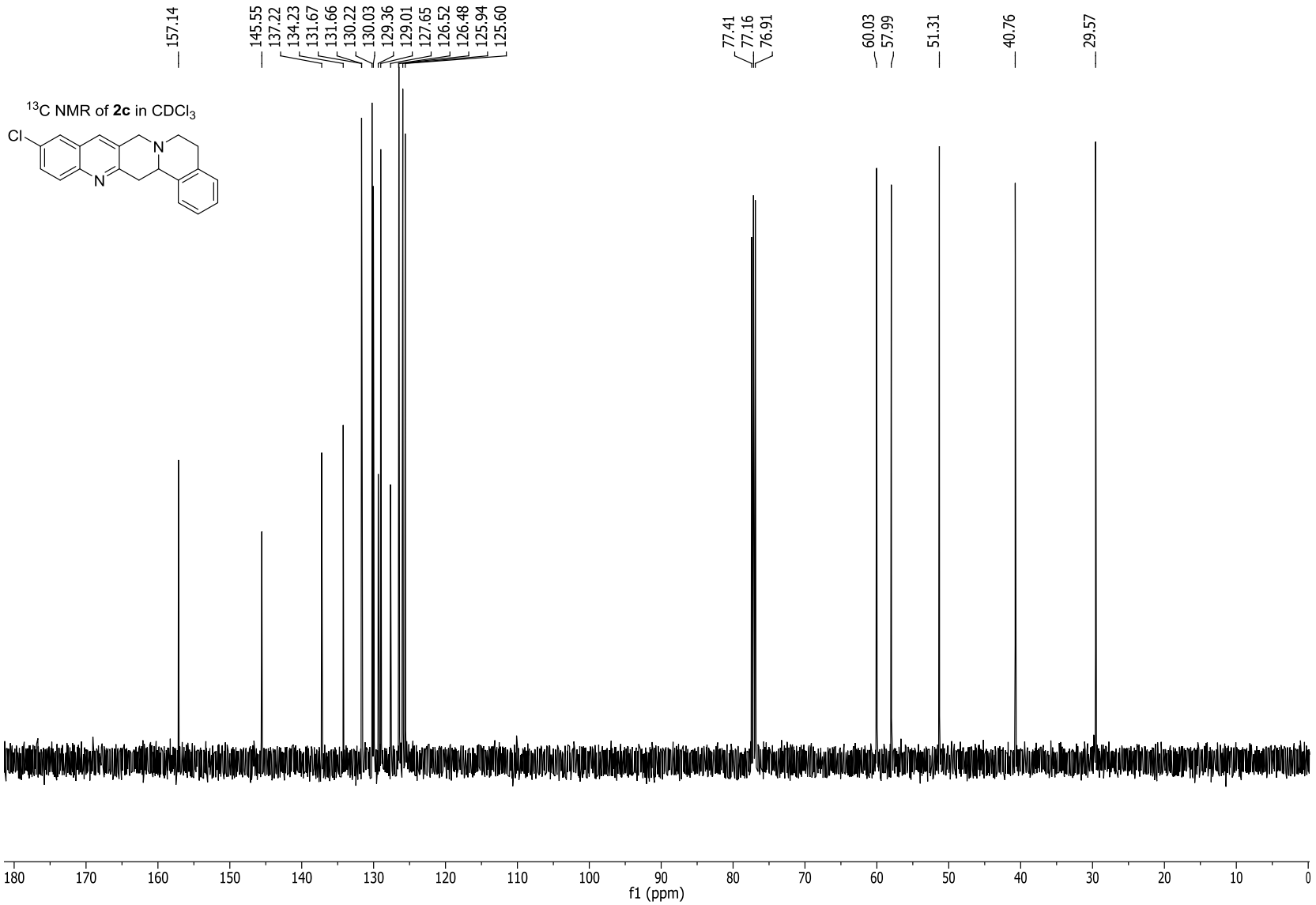

161.25
159.28
156.10
156.08
144.34
137.30
134.26
132.06
132.02
131.06
130.99
129.02
126.52
126.49
119.50
119.29
110.30
110.13
77.41
77.16
76.91
60.13
58.04
51.34
40.68
29.60
13C NMR of 2b in CDCl3
F
N
N
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
f1 (ppm)

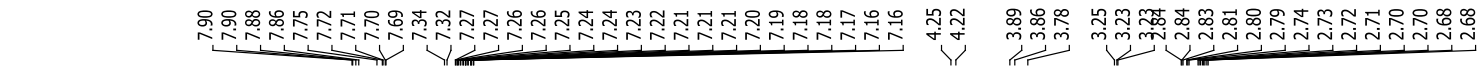


^{1}H NMR of **2c** in $CDCl_3$

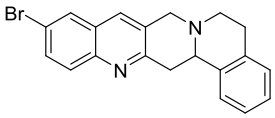


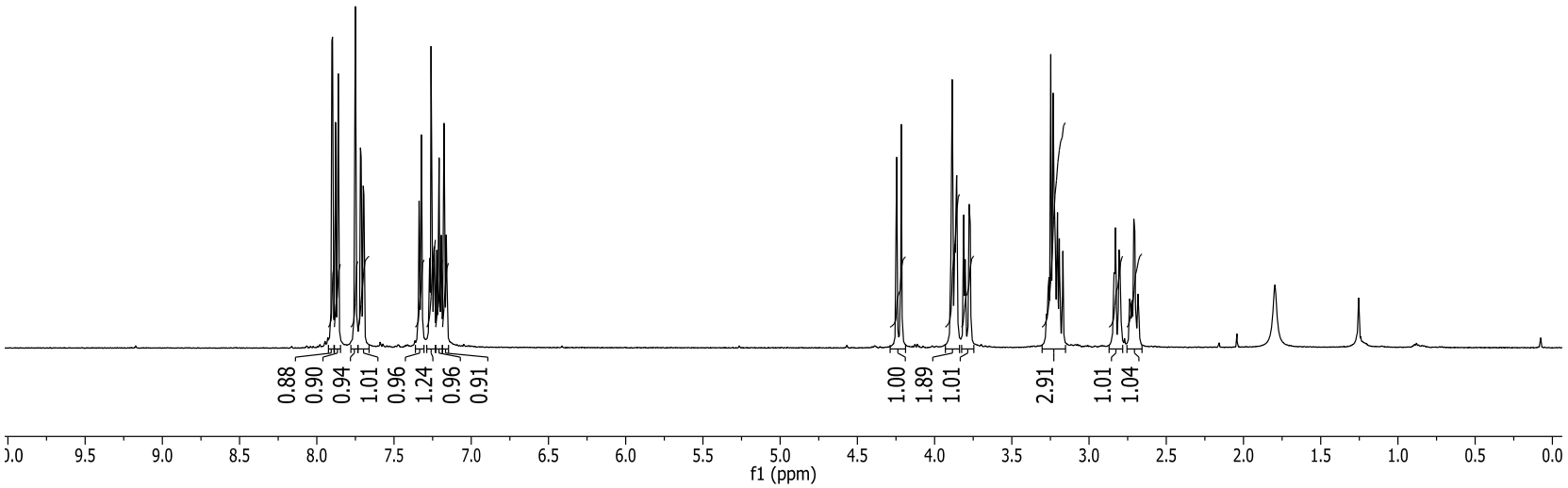


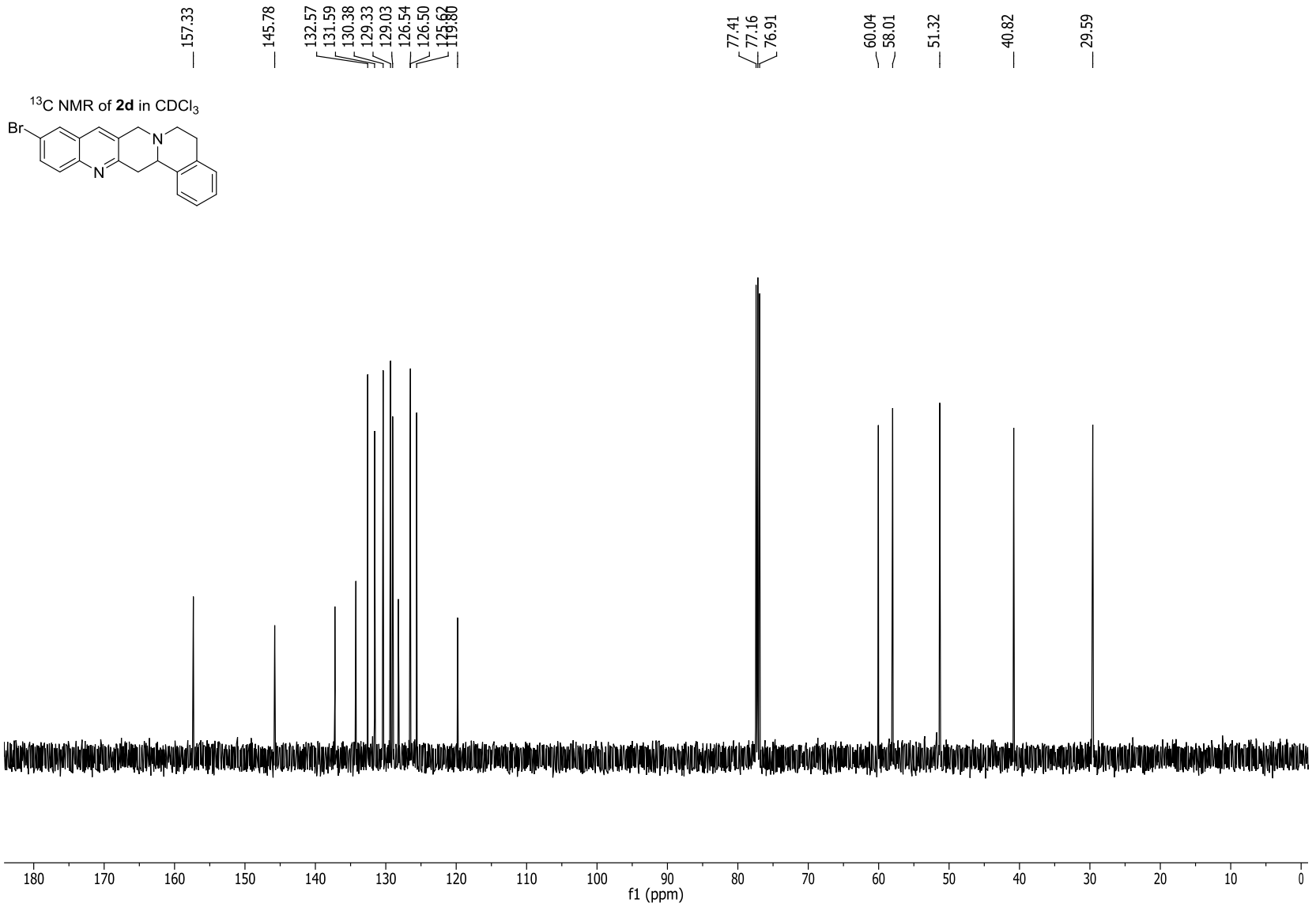

157.14
145.55
137.22
134.23
131.67
131.66
130.22
130.03
129.36
129.01
127.65
126.52
126.48
125.94
125.60
77.41
77.16
76.91
60.03
57.99
51.31
40.76
29.57
13C NMR of 2c in CDCl3
Cl
N
N
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

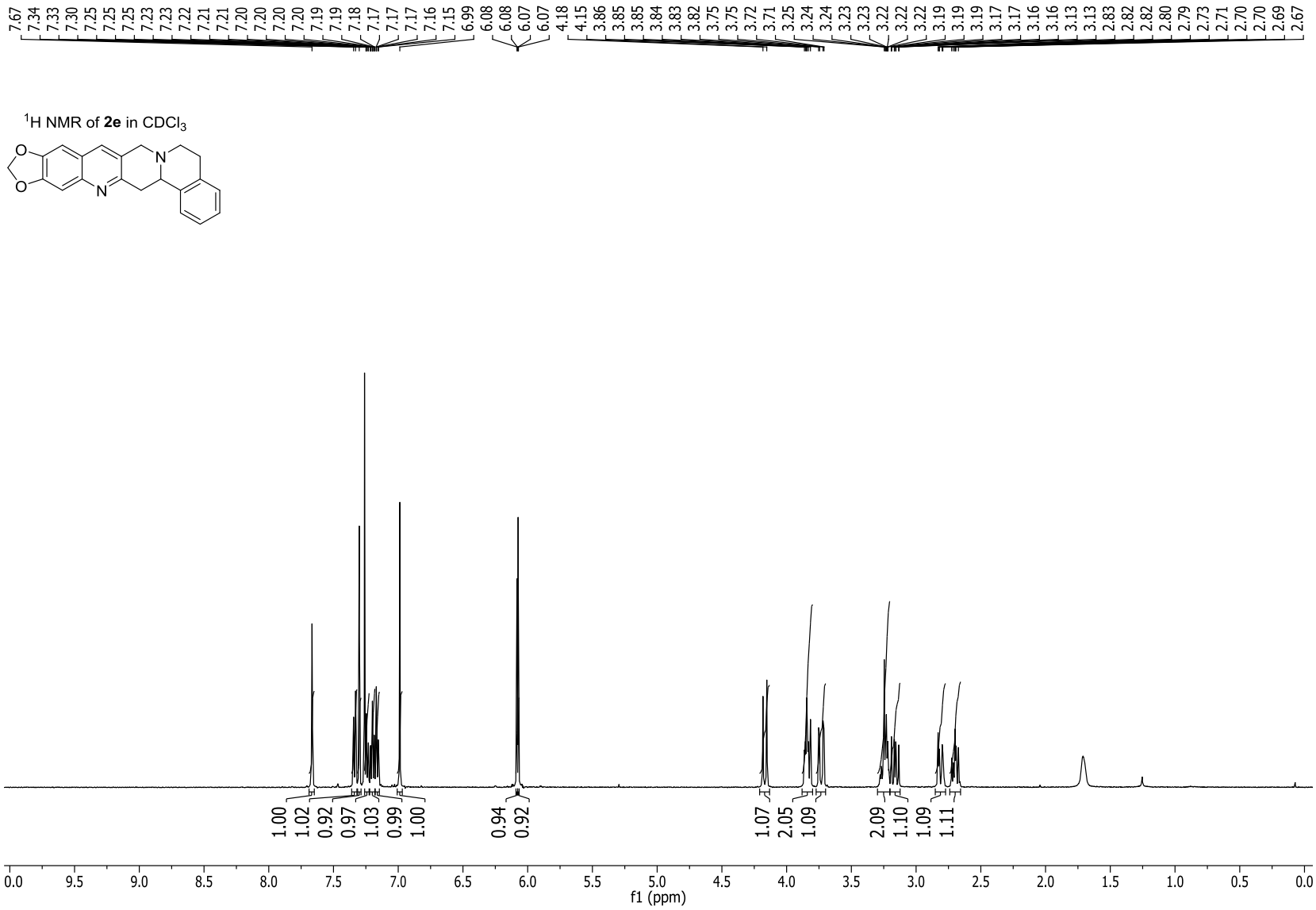


^{1}H NMR of **2d** in $CDCl_3$



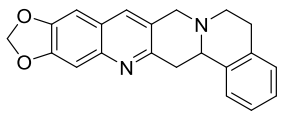


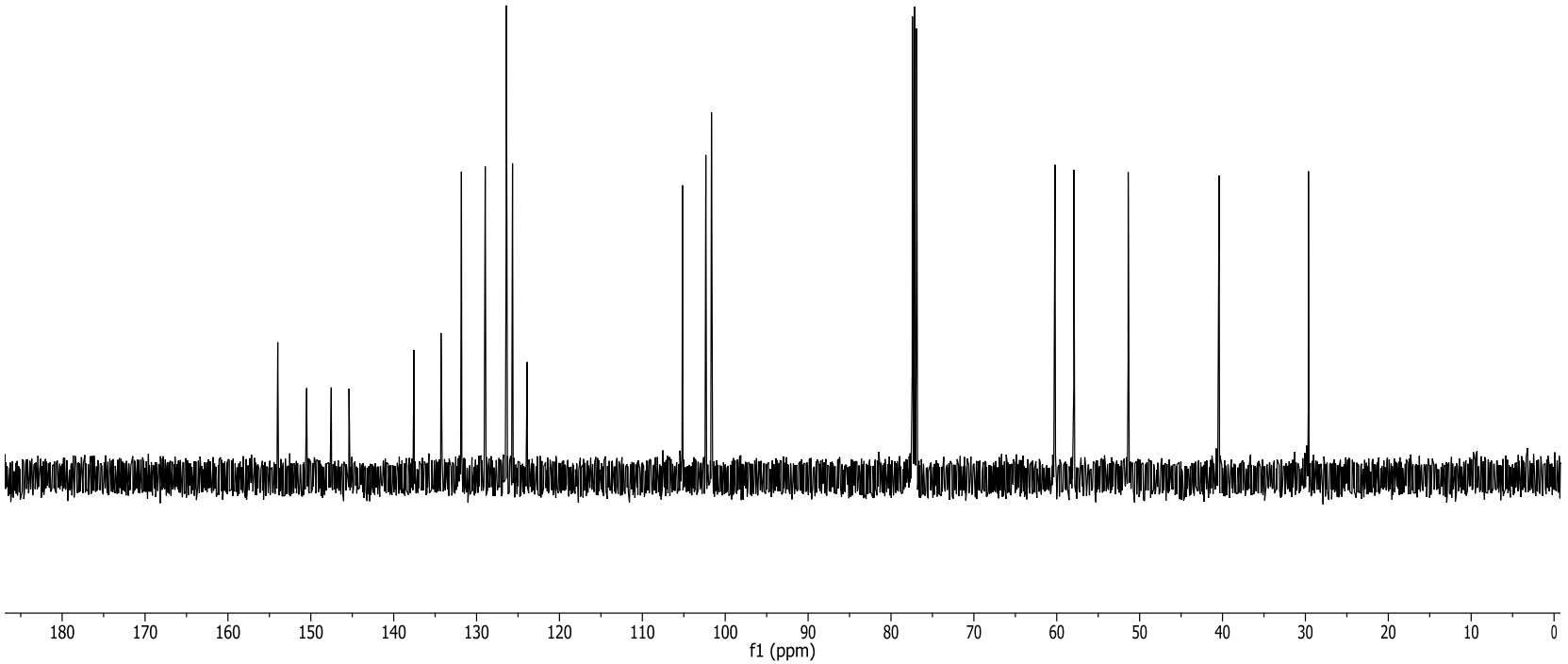

157.33
145.78
132.57
131.59
130.38
129.33
129.03
126.54
126.50
77.41
77.16
76.91
60.04
58.01
51.32
40.82
29.59
^{13}C NMR of **2d** in $CDCl_3$
Br
N
N
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


7.67
7.34
7.33
7.30
7.25
7.25
7.25
7.23
7.23
7.22
7.21
7.21
7.20
7.20
7.20
7.20
7.19
7.19
7.18
7.17
7.17
7.17
7.16
7.15
6.99
6.08
6.08
6.07
6.07
4.18
4.15
3.86
3.85
3.85
3.84
3.83
3.82
3.75
3.75
3.72
3.71
3.25
3.24
3.24
3.23
3.23
3.22
3.22
3.22
3.19
3.19
3.19
3.17
3.17
3.16
3.16
3.13
3.13
2.83
2.82
2.82
2.80
2.79
2.73
2.71
2.70
2.70
2.69
2.67
1H NMR of 2e in CDCl3
O
O
N
N
1.00
1.02
0.92
0.97
1.03
0.99
1.00
0.94
0.92
1.07
2.05
1.09
2.09
1.10
1.09
1.11
0.0
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)

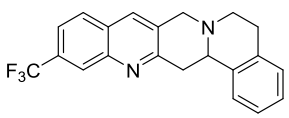
153.99
150.50
147.53
145.39
137.55
134.29
131.83
128.96
126.43
126.40
125.66
123.90
105.16
102.34
101.66
77.42
77.16
76.91
60.21
57.93
51.38
40.41
29.63

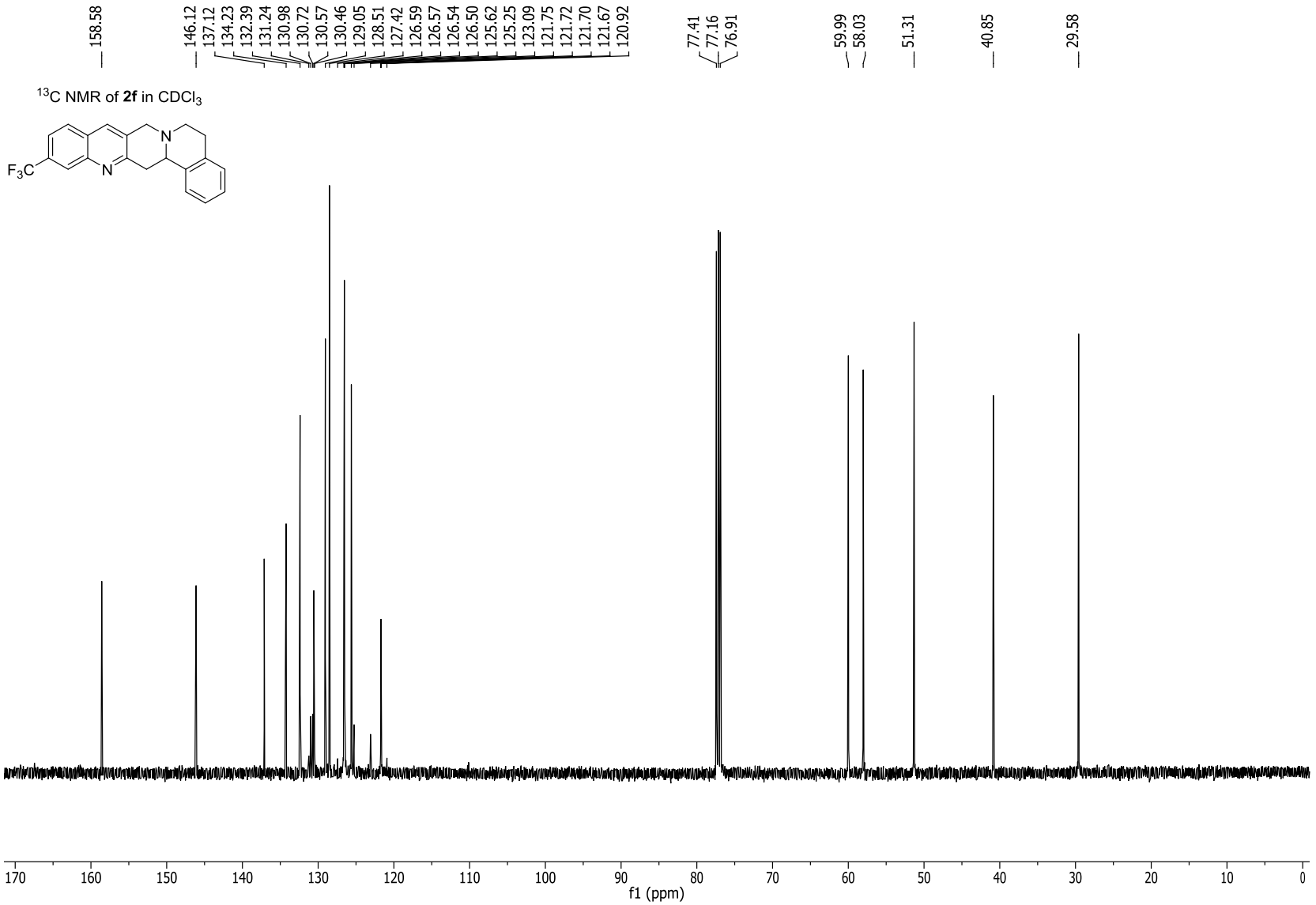
^{13}C NMR of **2e** in $CDCl_3$

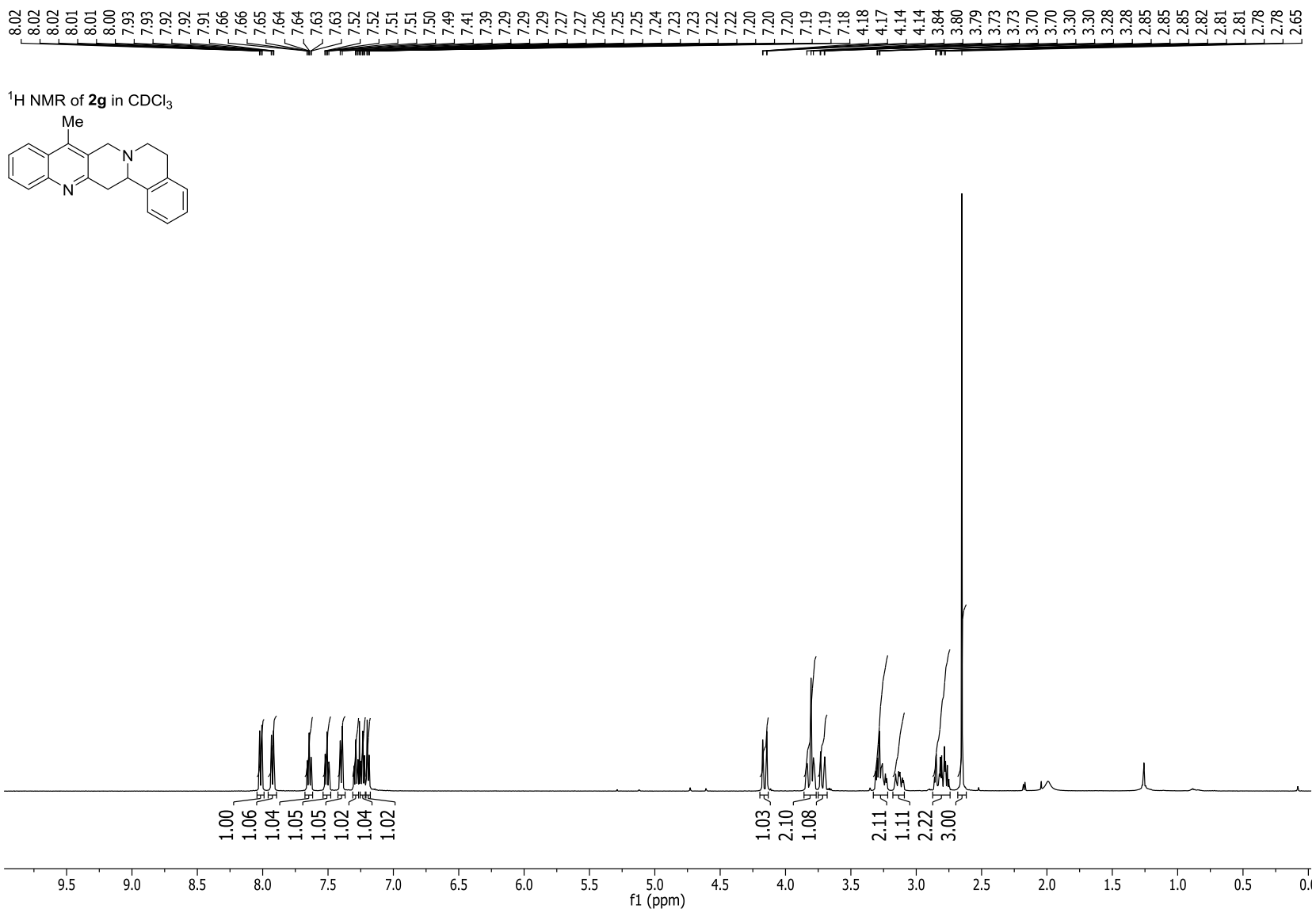


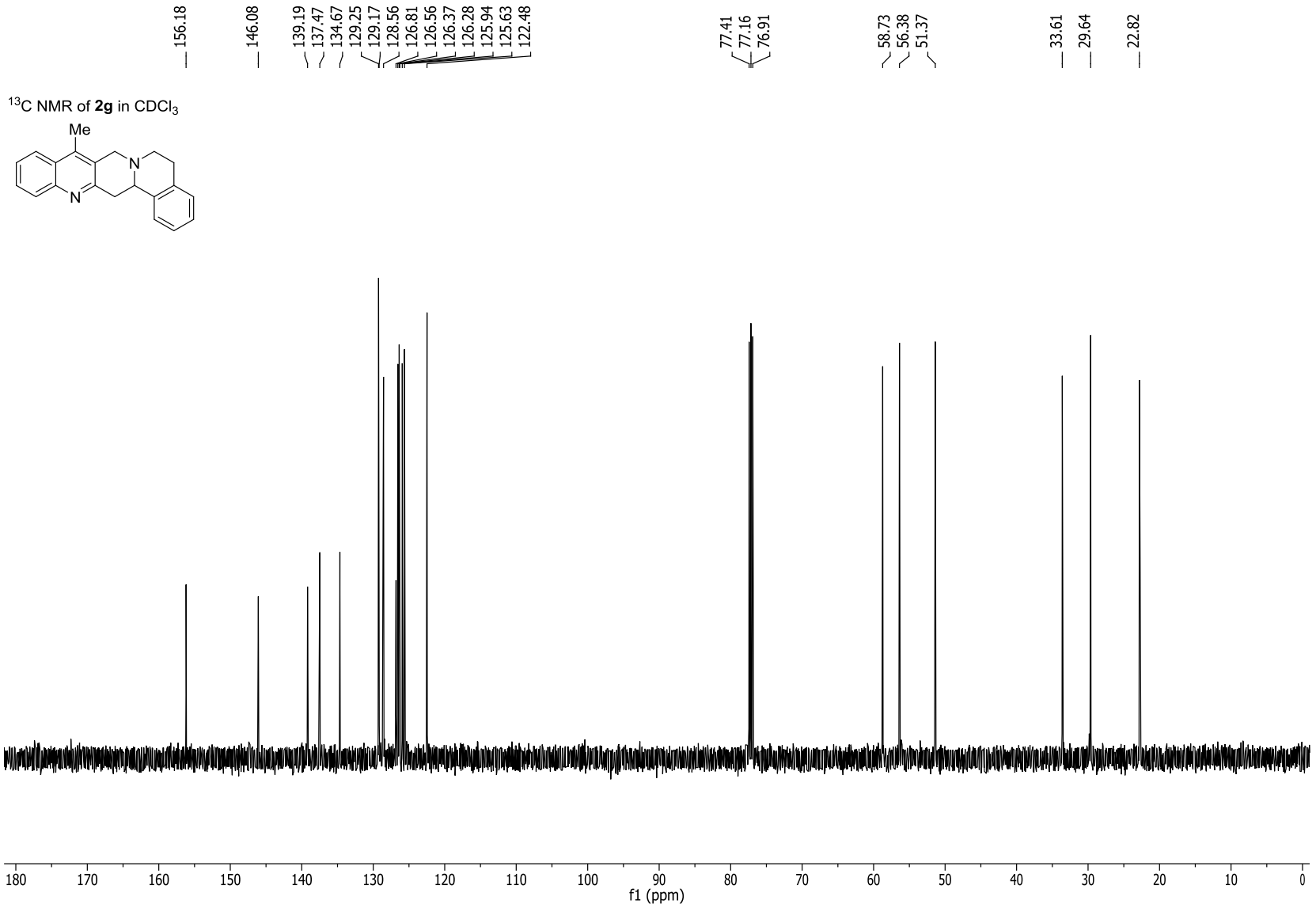


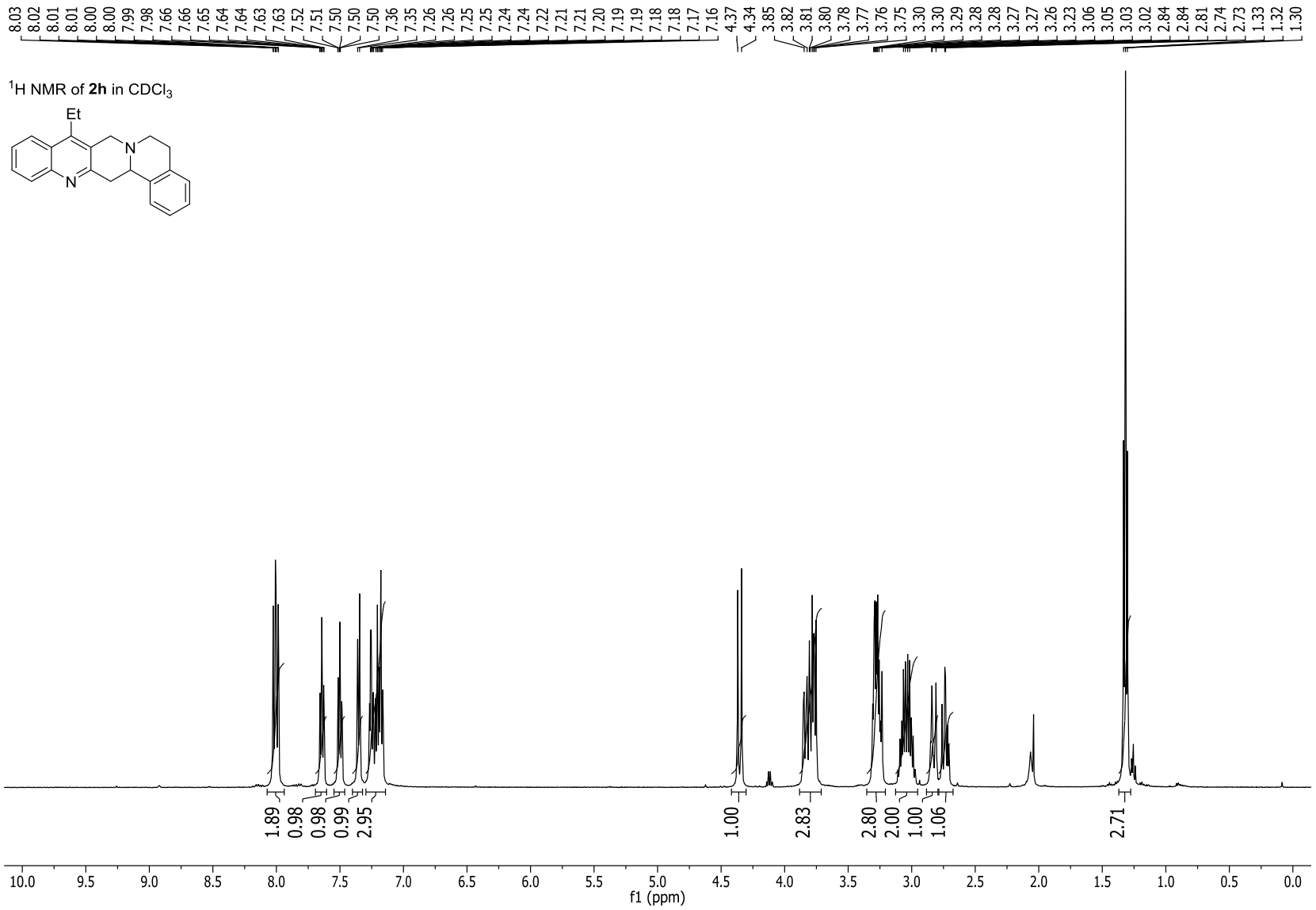
^{1}H NMR of **2f** in $CDCl_3$

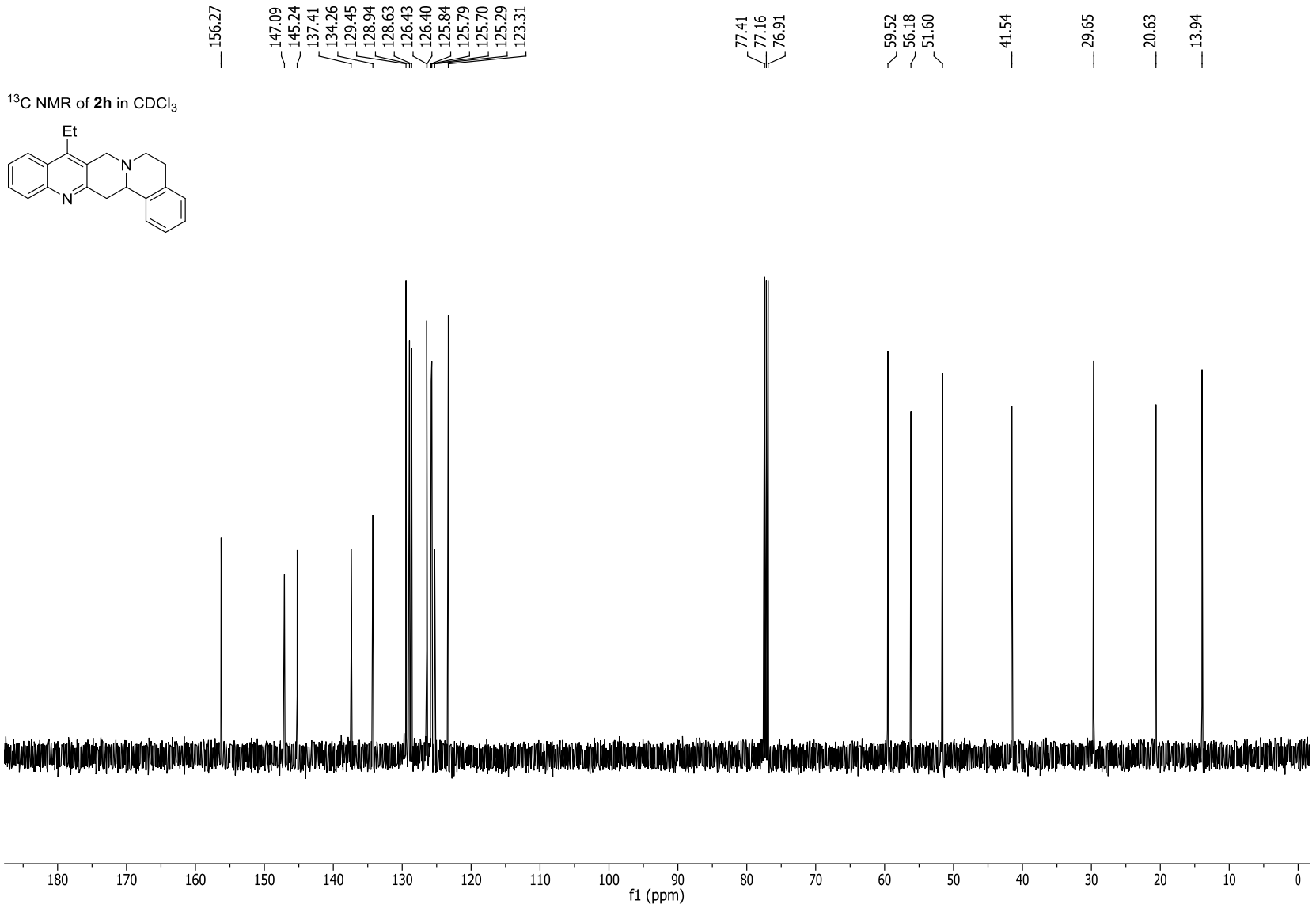


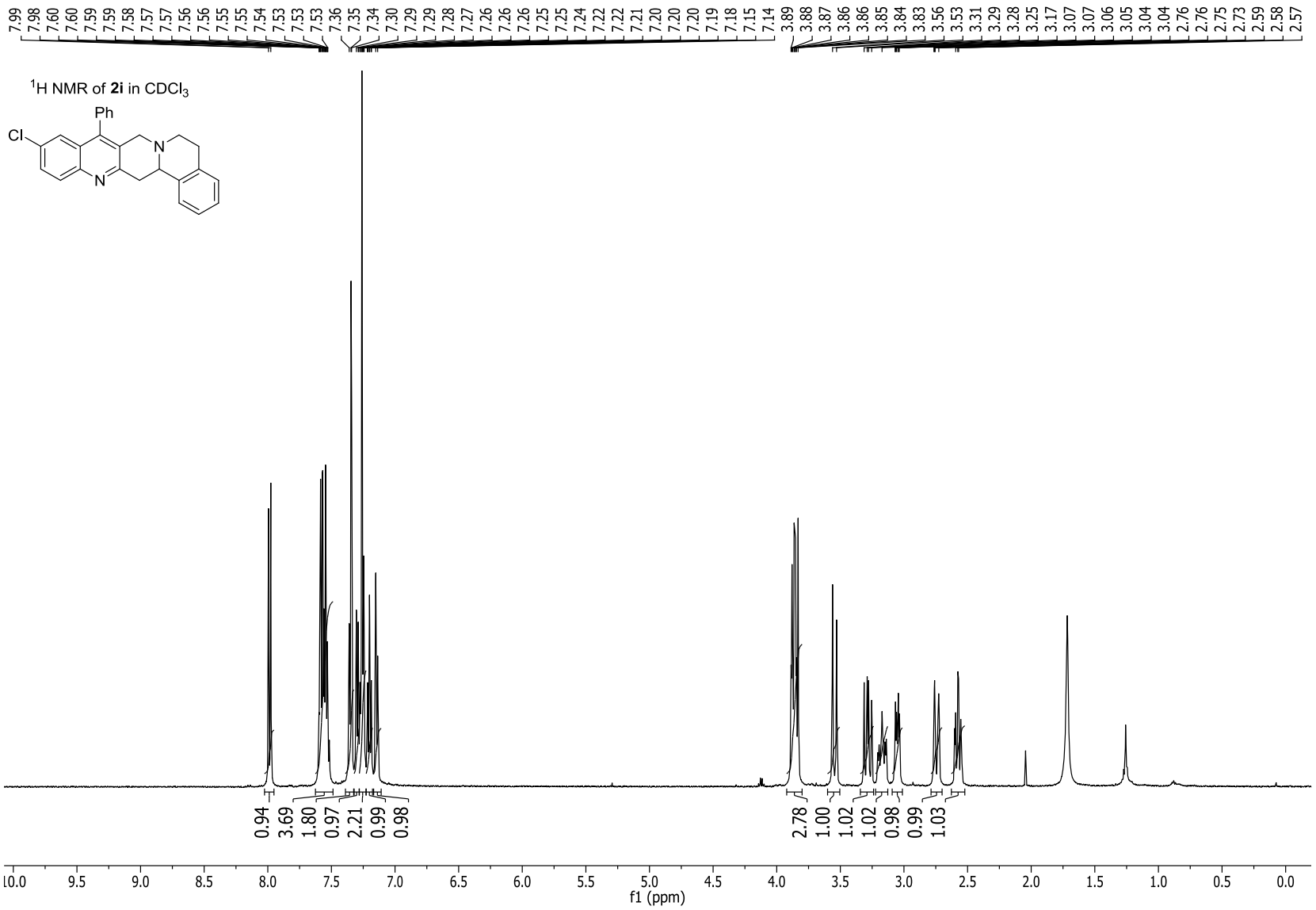

158.58
146.12
137.12
134.23
132.39
131.24
130.98
130.72
130.57
130.46
129.05
128.51
127.42
126.59
126.57
126.54
126.50
125.62
125.25
123.09
121.75
121.72
121.70
121.67
120.92
77.41
77.16
76.91
59.99
58.03
51.31
40.85
29.58
^{13}C NMR of **2f** in $CDCl_3$
F_3C
N
N
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

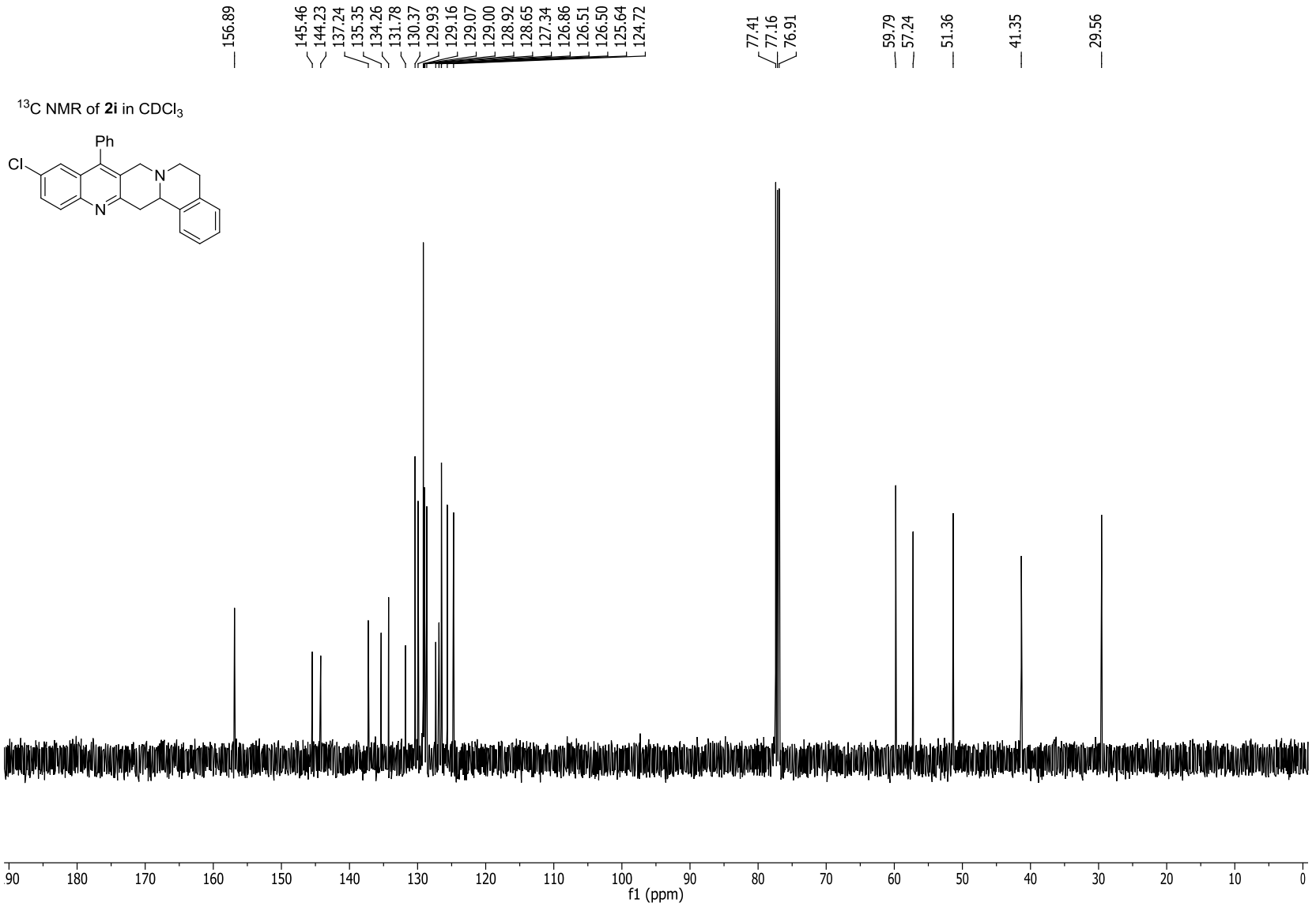

8.02 8.02 8.02 8.01 8.01 8.00 7.93 7.93 7.92 7.92 7.91 7.66 7.66 7.65 7.64 7.64 7.63 7.63 7.52 7.52 7.51 7.51 7.50 7.49 7.41 7.39 7.29 7.29 7.29 7.27 7.27 7.26 7.25 7.25 7.24 7.23 7.23 7.22 7.22 7.20 7.20 7.19 7.19 7.18 4.18 4.17 4.14 4.14 3.84 3.80 3.79 3.73 3.73 3.70 3.70 3.30 3.30 3.28 3.28 2.85 2.85 2.85 2.82 2.81 2.81 2.78 2.78 2.65
1H NMR of 2g in CDCl3
Me
N
N
1.00 1.06 1.04 1.05 1.05 1.02 1.04 1.02
1.03 2.10 1.08 2.11 1.11 2.22 3.00
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
f1 (ppm)

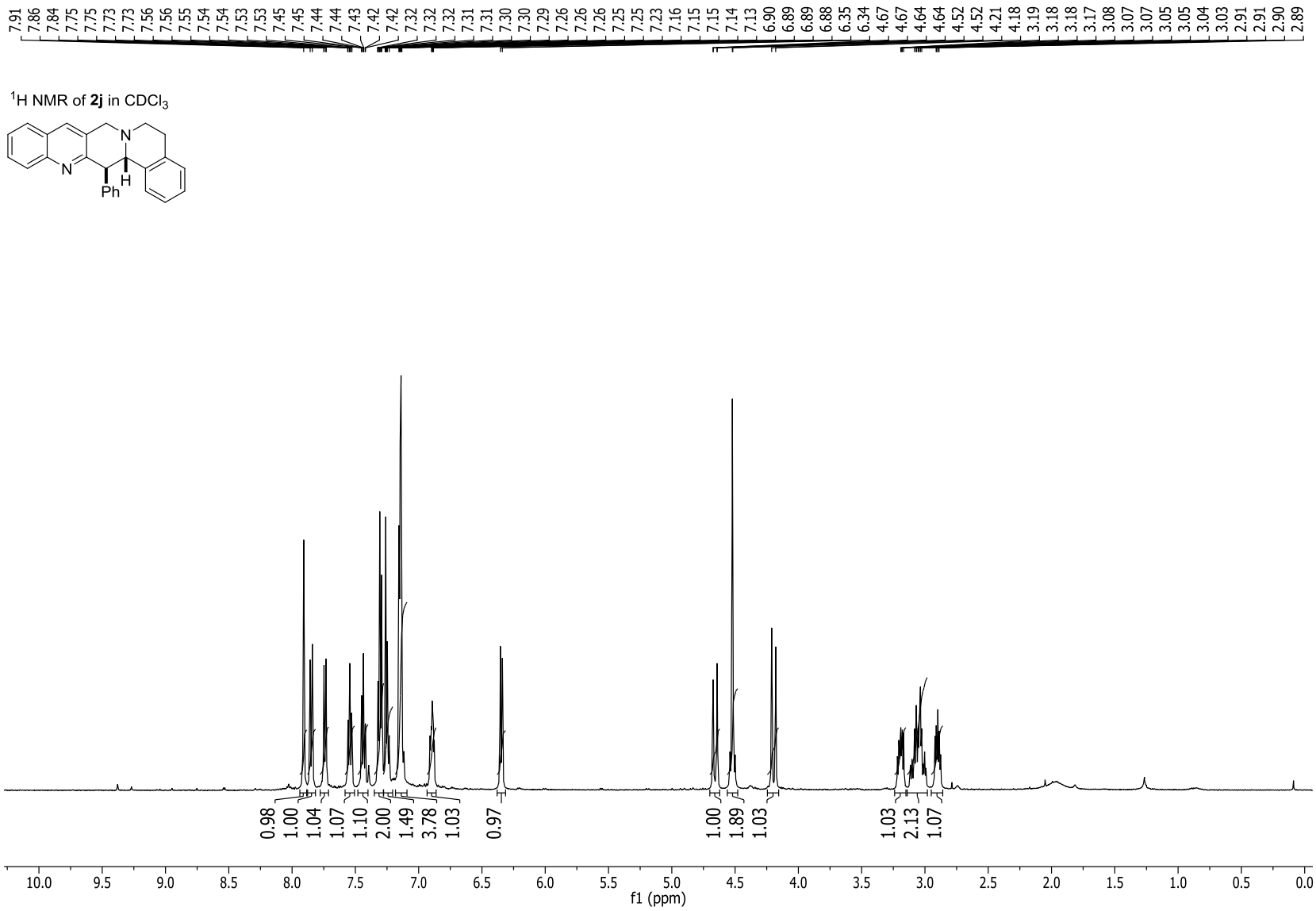

156.18
146.08
139.19
137.47
134.67
129.25
129.17
128.56
126.81
126.56
126.37
126.28
125.94
125.63
122.48
77.41
77.16
76.91
58.73
56.38
51.37
33.61
29.64
22.82
13C NMR of 2g in CDCl3
Me
N
N
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

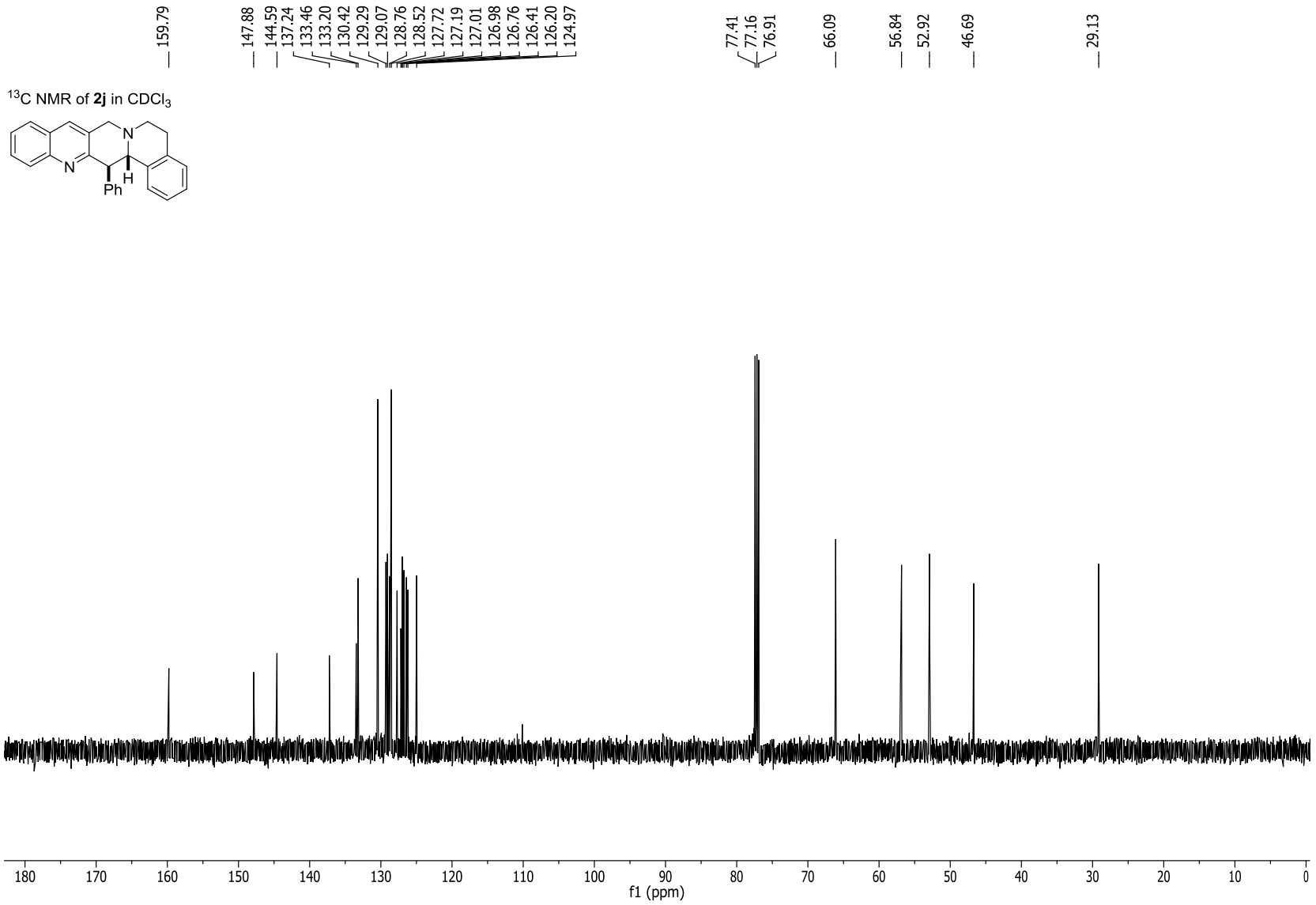

8.03 8.02 8.01 8.01 8.00 8.00 7.99 7.98 7.66 7.66 7.65 7.64 7.64 7.63 7.63 7.52 7.51 7.50 7.50 7.50 7.36 7.35 7.26 7.26 7.25 7.25 7.24 7.24 7.22 7.21 7.21 7.20 7.19 7.19 7.18 7.18 7.17 7.16 4.37 4.34 3.85 3.82 3.81 3.80 3.78 3.77 3.76 3.75 3.30 3.30 3.29 3.28 3.28 3.27 3.27 3.26 3.23 3.06 3.05 3.03 3.02 2.84 2.84 2.81 2.74 2.73 1.33 1.32 1.30
1H NMR of 2h in CDCl3
Et
N
N
1.89 0.98 0.98 0.99 2.95 1.00 2.83 2.80 2.00 1.00 1.06 2.71
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)


156.27
147.09
145.24
137.41
134.26
129.45
128.94
128.63
126.43
126.40
125.84
125.79
125.70
125.29
123.31
77.41
77.16
76.91
59.52
56.18
51.60
41.54
29.65
20.63
13.94
^{13}C NMR of **2h** in $CDCl_3$
Et
N
N
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

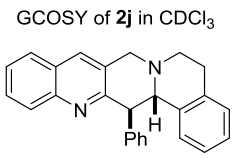

7.99 7.98 7.60 7.60 7.59 7.59 7.58 7.57 7.57 7.56 7.56 7.55 7.55 7.54 7.53 7.53 7.53 7.36 7.35 7.34 7.30 7.29 7.29 7.28 7.27 7.26 7.26 7.25 7.25 7.24 7.22 7.22 7.21 7.20 7.20 7.19 7.18 7.15 7.14
3.89 3.88 3.87 3.86 3.86 3.85 3.84 3.83 3.56 3.53 3.31 3.29 3.28 3.25 3.17 3.07 3.07 3.06 3.05 3.04 3.04 2.76 2.76 2.75 2.73 2.59 2.58 2.57
^{1}H NMR of **2i** in $CDCl_3$
Ph
Cl
N
N
0.94 3.69 1.80 0.97 2.21 0.99 0.98
2.78 1.00 1.02 1.02 0.98 0.99 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)

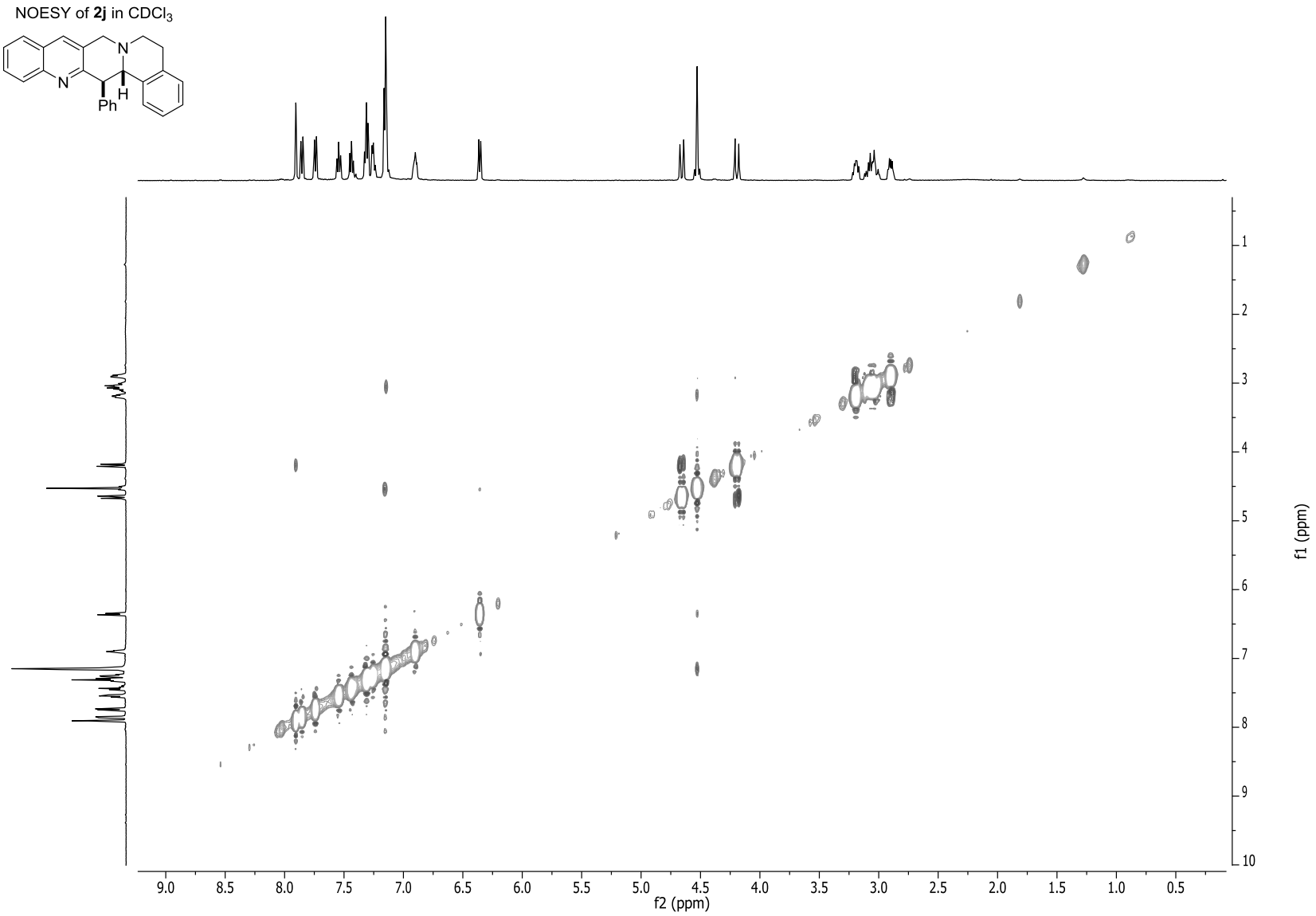

156.89
145.46
144.23
137.24
135.35
134.26
131.78
130.37
129.93
129.16
129.07
129.00
128.92
128.65
127.34
126.86
126.51
126.50
125.64
124.72
77.41
77.16
76.91
59.79
57.24
51.36
41.35
29.56
13C NMR of 2i in CDCl3
Ph
Cl
N
N
f1 (ppm)

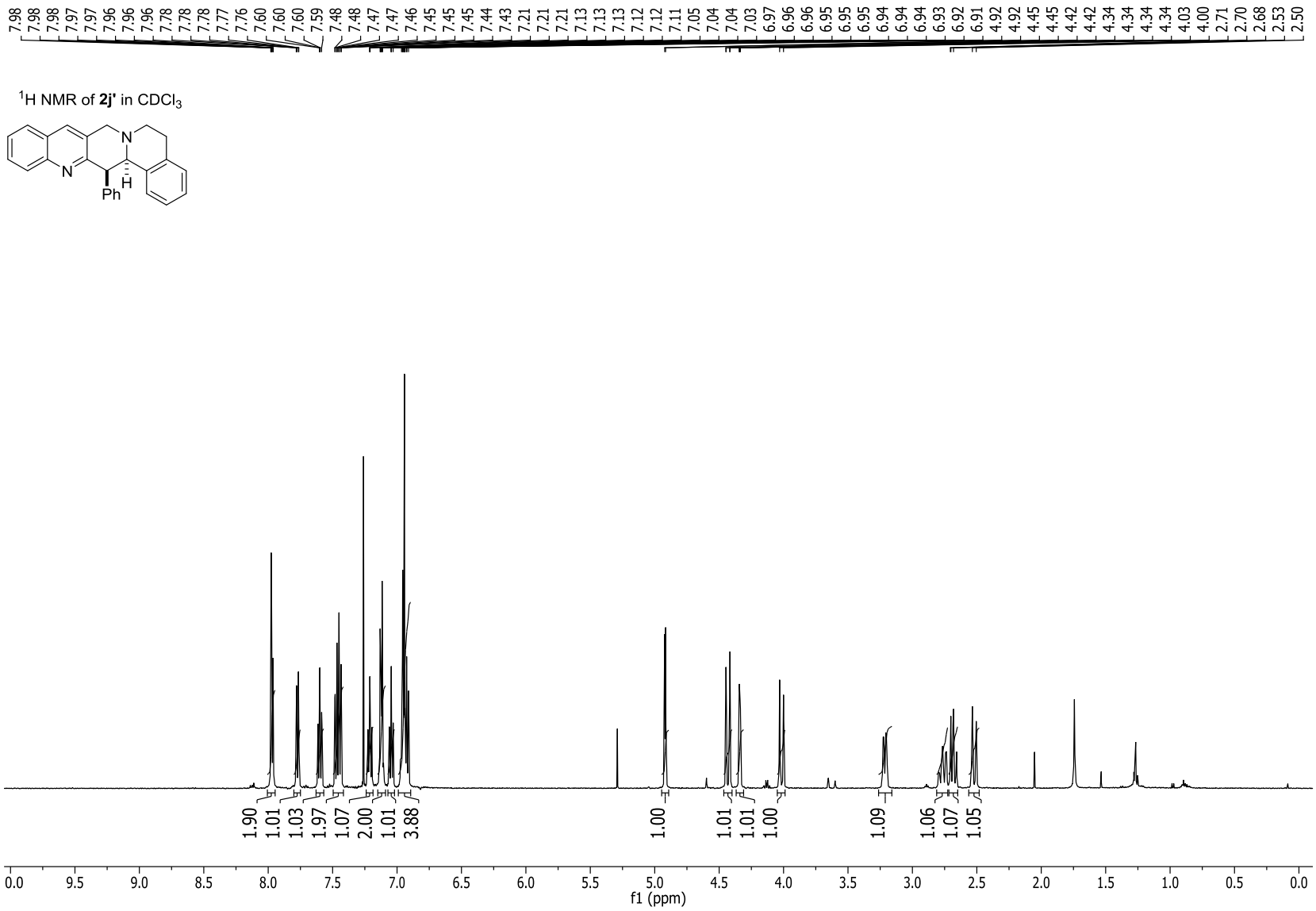

7.91 7.86 7.84 7.75 7.75 7.73 7.73 7.56 7.56 7.55 7.54 7.54 7.53 7.53 7.45 7.45 7.44 7.44 7.43 7.42 7.42 7.32 7.32 7.31 7.31 7.30 7.30 7.29 7.26 7.26 7.26 7.25 7.25 7.23 7.16 7.15 7.15 7.14 7.13 6.90 6.89 6.89 6.88 6.35 6.34 4.67 4.67 4.64 4.64 4.52 4.52 4.21 4.18 3.19 3.18 3.18 3.17 3.08 3.07 3.07 3.05 3.05 3.04 3.03 2.91 2.91 2.90 2.89
^{1}H NMR of **2j** in $CDCl_3$
N
N
H
Ph
0.98 1.00 1.04 1.07 1.10 2.00 1.49 3.78 1.03 0.97 1.00 1.89 1.03 1.03 2.13 1.07
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)

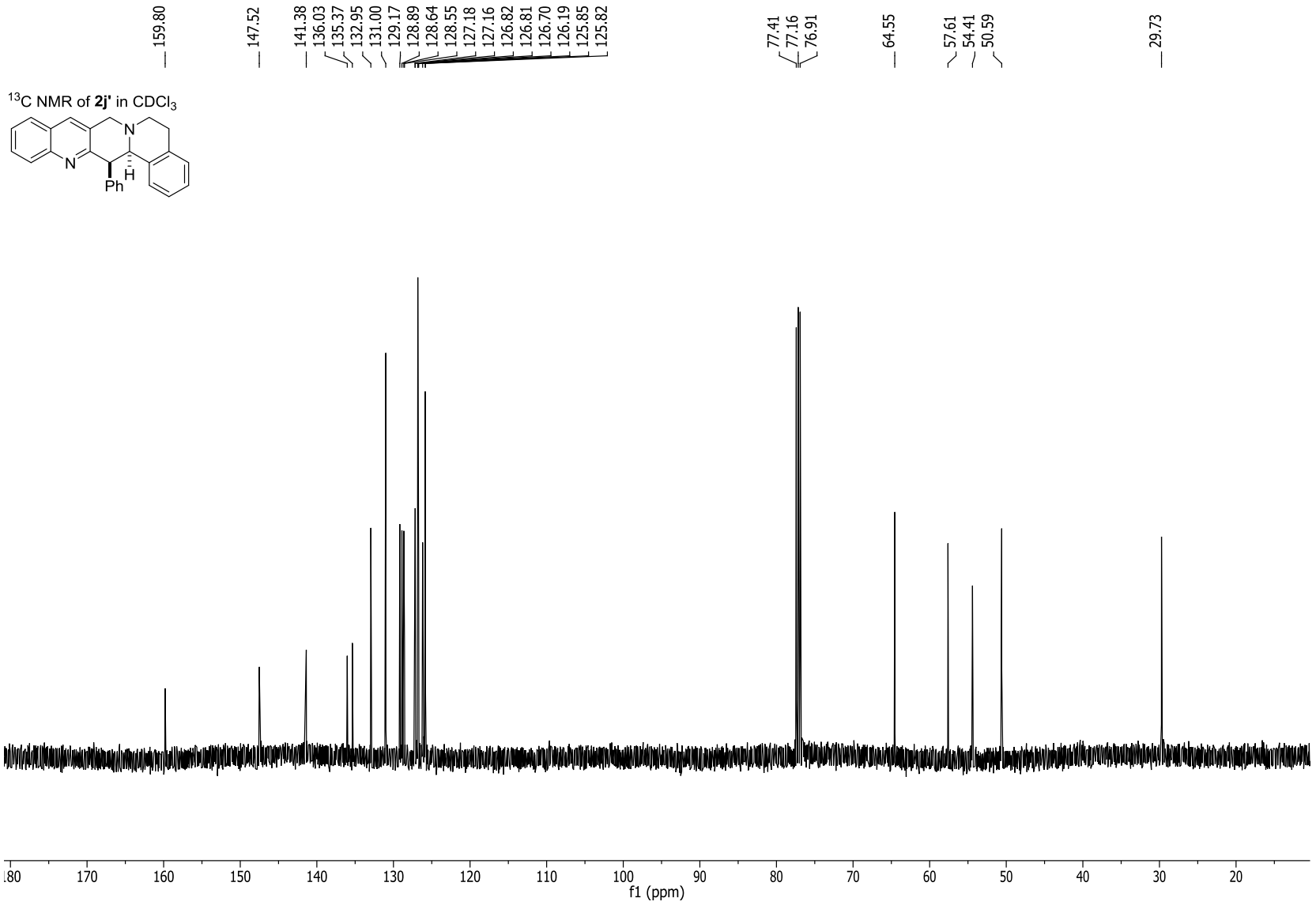

159.79
147.88
144.59
137.24
133.46
133.20
130.42
129.29
129.07
128.76
128.52
127.72
127.19
127.01
126.98
126.76
126.41
126.20
124.97
77.41
77.16
76.91
66.09
56.84
52.92
46.69
29.13
13C NMR of 2j in CDCl3
N
N
Ph
H
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

GCOSY of **2j** in $CDCl_3$

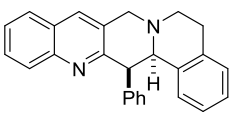



NOESY of **2j** in $CDCl_3$
N
N
H
Ph
f2 (ppm)
f1 (ppm)

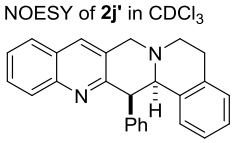

7.98 7.98 7.98 7.97 7.97 7.96 7.96 7.96 7.78 7.78 7.78 7.77 7.76 7.60 7.60 7.60 7.59 7.48 7.48 7.47 7.47 7.46 7.45 7.45 7.45 7.44 7.43 7.21 7.21 7.21 7.13 7.13 7.13 7.12 7.12 7.11 7.05 7.04 7.04 7.03 6.97 6.96 6.96 6.95 6.95 6.95 6.94 6.94 6.94 6.93 6.92 6.91 4.92 4.92 4.45 4.45 4.42 4.42 4.34 4.34 4.34 4.34 4.03 4.00 2.71 2.70 2.68 2.53 2.50
^{1}H NMR of **2j'** in $CDCl_3$
N
N
H
Ph
1.90 1.01 1.03 1.97 1.07 2.00 1.01 3.88 1.00 1.01 1.01 1.00 1.09 1.06 1.07 1.05
0.0 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)

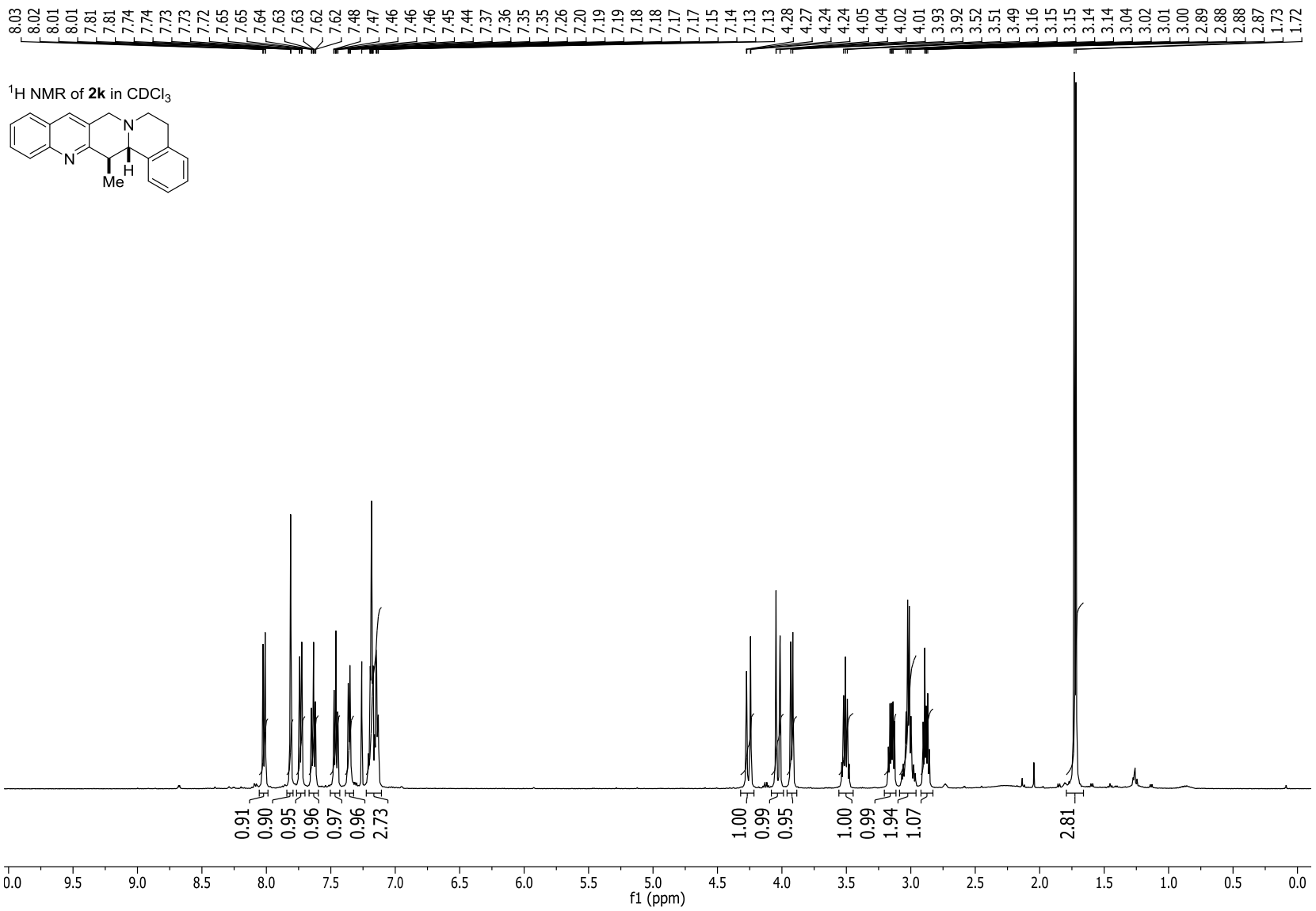

159.80
147.52
141.38
136.03
135.37
132.95
131.00
129.17
128.89
128.64
128.55
127.18
127.16
126.82
126.81
126.70
126.19
125.85
125.82
77.41
77.16
76.91
64.55
57.61
54.41
50.59
29.73
13C NMR of 2j' in CDCl3
N
N
H
Ph
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
f1 (ppm)

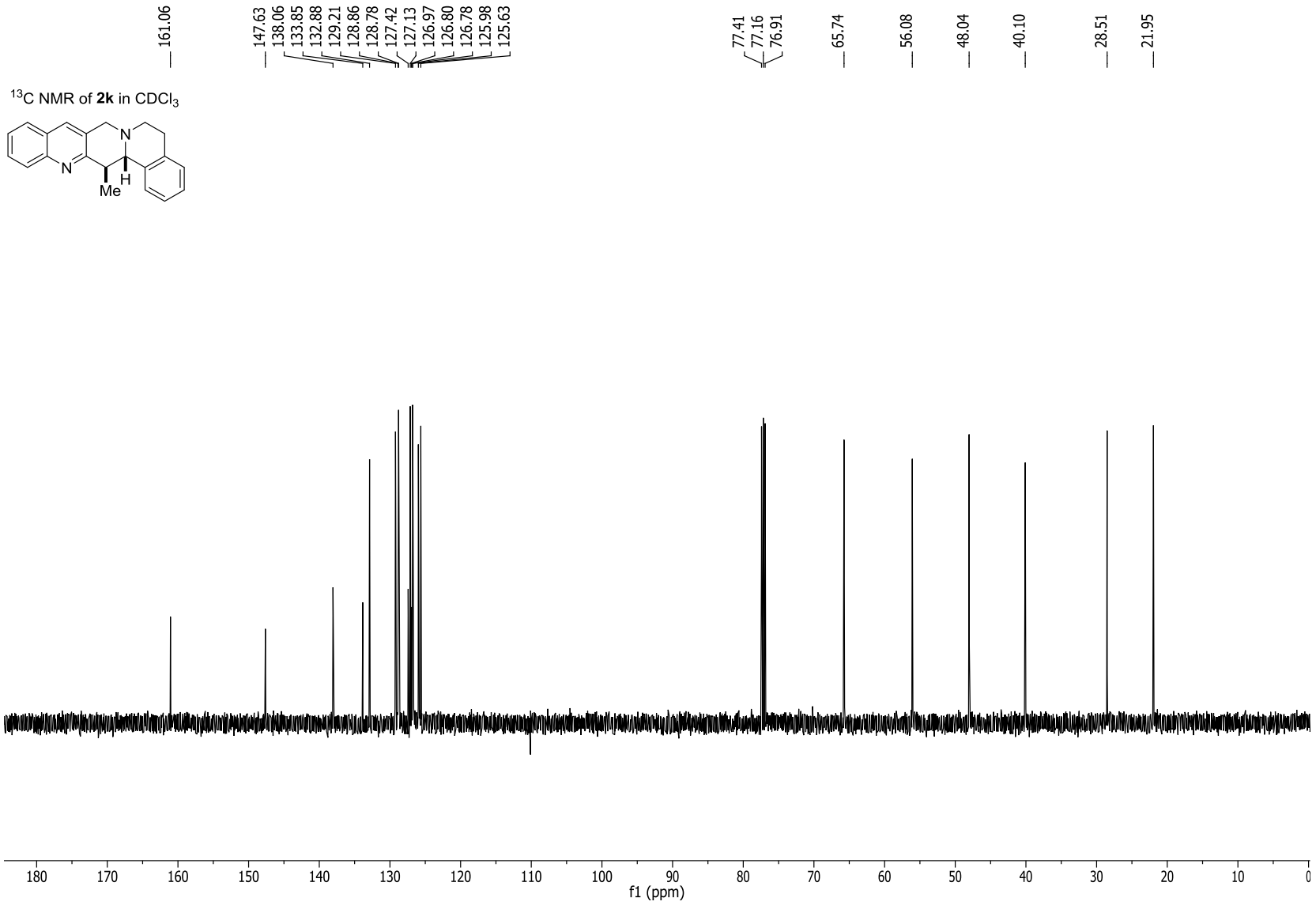
GCOSY of **2j'** in $CDCl_3$

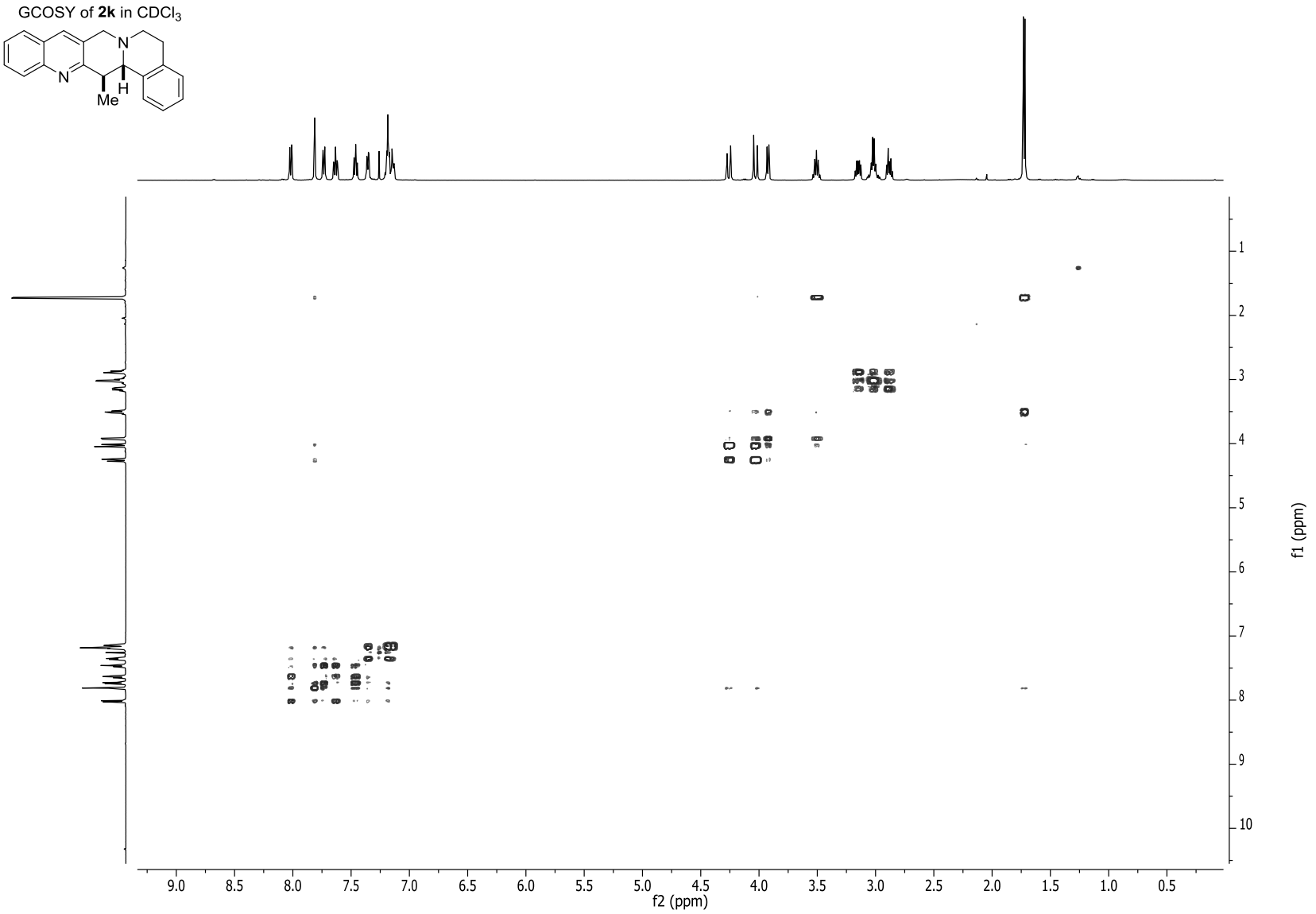


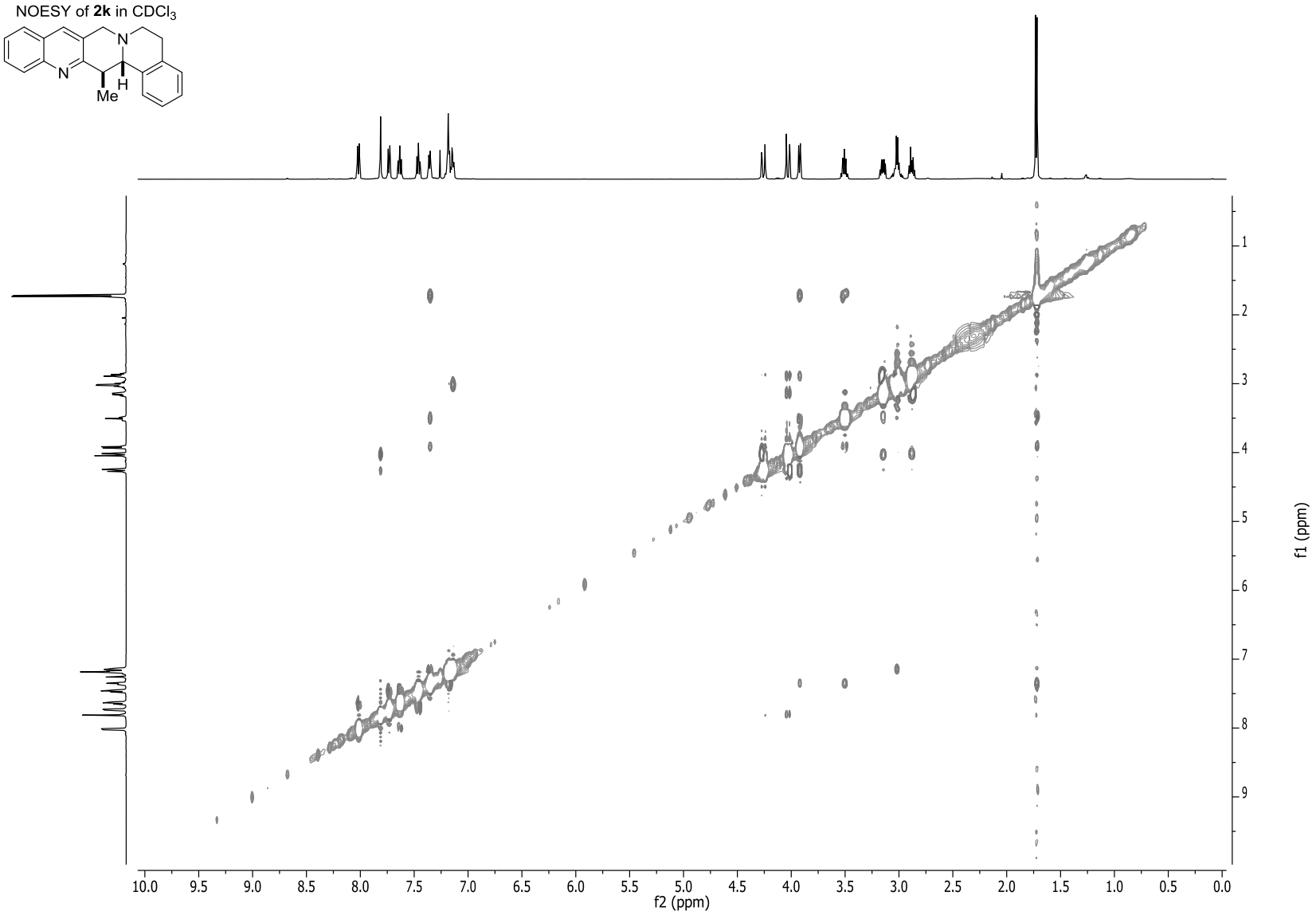
NOESY of **2j'** in $CDCl_3$

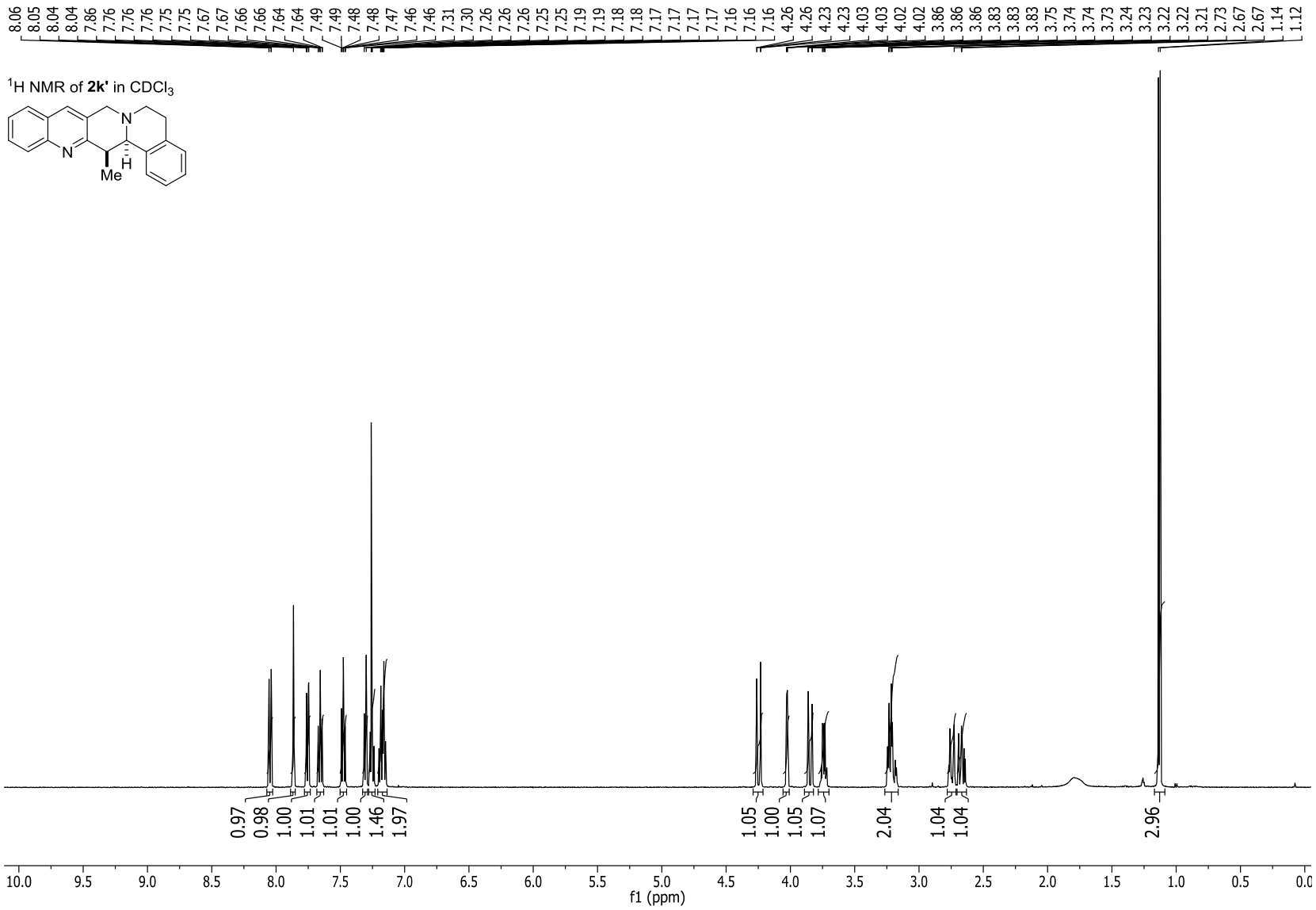


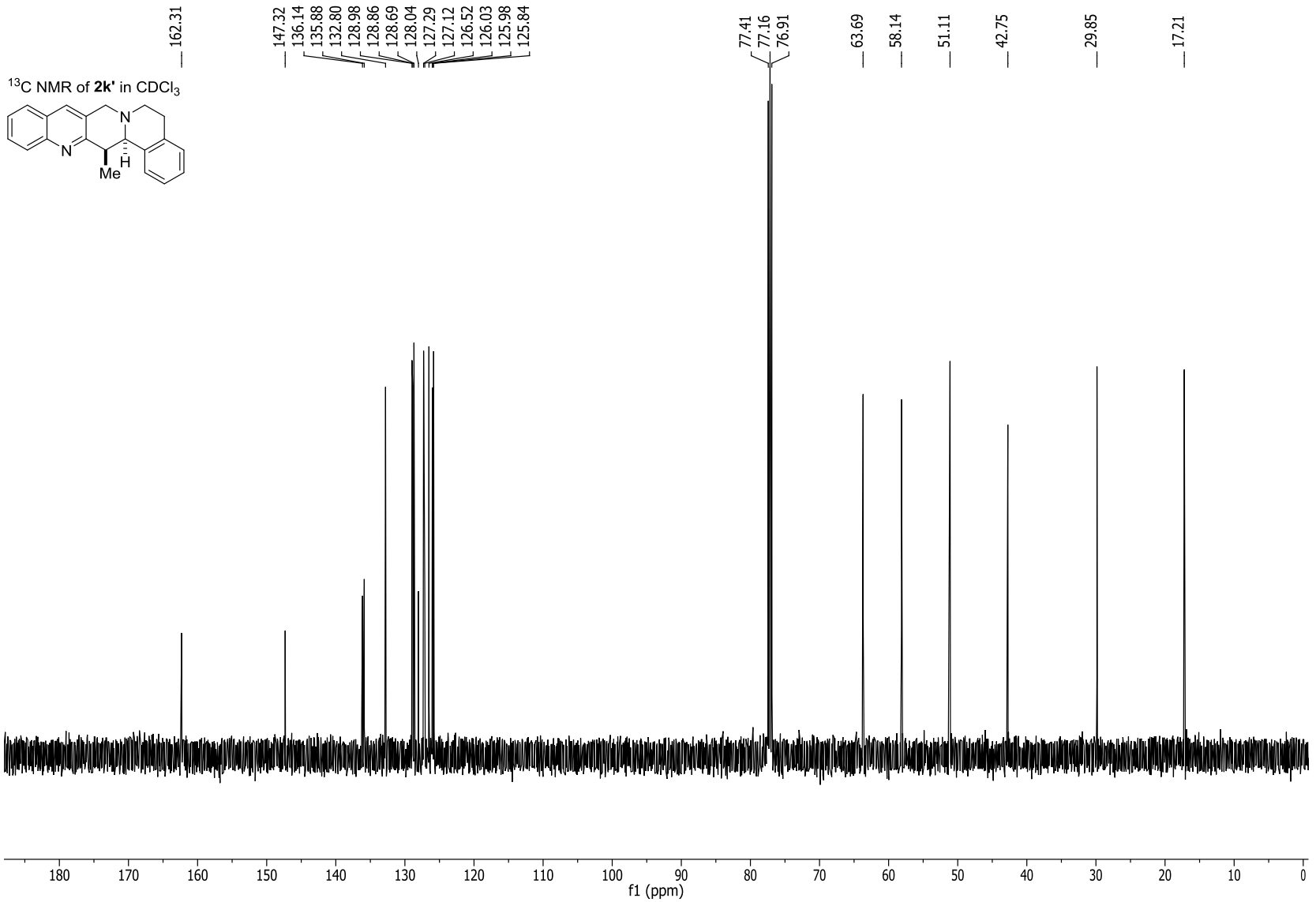

8.03 8.02 8.01 8.01 7.81 7.81 7.74 7.74 7.73 7.73 7.72 7.65 7.65 7.64 7.63 7.63 7.62 7.62 7.48 7.47 7.46 7.46 7.45 7.44 7.37 7.36 7.35 7.35 7.26 7.20 7.19 7.19 7.18 7.18 7.17 7.17 7.15 7.14 7.13 7.13 4.28 4.27 4.24 4.24 4.05 4.04 4.02 4.01 3.93 3.92 3.52 3.51 3.49 3.16 3.15 3.15 3.14 3.14 3.04 3.02 3.01 3.00 2.89 2.88 2.88 2.87 1.73 1.72
^{1}H NMR of **2k** in $CDCl_3$
N
N
H
Me
0.91 0.90 0.95 0.96 0.97 0.96 2.73
1.00 0.99 0.95 1.00 0.99 1.94 1.07 2.81
0.0 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)

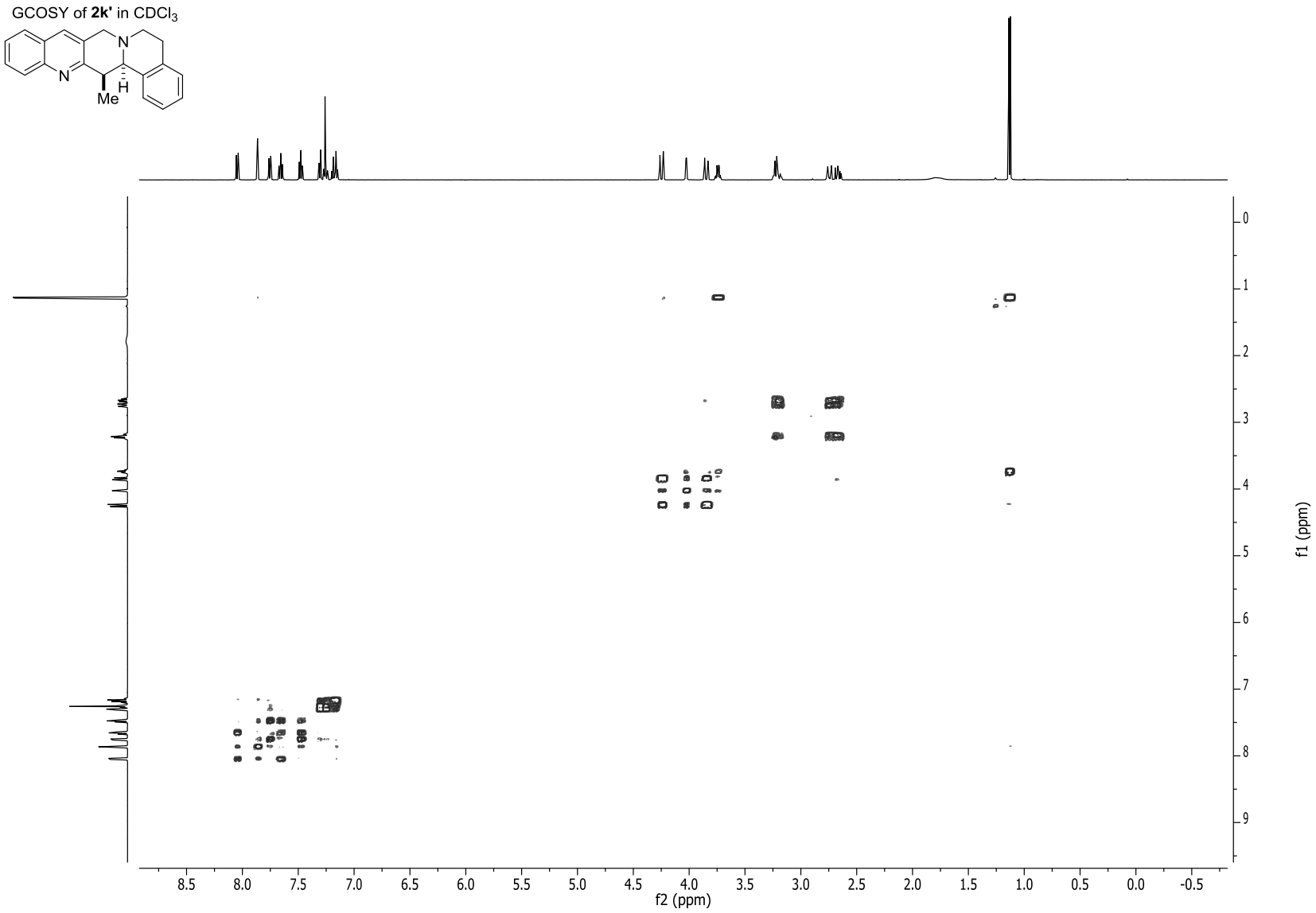

161.06
147.63
138.06
133.85
132.88
129.21
128.86
128.78
127.42
127.13
126.97
126.80
126.78
125.98
125.63
77.41
77.16
76.91
65.74
56.08
48.04
40.10
28.51
21.95
13C NMR of 2k in CDCl3
N
N
H
Me
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

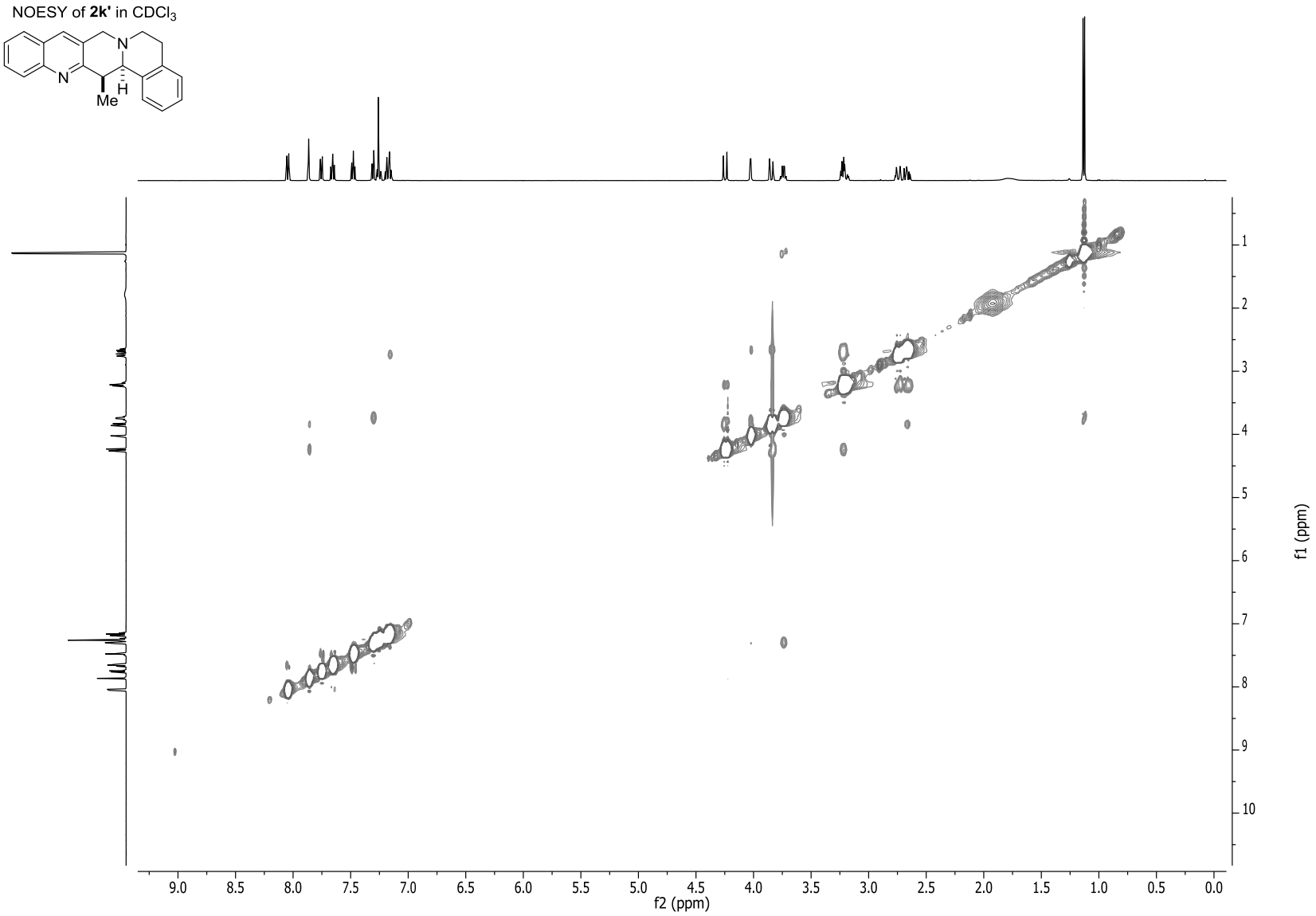

GCOSY of **2k** in $CDCl_3$
N
N
H
Me
f2 (ppm)
f1 (ppm)

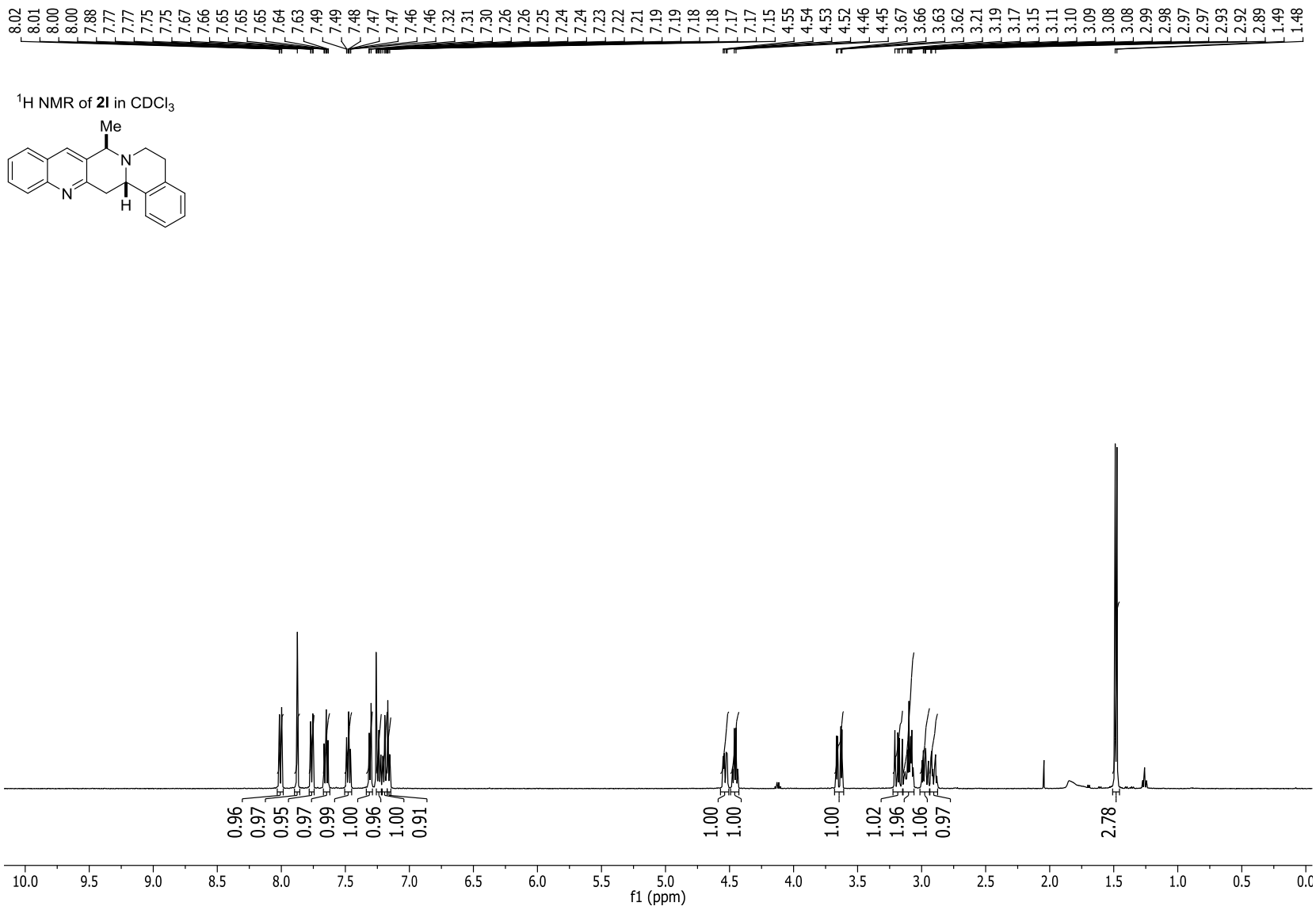

NOESY of **2k** in $CDCl_3$
N
N
H
Me
f2 (ppm)
f1 (ppm)

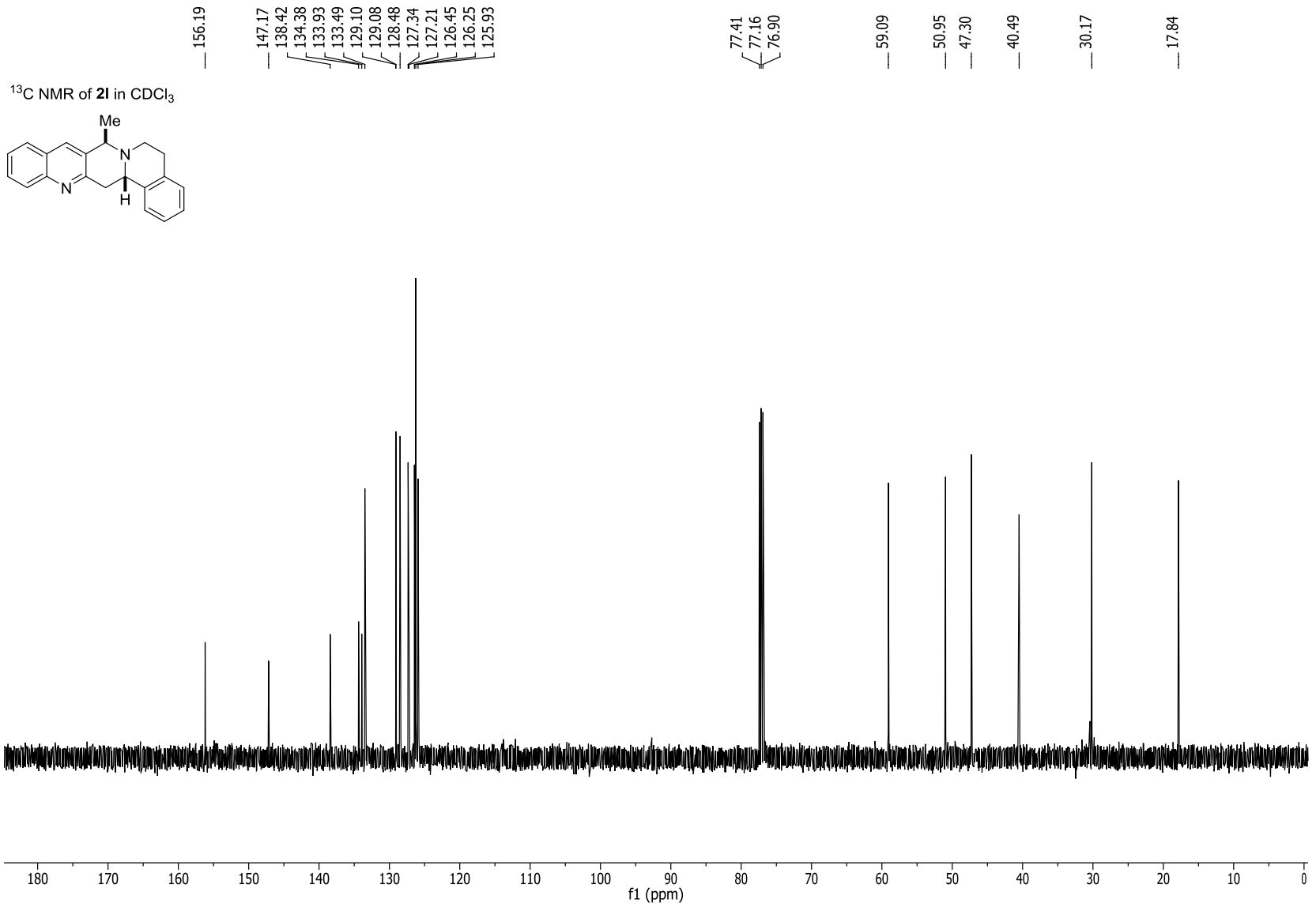

8.06 8.05 8.04 8.04 7.86 7.76 7.76 7.76 7.75 7.75 7.67 7.67 7.66 7.66 7.64 7.64 7.49 7.49 7.48 7.48 7.47 7.46 7.46 7.31 7.30 7.26 7.26 7.26 7.25 7.25 7.19 7.19 7.18 7.18 7.17 7.17 7.17 7.16 7.16 7.16 4.26 4.26 4.23 4.23 4.03 4.03 4.02 4.02 3.86 3.86 3.86 3.83 3.83 3.83 3.75 3.74 3.74 3.73 3.24 3.23 3.22 3.22 3.21 2.73 2.67 2.67 1.14 1.12
^{1}H NMR of **2k'** in $CDCl_3$
N
N
H
Me
0.97 0.98 1.00 1.01 1.01 1.00 1.46 1.97 1.05 1.00 1.05 1.07 2.04 1.04 1.04 2.96
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)

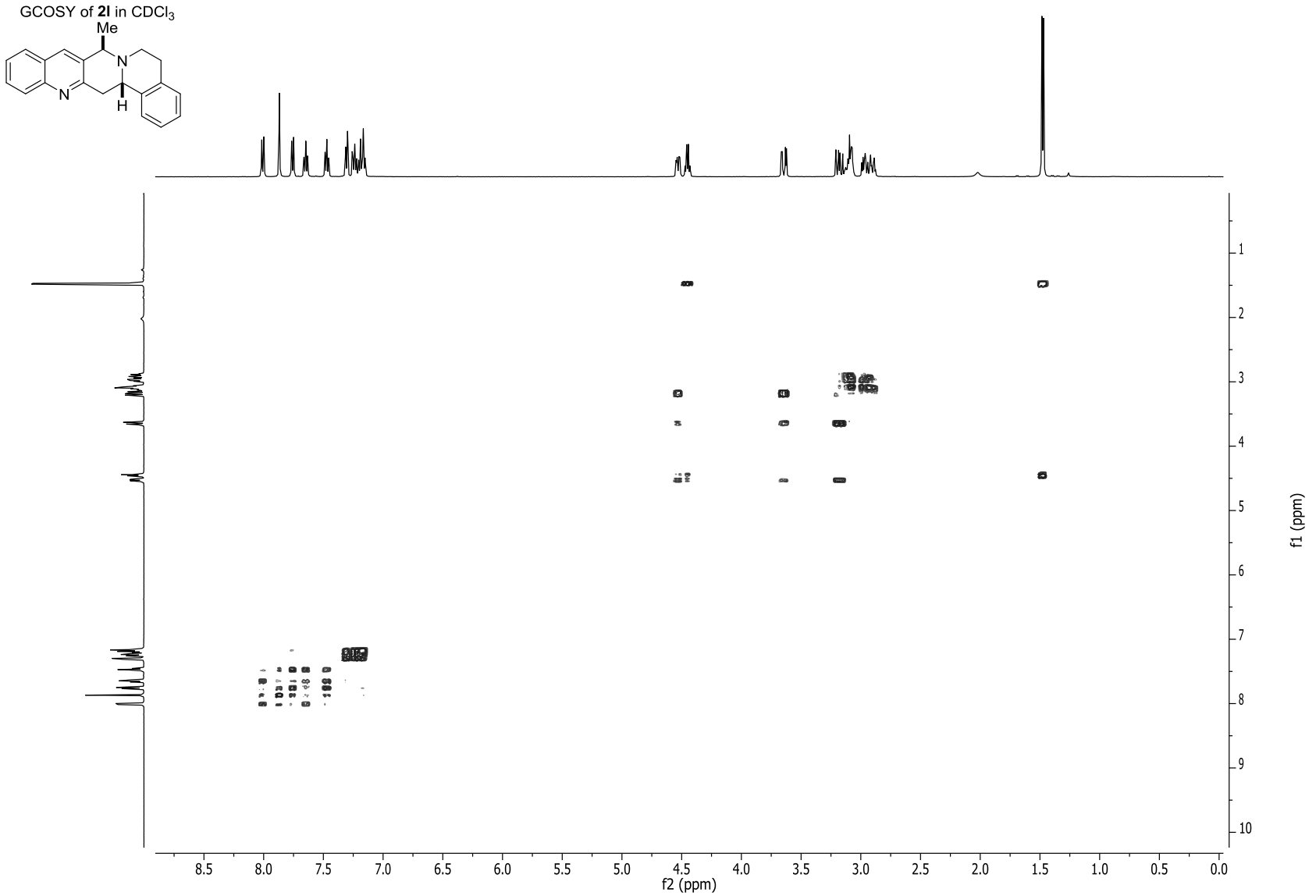

162.31
147.32
136.14
135.88
132.80
128.98
128.86
128.69
128.04
127.29
127.12
126.52
126.03
125.98
125.84
77.41
77.16
76.91
63.69
58.14
51.11
42.75
29.85
17.21
13C NMR of 2k' in CDCl3
N
N
H
Me
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

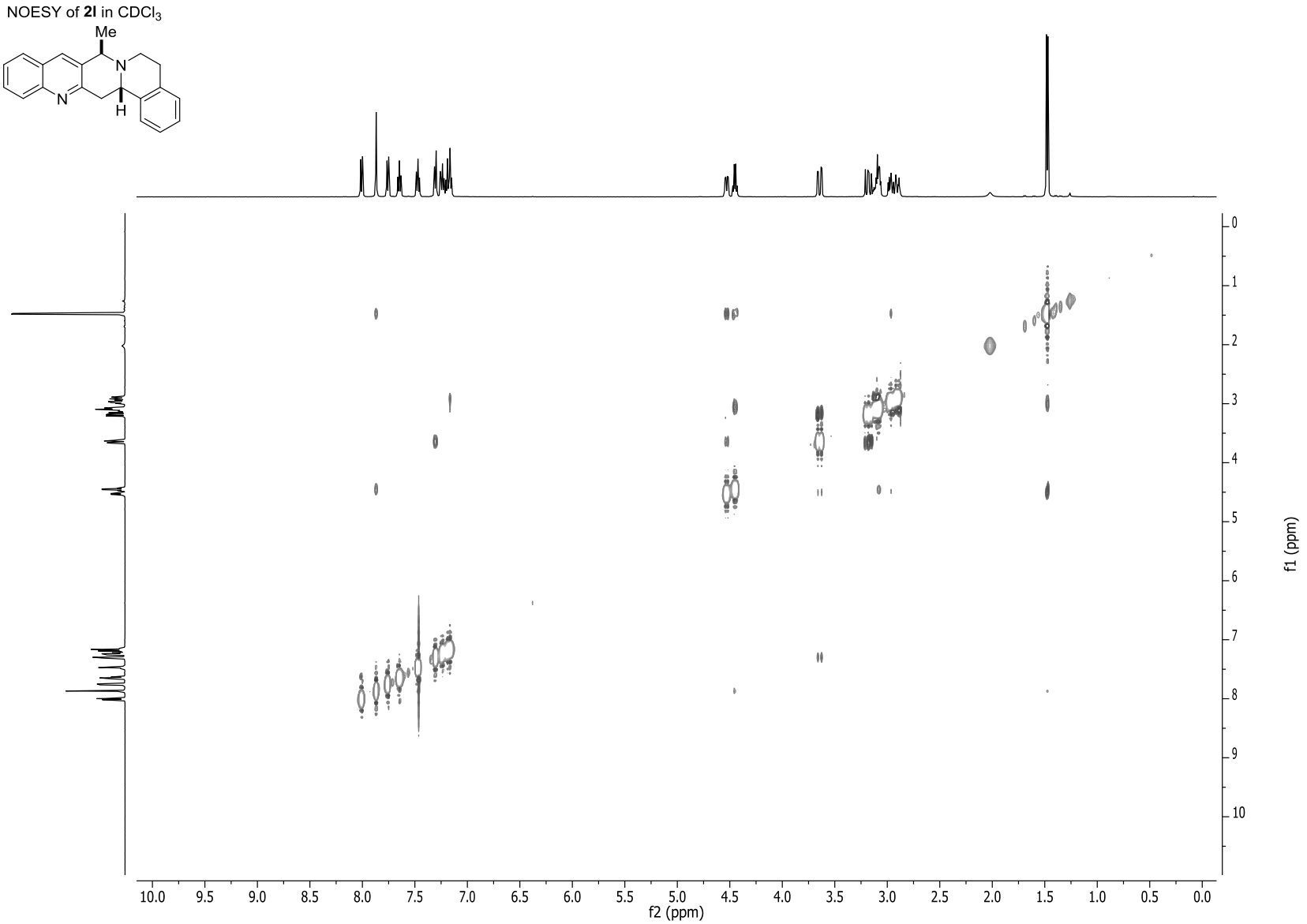

GCOSY of **2k'** in $CDCl_3$
N
N
H
Me
f2 (ppm)
f1 (ppm)

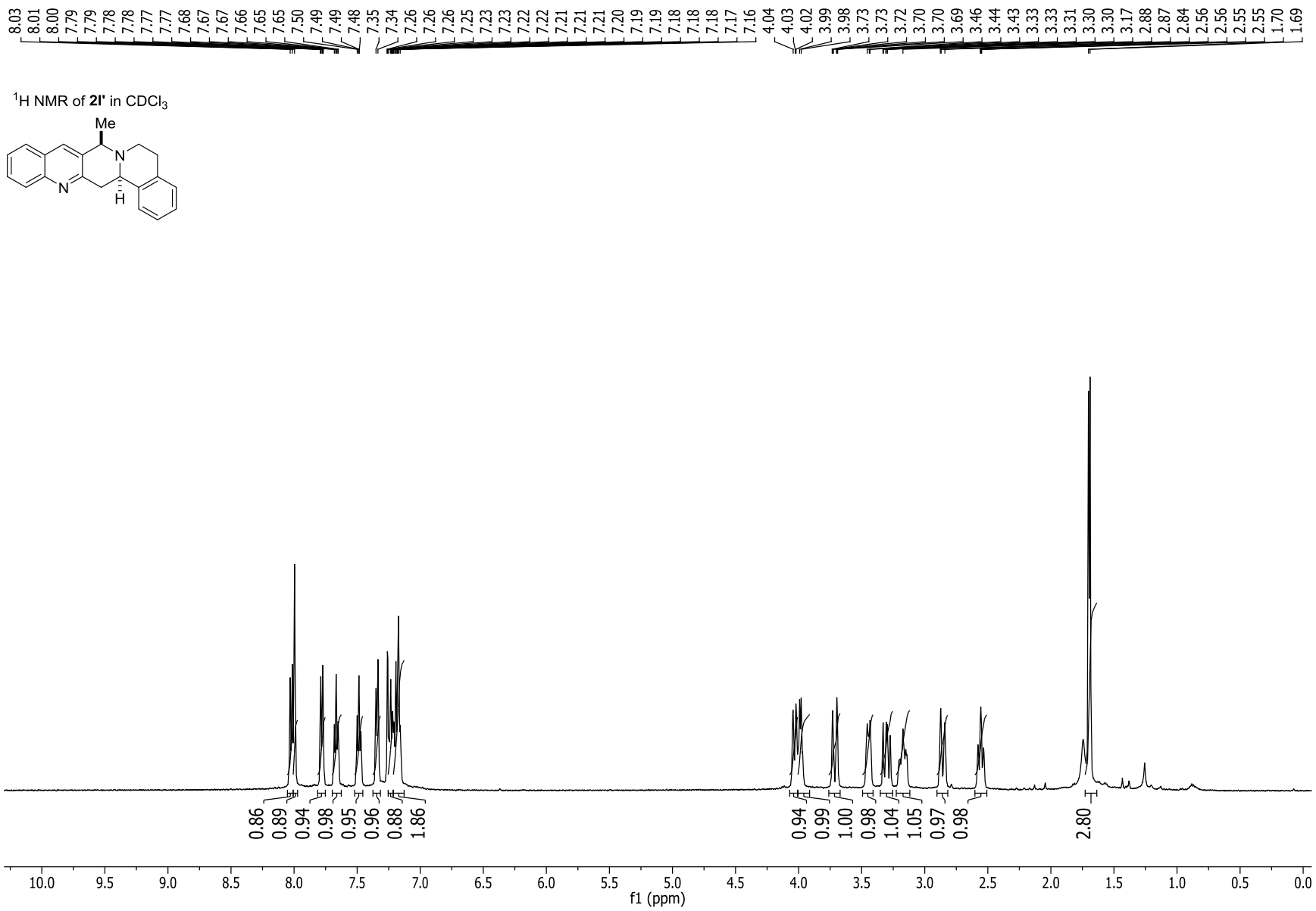

NOESY of **2k'** in $CDCl_3$
N
N
H
Me
f2 (ppm)
f1 (ppm)

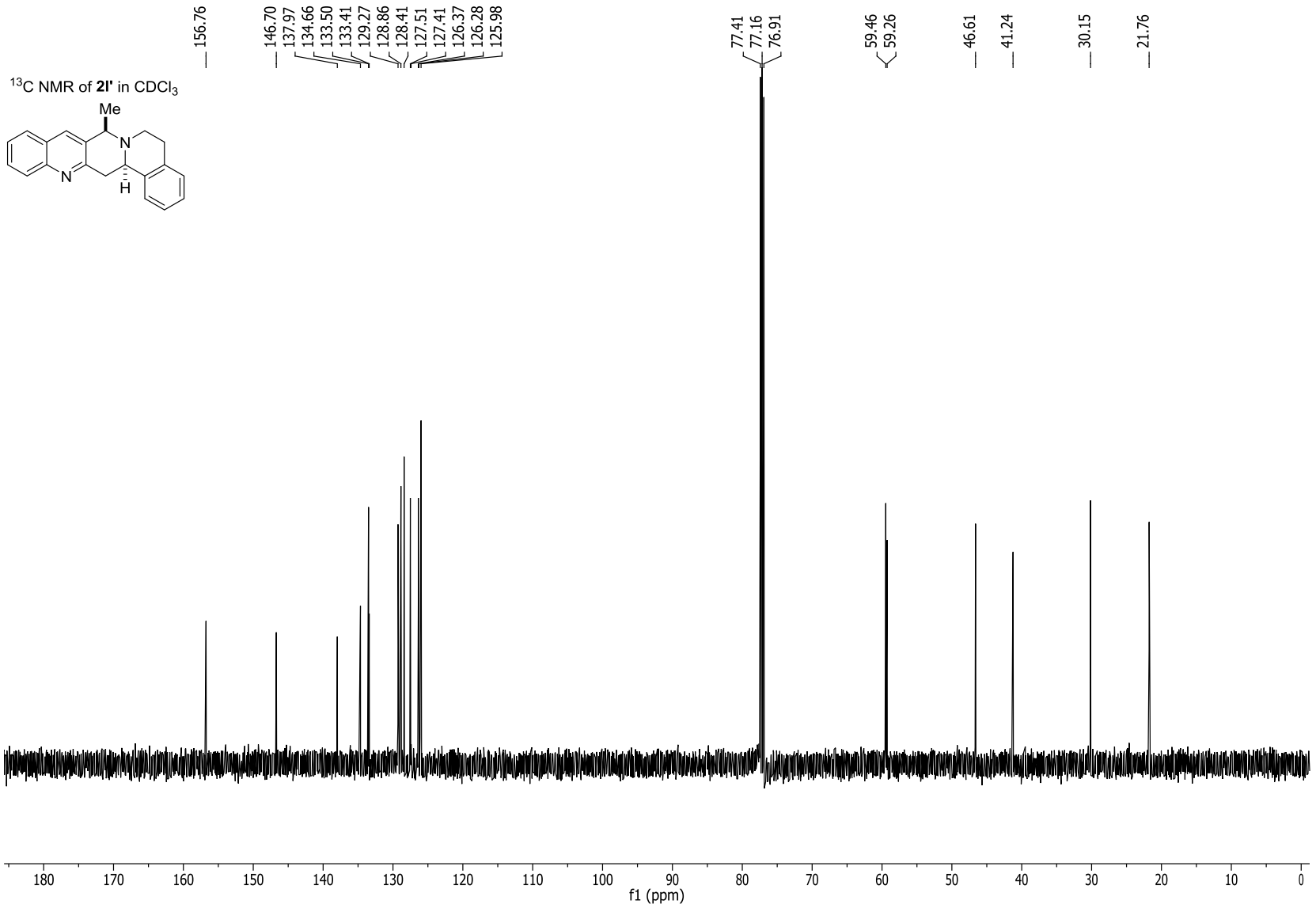

8.02 8.01 8.00 8.00 7.88 7.77 7.77 7.75 7.75 7.67 7.66 7.65 7.65 7.65 7.64 7.63 7.49 7.49 7.48 7.47 7.47 7.46 7.46 7.32 7.31 7.30 7.26 7.26 7.25 7.24 7.24 7.23 7.22 7.21 7.19 7.19 7.18 7.18 7.17 7.17 7.15 4.55 4.54 4.53 4.52 4.46 4.45 3.67 3.66 3.63 3.62 3.21 3.19 3.17 3.15 3.11 3.10 3.09 3.08 3.08 2.99 2.98 2.97 2.97 2.93 2.92 2.89 1.49 1.48
^{1}H NMR of **2l** in $CDCl_3$
Me
N
N
H
0.96 0.97 0.95 0.97 0.99 1.00 0.96 1.00 0.91
1.00 1.00
1.00
1.02 1.96 1.06 0.97
2.78
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)


156.19
147.17
138.42
134.38
133.93
133.49
129.10
129.08
128.48
127.34
127.21
126.45
126.25
125.93
77.41
77.16
76.90
59.09
50.95
47.30
40.49
30.17
17.84
13C NMR of 2l in CDCl3
Me
N
N
H
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

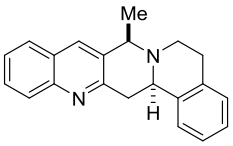

GCOSY of 2l in CDCl3
Me
N
N
H
f2 (ppm)
f1 (ppm)


NOESY of 2l in CDCl3
Me
N
N
H
f2 (ppm)
f1 (ppm)

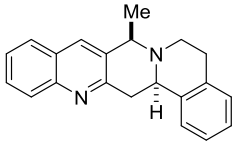

8.03 8.01 8.00 7.79 7.79 7.78 7.78 7.77 7.77 7.68 7.67 7.67 7.66 7.65 7.65 7.50 7.49 7.49 7.48 7.35 7.34 7.26 7.26 7.26 7.25 7.23 7.23 7.22 7.22 7.21 7.21 7.21 7.20 7.19 7.19 7.18 7.18 7.17 7.16 4.04 4.03 4.02 3.99 3.98 3.73 3.73 3.72 3.70 3.70 3.69 3.46 3.44 3.43 3.33 3.33 3.31 3.30 3.30 3.17 2.88 2.87 2.84 2.56 2.56 2.55 2.55 1.70 1.69
^{1}H NMR of **2l'** in $CDCl_3$
Me
N
N
H
0.86 0.89 0.94 0.98 0.95 0.96 0.88 1.86
0.94 0.99 1.00 0.98 1.04 1.05 0.97 0.98
2.80
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)


156.76
146.70
137.97
134.66
133.50
133.41
129.27
128.86
128.41
127.51
127.41
126.37
126.28
125.98
77.41
77.16
76.91
59.46
59.26
46.61
41.24
30.15
21.76
13C NMR of 2l' in CDCl3
Me
N
N
H
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

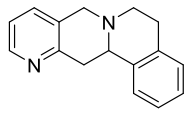
GCOSY of **2l'** in $CDCl_3$

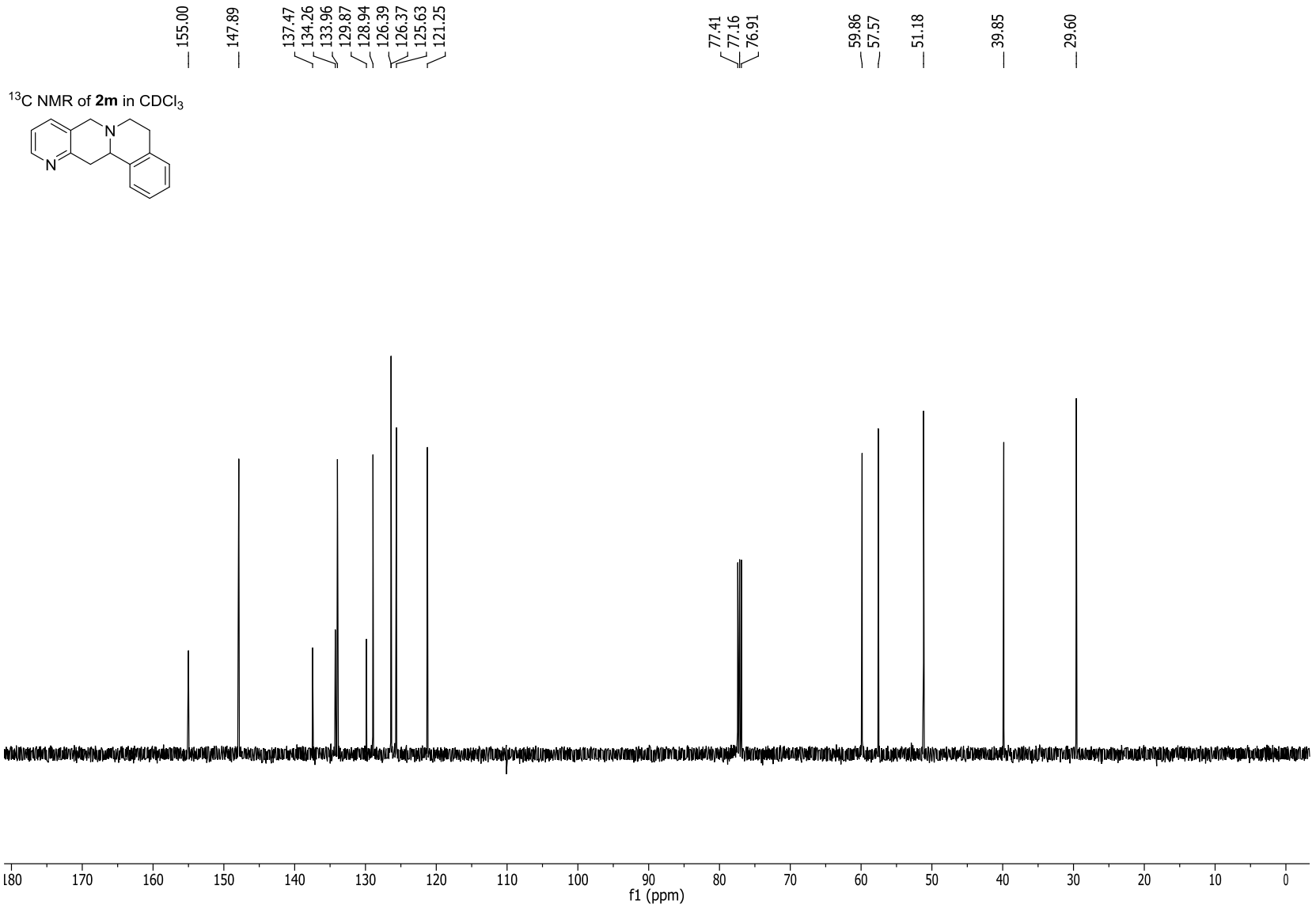


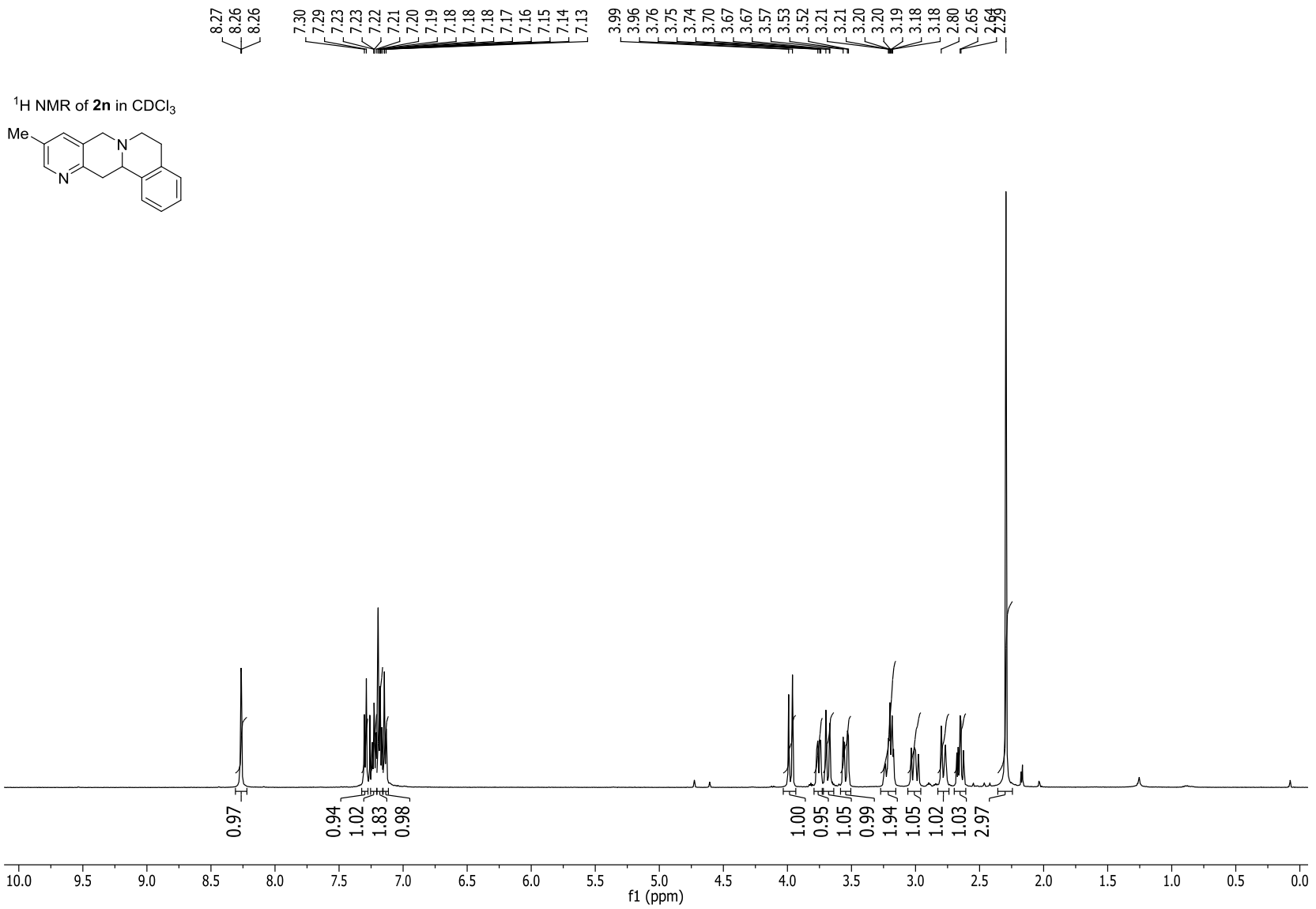
NOESY of **2l'** in $CDCl_3$

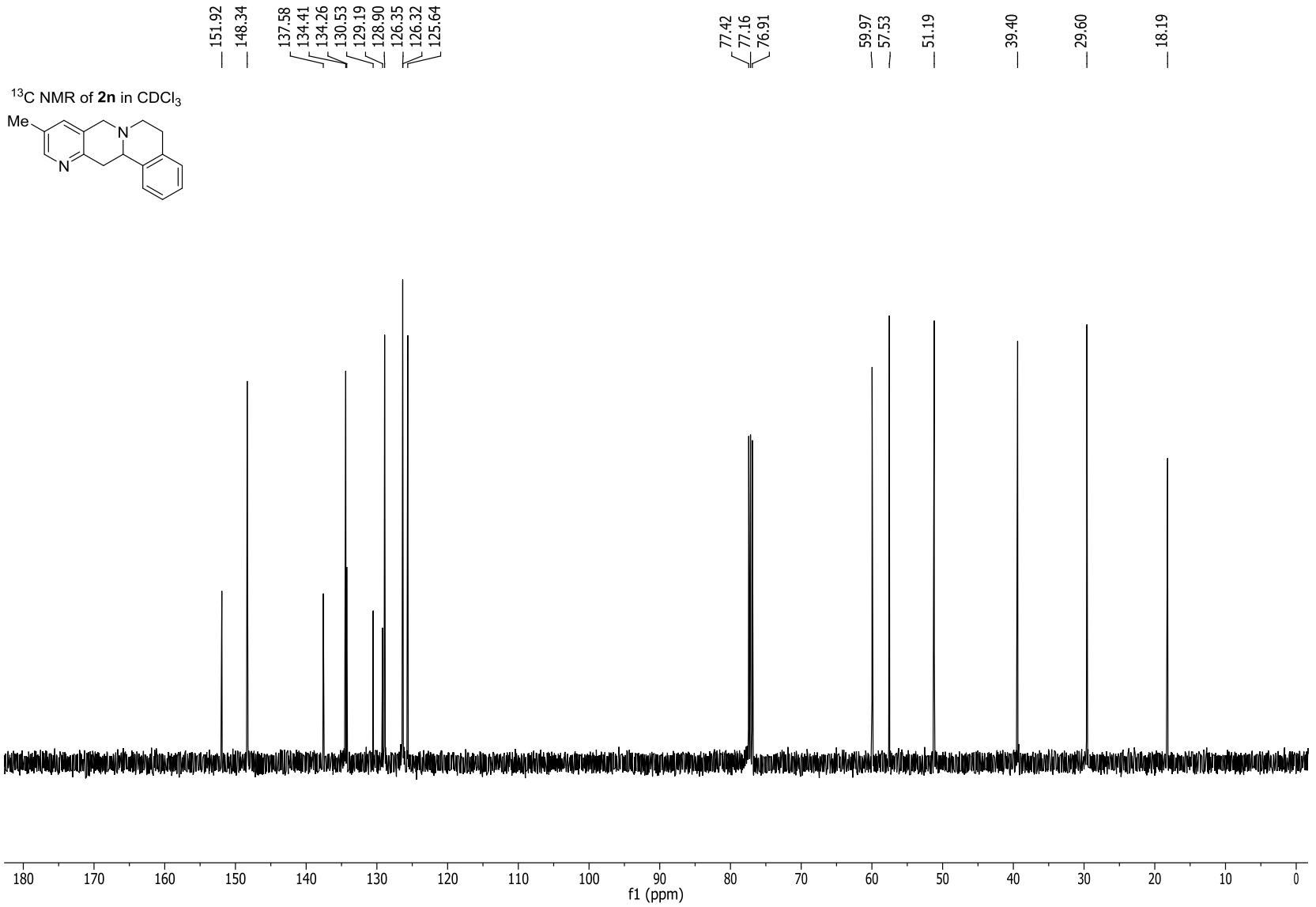


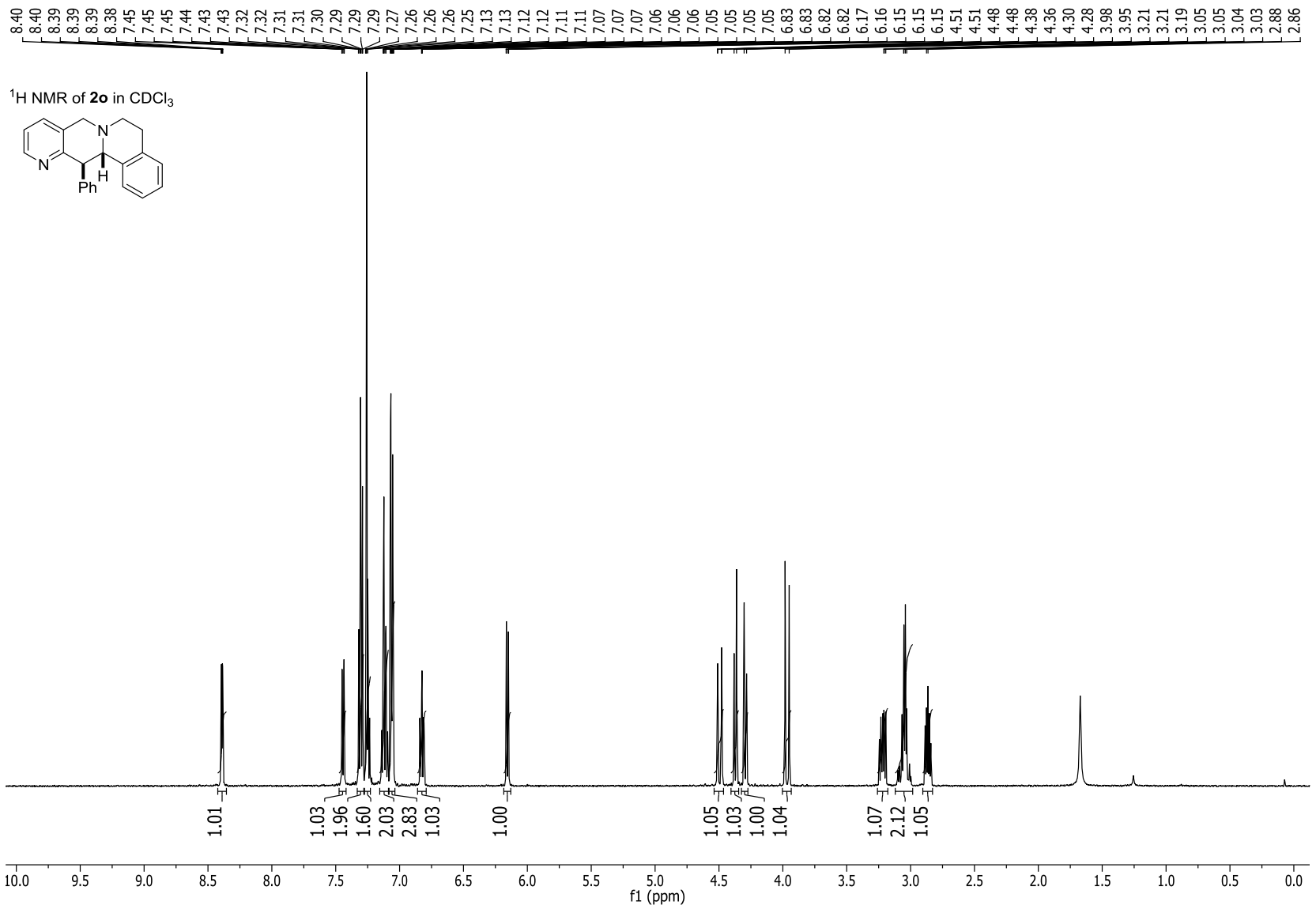
^{1}H NMR of **2m** in $CDCl_3$

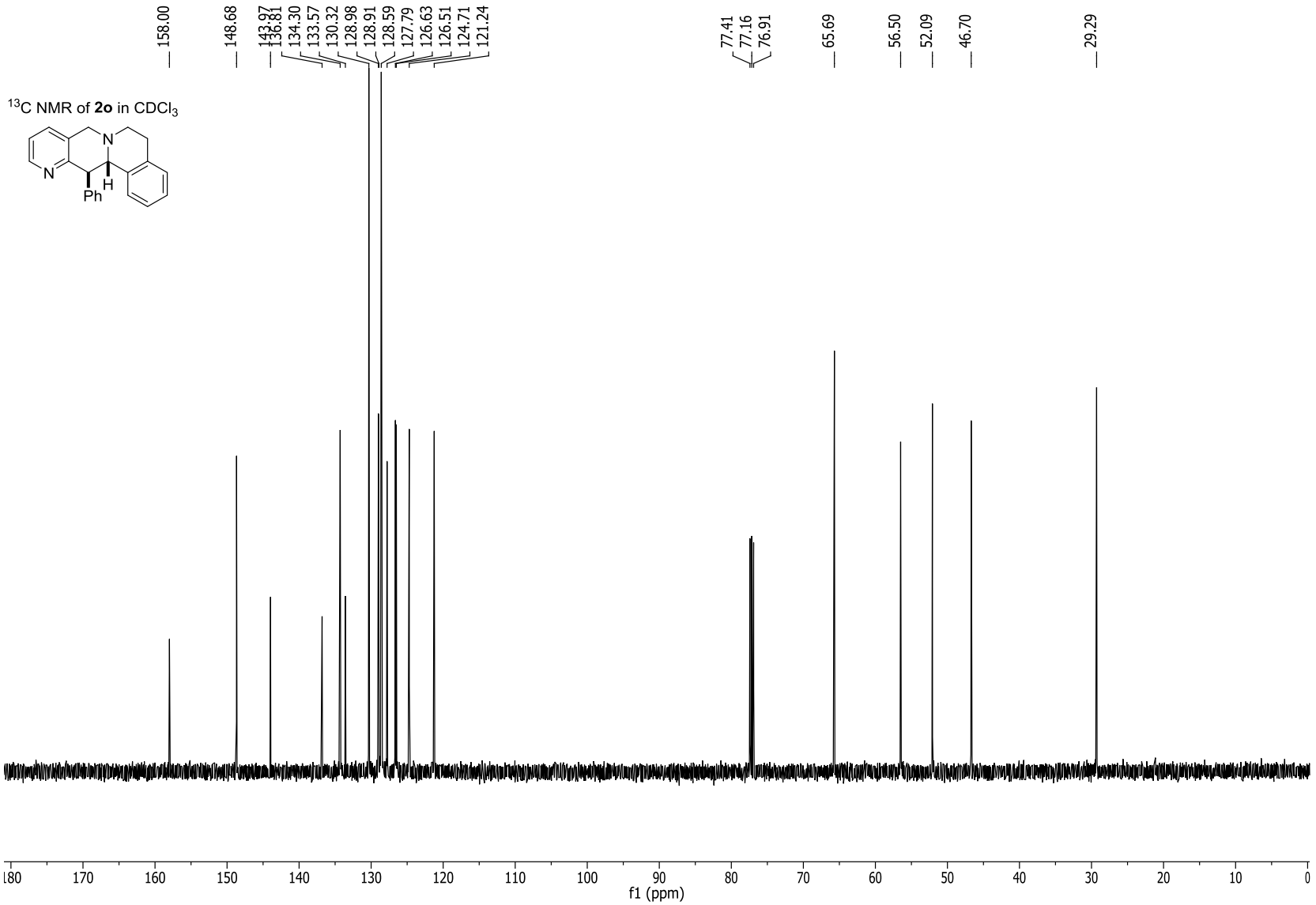



155.00
147.89
137.47
134.26
133.96
129.87
128.94
126.39
126.37
125.63
121.25
77.41
77.16
76.91
59.86
57.57
51.18
39.85
29.60
13C NMR of 2m in CDCl3
N
N
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

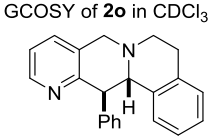

8.27
8.26
8.26
7.30
7.29
7.23
7.23
7.22
7.21
7.20
7.19
7.18
7.18
7.18
7.17
7.16
7.15
7.14
7.13
3.99
3.96
3.76
3.75
3.74
3.70
3.67
3.67
3.57
3.53
3.52
3.21
3.21
3.20
3.20
3.19
3.18
3.18
2.80
2.65
2.64
2.63
^{1}H NMR of **2n** in $CDCl_3$
Me
N
N
0.97
0.94
1.02
1.83
0.98
1.00
0.95
1.05
0.99
1.94
1.05
1.02
1.03
2.97
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)


151.92
148.34
137.58
134.41
134.26
130.53
129.19
128.90
126.35
126.32
125.64
77.42
77.16
76.91
59.97
57.53
51.19
39.40
29.60
18.19
13C NMR of 2n in CDCl3
Me
N
N
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

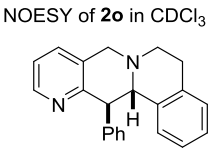

8.40 8.40 8.39 8.39 8.39 8.38 7.45 7.45 7.45 7.44 7.43 7.43 7.32 7.32 7.31 7.31 7.30 7.29 7.29 7.29 7.27 7.26 7.26 7.26 7.25 7.13 7.13 7.12 7.12 7.11 7.11 7.07 7.07 7.07 7.06 7.06 7.06 7.05 7.05 7.05 7.05 6.83 6.83 6.82 6.82 6.17 6.16 6.15 6.15 6.15 4.51 4.51 4.48 4.48 4.38 4.36 4.30 4.28 3.98 3.95 3.21 3.21 3.19 3.05 3.05 3.04 3.03 2.88 2.86
1H NMR of 2o in CDCl3
N
N
H
Ph
1.01
1.03
1.96
1.60
2.03
2.83
1.03
1.00
1.05
1.03
1.00
1.04
1.07
2.12
1.05
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)

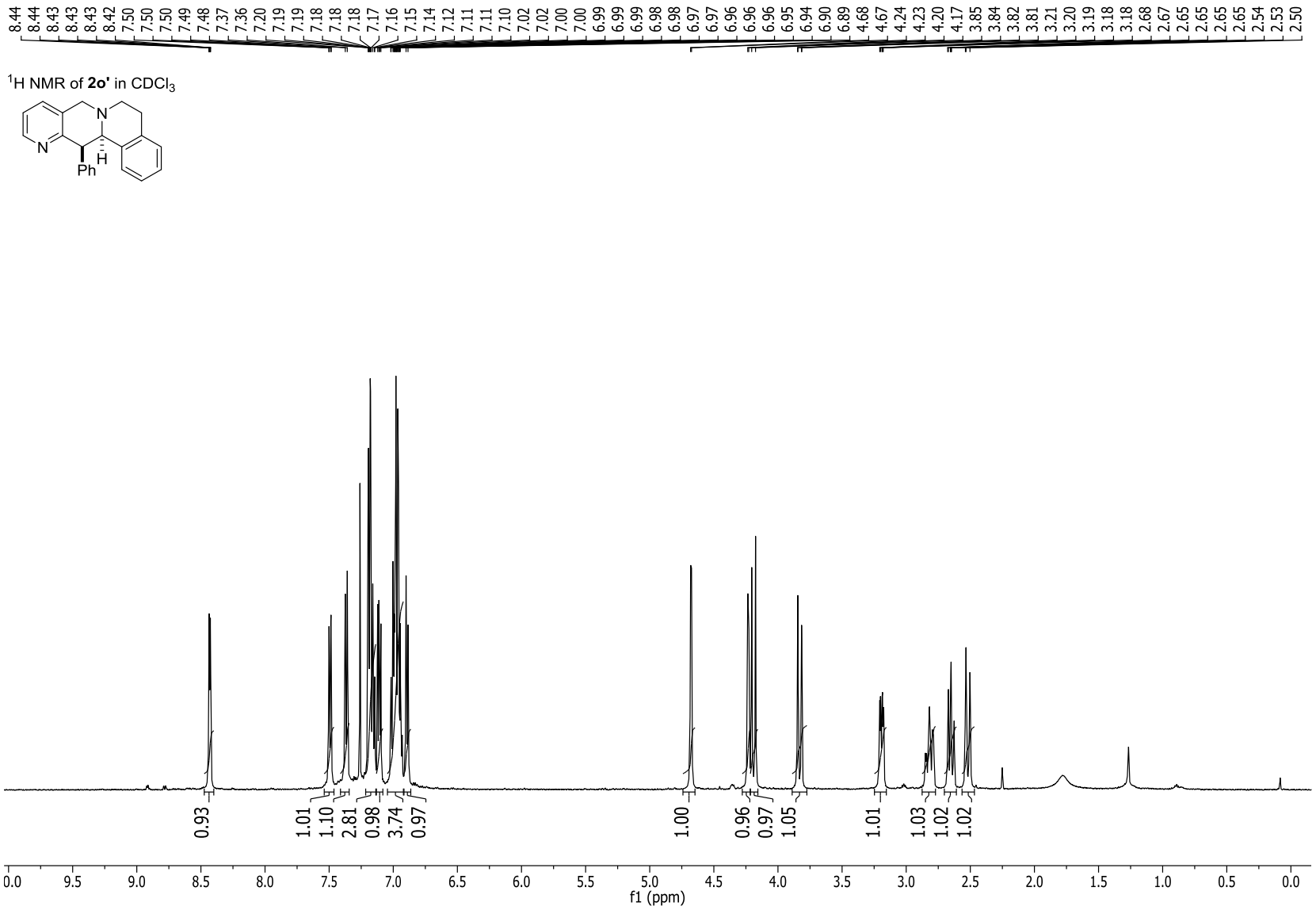

158.00
148.68
143.97
136.81
134.30
133.57
130.32
128.98
128.91
128.59
127.79
126.63
126.51
124.71
121.24
77.41
77.16
76.91
65.69
56.50
52.09
46.70
29.29
13C NMR of 2o in CDCl3
N
N
H
Ph
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

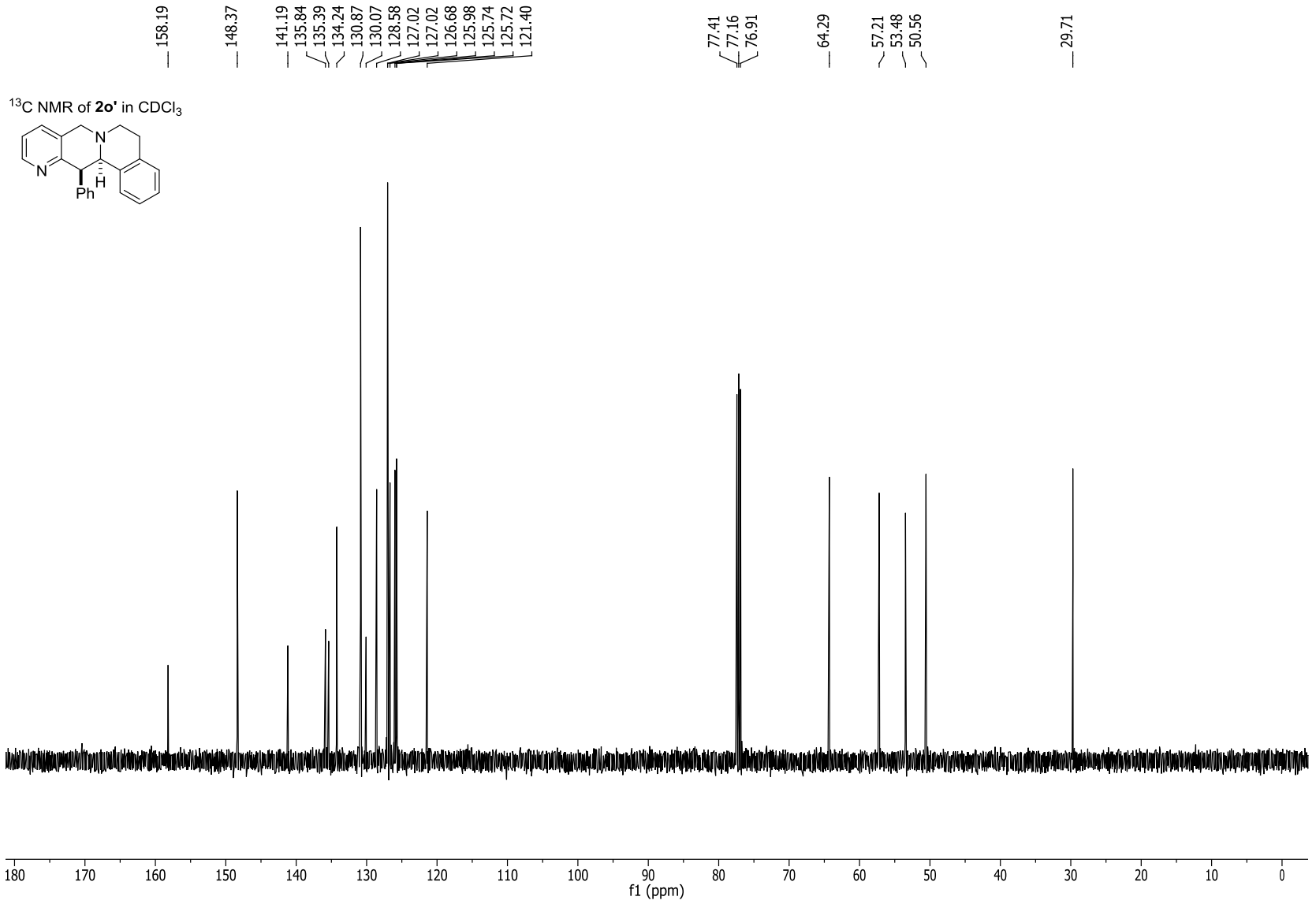
GCOSY of **2o** in $CDCl_3$



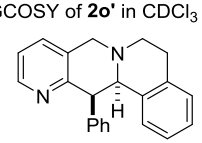
NOESY of **2o** in $CDCl_3$

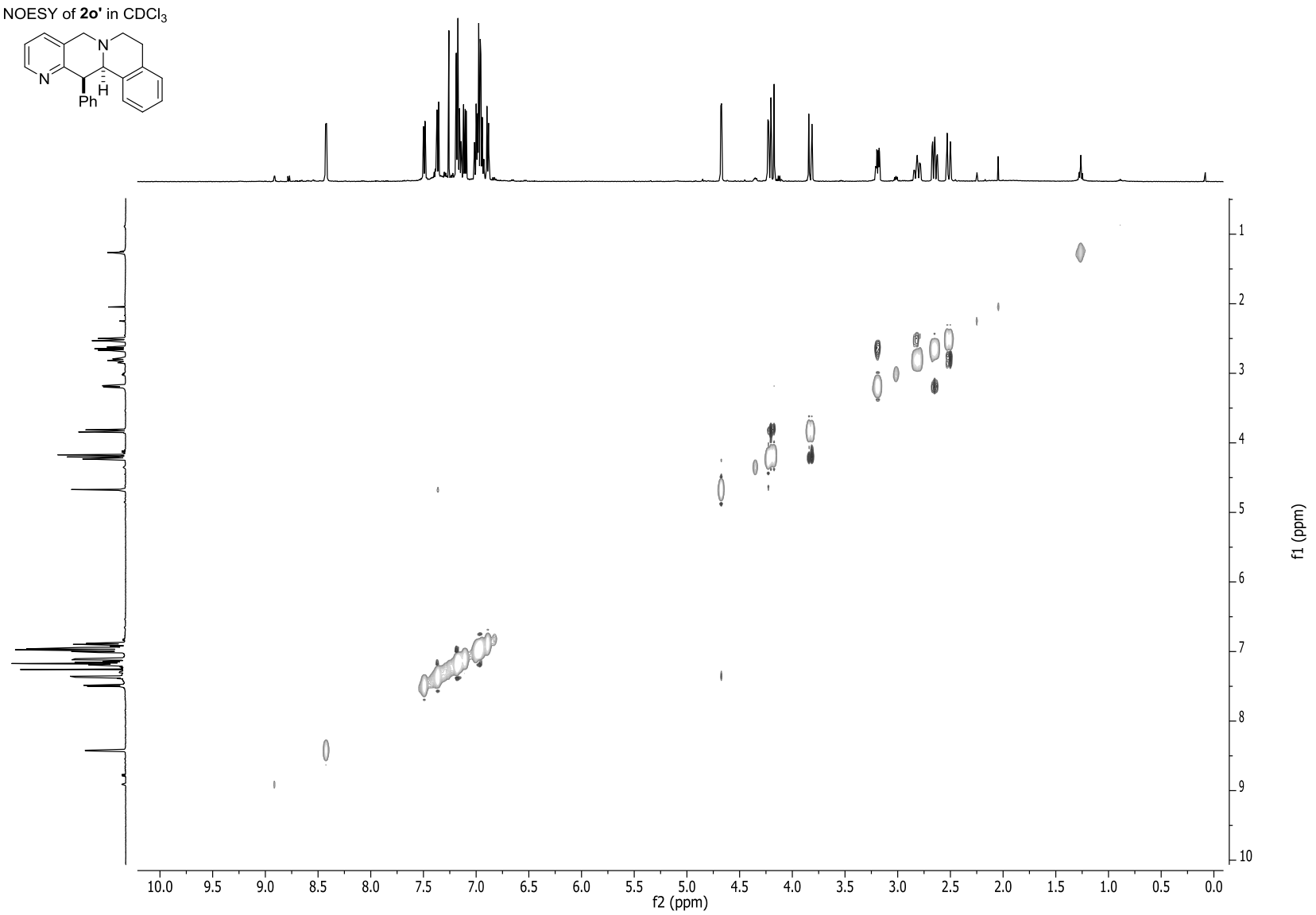


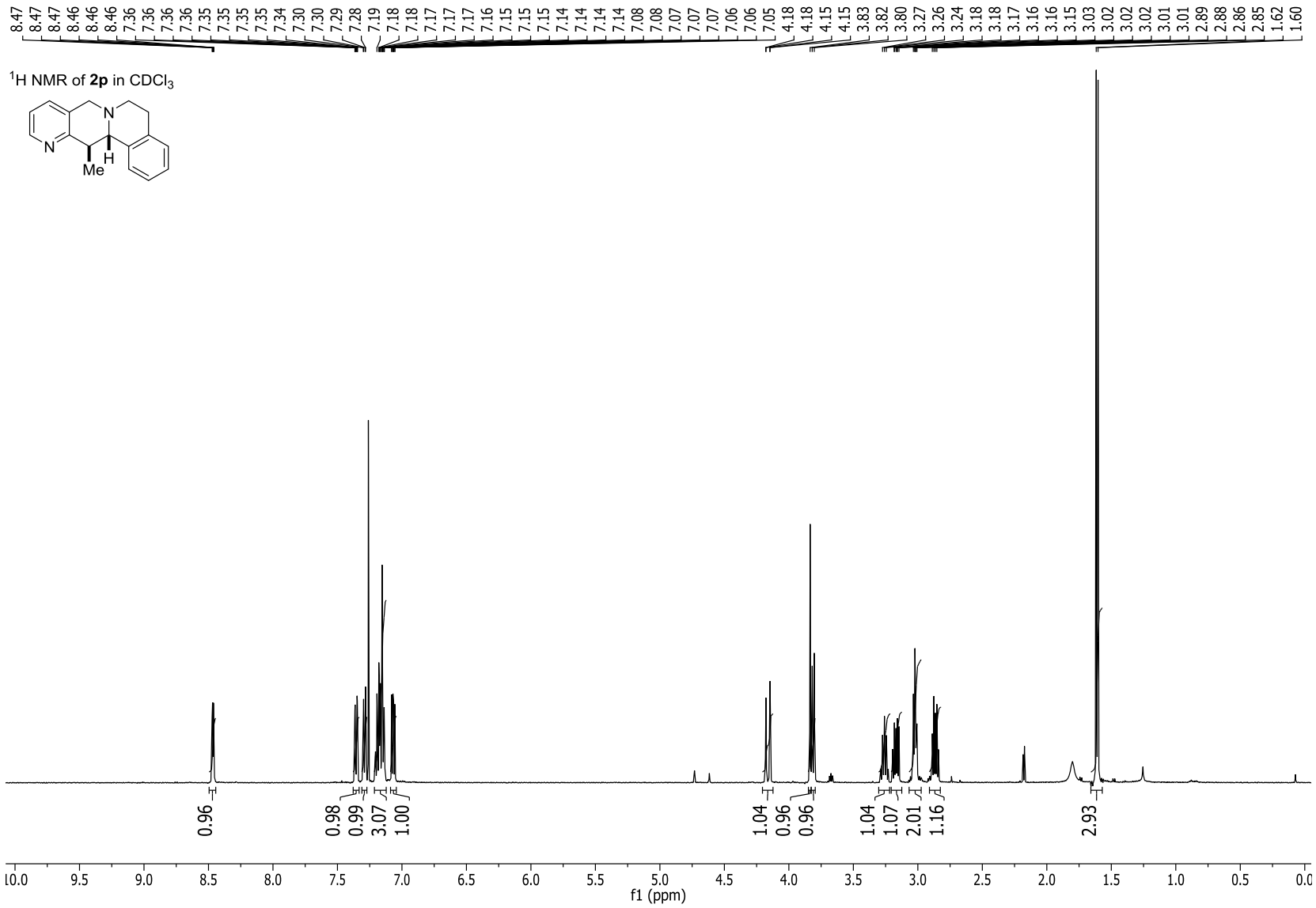

8.44 8.44 8.43 8.43 8.43 8.42 7.50 7.50 7.49 7.48 7.37 7.36 7.20 7.19 7.19 7.18 7.18 7.18 7.17 7.16 7.15 7.14 7.12 7.11 7.11 7.10 7.02 7.02 7.00 7.00 6.99 6.99 6.99 6.98 6.98 6.97 6.97 6.96 6.96 6.96 6.95 6.94 6.90 6.89 4.68 4.67 4.24 4.23 4.20 4.17 3.85 3.84 3.82 3.81 3.21 3.20 3.19 3.18 3.18 2.68 2.67 2.65 2.65 2.65 2.65 2.54 2.53 2.50
1H NMR of 2o' in CDCl3
N
N
H
Ph
0.93 1.01 1.10 2.81 0.98 3.74 0.97 1.00 0.96 0.97 1.05 1.01 1.03 1.02 1.02
0.0 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)


158.19
148.37
141.19
135.84
135.39
134.24
130.87
130.07
128.58
127.02
127.02
126.68
125.98
125.74
125.72
121.40
77.41
77.16
76.91
64.29
57.21
53.48
50.56
29.71
13C NMR of 2o' in CDCl3
N
N
Ph
H
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

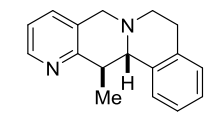
GCOSY of **2o'** in $CDCl_3$



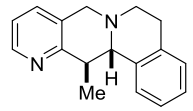

NOESY of **2o'** in $CDCl_3$
N
N
H
Ph
f2 (ppm)
f1 (ppm)


8.47 8.47 8.47 8.46 8.46 8.46 7.36 7.36 7.36 7.35 7.35 7.35 7.35 7.34 7.30 7.30 7.29 7.28 7.19 7.18 7.18 7.17 7.17 7.17 7.16 7.15 7.15 7.15 7.14 7.14 7.14 7.14 7.08 7.08 7.07 7.07 7.07 7.06 7.06 7.05 4.18 4.18 4.15 4.15 3.83 3.82 3.80 3.27 3.26 3.24 3.18 3.18 3.17 3.16 3.16 3.15 3.03 3.02 3.02 3.02 3.01 3.01 2.89 2.88 2.86 2.85 1.62 1.60
^{1}H NMR of **2p** in $CDCl_3$
N
N
Me
H
0.96
0.98
0.99
3.07
1.00
1.04
0.96
0.96
1.04
1.07
2.01
1.16
2.93
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)

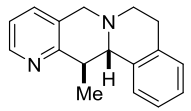
^{13}C NMR of **2p** in $CDCl_3$



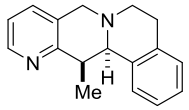
GCOSY of **2p** in $CDCl_3$

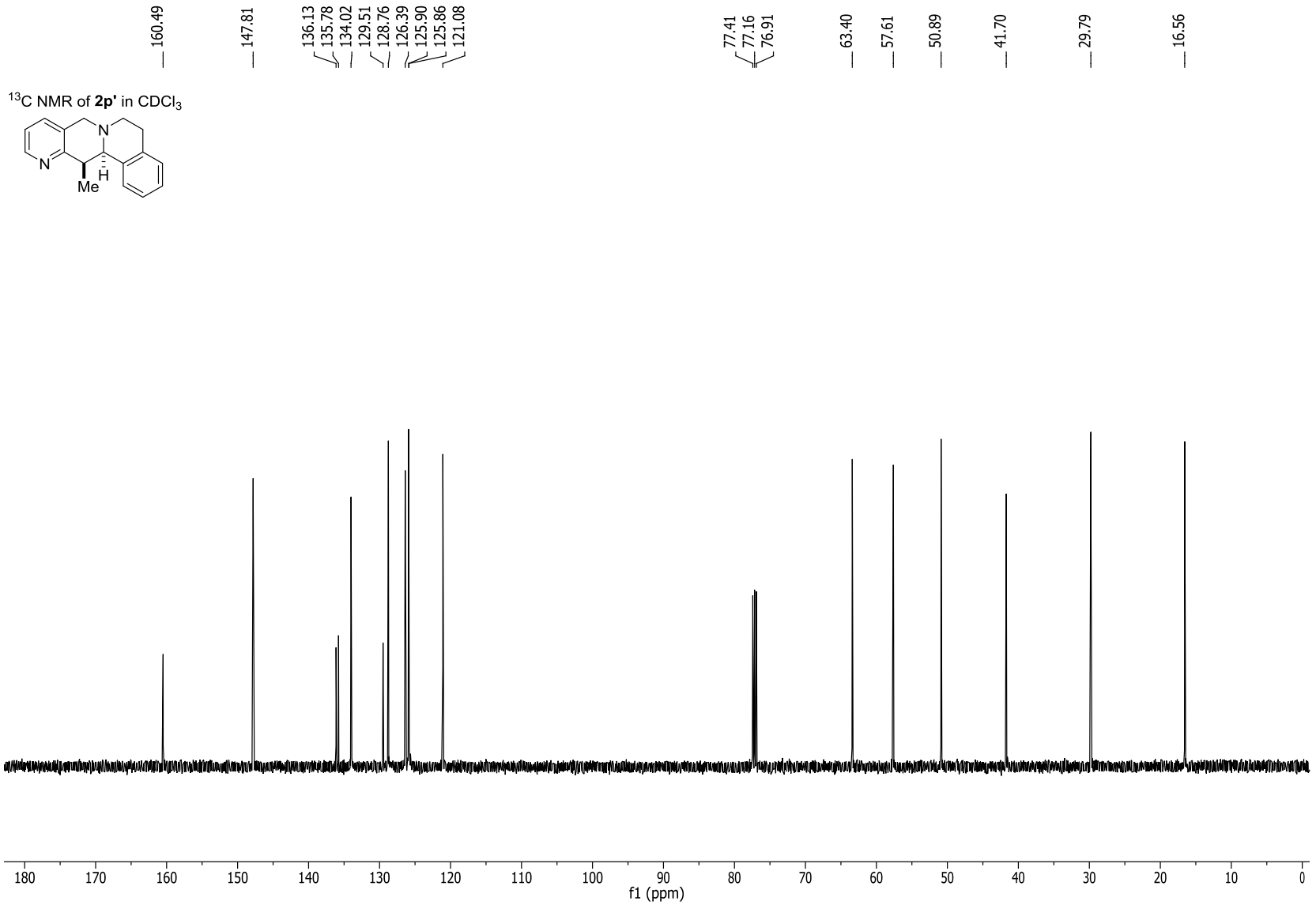


NOESY of **2p** in $CDCl_3$

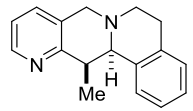


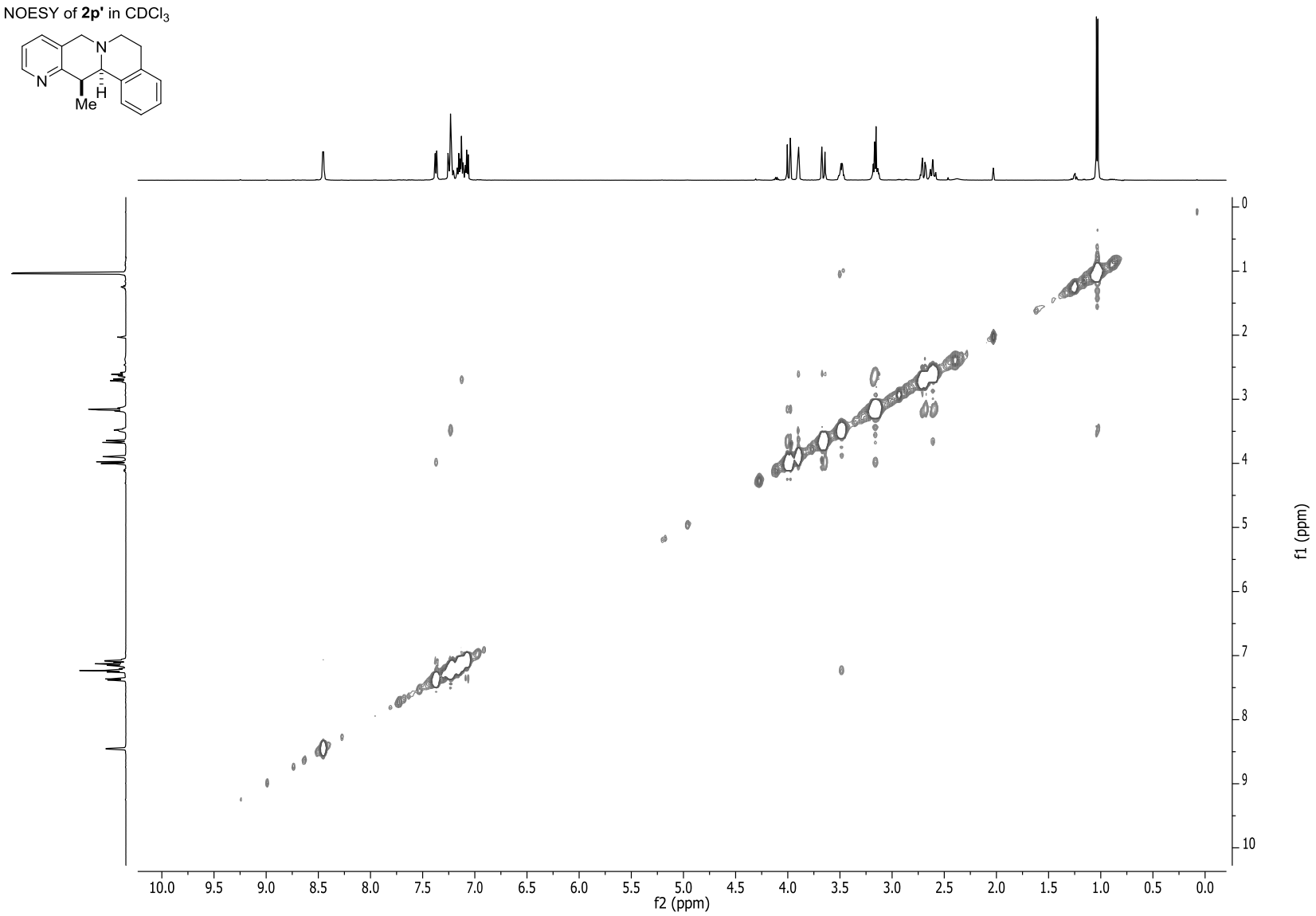
^{1}H NMR of **2p'** in $CDCl_3$

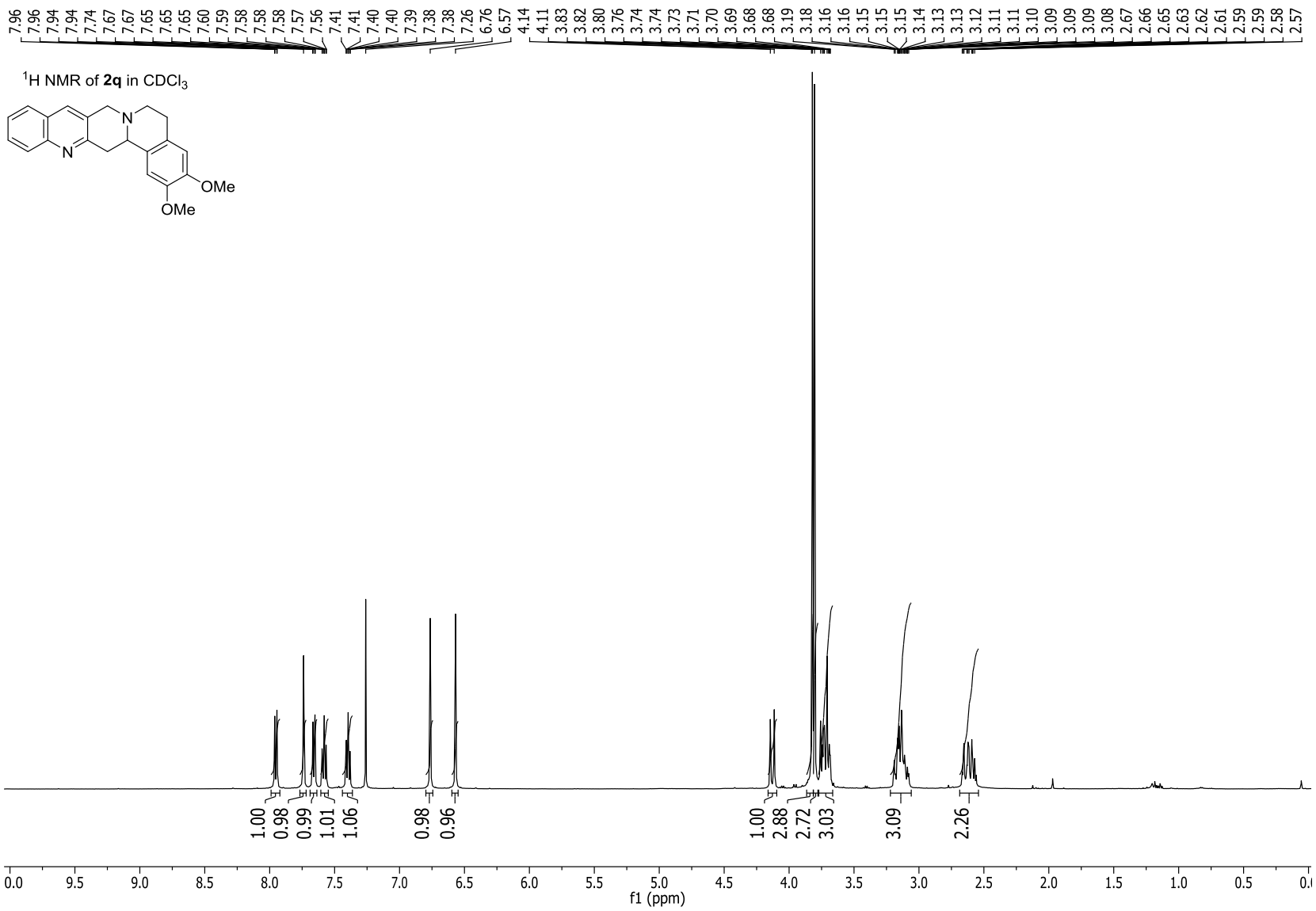


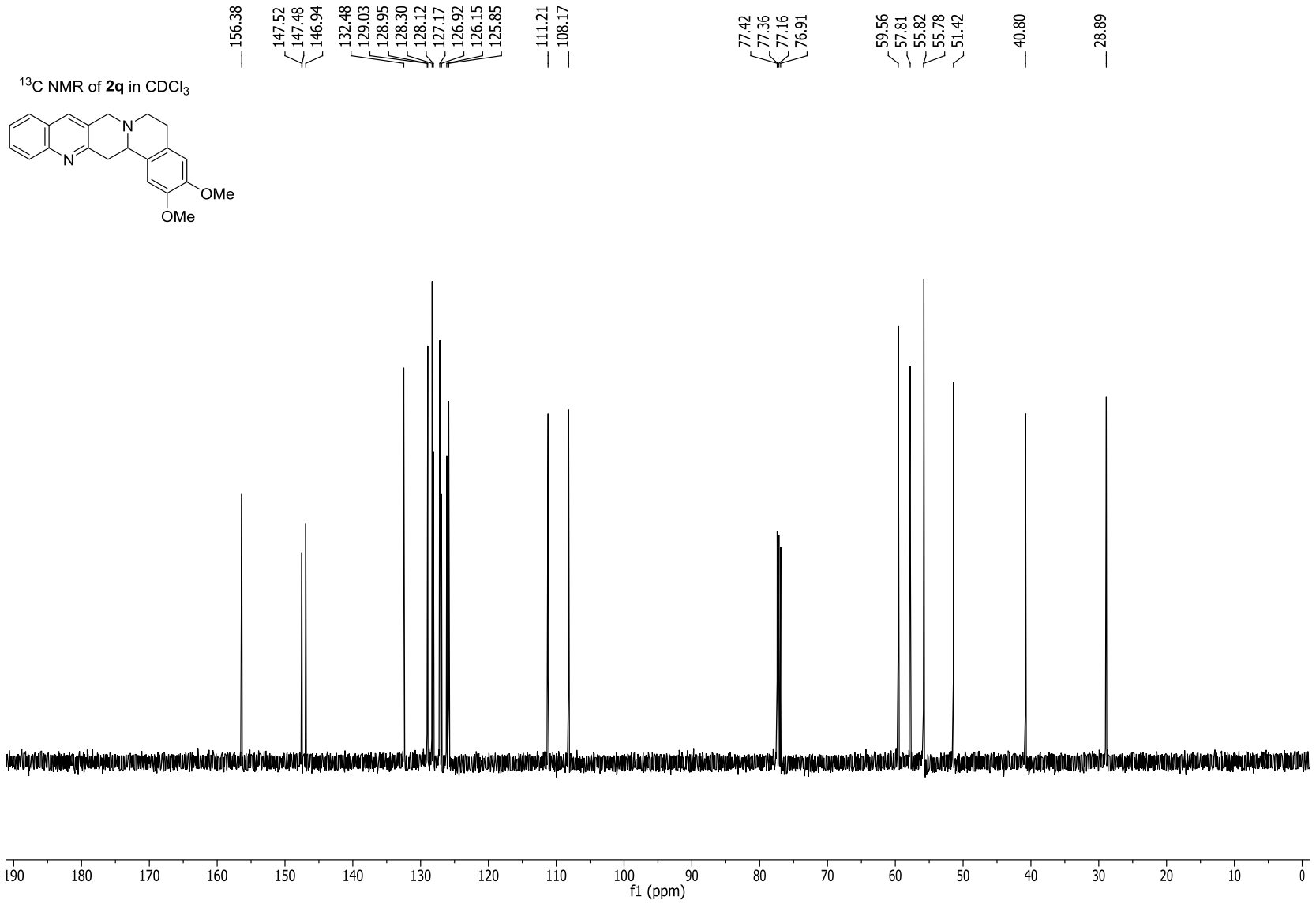

160.49
147.81
136.13
135.78
134.02
129.51
128.76
126.39
125.90
125.86
121.08
77.41
77.16
76.91
63.40
57.61
50.89
41.70
29.79
16.56
13C NMR of 2p' in CDCl3
N
N
H
Me
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

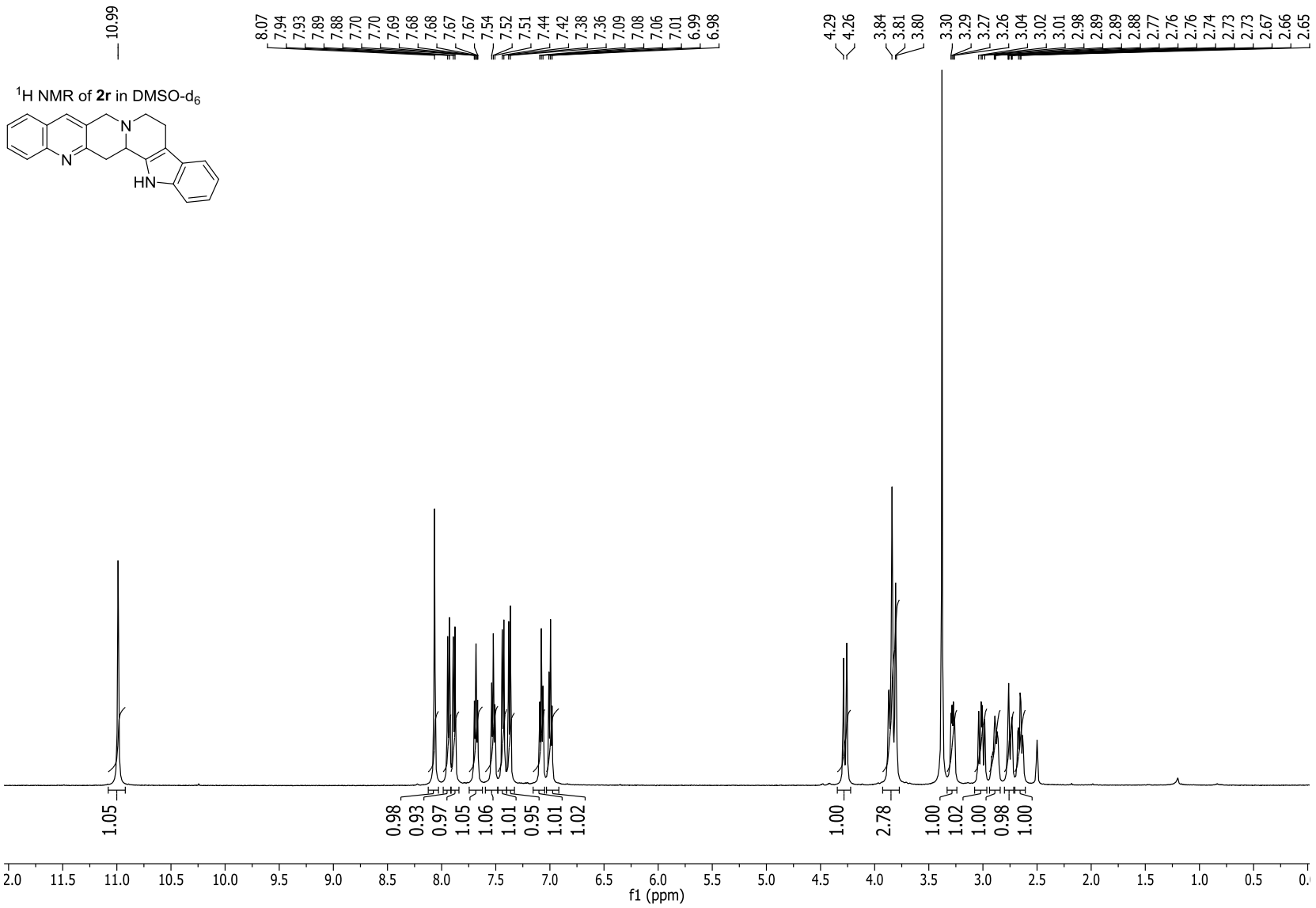
GCOSY of **2p'** in $CDCl_3$

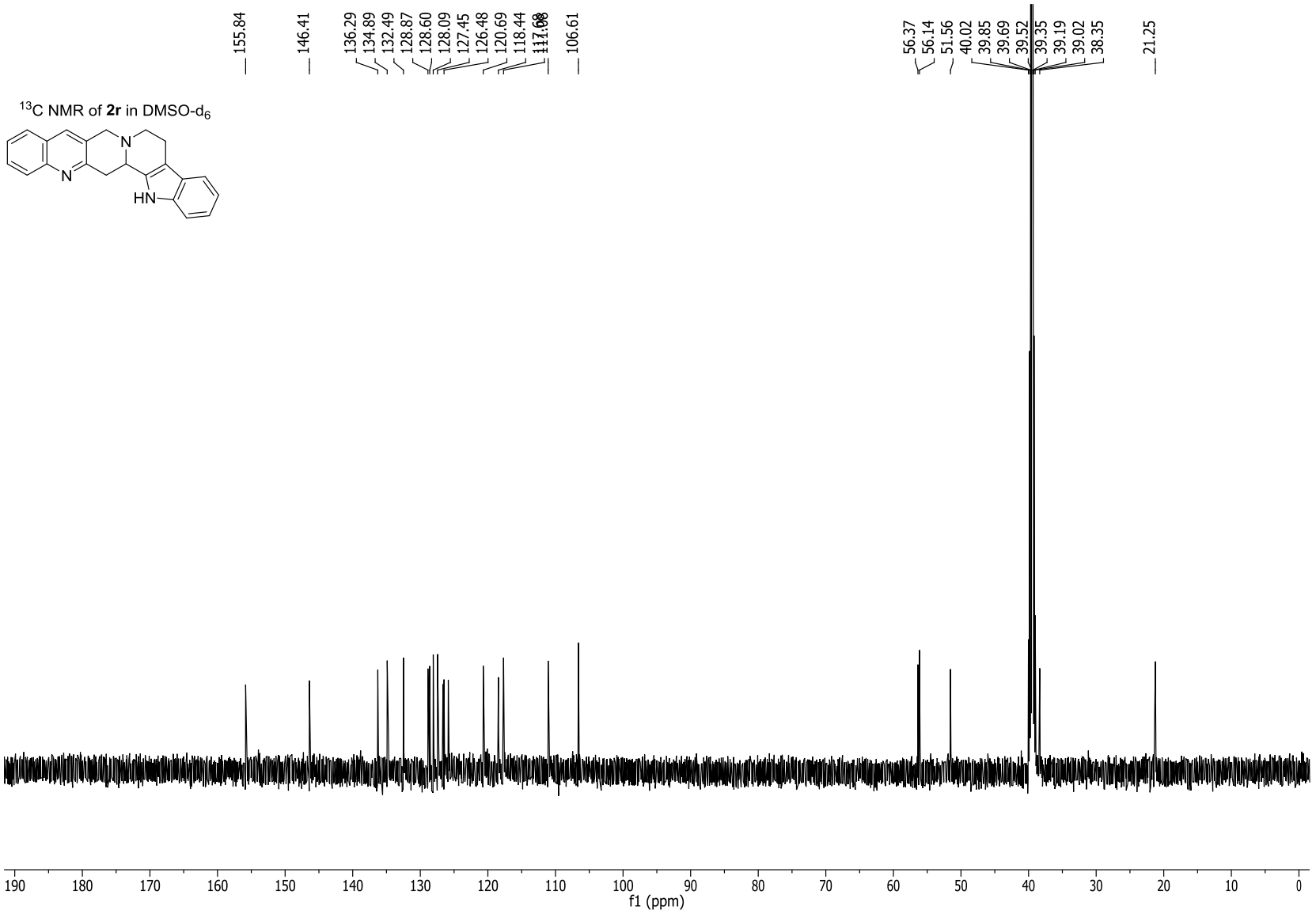


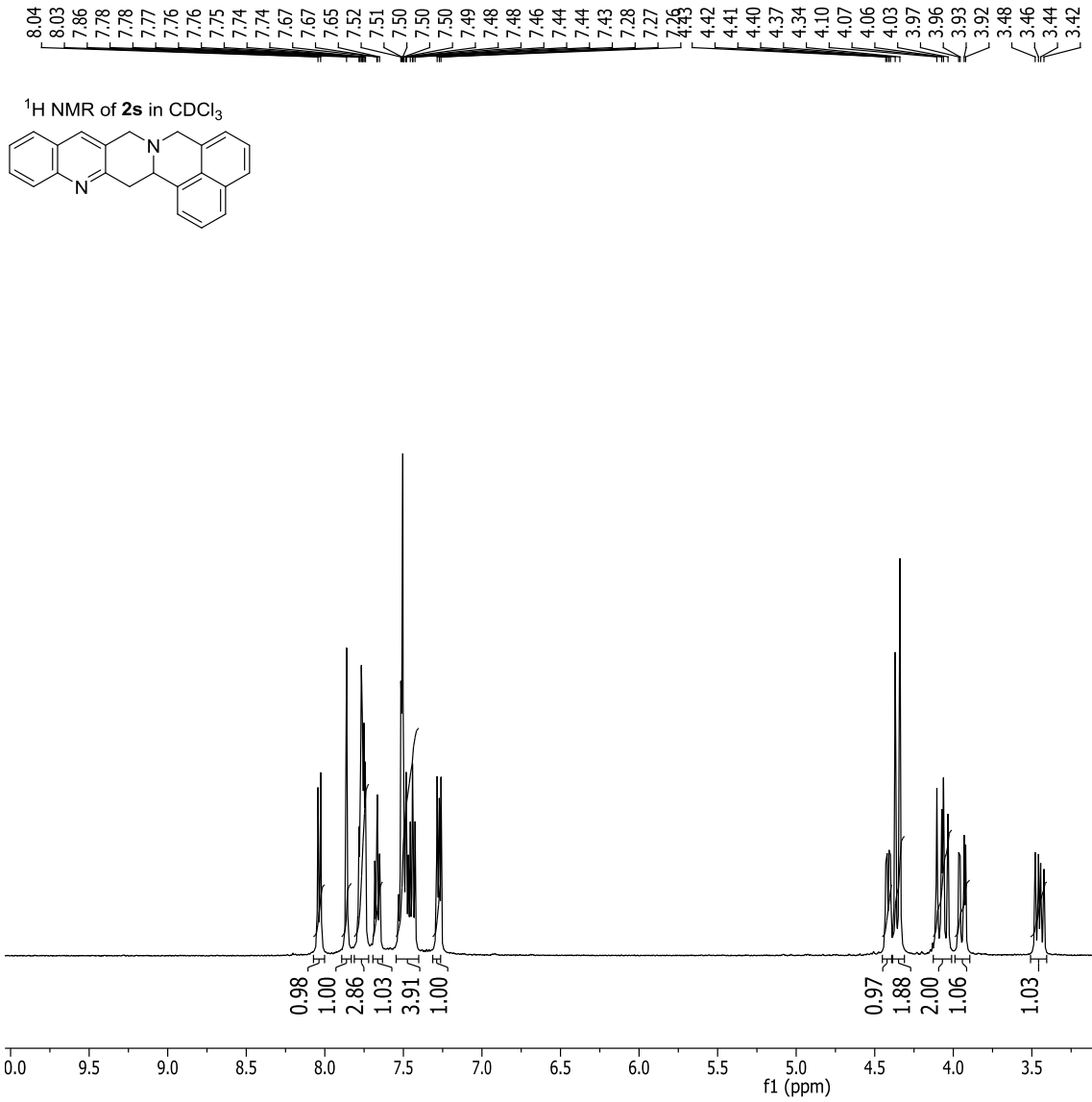

NOESY of **2p'** in $CDCl_3$
N
N
H
Me
f2 (ppm)
f1 (ppm)

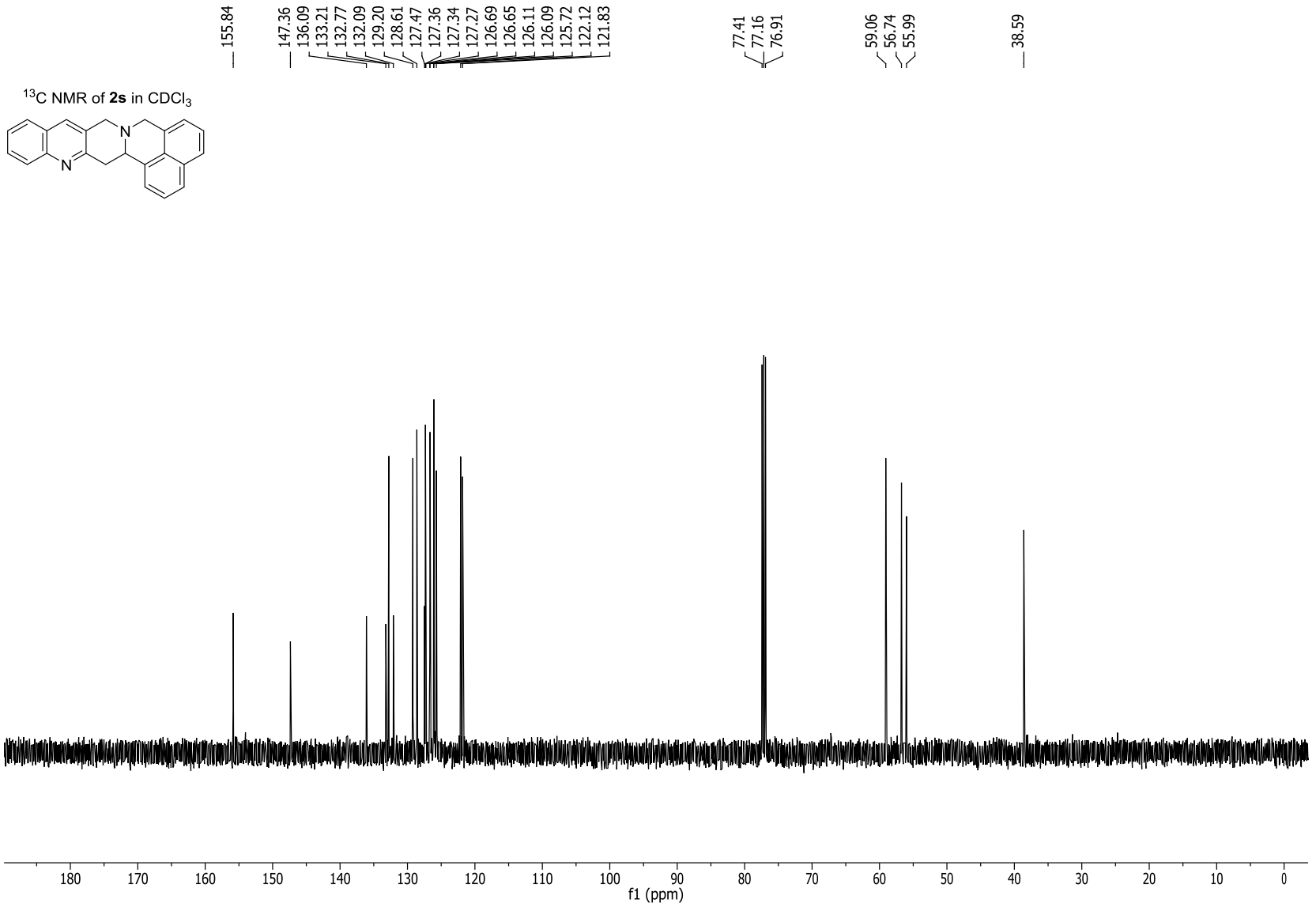

7.96 7.96 7.94 7.94 7.74 7.67 7.67 7.65 7.65 7.60 7.59 7.58 7.58 7.58 7.57 7.56 7.41 7.41 7.40 7.40 7.39 7.38 7.38 7.26 6.76 6.57
4.14 4.11 3.83 3.82 3.80 3.76 3.74 3.74 3.73 3.71 3.70 3.69 3.68 3.68 3.19 3.18 3.16 3.16 3.15 3.15 3.15 3.14 3.13 3.13 3.12 3.11 3.11 3.10 3.09 3.09 3.09 3.08 2.67 2.66 2.65 2.63 2.62 2.61 2.59 2.59 2.58 2.57
^{1}H NMR of **2q** in $CDCl_3$
N
N
OMe
OMe
1.00 0.98 0.99 1.01 1.06 0.98 0.96 1.00 2.88 2.72 3.03 3.09 2.26
0.0 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)

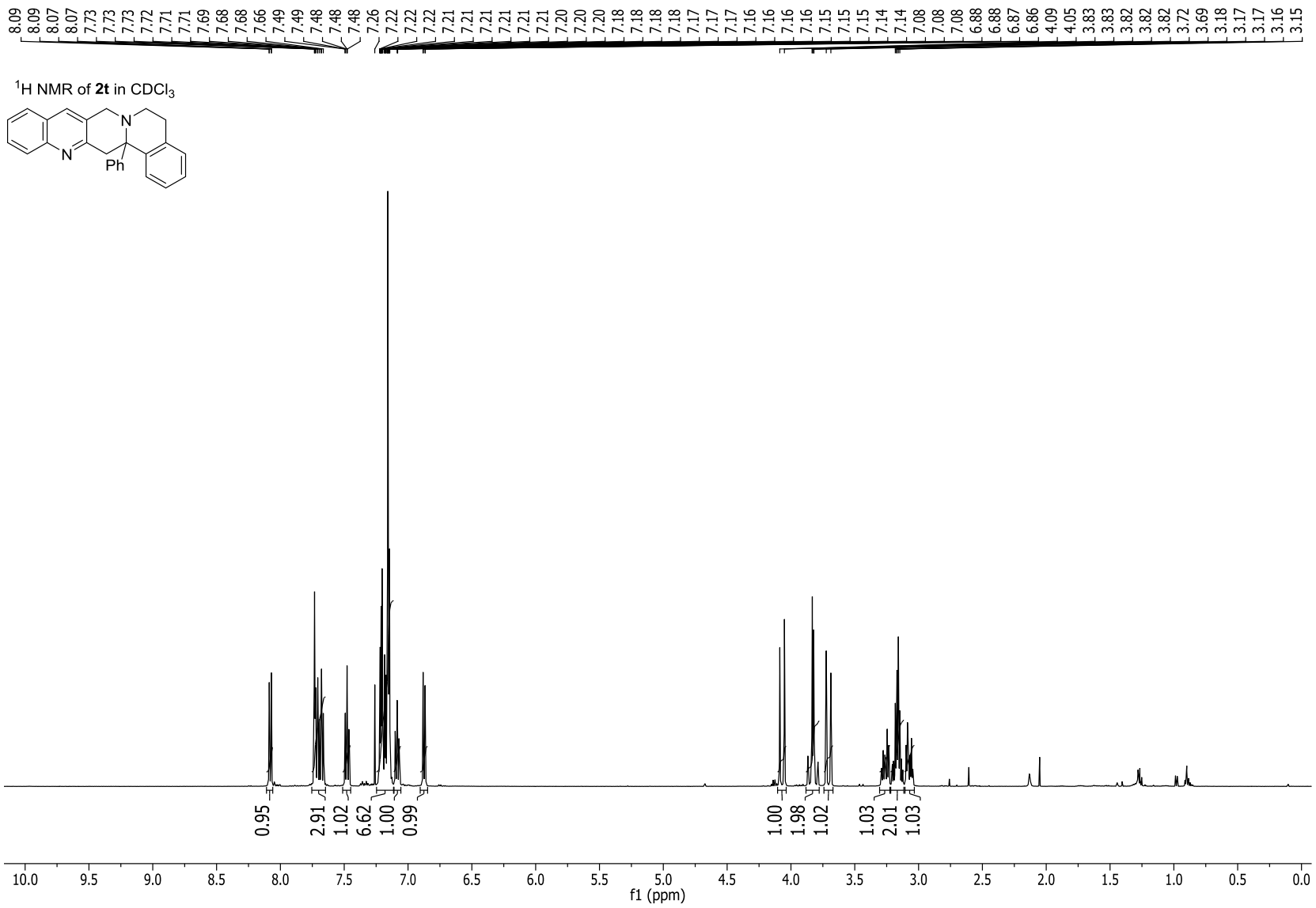

156.38
147.52
147.48
146.94
132.48
129.03
128.95
128.30
128.12
127.17
126.92
126.15
125.85
111.21
108.17
77.42
77.36
77.16
76.91
59.56
57.81
55.82
55.78
51.42
40.80
28.89
13C NMR of 2q in CDCl3
N
N
OMe
OMe
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

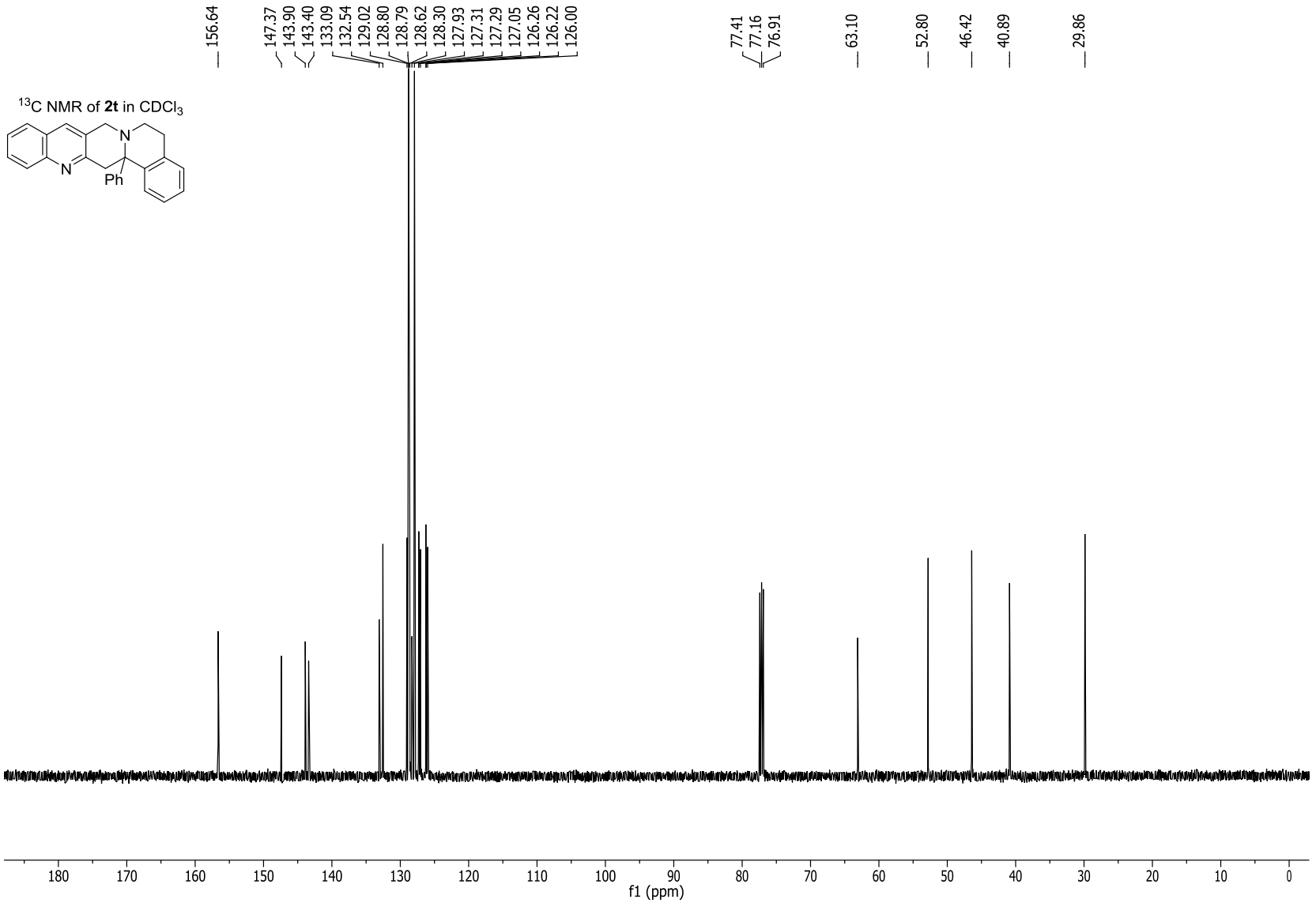

^{1}H NMR of **2r** in DMSO-d$_6$
10.99
8.07 7.94 7.93 7.89 7.88 7.70 7.70 7.69 7.68 7.68 7.67 7.67 7.54 7.52 7.51 7.44 7.42 7.38 7.36 7.09 7.08 7.06 7.01 6.99 6.98
4.29 4.26 3.84 3.81 3.80 3.30 3.29 3.27 3.26 3.04 3.02 3.01 2.98 2.89 2.89 2.88 2.77 2.76 2.76 2.74 2.73 2.73 2.67 2.66 2.65
1.05 0.98 0.93 0.97 1.05 1.06 1.01 0.95 1.01 1.02 1.00 2.78 1.00 1.02 1.00 0.98 1.00
f1 (ppm)

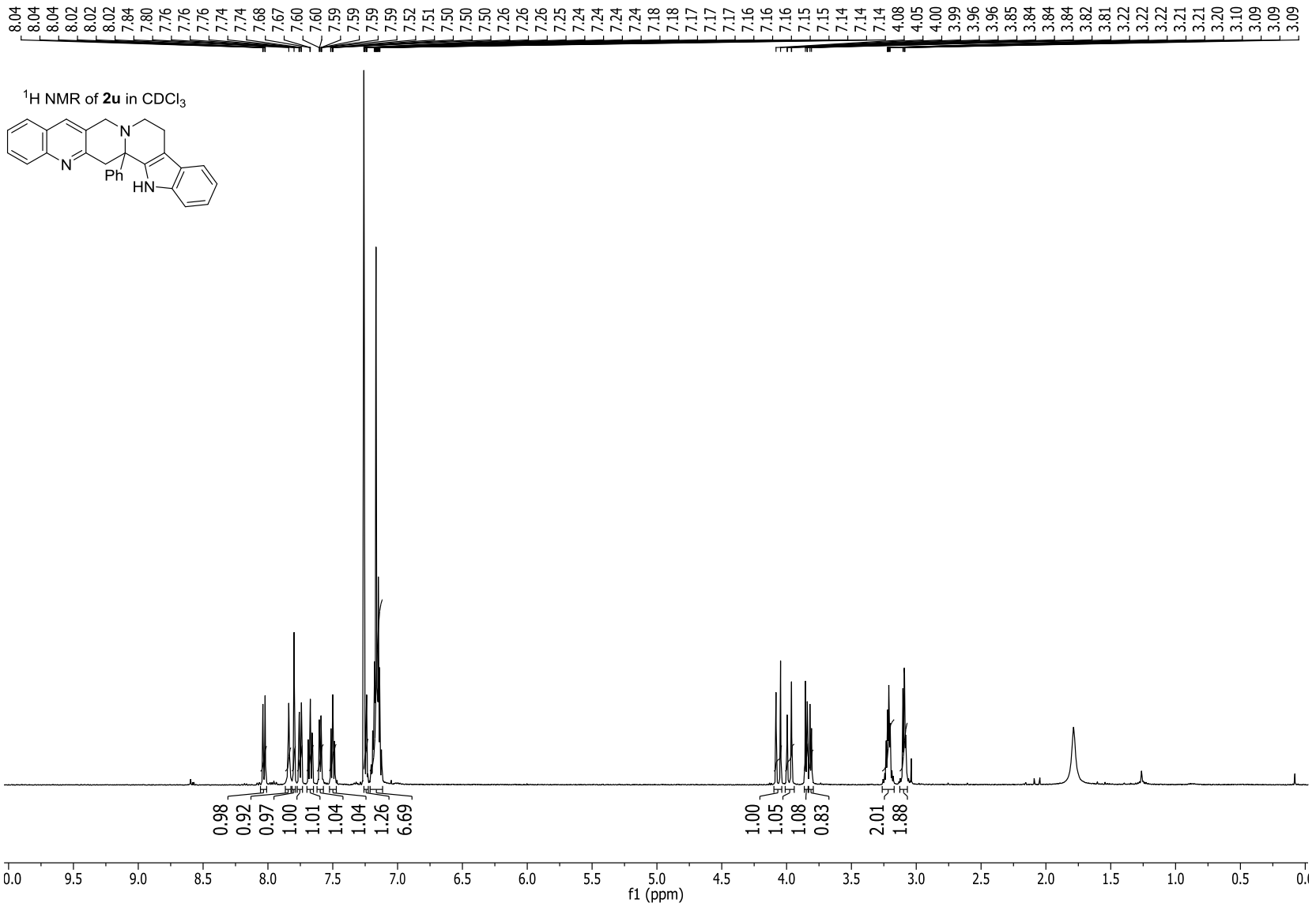

155.84
146.41
136.29
134.89
132.49
128.87
128.60
128.09
127.45
126.48
120.69
118.44
106.61
56.37
56.14
51.56
40.02
39.85
39.69
39.52
39.35
39.19
39.02
38.35
21.25
13C NMR of 2r in DMSO-d6
N
N
HN
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

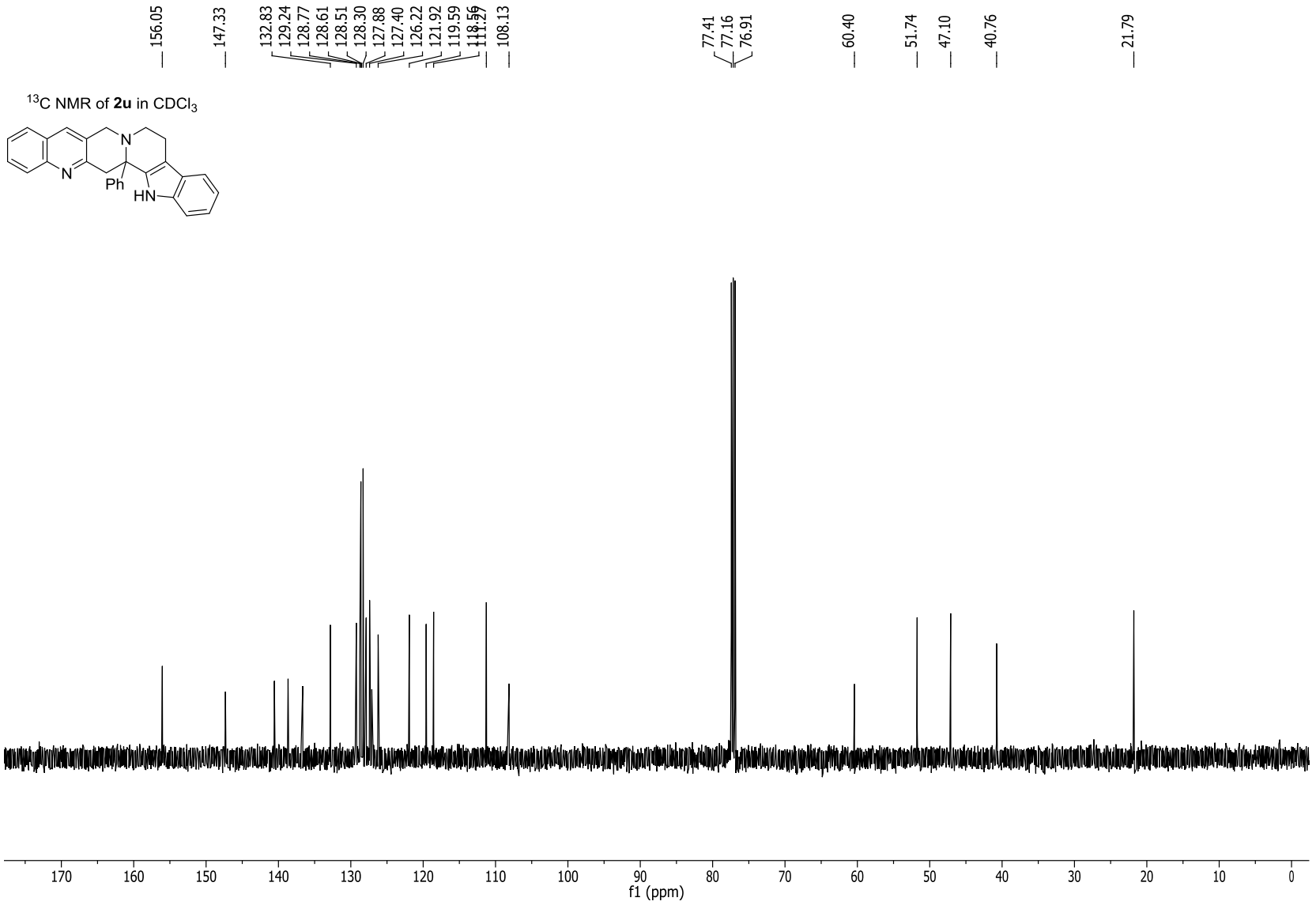

8.04
8.03
7.86
7.78
7.78
7.77
7.76
7.76
7.75
7.74
7.74
7.67
7.67
7.65
7.52
7.51
7.50
7.50
7.50
7.49
7.48
7.48
7.46
7.44
7.44
7.43
7.28
7.27
4.42
4.41
4.40
4.37
4.34
4.10
4.07
4.06
4.03
3.97
3.96
3.93
3.92
3.48
3.46
3.44
3.42
^{1}H NMR of **2s** in $CDCl_3$
N
N
0.98
1.00
2.86
1.03
3.91
1.00
0.97
1.88
2.00
1.06
1.03
0.0
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)

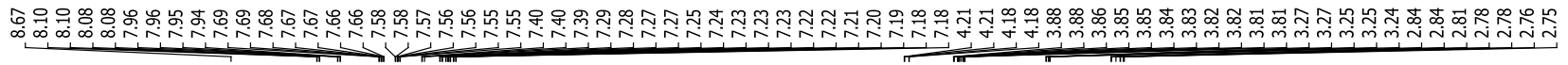

155.84
147.36
136.09
133.21
132.77
132.09
129.20
128.61
127.47
127.36
127.34
127.27
126.69
126.65
126.11
126.09
125.72
122.12
121.83
77.41
77.16
76.91
59.06
56.74
55.99
38.59
^{13}C NMR of **2s** in $CDCl_3$
N
N
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


8.09 8.09 8.07 8.07 7.73 7.73 7.73 7.72 7.71 7.71 7.69 7.68 7.68 7.66 7.49 7.49 7.48 7.48 7.48 7.26 7.22 7.22 7.21 7.21 7.21 7.21 7.21 7.20 7.20 7.20 7.18 7.18 7.18 7.18 7.17 7.17 7.17 7.16 7.16 7.16 7.15 7.15 7.15 7.14 7.14 7.08 7.08 7.08 6.88 6.88 6.87 6.86 4.09 4.05 3.83 3.83 3.82 3.82 3.82 3.72 3.69 3.18 3.17 3.17 3.16 3.15
1H NMR of 2t in CDCl3
N
N
Ph
0.95 2.91 1.02 6.62 1.00 0.99 1.00 1.98 1.02 1.03 2.01 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)

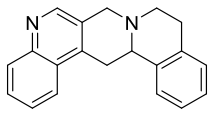

156.64
147.37
143.90
143.40
133.09
132.54
129.02
128.80
128.79
128.62
128.30
127.93
127.31
127.29
127.05
126.26
126.22
126.00
77.41
77.16
76.91
63.10
52.80
46.42
40.89
29.86
13C NMR of 2t in CDCl3
N
N
Ph
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

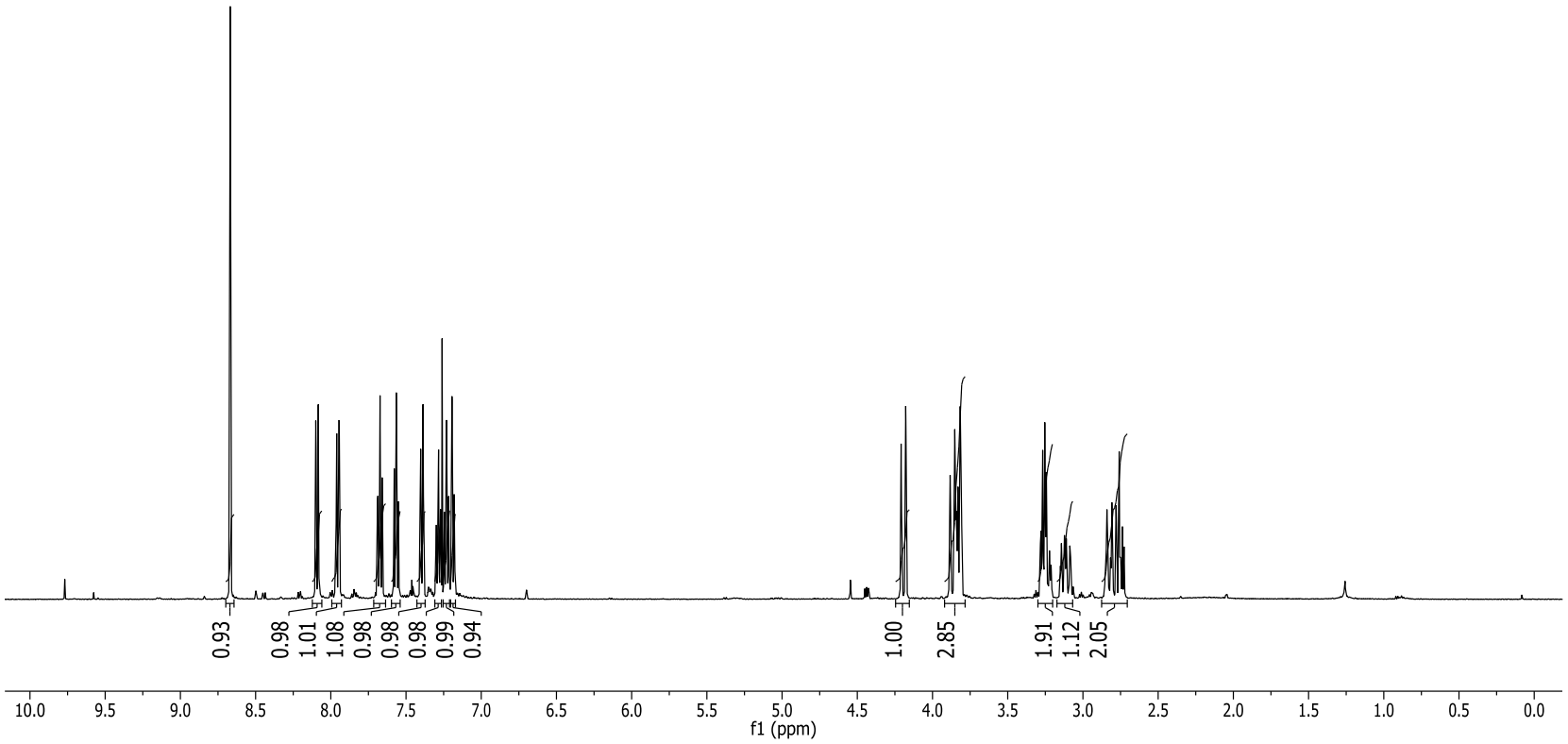

^{1}H NMR of **2u** in $CDCl_3$
N
N
Ph
HN
8.04 8.04 8.04 8.02 8.02 8.02 7.84 7.80 7.76 7.76 7.76 7.74 7.74 7.68 7.67 7.60 7.60 7.59 7.59 7.59 7.59 7.52 7.51 7.50 7.50 7.50 7.26 7.26 7.25 7.24 7.24 7.24 7.24 7.18 7.18 7.17 7.17 7.17 7.16 7.16 7.15 7.15 7.14 7.14 7.14 4.08 4.05 4.00 3.99 3.96 3.96 3.85 3.84 3.84 3.84 3.82 3.81 3.22 3.22 3.22 3.21 3.21 3.20 3.10 3.09 3.09 3.09
0.98 0.92 0.97 1.00 1.01 1.04 1.04 1.26 6.69
1.00 1.05 1.08 0.83
2.01 1.88
0.0 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)

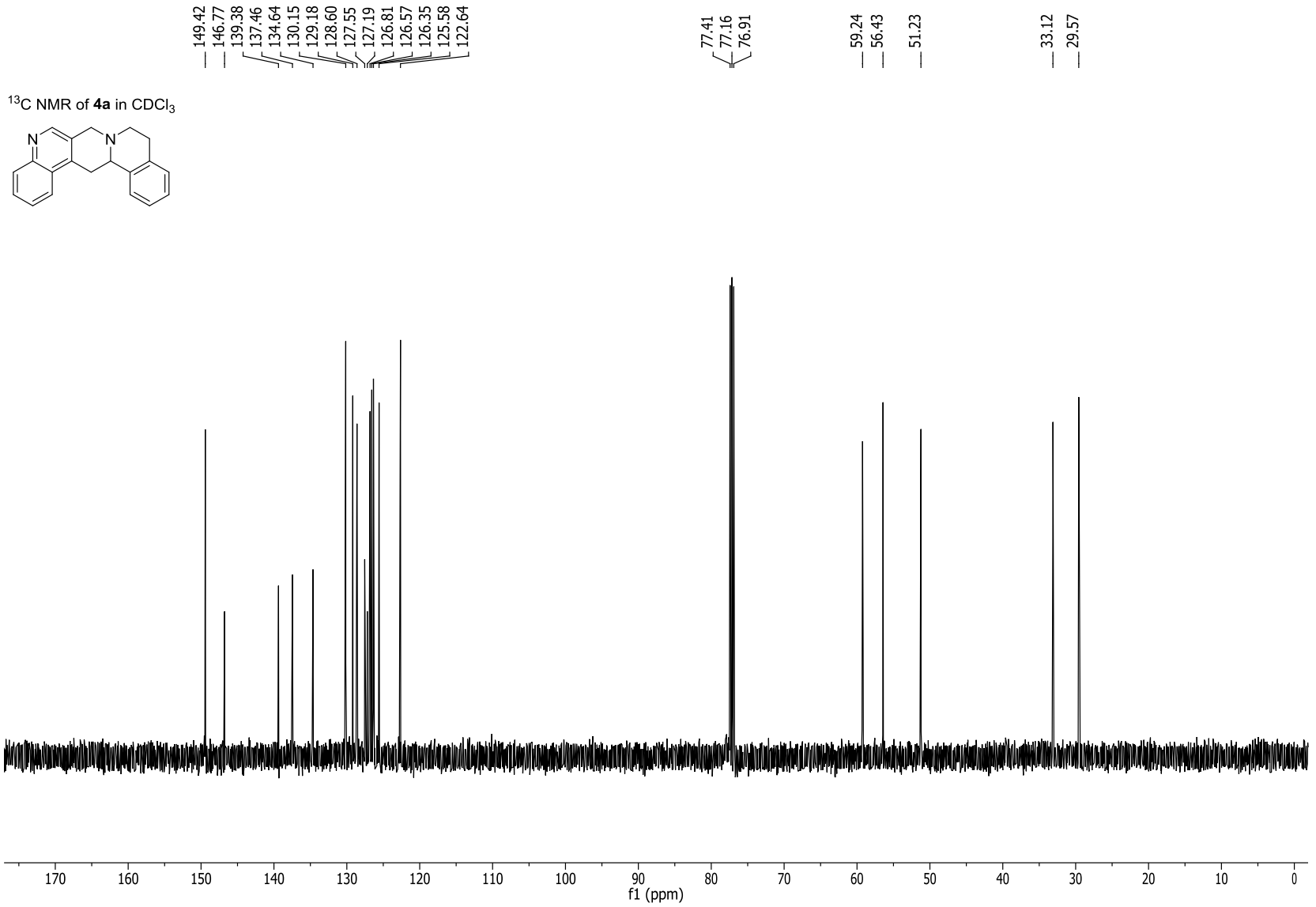

156.05
147.33
132.83
129.24
128.77
128.61
128.51
128.30
127.88
127.40
126.22
121.92
119.59
118.56
111.27
108.13
77.41
77.16
76.91
60.40
51.74
47.10
40.76
21.79
13C NMR of 2u in CDCl3
N
N
Ph
HN
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

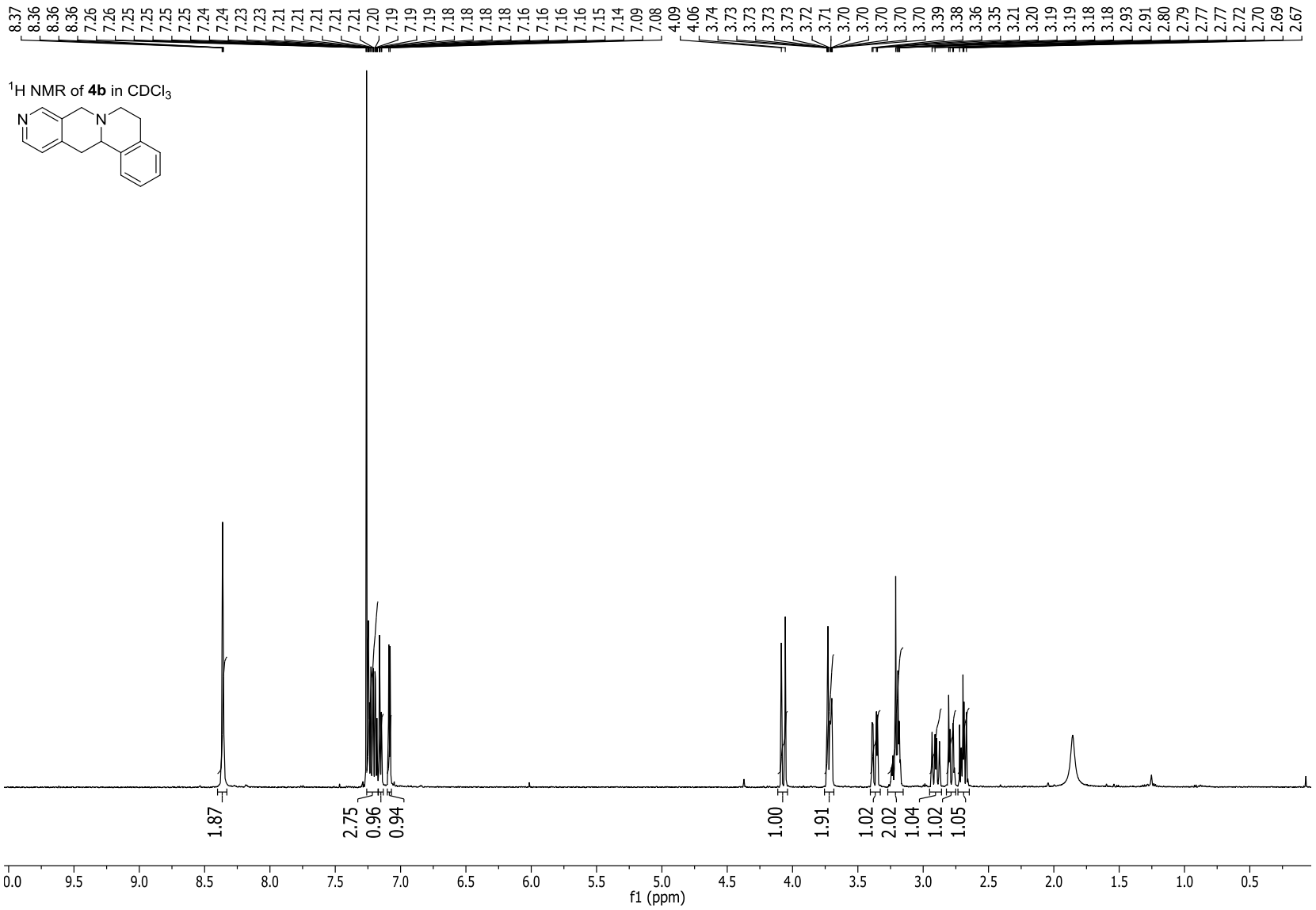


^{1}H NMR of **4a** in $CDCl_3$

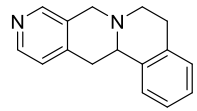


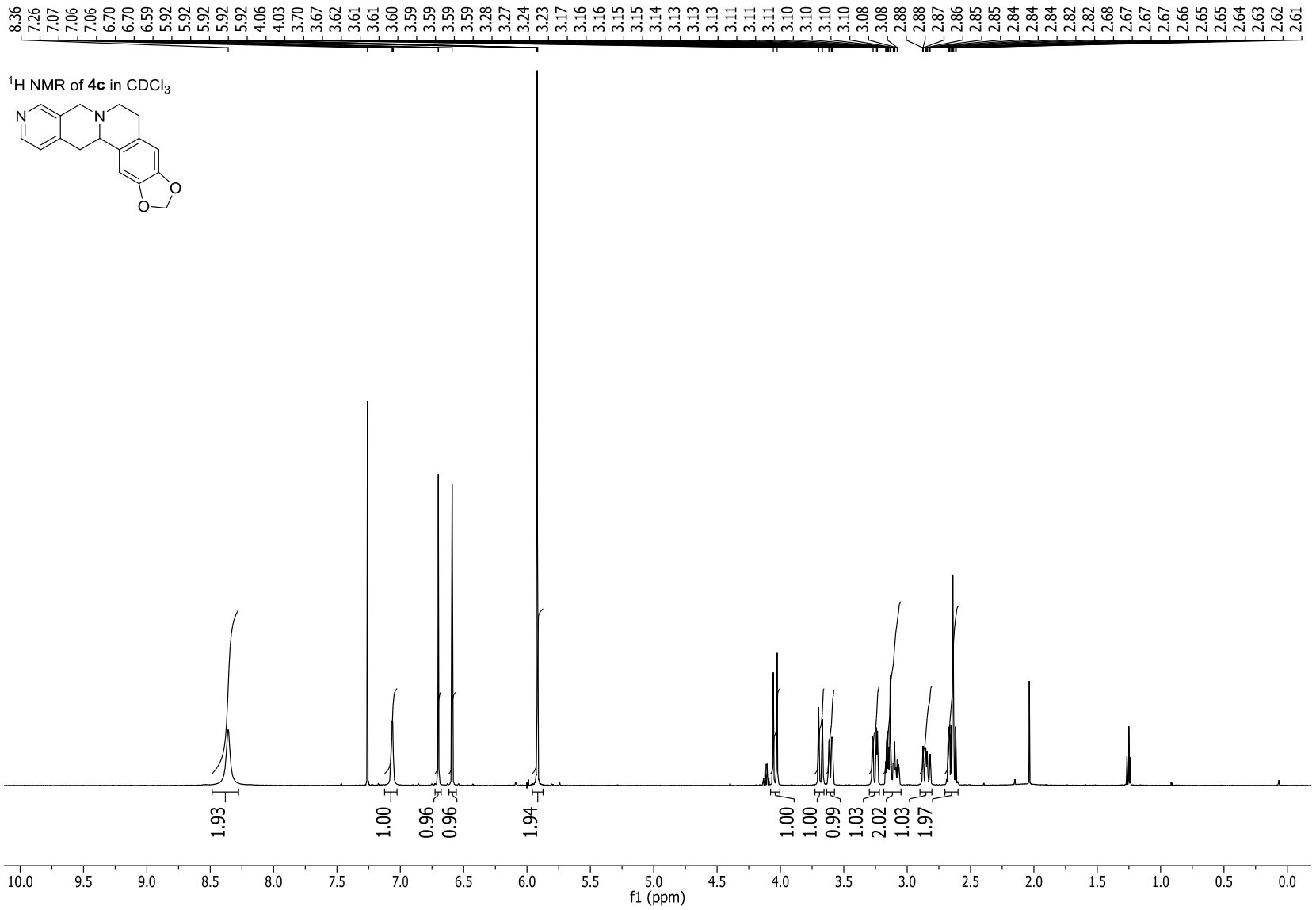


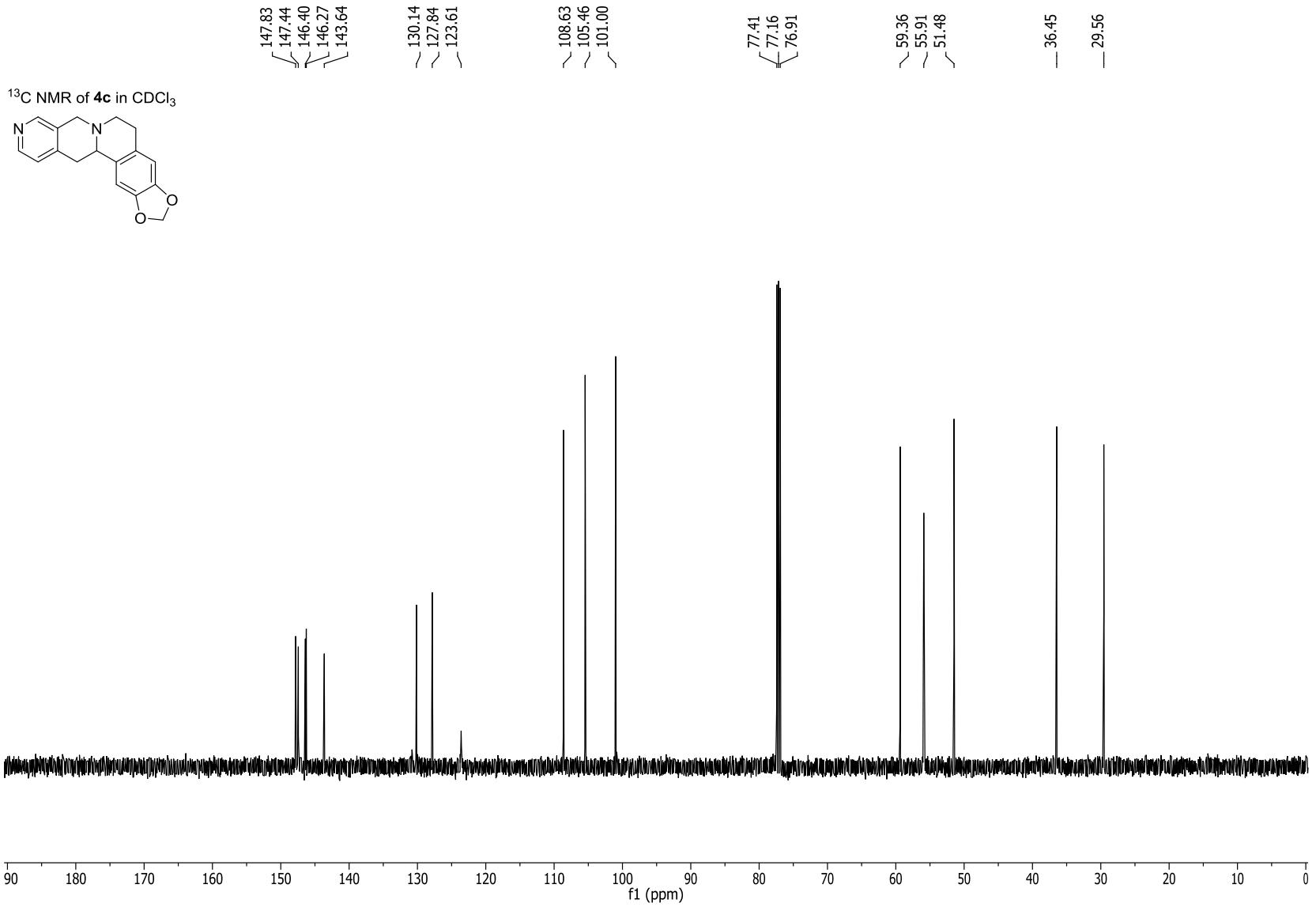

149.42
146.77
139.38
137.46
134.64
130.15
129.18
128.60
127.55
127.19
126.81
126.57
126.35
125.58
122.64
77.41
77.16
76.91
59.24
56.43
51.23
33.12
29.57
^{13}C NMR of **4a** in $CDCl_3$
N
N
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

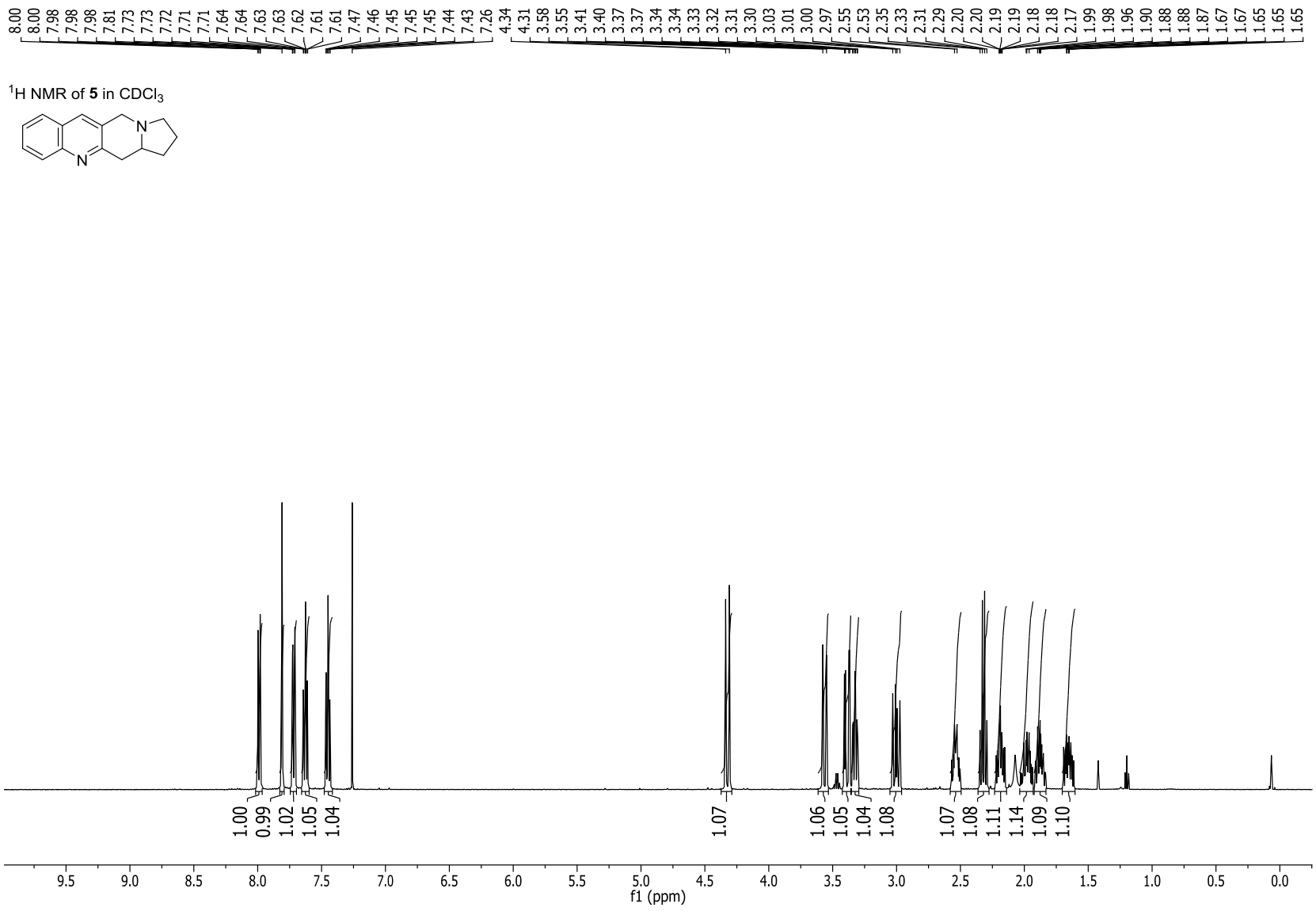

8.37 8.36 8.36 8.36 7.26 7.26 7.25 7.25 7.25 7.24 7.24 7.23 7.23 7.21 7.21 7.21 7.21 7.20 7.19 7.19 7.18 7.18 7.18 7.16 7.16 7.16 7.15 7.14 7.09 7.08
4.09 4.06 3.74 3.73 3.73 3.73 3.73 3.72 3.71 3.70 3.70 3.70 3.70 3.70 3.39 3.38 3.36 3.35 3.21 3.20 3.19 3.19 3.18 3.18 2.93 2.91 2.80 2.79 2.77 2.77 2.72 2.70 2.69 2.67
1H NMR of 4b in CDCl3
N
N
1.87
2.75
0.96
0.94
1.00
1.91
1.02
2.02
1.04
1.02
1.05
0.0 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)

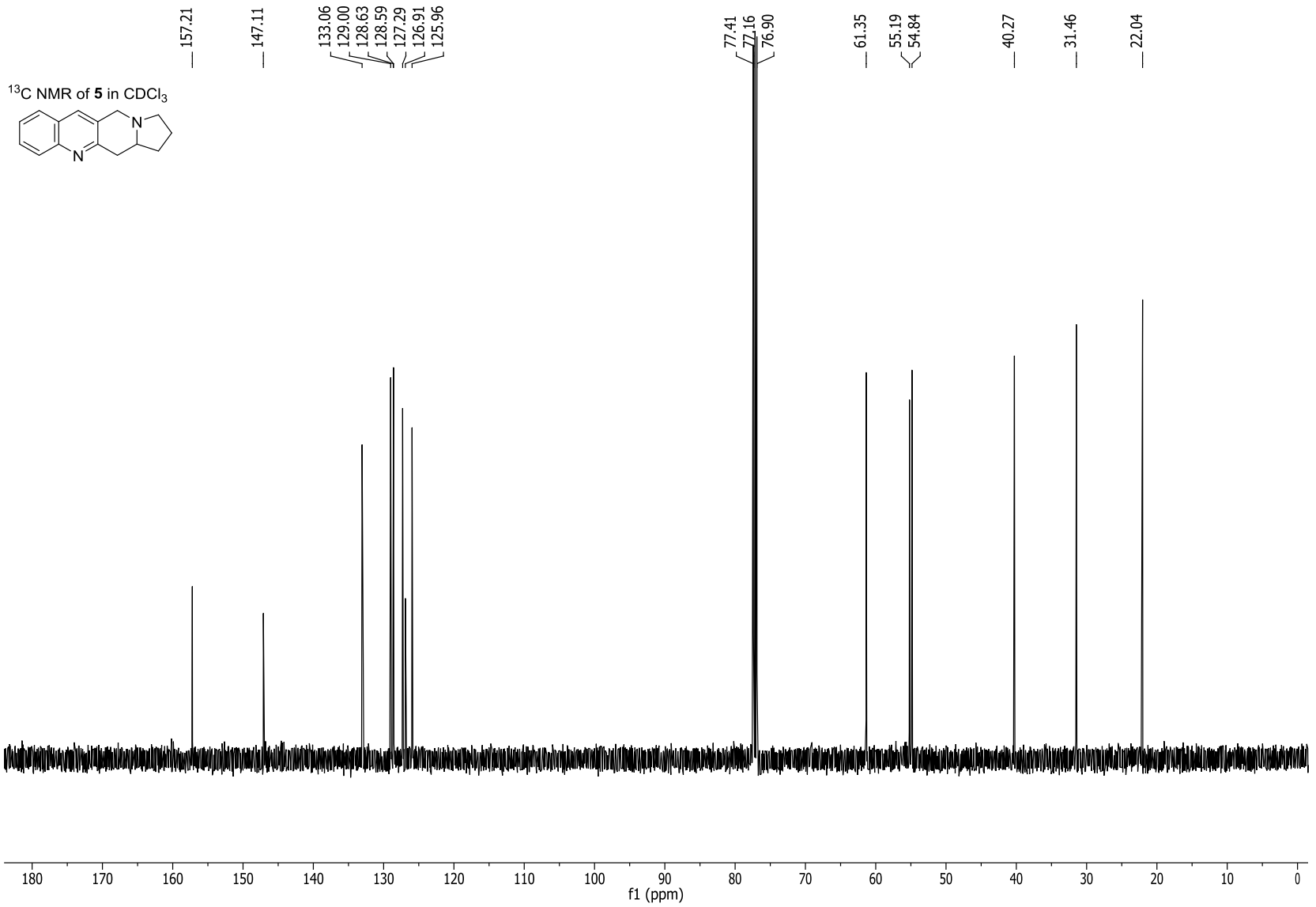
^{13}C NMR of **4b** in $CDCl_3$




8.36 7.26 7.07 7.06 7.06 6.70 6.70 6.59 5.92 5.92 5.92 5.92 5.92 4.06 4.03 3.70 3.67 3.62 3.61 3.61 3.60 3.59 3.59 3.59 3.59 3.28 3.27 3.24 3.23 3.17 3.16 3.16 3.15 3.15 3.14 3.13 3.13 3.13 3.11 3.11 3.11 3.10 3.10 3.10 3.10 3.08 3.08 2.88 2.88 2.87 2.86 2.85 2.85 2.84 2.84 2.84 2.82 2.82 2.68 2.67 2.67 2.67 2.66 2.65 2.65 2.64 2.63 2.62 2.61
1H NMR of 4c in CDCl3
N
N
O
O
1.93
1.00
0.96
0.96
1.94
1.00
1.00
0.99
1.03
2.02
1.03
1.97
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)


147.83
147.44
146.40
146.27
143.64
130.14
127.84
123.61
108.63
105.46
101.00
77.41
77.16
76.91
59.36
55.91
51.48
36.45
29.56
13C NMR of 4c in CDCl3
N
N
O
O
90
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


8.00 8.00 7.98 7.98 7.98 7.81 7.73 7.73 7.72 7.71 7.71 7.64 7.64 7.63 7.63 7.62 7.61 7.61 7.47 7.46 7.45 7.45 7.44 7.43 7.26
4.34 4.31 3.58 3.55 3.41 3.40 3.37 3.37 3.34 3.34 3.33 3.32 3.31 3.30 3.03 3.01 3.00 2.97 2.55 2.53 2.35 2.33 2.31 2.29 2.20 2.20 2.19 2.19 2.18 2.18 2.17 1.99 1.98 1.96 1.90 1.88 1.88 1.87 1.67 1.67 1.65 1.65 1.65
1H NMR of 5 in CDCl3
N
N
1.00 0.99 1.02 1.05 1.04 1.07 1.06 1.05 1.04 1.08 1.07 1.08 1.11 1.14 1.09 1.10
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)


157.21
147.11
133.06
129.00
128.63
128.59
127.29
126.91
125.96
77.41
77.16
76.90
61.35
55.19
54.84
40.27
31.46
22.04
13C NMR of 5 in CDCl3
N
N
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)