

Supporting Information

Environmentally Benign Decarboxylative N-, O-, and S-Acetylations and Acylations

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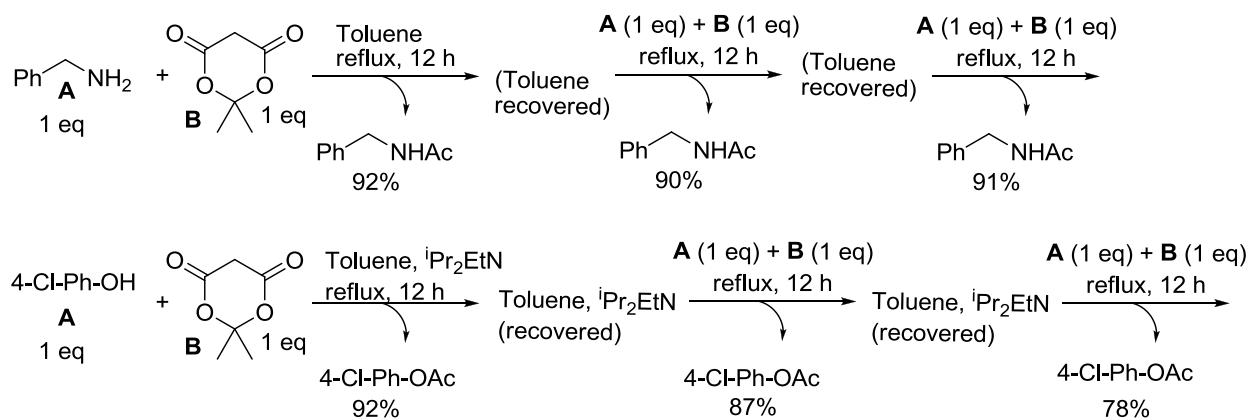
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Experimental:

General: All reactions involving air- or moisture-sensitive reagents or intermediates were carried out in oven-dried glassware under an argon atmosphere. Dichloromethane (CH_2Cl_2) was freshly distilled from phosphorus (V)oxide (P_2O_5). Commercial grade xylene, benzene and toluene were distilled over CaH_2 before use. All other solvents and reagents were purified according to standard procedures or were used as received from Aldrich, Acros, Merck and Spectrochem. ^1H , ^{13}C NMR spectroscopy: *Bruker 400 MHz*, *Bruker 600 MHz* (at 298 K). Chemical shifts, δ (in ppm), are reported relative to TMS (^1H) 0.0 ppm, δ (^{13}C) 0.0 ppm) which was used as the inner reference. Otherwise the solvents residual proton resonance and carbon resonance (CHCl_3 , δ (^1H) 7.26 ppm, δ (^{13}C) 77.2 ppm; CD_3OD , (^1H) 3.31 ppm, δ (^{13}C) 49.0 ppm) were used for calibration. Column chromatography: Merck or Spectrochem silica gel 60-120 under gravity.. MS (ESI-HRMS): Mass spectra were recorded on an Agilent Accurate-Mass Q-TOF LC/MS 6520, and peaks are given in m/z (% of basis peak).

Scheme s1: Solvent and catalyst recycling



General procedure for the acetylation of amine (GP I): Meldrum's acid (1eq.) was added to a solution of amine (1eq) in toluene (2 - 4 mL) and the mixture was refluxed for 12 h. After disappearance of starting materials (indicated by TLC) solvent was evaporated under reduced pressure to obtain the analytically pure product.

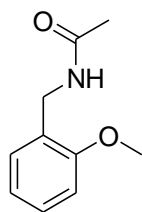
***N*-benzylacetamide (3a)¹²:** According to GP I: Benzylamine (0.2 g, 1.86 mmol) and Meldrum's

acid **2** (0.27 g, 1.86 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product **3a** as pale yellow liquid (0.25 g, 90%). ¹H NMR (400 MHz, CDCl₃) δ = 7.18 (br. s, 1H), 7.17 - 7.07 (m, 5H), 4.16 (d, *J* = 5.8 Hz, 2H), 1.75 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 170.7, 138.5, 128.5, 127.6, 127.6, 127.2, 43.4, 22.9 ppm.

***N*-((*R*)-1-phenylethyl)acetamide (3b)⁹:** According to GP I: (*S*)-(-)- α -Methylbenzylamine (0.20

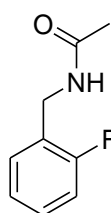
g, 1.65 mmol) and Meldrum's acid **2** (0.24 g, 1.65 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product **3b** as pale yellow liquid (0.26 g, 96%). ¹H NMR (600 MHz, CDCl₃) δ = 7.37 - 7.33 (m, 4H), 7.30 - 7.27 (m, 1H), 6.21 (br. s, 1H), 5.15 - 5.10 (m, 1H), 2.01 (s, 3H), 1.51 (d, *J* = 6.6 Hz, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 169.5, 143.0, 128.7, 127.5, 126.2, 49.0, 23.2, 21.7 ppm.

N-benzylacetamide (3c)¹²: According to GP I: 2-Methoxybenzylamine (0.20 g, 1.46 mmol) and



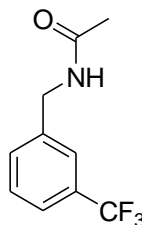
Meldrum's acid **2** (0.21 g, 1.46 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product **3c** as pale yellow liquid (0.24 g, 91%). ¹H NMR (600 MHz, CDCl₃) δ = 7.21 - 7.17 (m, 2H), 6.86 - 6.83 (m, 1H), 6.80 (d, *J* = 8.0 Hz, 2H, one NH is there, not distinguishable), 4.34 (d, *J* = 6.0 Hz, 2H), 3.77 (s, 3H), 1.89 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 170.3, 157.3, 129.2, 128.6, 126.3, 120.5, 110.2, 55.2, 39.0, 23.0 ppm.

N-(2-fluorobenzyl)acetamide (3d)⁷: According to GP I: 2-Fluorobenzylamine (0.20 g, 1.60



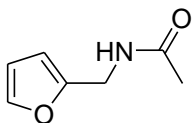
mmol) and Meldrum's acid **2** (0.23 g, 1.60 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product **3d** as pale yellow liquid (0.25 g, 92%). ¹H NMR (600 MHz, CDCl₃) δ = 7.24 - 7.21 (m, 1H), 7.18 - 7.15 (m, 1H), 7.02 - 6.99 (m, 2H, one NH, not distinguishable), 6.96 - 6.93 (m, 1H), 4.36 (d, *J* = 5.4 Hz, 2H), 1.91 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 170.7, 161.6, 160.0, 129.96, 129.94, 129.1, 129.0, 125.35, 125.25, 124.17, 124.15, 115.3, 115.1, 37.29, 37.26, 22.83, 22.81 ppm.

N-(3-(trifluoromethyl)benzyl)acetamide (3e)¹²: According to GP I: 3-(Trifluoromethyl)benzylamine (0.20 g, 1.14 mmol) and Meldrum's acid **2** (0.16 g, 1.14 mmol) was reacted for 12



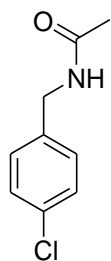
h and evaporation of the solvent gave analytically pure product **3e** as white solid (0.23 g, 94%). ¹H NMR (400 MHz, CDCl₃) δ = 7.39 - 7.36 (m, 2H), 7.32 - 7.28 (m, 2H), 6.97 (br. s, 1H), 4.16 (d, *J* = 5.8 Hz, 2H), 1.75 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 170.8, 139.5, 131.0, 130.9, 130.9, 130.6, 129.1, 129.0, 124.9, 124.2 (CF₃), 124.15 (CF₃), 124.12 (CF₃), 124.09 (CF₃), 124.06 (CF₃), 124.04 (CF₃), 123.1, 42.9, 42.9, 22.8 ppm.

N-((furan-2-yl)methyl)acetamide (3f)¹¹: According to GP I: Furfurylamine (0.20 g, 2.05 mmol) and Meldrum's acid **2** (0.30 g, 2.05 mmol) was reacted for 12 h and evaporation of the solvent



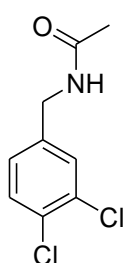
gave analytically pure product **3f** as pale yellow liquid (0.25 mg, 88%). ¹H NMR (400 MHz, CDCl₃) δ = 7.30 - 7.29 (m, 1H), 6.85 (br. s, 1H), 6.27 - 6.26 (m, 1H), 6.17 - 6.16 (m, 1H), 4.34 (d, *J* = 5.4 Hz, 2H), 1.94 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 170.5, 151.4, 142.0, 110.4, 107.3, 36.5, 22.9 ppm.

N-(4-chlorobenzyl)acetamide (3g)¹²: According to GP I: 4-Chlorobenzylamine (0.20 g, 1.42



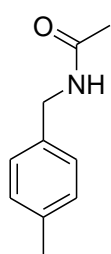
mmol) and Meldrum's acid **2** (0.20 g, 1.42 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product **3g** as pale yellow liquid (0.24 g, 92%). ¹H NMR (600 MHz, CDCl₃) δ = 7.28 (d, *J* = 7.8 Hz, 2H), 7.19 (d, *J* = 8.4 Hz, 2H), 6.10 (br. s, 1H), 4.36 (d, *J* = 5.8 Hz, 2H), 2.01 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 170.2, 136.7, 133.3, 129.2, 128.8, 43.0, 23.2 ppm.

N-(3,4-dichlorobenzyl)acetamide (3h)²⁰: According to GP I: 3,4-Dichlorobenzylamine (0.20 g,



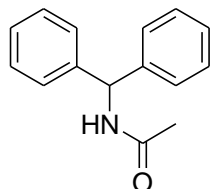
1.14 mmol) and Meldrum's acid **2** (0.16 g, 1.14 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product **3h** as white solid (0.24 mg, 95%). ¹H NMR (600 MHz, CDCl₃) δ = 7.35 (d, *J* = 8.4 Hz, 1H), 7.32 (d, *J* = 1.8 Hz, 1H), 7.32 (dd, *J* = 7.8 Hz, *J* = 1.8 Hz, 1H), 6.57 (br. s, 1H), 4.31 (d, *J* = 6.0 Hz, 2H), 2.00 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 170.5, 138.6, 132.6, 131.4, 130.6, 129.5, 127.0, 42.5, 23.1 ppm.

N-(4-methylbenzyl)acetamide (3i)¹²: According to GP I: 4-Methylbenzylamine (0.20 g, 1.46



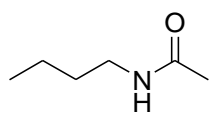
mmol) and Meldrum's acid **2** (0.21 g, 1.46 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product **3i** as pale yellow liquid (0.24 g, 93%). ¹H NMR (600 MHz, CDCl₃) δ = 7.15 - 7.11 (m, 4H), 6.21 (br. s, 1H), 4.33 (d, *J* = 5.4 Hz, 2H), 2.32 (s, 3H), 1.97 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 170.7, 138.5, 128.5, 127.6, 127.2, 43.4, 22.9 ppm.

N-benzhydrylacetamide (3j)²: According to GP I: Benzhydrylamine (0.20 g, 1.09 mmol) and



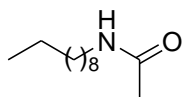
Meldrum's acid **2** (0.16 g, 1.09 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product **3j** as white solid (0.24 g, 96%). ¹H NMR (600 MHz, CDCl₃) δ = 7.36 - 7.33 (m, 4H), 7.30 - 7.28 (m, 2H), 7.26 - 7.23 (m, 4H), 6.49 (br. s, 1H), 6.25 (d, *J* = 8.4 Hz, 1H), 2.06 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 169.5, 141.4, 128.7, 127.5, 127.4, 57.2, 23.2 ppm.

N-butylacetamide (3k)⁵: According to GP I: Butylamine (0.1 g, 0.136 mmol) and Meldrum's acid **2** (0.20 g, 1.36 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure



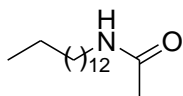
product **3k** as pale yellow liquid (0.14 g, 92%). ¹H NMR (600 MHz, CDCl₃) δ = 6.52 (br. s, 1H), 3.14 - 3.11 (m, 2H), 1.89 (s, 3H), 1.42 - 1.35 (m, 2H), 1.28 - 1.22 (m, 2H), 0.82 (t, *J* = 7.8 Hz, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 170.5, 39.3, 31.5, 23.0, 20.0, 13.7 ppm.

N-decylacetamide (3l)²⁵: According to GP I: Decylamine (0.20 g, 1.08 mmol) and Meldrum's acid **2** (0.16 g, 1.08 mmol) was reacted for 12 h and evaporation of the solvent



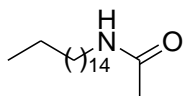
gave analytically pure product **3l** as pale yellow gum (0.19 g, 90%). ¹H NMR (400 MHz, CDCl₃) δ = 6.28 (br. s, 1H), 3.17 - 3.12 (m, 2H), 1.91 (s, 3H), 1.44 - 1.41 (m, 2H), 1.25 - 1.19 (m, 14H), 0.80 (t, *J* = 6.6 Hz, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 170.2, 39.7, 31.8, 29.5, 29.3, 29.3, 26.9, 23.1, 22.6, 14.1 ppm (Total count of ¹³C is less than expected due to the merging of signal in the aliphatic region.).

N-tetradecylacetamide (3m)²⁵: According to GP I: Tetradecylamine (0.20 g, 0.94 mmol) and Meldrum's acid **2** (0.14 g, 0.94 mmol) was reacted for 12 h and evaporation of the solvent gave



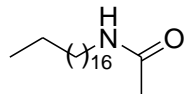
analytically pure product **3m** as white solid (0.21 g, 87%). ¹H NMR (400 MHz, CDCl₃) δ = 5.85 (br. s, 1H), 3.22 - 3.17 (m, 2H), 1.96 (s, 3H), 1.50 - 1.44 (m, 2H), 1.27 - 1.21 (m, 22H), 0.85 (t, *J* = 6.8 Hz, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 170.2, 39.8, 31.9, 29.7, 29.7, 29.64, 29.58, 29.56, 29.54, 29.34, 29.30, 26.9, 23.2, 22.7, 14.1 ppm. Total count of ¹³C is less than expected due to the merging of signal in the aliphatic region.

N-hexadecylacetamide (3n)²⁶: According to GP I: Hexadecylamine (0.20 g, 0.83 mmol) and Meldrum's acid **2** (0.12 g, 0.83 mmol) was reacted for 12 h and evaporation of the solvent gave



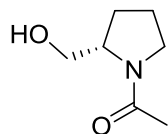
analytically pure product **3n** as white solid (0.22 mg, 92%). ¹H NMR (600 MHz, CDCl₃) δ = 5.90 (br. s, 1H), 3.21 - 3.18 (m, 2H), 1.96 (s, 3H), 1.49 - 1.44 (m, 2H), 1.28 - 1.21 (m, 26H), 0.85 (t, *J* = 7.0 Hz, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 170.22, 39.76, 31.93, 29.71, 29.69, 29.67, 29.61, 29.56, 29.38, 29.33, 26.95, 23.24, 22.70, 14.14 ppm. Total count of ¹³C is less than expected due to the merging of signal in the aliphatic region.

N-octadecylacetamide (3o)²⁷: According to GP I: Octadecylamine (0.30 g, 1.12 mmol) and Meldrum's acid **2** (0.16 g, 1.12 mmol) was reacted for 12 h and evaporation of the solvent gave



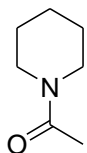
analytically pure product **3o** as white solid (0.33 g, 95%). ¹H NMR (400 MHz, CDCl₃) δ = 5.89 (br. s, 1H), 3.20 - 3.17 (m, 2H), 1.95 (s, 3H), 1.48 - 1.45 (m, 2H), 1.29 - 1.22 (s, 30H), 0.85 (t, *J* = 6.8 Hz, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 170.2, 39.7, 31.9, 29.69, 29.67, 29.65, 29.59, 29.56, 29.55, 29.35, 29.31, 26.9, 23.2, 22.7, 14.1 ppm. Total count of ¹³C is less than expected due to the merging of signal in the aliphatic region.

((S)-1-acetylpyrrolidin-2-yl)methyl acetate (3p)¹⁴: According to GP I: (S)-Prolinol (0.20 g, 0.99 mmol) and Meldrum's acid **2** (0.14 g, 0.99 mmol) was reacted for 12 h and



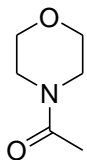
evaporation of the solvent gave analytically pure product **3p** as liquid oil (0.13 g, 92%). ¹H NMR (400 MHz, CDCl₃) δ = 4.23 (br. s, 1H), 4.15 - 4.09 (m, 1H), 3.60 - 3.50 (m, 2H), 3.46 - 3.42 (m, 2H), 2.04 (s, 3H), 2.00 - 1.82 (m, 3H), , 1.63 - 1.55 (m, 1H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 172.3, 67.4, 61.3, 49.2, 28.6, 24.5, 23.1 ppm.

1-(piperidin-1-yl)ethanone (3q)¹¹: According to GP I: Piperidine (0.10 g, 1.18 mmol) and Meldrum's acid **2** (0.17 g, 1.18 mmol) was reacted for 12 h and evaporation of the solvent gave



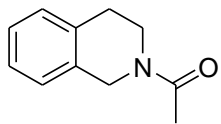
analytically pure product **3q** as pale yellow oil (0.14 g, 94%). ¹H NMR (600 MHz, CDCl₃) δ = 3.31 - 3.29 (m, 2H), 3.18 - 3.16 (m, 2H), 1.84 (s, 3H), 1.42 - 1.39 (m, 2H), 1.36 - 1.34 (m, 2H), 1.31 - 1.28 (m, 2H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 168.5, 47.2, 42.2, 26.2, 25.3, 24.3, 21.3 ppm.

1-Morpholinoethanone (3r)¹¹: According to GP I: Morpholine (0.1 g, 1.15 mmol) and Meldrum's acid **2** (0.17 g, 1.15 mmol) was reacted for 12 h and evaporation of the solvent gave



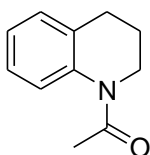
analytically pure product **3r** as pale yellow oil (0.14 g, 96%). ¹H NMR (600 MHz, CDCl₃) δ = 3.57 - 3.52 (m, 4H), 3.48 - 3.46 (m, 2H), 3.54 - 3.33 (m, 2H), 1.86 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 169.1, 66.7, 66.5, 46.5, 41.6, 21.0 ppm.

1-(3,4-dihydroisoquinolin-2(1H)-yl)ethanone (3s)¹⁵: According to GP I: Tetrahydroisoquinilone (0.20 g, 1.50 mmol) and Meldrum's acid **2** (0.22 g, 1.50 mmol) was reacted



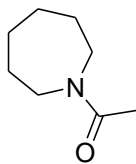
for 12 h and evaporation of the solvent gave analytically pure product **3s** as colourless oil with a mixture (1.3:1) of two rotamers (0.26 mg, 99%). ¹H NMR (600 MHz, CDCl₃) δ = 7.12 - 7.08 (m, 2H), 7.07 - 7.03 (m, 2H), 4.63 (s, 2H, major), 4.51 (s, 1H, minor), 3.71 (t, *J* = 6.0 Hz, 1H, minor), 3.56 (t, *J* = 5.9 Hz, 2H, major), 2.80 (t, *J* = 5.9 Hz, 2H, major), 2.74 (t, *J* = 5.9 Hz, 1H, minor), 2.09 (s, 2H, minor), 2.08 (s, 3H, major) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 169.5, 169.4, 135.0, 134.0, 133.4, 132.6, 128.9, 128.3, 126.9, 126.6, 126.5, 126.5, 126.3, 126.0, 48.0, 44.0, 43.9, 39.4, 29.4, 28.5, 22.0, 21.7 ppm.

1-(3,4-dihydroquinolin-1(2H)-yl)ethanone (3t)¹⁶: According to GP I: Tetrahydroquinoline (0.10 g, 0.75 mmol) and Meldrum's acid **2** (0.11 g, 0.75 mmol) was reacted for 12 h and



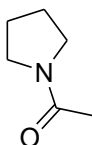
evaporation of the solvent gave analytically pure product **3t** as colourless oil (0.12 g, 93%). ¹H NMR (600 MHz, CDCl₃) δ = 7.17 - 7.08 (m, 4H), 3.80 - 3.73 (m, 2H), 2.71 - 2.68 (m, 2H), 2.20 (s, 3H), 1.95 - 1.91 (m, 2H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 170.1, 139.3, 133.6, 128.4, 126.1, 125.2, 124.6, 42.8, 26.9, 24.1, 23.2 ppm.

1-(azepan-1-yl)ethanone (3u)²⁴: According to GP I: Azepane (0.1 g, 1.01 mmol) and Meldrum's acid **2** (1.5 g, 1.01 mmol) was reacted for 12 h and evaporation the solvent gave



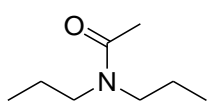
analytically pure product **3u** as pale yellow oil (0.14 mg, 95%). ¹H NMR (600 MHz, CDCl₃) δ = 3.41 - 3.39 (m, 2H), 3.32 - 3.30 (m, 2H), 1.98 (s, 3H), 1.62 - 1.59 (m, 4H), 1.45 - 1.44 (m, 4H). ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 170.3, 48.6, 45.7, 28.9, 27.5, 27.0, 26.8, 21.5 ppm.

1-(pyrrolidin-1-yl)ethanone (3v)¹¹: According to GP I: Pyrrolidine (0.20 g, 2.82 mmol) and Meldrum's acid **2** (0.41 g, 2.82 mmol) was reacted for 12 h and evaporation of the



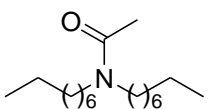
solvent gave get analytically pure product **3v** as pale yellow oil (0.28 g, 98%). ¹H NMR (400 MHz, CDCl₃) δ = 3.42 - 3.36 (m, 4H), 2.01 (s, 3H), 1.93 - 1.88 (m, 2H), 1.85 - 1.80 (m, 2H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 169.2, 47.4, 45.6, 26.1, 24.6, 22.4 ppm.

***N,N*-dipropylacetamide (3w)**²⁸: According to GP I: Dipropylamine (0.10 g, 0.99 mmol) and Meldrum's acid **2** (0.14 g, 0.99 mmol) was reacted for 12 h and evaporation the solvent gave



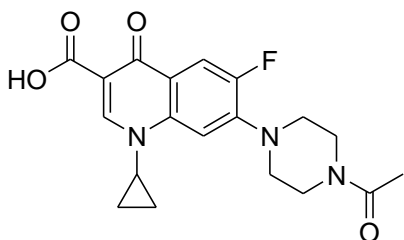
analytically pure product **3w** as pale yellow liquid (0.13 g, 93%). ¹H NMR (400 MHz, CDCl₃) δ = 3.20 (t, *J* = 8.0 Hz, 2H), 3.12 (t, *J* = 7.6 Hz, 2H), 2.01 (s, 3H), 1.55 - 1.45 (m, 4H), 0.87 - 0.79 (m, 6H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 170.1, 50.4, 47.3, 22.0, 21.5, 20.8, 11.3, 11.1 ppm.

***N,N*-dioctylacetamide (3x)**²⁹: According to GP I: Dioctylamine (0.20 g, 1.55 mmol) and Meldrum's acid **2** (0.22 g, 1.55 mmol) was reacted for 12 h and evaporation



the solvent gave analytically pure product **3x** as pale yellow liquid (0.26 g, 97%). ¹H NMR (600 MHz, CDCl₃) δ = 3.21 - 3.18 (m, 2H), 3.13 - 3.10 (m, 2H), 2.44 - 2.42 (m, 1H), 1.99 (s, 3H), 1.65 - 1.62 (m, 1H), 1.50 - 1.40 (m, 4H), 1.22 - 1.16 (m, 18H), 0.80 - 0.76 (m, 6H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 170.2, 48.8, 45.8, 31.8, 31.7, 29.3, 29.3, 29.18, 29.15, 28.8, 27.7, 27.03, 26.97, 26.8, 26.6, 22.6, 21.4, 14.0 ppm.

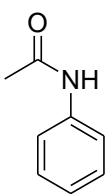
7-(4-acetylpiperazin-1-yl)-1-cyclopropyl-6-fluoro-1,4-dihydro-4-oxoquinoline-3-carboxylic acid (3y)³³: According to GP I: Ciprofloxacin (0.20 g, 0.60 mmol) and Meldrum's acid **2** (87 mg,



0.60 mmol) was reacted for 12 h and after evaporation of the solvent and washing with ethyl acetate gave analytically pure product **3y** as white solid (0.21 g, 93%). ¹H NMR (600 MHz, DMSO-*d*₆) δ = 8.61 (s, 1H), 7.82 (d, *J* = 13.2 Hz, 1H), 7.53 (d, *J* = 7.2 Hz, 1H), 3.79 - 3.78 (m, 1H), 3.68 - 3.66 (m, 4H), 3.36 - 3.34 (m, 2H), 3.29 - 3.27 (m, 2H), 2.07 (s, 3H), 1.33 - 1.32 (m, 2H), 1.18 - 1.17 (m, 2H) ppm. ¹³C NMR (151 MHz, DMSO-*d*₆) δ = 176.7, 169.0, 166.4, 154.2, 152.5, 148.4, 145.3, 139.4, 119.1, 119.1, 111.4, 111.3, 107.1, 106.9, 50.0, 49.6, 45.8, 40.9, 36.3, 21.6, 8.0 ppm. HRMS: Exact mass calculated for C₁₉H₂₀FN₃O₄ ([M+H]⁺): 374.1516, Found:374.1540.

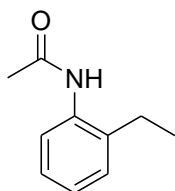
General procedure for the acetylation of aniline (GP II): Meldrum's acid (1eq.) was added to a solution of aniline (1eq) in toluene (2 - 4 mL) and the mixture was refluxed for 12 h. After cooling the reaction mixture, solid product was filtered and washed to get analytically pure product.

N-phenylacetamide (5a)¹: According to GP II: Aniline (0.20 g, 2.15 mmol) and Meldrum's acid



2 (0.31 g, 2.15 mmol) was reacted for 12 h and filtration and washing (EtOAc : hexane, 1:5) gave **5a** as white solid (0.27 g, 94%). ¹H NMR (400 MHz, CDCl₃) δ = 8.31 (br. s, 1H), 7.43 (d, *J* = 8.0 Hz, 2H), 7.21 - 7.16 (m, 2H), 7.01 - 6.98 (m, 1H), 2.05 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 169.4, 138.1, 128.9, 124.3, 120.3, 24.4 ppm.

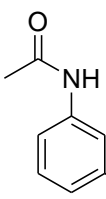
N-(2-ethylphenyl)acetamide (5b)²³: According to GP II: 2-Ethylaniline (0.20 g, 1.65 mmol) and



Meldrum's acid **2** (0.24 g, 1.65 mmol) was reacted for 12 h and filtration and washing (EtOAc : hexane, 1:5) gave **5b** as white solid (0.26 g, 98%). ¹H NMR (400 MHz, CDCl₃) δ = 7.58 - 7.54 (m, 1H), 7.28 (br. s, 1H), 7.19 - 7.10 (m, 3H), 2.48 (q, *J* = 7.6 Hz, 2H), 2.05 (s, 3H), 1.10 (t, *J* = 7.6 Hz, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 169.2, 136.3, 134.9, 128.5, 126.5, 126.0, 124.9, 24.2, 23.9, 14.0

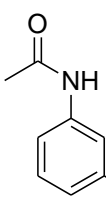
ppm.

N-(3-bromophenyl)acetamide (5c)⁸: According to GP II: 3-Bromoaniline (0.20 g, 1.16 mmol)



and Meldrum's acid **2** (0.17 g, 1.16 mmol) was reacted for 12 h and filtration and washing (EtOAc : hexane, 1:5) gave **5c** as white solid (0.24 g, 95%). ¹H NMR (400 MHz, CDCl₃) δ = 8.60 (br. s, 1H), 7.78 (s, 1H), 7.38 (d, *J* = 8.0 Hz, 1H), 7.19 (d, *J* = 8.0 Hz, 1H), 7.11 (t, *J* = 8.0 Hz, 1H), 2.16 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 169.6, 139.3, 130.2, 127.3, 123.2, 122.5, 118.7, 24.4 ppm.

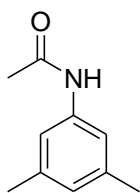
N-(3-fluorophenyl)acetamide (5d)¹: According to GP II: 3-Fluoroaniline (0.20 g, 1.80 mmol)



and Meldrum's acid **2** (0.26 g, 1.80 mmol) was reacted for 12 h and filtration and washing (EtOAc : hexane, 1:5) gave **5d** as white solid (0.26 g, 93%). ¹H NMR (600 MHz, CDCl₃) δ = 8.96 (br. s, 1H), 7.51 - 7.47 (m, 1H), 7.22 - 7.17 (m, 2H), 6.78 - 6.75 (m, 1H), 2.16 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 169.8, 169.8,

163.7, 162.1, 139.7, 139.6, 130.02, 129.96, 115.6, 111.0, 110.9, 107.7, 107.6, 24.3 ppm.

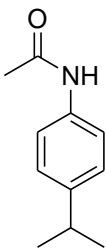
***N*-(3,5-dimethylphenyl)acetamide (5e)**²¹: According to GP II: 3,5-Dimethylaniline (0.20 g, 1.66 mmol) and Meldrum's acid **2** (0.24 g, 1.65 mmol) was reacted for 12 h and filtration



and washing (EtOAc : hexane, 1:5) gave **5e** as white solid (0.25 g, 91%). ¹H NMR (600 MHz, CDCl₃) δ = 7.64 (br. s, 1H), 7.13 (s, 2H), 6.74 (s, 1H), 2.26 (s, 6H), 2.14 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 168.7, 138.6, 137.8, 126.1, 117.8, 24.5,

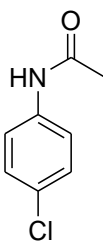
21.4 ppm.

***N*-(4-isopropylphenyl)acetamide (5f)**²¹: According to GP II: 4-Isopropylaniline (0.20 g, 1.48 mmol) and Meldrum's acid **2** (0.14 g, 1.48 mmol) was reacted for 12 h and filtration



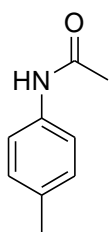
and washing (EtOAc : hexane, 1:5) gave **5f** as white solid (0.25 g, 95%). ¹H NMR (600 MHz, CDCl₃) δ = 8.15 (br. s, 1H), 7.44 - 7.41 (m, 2H), 7.16 - 7.12 (m, 2H), 2.88 - 2.84 (m, 1H), 2.12 (s, 3H), 1.22 (d, *J* = 7.2 Hz, 6H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 169.2, 145.0, 135.8, 126.8, 120.5, 33.6, 24.3, 24.1 ppm.

***N*-(4-chlorophenyl)acetamide (5g)**¹: According to GP II: 4-Chloroaniline (0.2 g, 1.57 mmol) and



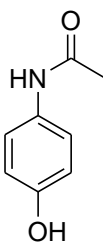
Meldrum's acid **2** (0.23 g, 1.57 mmol) was reacted for 12 h and filtration and washing (EtOAc : hexane, 1:5) gave **5g** as white solid (0.24 g, 90%). ¹H NMR (600 MHz, DMSO-*d*₆) δ = 10.09 (br. s, 1H), 7.61 (d, *J* = 8.8 Hz, 2H), 7.33 (d, *J* = 8.8 Hz, 2H), 2.04 (s, 3H). ppm. ¹³C NMR (151 MHz, DMSO-*d*₆) δ = 168.9, 138.7, 129.0, 126.0, 120.9, 24.4 ppm.

***N*-*p*-tolylacetamide (5h)**¹: According to GP II: 4-Methylaniline (0.30 g, 2.80 mmol) and



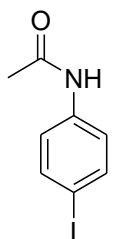
Meldrum's acid **2** (0.40 g, 2.80 mmol) was reacted for 12 h and filtration and washing (EtOAc : hexane, 1:5) gave **5h** as white solid (0.39 g, 93%). ¹H NMR (400 MHz, CDCl₃) δ = 7.90 (br. s, 1H), 7.37 (d, *J* = 8.4 Hz, 2H), 7.09 (d, *J* = 8.4 Hz, 2H), 2.30 (s, 3H), 2.14 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 168.8, 135.4, 134.0, 129.4, 120.3, 24.3, 20.9 ppm.

***N*-(4-hydroxyphenyl)acetamide (5i)**¹: According to GP II: 4-Hydroxyaniline (0.3 g, 2.75 mmol)



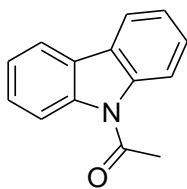
and Meldrum's acid **2** (0.40 g, 2.75 mmol) was reacted for 12 h and filtration and washing (EtOAc : hexane, 1:3) gave **5i** as white solid (0.31 g, 75%). ¹H NMR (400 MHz, DMSO-d₆) δ = 9.42 (br. s, 1H), 8.95 (br. s, 1H), 7.08 (d, *J* = 8.8 Hz, 2H), 6.42 (d, *J* = 8.8 Hz, 2H), 1.72 (s, 3H) ppm. ¹³C NMR (101 MHz, DMSO-d₆) δ = 168.2, 153.6, 131.4, 121.4, 115.5, 24.1 ppm.

***N*-(4-iodophenyl)acetamide (5j)**²²: According to GP II: 4-Iodoaniline (0.2 g, 0.91 mmol) and



Meldrum's acid **2** (1.3 g, 0.91 mmol) was reacted for 12 h and filtration and washing (EtOAc : hexane, 1:5) gave **5j** as white solid (0.23 g, 98%). ¹H NMR (400 MHz, CDCl₃) δ = 7.60 (d, *J* = 8.8 Hz, 2H), 7.53 (br. s, 1H), 7.26 (d, *J* = 8.4 Hz, 2H), 2.17 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 168.6, 137.9, 137.6, 121.8, 87.6, 24.6 ppm.

1-(acridin-10(9*H*)-yl)ethanone (5k)⁶: According to GP II: Carbazole (0.20 g, 1.20 mmol) and

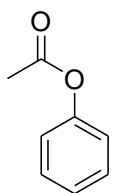


Meldrum's acid **2** (0.17 g, 1.20 mmol) was reacted for 12 h and filtration and washing (EtOAc : hexane, 1:5) gave **5k** as white solid (0.22 mg, 86%). ¹H NMR (600 MHz, CDCl₃) δ = 7.87 (d, *J* = 8.4 Hz, 2H), 7.62 (d, *J* = 8.0 Hz, 2H), 7.21 - 7.16 (m, 2H), 7.11 - 7.07 (m, 2H), 2.87 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃)

δ = 170.2, 138.6, 127.4, 126.4, 123.7, 119.9, 116.3, 27.8 ppm.

General procedure for the acetylation of alcohol (GP III): Triethylamine (20 mol%) was added to a solution of alcohol/thiol (1 eq.) and Meldrum's acid (1.5 eq.) in toluene (2 - 4 mL) and the mixture was refluxed for 12 h. After disappearance of the starting materials (indicated by TLC) solvent was evaporated under reduced pressure. The crude mixture was subjected to column chromatography (silica) to afford analytically pure products.

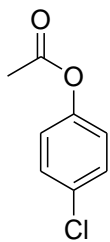
Phenyl acetate (11a)¹³: According to GP III: Phenol (0.20 g, 2.13 mmol), Meldrum's acid **2** (0.31



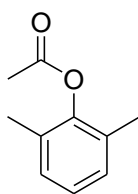
g, 3.19 mmol) and triethylamine (43 mg, 0.43 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **11a** as colourless liquid (0.27 g, 92%). ¹H NMR (400 MHz, CDCl₃) δ = 7.27 - 7.23 (m, 2H), 7.12 - 7.11 (m, 1H), 6.99 - 6.96 (m, 2H), 2.15 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 169.5, 150.8, 129.5,

125.9, 121.6, 21.1 ppm.

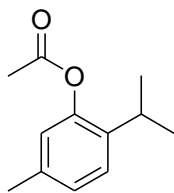
4-chlorophenyl acetate (11b)¹³: According to GP III: 4-Chlorophenol (0.1 g, 0.78 mmol), Meldrum's acid **2** (0.17 g, 1.18mmol), and triethylamine (16 mg, 0.16 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **11b** as colourless liquid (1.2 g, 91%). ¹H NMR (600 MHz, CDCl₃) δ = 7.36 (d, *J* = 9.0 Hz, 2H), 7.06 (d, *J* = 9.0 Hz, 2H), 2.31 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 169.3, 149.1, 131.2, 129.5, 123.0, 21.1 ppm.



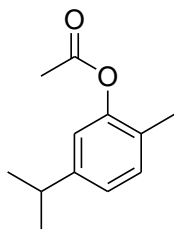
2,6-dimethylphenyl acetate (11c)¹⁹: According to GP III: 2,6-Dimethylphenol (0.1 g, 0.82 mmol), Meldrum's acid **2** (0.18 g, 1.23 mmol) and triethylamine (17 mg, 0.16 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **11c** as colourless liquid (0.13 g, 95%). ¹H NMR (600 MHz, CDCl₃) δ = 7.15 - 7.12 (m, 3H), 2.40 (s, 3H), 2.23 (s, 6H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 168.9, 148.3, 130.1, 128.6, 125.9, 20.5, 16.4 ppm.



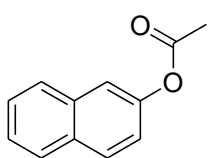
2-isopropyl-5-methylphenyl acetate (11d)¹⁸: According to GP III: Thymol (0.1 g, 0.67 mmol), Meldrum's acid **2** (0.14 g, 0.10 mmol) and triethylamine (14 mg, 0.13 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **11d** as colourless liquid (0.12 g, 90%). ¹H NMR (400 MHz, CDCl₃) δ = 7.10 (d, *J* = 8.0 Hz, 1H), 6.93 (d, *J* = 8.0 Hz, 1H), 6.72 (s, 1H), 2.92 - 2.85 (m, 1H), 2.22 (s, 3H), 2.21 (s, 3H), 1.10 (d, *J* = 7.2 Hz, 6H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 169.8, 147.9, 137.0, 136.6, 127.2, 126.5, 122.8, 27.2, 23.1, 21.0, 20.9 ppm.



5-isopropyl-2-methylphenyl acetate (11e)³¹: According to GP III: Carvacol (0.1 g, 0.67 mmol), Meldrum's acid **2** (0.14 g, 1.0 mmol) and triethylamine (14 mg, 0.13 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **11e** as colourless liquid (0.12 g, 94%). ¹H NMR (400 MHz, CDCl₃) δ = 7.19 (d, *J* = 7.6 Hz, 1H), 7.06 (d, *J* = 7.6 Hz, 1H), 6.92 (s, 1H), 2.96 - 2.89 (m, 1H), 2.34 (s, 3H), 2.19 (s, 3H), 1.28 (d, *J* = 6.8 Hz, 6H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 169.3, 149.4, 148.1, 130.9, 127.2, 124.2, 119.8, 33.6, 23.9, 20.8, 15.8 ppm.



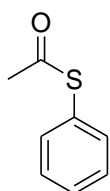
Naphthalen-6-yl acetate (11f)¹³: According to GP III: 2-Napthol (0.1 g, 0.69 mmol), Meldrum's acid **2** (0.15 g, 1.04 mmol) and triethylamine (14 mg, 0.14 mmol) was reacted for 12 h and column



chromatography (silica gel; EtOAc: hexane, 1:15) gave **11f** as colourless liquid (0.11 g, 88%). ¹H NMR (400 MHz, CDCl₃) δ = 7.69 - 7.62 (m, 3H), 7.41 (s, 1H), 7.34 - 7.27 (m, 2H), 7.10 - 7.07 (m, 1H), 2.17 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 169.7, 148.5, 133.9, 131.6, 129.5, 127.9, 127.7, 126.7,

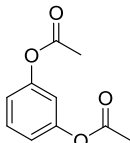
125.8, 121.3, 118.6, 21.2 ppm.

S-phenyl ethanethioate (11g)¹⁹: According to GP III: Thiophenol (0.1 g, 0.91 mmol), Meldrum's



acid **2** (0.20 g, 1.36 mmol) and triethylamine (18 mg, 0.18 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:30) gave **11g** as colourless liquid (0.15 g, 92%). ¹H NMR (400 MHz, CDCl₃) δ = 7.42 - 7.41 (m, 5H), 2.42 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 193.1, 133.4, 128.4, 128.2, 126.9, 29.2 ppm.

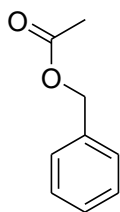
1,3-phenylene diacetate (11h)¹³: According to GP III: Resorcinol (0.1 g, 0.91 mmol), Meldrum's acid **2** (0.39 g, 2.72 mmol) and triethylamine (18 mg, 0.18 mmol) was reacted for 12



h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **11h** as colourless liquid (0.13 g, 75%). ¹H NMR (600 MHz, CDCl₃) δ = 7.39 - 7.36 (m, 1H), 7.00 - 6.98 (m, 2H), 6.94 - 6.92 (m, 1H), 2.28 (s, 6H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ =

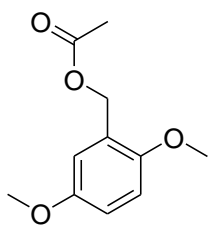
169.0, 151.1, 129.7, 119.0, 115.5, 21.1 ppm.

Benzyl acetate (11i)⁴: According to GP III: Benzyl alcohol (0.1 g, 0.92 mmol), Meldrum's acid **2**



(0.2 g, 1.38 mmol) and triethylamine (18 mg, 0.18 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:30) gave **11i** as colourless liquid (0.12 g, 86%). ¹H NMR (400 MHz, CDCl₃) δ = 7.27 - 7.22 (m, 5H), 5.01 (s, 2H), 2.00 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 170.8, 136.0, 128.6, 128.3, 128.2, 66.3, 21.0 ppm.

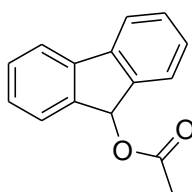
2,5-dimethoxybenzyl acetate (11j)³⁰: According to GP III: 2,5-Dimethoxy benzylalcohol (0.1 g,



0.59 mmol), Meldrum's acid **2** (0.13 g, 0.89 mmol) and triethylamine (12 mg, 0.12 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:10) gave **11j** as colourless liquid (0.10 g, 88%). ¹H NMR (400 MHz, CDCl₃) δ = 6.91 (s, 1H), 6.82 - 6.81 (m, 2H), 5.13 (s, 2H), 3.79 (s, 3H), 3.76 (s, 3H), 2.10 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 171.0, 153.4, 151.6,

125.2, 115.7, 113.7, 111.5, 61.6, 56.0, 55.7, 21.1 ppm.

9H-fluoren-9-yl acetate (11k)³: According to GP III: 9H-fluoren-9-ol (0.1 g, 0.55 mmol),

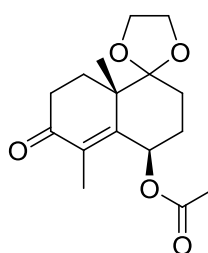


Meldrum's acid **2** (0.12 g, 0.82 mmol) and triethylamine (11 mg, 0.11 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:10) gave **11k** as colourless liquid (0.10 g, 91%). ¹H NMR (400 MHz, CDCl₃) δ = 7.51 (d, *J* = 7.4 Hz, 2H), 7.43 (d, *J* = 7.4 Hz, 2H), 7.26 (t, *J* = 7.4 Hz, 2H),

7.16 (t, *J* = 7.4 Hz, 2H), 6.67 (s, 1H), 2.05 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 171.8, 142.1, 141.1, 129.5, 127.9, 125.9, 120.1, 75.2, 21.3 ppm.

(4R,8aS)-5,8a-dimethyl-6-oxo-3,4,6,7,8,8a-hexahydro-2H-spiro[naphthalene-1,2'

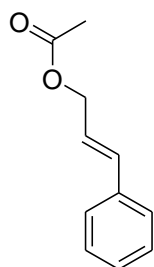
[1,3]dioxolan]-4-yl acetate (11l): According to GP III: (4R,8aS)-4-hydroxy-5,8a-dimethyl-



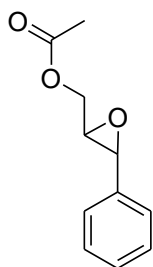
3,4,8,8a-tetrahydro-2H-spiro[naphthalene-1,2'-[1,3]dioxolan]-6(7H)-one (0.1 g, 0.42 mmol), Meldrum's acid **2** (91 mg, 0.63 mmol) and triethylamine (8 mg, 0.08 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:5) gave **11l** as colourless liquid (99 mg, 84%). ¹H NMR (600 MHz, CDCl₃) δ = 5.97 - 5.96 (m, 1H), 3.97 - 3.89 (m, 4H), 2.45 - 2.44 (m, 2H), 2.26 - 2.19 (m, 1H), 2.13 - 2.07 (m, 1H), 2.01 (s, 3H), 1.91 - 1.89 (m, 2H), 1.81

(s, 3H), 1.66 - 1.62 (m, 1H), 1.58 - 1.55 (m, 1H), 1.41 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 199.4, 169.7, 153.0, 135.0, 112.0, 68.3, 65.4, 65.1, 44.6, 33.6, 27.4, 27.2, 25.2, 21.5, 21.3, 11.5 ppm. HRMS: Exact mass calculated for C₁₆H₂₂O₅ ([M+Na]⁺): 317.1359, Found: 317.1366.

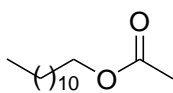
Cinnamyl acetate (11m)⁴: According to GP III: Cinnamyl alcohol (0.1 g, 0.75 mmol), Meldrum's acid **2** (0.16 g, 1.12 mmol) and triethylamine (15 mg, 0.15 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:10) gave **11m** as colourless liquid (0.10 g, 85%). ¹H NMR (600 MHz, CDCl₃) δ = 7.43 - 7.41 (m, 2H), 7.37 - 7.34 (m, 2H), 7.30 - 7.28 (m, 1H), 6.68 (d, *J* = 16.2 Hz, 1H), 6.34 - 6.30 (m, 1H), 4.76 (d, *J* = 6.6 Hz, 2H), 2.13 (s, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 170.9, 136.2, 134.2, 128.6, 128.1, 126.6, 123.2, 65.1, 21.0 ppm.



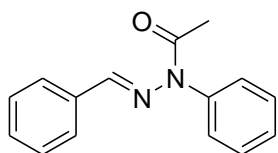
(3-phenyloxiran-2-yl)methyl acetate (11n)³⁴: According to GP III: (3-phenyloxiran-2-yl)methanol (0.1 g, 0.67 mmol), Meldrum's acid **2** (0.14 g, 0.99 mmol) and triethylamine (14 mg, 0.13 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:5) gave **11n** as colourless liquid (0.11 g, 82%). ¹H NMR (400 MHz, CDCl₃) δ = 7.38 - 7.23 (m, 3H), 7.29 - 7.27 (m, 2H), 4.49 (dd, *J* = 12.4, 3.2 Hz, 1H), 4.09 (dd, *J* = 12.4, 6.0 Hz, 1H), 3.82 (d, *J* = 2.0 Hz, 1H), 3.29 - 3.26 (m, 1H), 2.12 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 170.7, 136.3, 128.6, 128.5, 125.7, 64.2, 59.3, 56.4, 20.8 ppm.



dodecyl acetate: (11o)³²: According to GP III: Dodecyl alcohol (0.2 g, 1.07 mmol), Meldrum's acid **2** (2.30 g, 1.61 mmol) and triethylamine (22 mg, 0.21 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **11o** as colourless liquid (0.25 g, 80%). ¹H NMR (600 MHz, CDCl₃) δ = 4.04 (t, *J* = 6.8 Hz, 2H), 2.04 (s, 3H), 1.64 - 1.57 (m, 2H), 1.34 - 1.24 (m, 18H), 0.87 (t, *J* = 6.8 Hz, 3H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 171.5, 64.9, 31.1, 29.9, 29.83, 29.77, 29.72, 29.55, 29.46, 28.8, 26.1, 22.9, 21.2, 14.3 ppm.

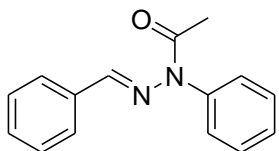


(E)-N'-benzylidene-N-phenylacetohydrazide (15a): According to GP II: (*E*)-1-benzylidene-2-phenylhydrazone (0.1 g, 0.51 mmol) and Meldrum's acid **2** (73 mg, 0.51 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:5) gave **15a** as colourless gum (0.1 g, 82%). ¹H NMR (600 MHz, CDCl₃) δ = 7.60 - 7.56 (m, 4H), 7.50 - 7.48 (m, 1H), 7.37 -



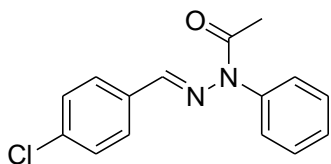
7.36 (m, 3H), 7.23 (s, 1H), 7.19 (d, $J = 7.4$ Hz, 2H), 2.64 (s, 3H) ppm. ^{13}C NMR (151 MHz, CDCl_3) $\delta = 173.0, 141.5, 135.9, 134.3, 130.3, 129.9, 129.4, 129.3, 128.7, 127.2, 22.3$ ppm.

(E)-N'-benzylidene-N-phenylacetohydrazide (15a): According to GP II: phenylhydrazine (0.1 g, 0.93 mmol), benzaldehyde (98 mg, 0.93 mmol) and Meldrum's acid **2** (0.13 g, 0.93 mmol) was



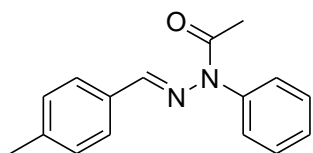
reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:5) gave **15a** as colourless gum (0.17 g, 76 %). ^1H NMR (600 MHz, CDCl_3) $\delta = 7.60 - 7.56$ (m, 4H), 7.50 - 7.48 (m, 1H), 7.37 - 7.36 (m, 3H), 7.23 (s, 1H), 7.19 (d, $J = 7.4$ Hz, 2H), 2.64 (s, 3H) ppm. ^{13}C NMR (151 MHz, CDCl_3) $\delta = 173.0, 141.5, 135.9, 134.3, 130.3, 129.9, 129.4, 129.3, 128.7, 127.2, 22.3$ ppm. HRMS: Exact mass calculated for $\text{C}_{15}\text{H}_{14}\text{N}_2\text{O}$ ($[\text{M}+\text{H}]^+$): 239.1179, Found: 239.1201.

(E)-N'-(4-chlorobenzylidene)-N-phenylacetohydrazide (15b): According to GP II: phenylhydrazine (77 mg, 0.71 mmol), 4-chlorobenzaldehyde (0.1 g, 0.71 mmol) and Meldrum's



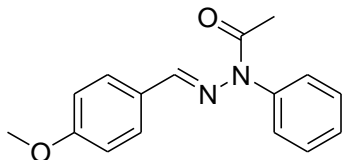
acid **2** (0.10 g, 0.71 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:5) gave **15b** as colourless gum (0.15 g, 75%). ^1H NMR (400 MHz, CDCl_3) $\delta = 7.58 - 7.46$ (m, 5H), 7.35 - 7.32 (m, 2H), 7.18 - 7.16 (m, 3H), 2.61 (s, 3H) ppm. ^{13}C NMR (101 MHz, CDCl_3) $\delta = 172.9, 140.2, 135.8, 135.6, 132.8, 130.3, 129.5, 129.2, 129.0, 128.3, 22.20$ ppm. HRMS: Exact mass calculated for $\text{C}_{15}\text{H}_{13}\text{N}_2\text{O}$ ($[\text{M}+\text{H}]^+$): 273.0789, Found: 273.0796.

(E)-N'-(4-methylbenzylidene)-N-phenylacetohydrazide (15c): According to GP II: phenylhydrazine (0.1 g, 0.93 mmol), 4-methylbenzaldehyde (0.11 g, 0.93 mmol) and Meldrum's



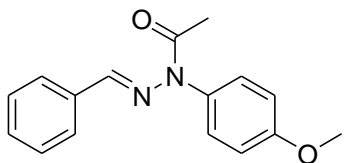
acid **2** (0.13 g, 0.93 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:5) gave **15c** as colourless gum (0.16 g, 70%). ^1H NMR (400 MHz, CDCl_3) $\delta = 7.58 - 7.54$ (m, 2H), 7.50 - 7.47 (m, 3H), 7.21 - 7.17 (m, 5H), 2.63 (s, 3H), 2.37 (s, 3H) ppm. ^{13}C NMR (101 MHz, CDCl_3) $\delta = 172.9, 141.7, 140.1, 136.1, 131.6, 130.2, 129.4, 129.31, 129.28, 127.1, 22.3, 21.5$ ppm. HRMS: Exact mass calculated for $\text{C}_{16}\text{H}_{16}\text{N}_2\text{O}$ ($[\text{M}+\text{H}]^+$): 253.1335, Found: 253.1339.

(E)-N'-(4-methoxybenzylidene)-N-phenylacetohydrazide(15d): According to GP II: phenylhydrazine (80 mg, 0.74 mmol), 4-methoxybenzaldehyde (0.1 g, 0.74 mmol) and Meldrum's



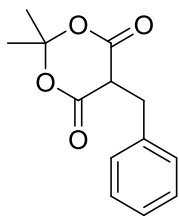
acid **2** (0.11 g, 0.74 mmol) was reacted for 12 h and column chromatography (slica gel; EtOAc: hexane, 1:5) gave **15d** as colourless gum (0.15 g, 74%). $^1\text{H NMR}$ (400 MHz, CDCl_3) δ = 7.45 - 7.41 (m, 4H), 7.37 - 7.34 (m, 1H), 7.08 - 7.06 (m, 3H), 6.77 (d, J = 8.8 Hz, 2H), 3.69 (s, 3H), 2.51 (s, 3H) ppm. $^{13}\text{C NMR}$ (101 MHz, CDCl_3) δ = 171.7, 160.0, 140.4, 135.1, 129.1, 128.3, 128.2, 127.6, 125.9, 113.1, 54.3, 21.2 ppm. HRMS: Exact mass calculated for $\text{C}_{16}\text{H}_{16}\text{N}_2\text{O}_2$ ($[\text{M}+\text{H}]^+$): 269.1285, Found: 269.1286.

(E)-N'-benzylidene-N-(4-methoxyphenyl)acetohydrazide (15e): According to GP II: 4-methoxyphenylhydrazine (0.1 g, 0.72 mmol), benzaldehyde (77 mg, 0.72 mmol) and Meldrum's



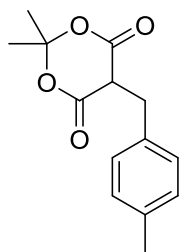
acid **2** (0.10 g, 0.72 mmol) was reacted for 12 h and column chromatography (slica gel; EtOAc: hexane, 1:5) gave **15e** as colourless gum (0.15 g, 78%). $^1\text{H NMR}$ (400 MHz, CDCl_3) δ = 7.63 - 7.61 (m, 2H), 7.42 - 7.38 (m, 3H), 7.29 (s, 1H), 7.13 - 7.07 (m, 4H), 3.89 (s, 3H), 2.65 (s, 3H) ppm. $^{13}\text{C NMR}$ (101 MHz, CDCl_3) δ = 173.2, 160.0, 141.4, 134.4, 130.3, 129.8, 128.7, 128.2, 127.2, 115.5, 55.5, 22.2 ppm. HRMS: Exact mass calculated for $\text{C}_{16}\text{H}_{16}\text{N}_2\text{O}_2$ ($[\text{M}+\text{H}]^+$): 269.1285, Found: 269.1300.

5-benzyl-2,2-dimethyl-1,3-dioxane-4,6-dione (19a)^{7a}: Compound **19a** was prepared by



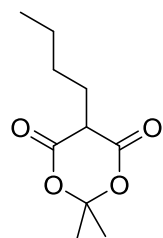
following the exactly same procedure mentioned in the reference 7a. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ = 7.31 - 7.10 (m, 5H), 3.70 (t, J = 5.0 Hz, 1H), 3.42 (d, J = 5.0 Hz, 2H), 1.66 (s, 3H), 1.42 (s, 3H) ppm. $^{13}\text{C NMR}$ (101 MHz, CDCl_3) δ = 165.34, 137.3, 129.8, 128.7, 127.2, 105.3, 48.2, 32.2, 28.5, 27.3 ppm.

2,2-dimethyl-5-(4-methylbenzyl)-1,3-dioxane-4,6-dione (19b)^{7a}: Compound was prepared by



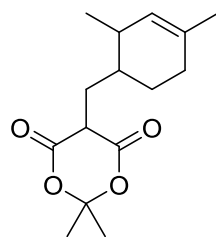
following the procedure mentioned in the reference 7a. ¹H NMR (400 MHz, CDCl₃) δ = 7.20 (d, J = 8.0 Hz, 2H), 7.09 (d, J = 8.0 Hz, 2H), 3.75 (t, J = 5.0 Hz, 1H), 3.44 (d, J = 4.9 Hz, 2H), 2.30 (s, 3H), 1.73 (s, 3H), 1.50 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 160.7, 132.1, 129.4, 124.9, 124.5, 100.5, 43.5, 27.0, 23.7, 22.5, 16.3 ppm.

5-butyl-2,2-dimethyl-1,3-dioxane-4,6-dione (19c)^{7a}: Compound was prepared by following the



exactly same procedure mentioned in the reference 7a. ¹H NMR (400 MHz, CDCl₃) δ = 3.58 – 3.41 (m, 1H), 2.20 – 1.99 (m, 2H), 1.75 (d, J = 12.0 Hz, 6H), 1.43 – 1.32 (m, 4H), 0.91 (t, J = 7.6 Hz, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 165.7, 104.8, 46.1, 28.6, 28.4, 26.9, 26.4, 22.6, 13.7 ppm.

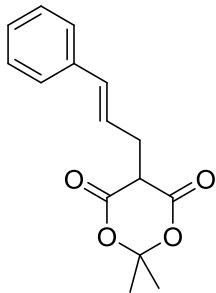
5-((2,4-dimethylcyclohex-3-en-1-yl)methyl)-2,2-dimethyl-1,3-dioxane-4,6-dione (19d): In a round-bottom flask, 2,4-dimethylcyclohex-3-ene-1-carbaldehyde (1.3 g, 10 mmol) were dissolved in TEAF solution (10 mL) prepared by mixing triethylamine and aqueous formic acid (85%) (5:2



mol/mol). After addition of Meldrum's acid (1.4 g, 10 mmol), the mixture was stirred at 45 °C (oil bath), and the reaction was monitored by TLC analysis (for around 6 hours). Then, the mixture was cooled to room temperature, and poured into ice water (pH = 2-3), leading to the precipitation of 5-alkyl Meldrum's acid **19d** as white solid (1.8 g, 68%). FTIR: $\tilde{\nu}$ = 2959, 2915, 2868, 1785, 1745, 1448,

1394, 1382, 1300, 1203, 1062, 1006, 983, 856 cm⁻¹. ¹H NMR (400 MHz, CDCl₃) δ = 5.13 – 5.11 (m, 1H), 3.44 – 3.41 (1, 1H), 2.22 – 2.16 (m, 1H), 1.93 – 1.89 (m, 1H), 1.89 – 1.78 (m, 3H), 1.75 (s, 3H), 1.70 (s, 3H), 1.58 (s, 3H), 1.51 – 1.45 (m, 1H), 1.28 – 1.22 (m, 1H), 0.97 (d, J = 6.8 Hz, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 166.2, 166.0, 132.8, 126.8, 104.9, 44.2, 37.9, 35.9, 30.7, 28.9, 28.7, 26.8, 26.2, 23.5, 20.6 ppm. HRMS: Exact mass calculated for C₁₅H₂₂O₄ ([M+H]⁺): 267.1591, Found: 267.1595.

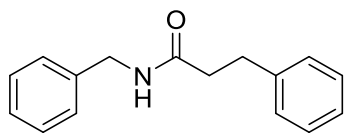
5-cinnamyl-2,2-dimethyl-1,3-dioxane-4,6-dione (19e)⁴⁶: In a round-bottom flask,



cinnamaldehydes (1.3 g, 10 mmol) were dissolved in TEAF solution (10 mL) prepared by mixing triethylamine and aqueous formic acid (85%) (5:2 mol/mol). After addition of Meldrum's acid (1.4 g, 10 mmol), the mixture was stirred at 45 °C (oil bath), and the reaction was monitored by TLC analysis (for around 6 hours). Then, the mixture was cooled to room temperature, and then poured into ice water (pH = 2-3), leading to the precipitation of 5-alkyl

Meldrum's acid derivative **19e** as white solid (1.7 g, 65%). ¹H NMR (400 MHz, CDCl₃) δ = 7.28 (d, *J* = 7.2 Hz, 2H), 7.25 – 7.18 (m, 2H), 7.14 (t, *J* = 7.2 Hz, 1H), 6.53 (d, *J* = 15.8 Hz, 1H), 6.23 – 6.13 (m, 1H), 3.59 (t, *J* = 5.0 Hz, 1H), 2.97 – 2.94 (m, 2H), 1.71 (s, 3H), 1.67 (s, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 165.0, 136.8, 134.7, 128.5, 127.6, 126.4, 123.9, 105.1, 46.7, 29.6, 28.4, 27.0. ppm.

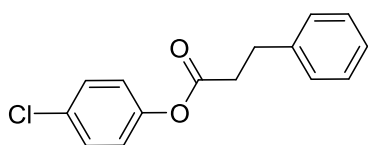
N-benzyl-3-phenylpropanamide (20a)³⁸: According to GP I: benzylamine (80 mg, 0.34 mmol)



and 5-benzyl-2,2-dimethyl-1,3-dioxane-4,6-dione **19a** (37 mg, 0.34 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product **20a** as colorless gum (68 mg, 83%). ¹H

NMR (400 MHz, CDCl₃) δ = 7.18 – 7.15 (m, 5H), 7.14 – 7.06 (m, 3H), 7.02 – 7.00 (m, 2H), 6.08 (s, 1H), 4.24 (d, *J* = 6.0 Hz, 2H), 2.85 (t, *J* = 7.6 Hz, 2H), 2.38 (t, *J* = 7.6 Hz, 2H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 172.2, 140.8, 138.2, 128.63, 128.56, 128.4, 127.7, 127.4, 126.3, 43.5, 38.4, 31.8 ppm.

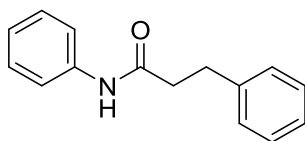
4-chlorophenyl 3-phenylpropanoate (20b)⁴⁰: According to GP III: 4-chloro phenol (37 mg, 0.28 mmol), 5-benzyl-2,2-dimethyl-1,3-dioxane-4,6-dione **19a** (0.1 g, 0.43 mmol) and triethylamine (6



mg, 0.06 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **20b** as colorless liquid (60 mg, 80%). ¹H NMR (400 MHz, CDCl₃) δ = 7.23 – 7.17 (m, 4H),

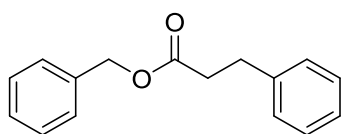
7.16 – 7.10 (m, 3H), 6.83 (d, *J* = 8.8 Hz, 2H), 2.95 (t, *J* = 7.8 Hz, 2H), 2.76 (t, *J* = 7.6 Hz, 2H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 170.1, 148.0, 138.9, 130.1, 128.4, 127.6, 127.3, 125.5, 121.9, 34.8, 29.8 ppm.

N,3-diphenylpropanamide (20c)^{7a}: According to GP II: aniline (40 mg, 0.43 mmol) and 5-benzyl-2,2-dimethyl-1,3-dioxane-4,6-dione **19a** (0.1 g, 0.43 mmol) was reacted for 12 h and



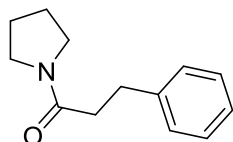
column chromatography (silica gel; EtOAc: hexane, 1:1) gave **20c** as colourless gum (89 mg, 92%). ¹H NMR (400 MHz, CDCl₃) δ = 7.35 (d, *J* = 7.8 Hz, 2H), 7.24 – 7.21 (m, 3H), 7.19 – 7.15 (m, 3H), 7.07 – 6.97 (m, 2H), 2.98 (t, *J* = 7.2 Hz, 2H), 2.59 (t, *J* = 7.2 Hz, 2H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 170.6, 140.6, 137.7, 129.0, 128.7, 128.4, 126.4, 124.3, 120.0, 39.4, 31.6 ppm.

benzyl 3-phenylpropanoate (20d)³⁹: According to GP III: benzyl alcohol (80 mg, 0.34 mmol),



5-benzyl-2,2-dimethyl-1,3-dioxane-4,6-dione **19a** (0.2 g, 0.85 mmol) and triethylamine (12 mg, 0.11 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product gave **20d** as colourless liquid (0.12 g, 87%). ¹H NMR (400 MHz, CDCl₃) δ = 7.29 – 7.14 (m, 7H), 7.14 – 7.07 (m, 3H), 5.02 (s, 2H), 2.88 (t, *J* = 7.8 Hz, 2H), 2.59 (t, *J* = 7.8 Hz, 2H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 172.8, 140.5, 136.0, 128.59, 128.55, 128.4, 128.3, 126.3, 66.3, 35.9, 31.0 ppm.

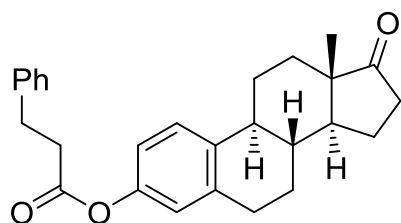
3-phenyl-1-(pyrrolidin-1-yl)propan-1-one (20e)³⁸: According to GP I: pyrrolidine (30 mg, 0.43 mmol) and 5-benzyl-2,2-dimethyl-1,3-dioxane-4,6-dione **19a** (0.1 g, 0.34 mmol) was reacted for



12 h and evaporation of the solvent gave analytically pure product **20e** as colorless gum (73 mg, 84%). ¹H NMR (400 MHz, CDCl₃) δ = 7.23 – 7.17 (m, 2H), 7.16 – 7.07 (m, 3H), 3.37 (t, *J* = 6.7 Hz, 2H), 3.18 (t, *J* = 6.6 Hz, 2H), 2.94 – 2.83 (m, 2H), 2.52 – 2.40 (m, 2H), 1.82 – 1.69 (m, 4H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 170.9, 141.4, 128.4, 126.1, 46.6, 45.7, 36.7, 31.2, 26.0, 24.4 ppm (Less number of ¹³C observed due to overlap in the aromatic region).

(8R,9S,13S,14S)-13-methyl-17-oxo-7,8,9,11,12,13,14,15,16,17-decahydro-6H-

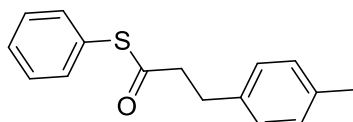
cyclopenta[a]phenanthren-3-yl 3-phenylpropanoate (20f): According to GP III: estrone (77



mg, 0.28 mmol), 5-benzyl-2,2-dimethyl-1,3-dioxane-4,6-dione **19a** (0.1 g, 0.43 mmol) and triethylamine (6 mg, 0.06 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **20f** as colorless liquid (90 mg, 78%). FTIR: $\tilde{\nu}$ = 3030, 2931, 2861, 1738, 1493, 1453, 1221, 1208, 1150, 1127, 1077, 1052, 1007, 733, 698 cm⁻¹. ¹H NMR (400 MHz, CDCl₃) δ = 7.27 – 7.21 (m, 2H), 7.21 –

7.13 (m, 4H), 6.72 – 6.69 (m, 1H), 6.66 (s, 1H), 2.99 (t, $J = 7.6$ Hz, 2H), 2.84 – 2.77 (m, 4H), 2.45 – 2.39 (m, 1H), 2.35 – 2.29 (m, 1H), 2.34 – 2.29 (m, 1H), 2.11 – 2.04 (m, 1H), 2.02 – 1.86 (m, 3H), 1.58 – 1.33 (m, 6H), 0.83 (s, 3H) ppm. ^{13}C NMR (101 MHz, CDCl_3) $\delta = 220.8, 171.7, 148.5, 140.2, 138.0, 137.4, 128.6, 128.4, 126.44, 126.39, 121.6, 118.7, 50.4, 48.0, 44.2, 38.0, 36.0, 35.9, 31.6, 31.0, 29.4, 26.4, 25.8, 21.6, 13.9$ ppm. HRMS: Exact mass calculated for $\text{C}_{27}\text{H}_{30}\text{O}_3$ ($[\text{M}+\text{H}]^+$): 403.2268, Found: 403.2265.

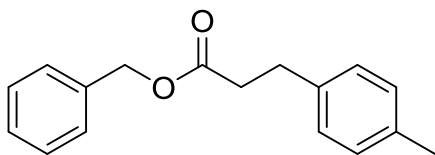
S-phenyl 3-(p-tolyl)propanethioate (20g): According to GP III: thiophenol (44 mg, 0.40 mmol), 2,2-dimethyl-5-(4-methylbenzyl)-1,3-dioxane-4,6-dione **19b** (0.1 g, 0.40 mmol) and triethylamine



(8 mg, 0.08 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **20g** as colorless liquid (86 mg, 83%). FTIR: $\tilde{\nu} = 3025, 2923, 2861, 1704, 1515, 1477, 1440, 1042,$

$1023, 961, 744, 688$ cm^{-1} . ^1H NMR (400 MHz, CDCl_3) $\delta = 7.29$ (s, 5H) (multiplet but looks like singlet), 7.00 (s, 4H) (multiplet but looks like singlet), 2.91 – 2.81 (m, 4H), 2.22 (s, 3H). ppm. ^{13}C NMR (101 MHz, CDCl_3) $\delta = 196.8, 136.9, 136.0, 134.6, 129.4, 129.31, 129.25, 128.3, 127.8, 45.4, 31.1, 21.1$ ppm. HRMS: Exact mass calculated for $\text{C}_{16}\text{H}_{16}\text{OS}$ ($[\text{M}+\text{H}]^+$): 257.0995, Found: 257.1466.

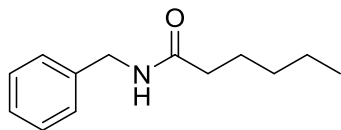
benzyl (E)-5-phenylpent-4-enoate (20h): According to GP III: benzyl alcohol (43 mg, 0.38 mmol), 2,2-dimethyl-5-(4-methylbenzyl)-1,3-dioxane-4,6-



dione **19b** (0.1 g, 0.40 mmol) and triethylamine (8 mg, 0.08 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **20h** as colorless liquid

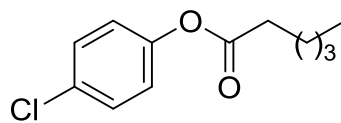
(83 mg, 82%). FTIR: $\tilde{\nu} = 3035, 2923, 2861, 1732, 1515, 1464, 1380, 1352, 1288, 1212, 1147, 811, 697$ cm^{-1} . ^1H NMR (400 MHz, CDCl_3) $\delta = 7.40 - 7.31$ (m, 5H), 7.10 (s, 4H) (multiplet but looks like singlet), 5.13 (s, 2H), 2.96 (t, $J = 7.8$ Hz, 2H), 2.69 (t, $J = 7.8$ Hz, 2H), 2.34 (s, 3H) ppm. ^{13}C NMR (101 MHz, CDCl_3) $\delta = 172.8, 137.4, 136.0, 135.8, 129.2, 128.6, 128.3, 128.22, 128.21, 66.3, 36.1, 30.6, 21.1$ ppm. HRMS: Exact mass calculated for $\text{C}_{17}\text{H}_{18}\text{O}_2$ ($[\text{M}+\text{H}]^+$): 255.1380, Found: 255.1380.

N-benzylhexanamide (20i)³⁸: According to GP I: benzylamine (54 mg, 0.5 mmol) and 5-butyl-2,2-dimethyl-1,3-dioxane-4,6-dione **19c** (0.1 g, 0.34 mmol) was reacted for 12 h and evaporation



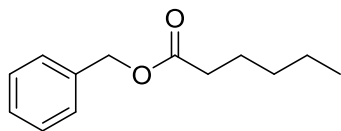
of the solvent gave analytically pure product **20i** as colorless gum (88 mg, 86%). ¹H NMR (400 MHz, CDCl₃) δ = 7.29 – 7.23 (m, 2H), 7.22 – 7.18 (m, 3H), 5.84 (s, 1H), 4.35 (d, *J* = 5.6 Hz, 2H), 2.17 – 2.08 (m, 2H), 1.62 – 1.54 (m, 2H), 1.25 – 1.22 (m, 4H), 0.82 (t, *J* = 7.6 Hz, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 173.1, 138.4, 128.7, 127.8, 127.5, 43.6, 36.8, 31.5, 25.5, 22.4, 14.0 ppm.

4-chlorophenyl hexanoate (20j)⁴⁴: According to GP III: 4-chloro phenol (43 mg, 0.33 mmol), 5-butyl-2,2-dimethyl-1,3-dioxane-4,6-dione **19c** (0.1 g, 0.5 mmol) and triethylamine (7 mg, 0.07



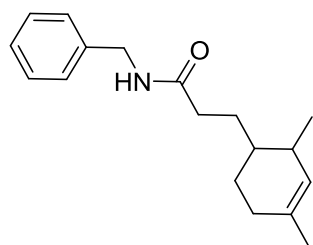
mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **20j** as colorless liquid (61 mg, 81%). ¹H NMR (400 MHz, CDCl₃) δ = 7.28 – 7.24 (m, 2H), 6.97 – 6.92 (m, 2H), 2.47 (t, *J* = 7.6 Hz, 2H), 1.71- 1.64 (m, 2H), 1.34 – 1.28 (m, 4H), 0.86 (t, *J* = 6.8 Hz, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 172.1, 149.2, 131.1, 129.4, 123.0, 34.3, 31.3, 24.6, 22.3, 13.9 ppm.

N-benzyl-3-phenylpropanamide (20k)⁴⁵: benzyl alcohol (54 mg, 0.34 mmol), 5-butyl-2,2-dimethyl-1,3-dioxane-4,6-dione **19c** (0.1 g, 0.5 mmol) and triethylamine (5 mg, 0.05 mmol) was



reacted for 12 h and evaporation of the solvent gave analytically pure product **20k** as colorless liquid (64 mg, 91%). ¹H NMR (400 MHz, CDCl₃) δ = 7.30 – 7.18 (m, 5H), 5.03 (s, 2H), 2.27 (t, *J* = 7.6 Hz, 2H), 1.60 – 1.53 (m, 2H), 1.26 – 1.18 (m, 4H), 0.80 (t, *J* = 7.0 Hz, 3H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 173.7, 136.2, 128.6, 128.18, 128.16, 66.1, 34.3, 31.3, 24.7, 22.3, 13.9 ppm.

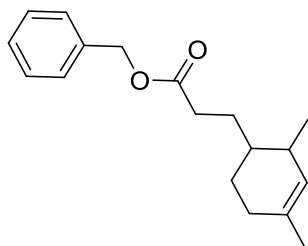
N-benzyl-3-(2,4-dimethylcyclohex-3-en-1-yl)propanamide (20l): According to GP I:



benzylamine (41 mg, 0.38 mmol) and 5-((2,4-dimethylcyclohex-3-en-1-yl)methyl)-2,2-dimethyl-1,3-dioxane-4,6-dione **19d** (0.1 g, 0.38 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:2) gave **20l** as colorless liquid (88 mg, 84%). FTIR: $\tilde{\nu}$ = 3282, 2957, 2923, 2868, 1643, 1539, 1453, 1260, 1080, 1028, 802, 730, 696 cm⁻¹. ¹H NMR (400 MHz, CDCl₃) δ = 7.29 – 7.25 (m, 2H), 7.24 – 7.19 (m, 3H), 5.68 (s, 1H), 5.10 – 5.09 (m, 1H), 4.37 (d, *J* = 5.6 Hz, 2H), 2.29 – 2.22 (m, 1H), 2.14 – 2.06 (m, 1H), 1.88

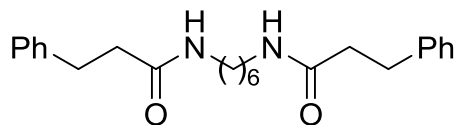
– 1.79 (m, 2H), 1.78 – 1.75 (m, 1H), 1.72 – 1.65 (m, 3H), 1.56 (s, 3H), 1.45 – 1.34 (m, 1H), 1.08 – 1.01 (m, 1H), 0.90 (d, $J = 7.2$ Hz, 3H) ppm. ^{13}C NMR (101 MHz, CDCl_3) $\delta = 173.2, 138.4, 132.9, 128.7, 127.9, 127.5, 127.2, 43.7, 39.9, 35.4, 34.4, 29.3, 29.2, 26.5, 23.5, 20.7$ ppm. HRMS: Exact mass calculated for $\text{C}_{18}\text{H}_{25}\text{NO}$ ($[\text{M}+\text{H}]^+$): 272.2009, Found: 272.2008.

benzyl 3-(2,4-dimethylcyclohex-3-en-1-yl)propanoate (20m): benzyl alcohol (28 mg, 0.26 mmol), 5-((2,4-dimethylcyclohex-3-en-1-yl)methyl)-2,2-dimethyl-1,3-dioxane-4,6-dione **19d**



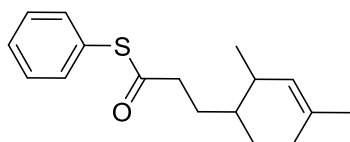
(0.1 g, 0.38 mmol) and triethylamine (5 mg, 0.05 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:20) gave **20m** as colorless liquid (61 mg, 87%). FTIR: $\tilde{\nu} = 2956, 2925, 2869, 1734, 1454, 1379, 1257, 1150, 966, 748, 734, 696$ cm^{-1} . ^1H NMR (400 MHz, CDCl_3) $\delta = 7.30 - 7.27$ (m, 5H), 5.09 – 5.08 (m, 1H), 5.04 (s, 2H), 2.41 – 2.34 (m, 1H), 2.30 – 2.22 (m, 1H), 1.86 – 1.78 (m, 3H), 1.77 – 1.72 (m, 1H), 1.70 – 1.64 (m, 1H), 1.55 (s, 3H), 1.43 – 1.34 (m, 1H), 1.21 – 1.12 (m, 1H), 1.05 – 0.97 (m, 1H), 0.88 (d, $J = 6.8$ Hz, 3H). ppm. ^{13}C NMR (101 MHz, CDCl_3) $\delta = 173.9, 136.1, 132.9, 128.6, 128.2, 128.2, 127.2, 66.2, 39.8, 35.4, 32.1, 29.3, 28.5, 26.5, 23.5, 20.6$ ppm. HRMS: Exact mass calculated for $\text{C}_{18}\text{H}_{24}\text{O}_2$ ($[\text{M}+\text{H}]^+$): 273.1849, Found: 273.1840.

***N,N'*-(hexane-1,6-diyl)bis(3-phenylpropanamide) (20n)**⁴³: According to GP I: hexamethylene diamine (25 mg, 0.21 mmol) and 5-benzyl-2,2-dimethyl-1,3-dioxane-4,6-dione **19a** (0.1 g, 0.43



mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product **20n** as yellow gum (74 mg, 91%). FTIR: $\tilde{\nu} = 3006, 2935, 2857, 1635, 1537, 1496, 1371, 1236, 698$ cm^{-1} . ^1H NMR (400 MHz, CDCl_3) $\delta = 7.20 - 7.18$ (m, 4H), 7.13 – 7.09 (m, 6H), 5.56 (s, 2H), 3.13 – 3.08 (m, 4H), 2.88 (t, $J = 7.7$ Hz, 4H), 2.40 (d, $J = 7.6$ Hz, 4H), 1.35 – 1.29 (m, 4H), 1.16 – 1.12 (m, 4H) ppm. ^{13}C NMR (101 MHz, CDCl_3) $\delta = 172.2, 140.9, 128.5, 128.4, 126.2, 39.0, 38.5, 31.8, 29.3, 25.8$ ppm. HRMS: Exact mass calculated for $\text{C}_{24}\text{H}_{32}\text{N}_2\text{O}_2$ ($[\text{M}+\text{H}]^+$): 381.2537, Found: 381.2537.

S-phenyl 3-(2,4-dimethylcyclohex-3-en-1-yl)propanethioate (20o): According to GP III:

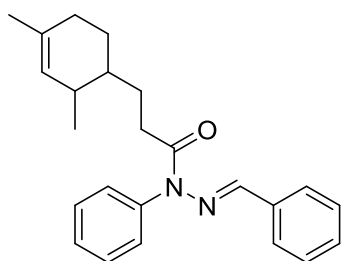


thiophenol (42 mg, 0.38 mmol), 5-((2,4-dimethylcyclohex-3-en-1-yl)methyl)-2,2-dimethyl-1,3-dioxane-4,6-dione **19d** (0.1 g, 0.38 mmol) and triethylamine (8 mg, 0.08 mmol) was reacted for 12 h and

column chromatography (slica gel; EtOAc: hexane, 1:20) gave **20o** as colorless liquid (78 mg, 76%). FTIR: $\tilde{\nu}$ = 2956, 2923, 2868, 1706, 1477, 1440, 1036, 1023, 965, 742, 704, 694 cm^{-1} . ^1H NMR (400 MHz, CDCl_3) δ = 7.34 – 7.31 (m, 5H), 5.10 – 5.09 (m, 1H), 2.71 – 2.63 (m, 1H), 2.60 – 2.52 (m, 1H), 1.94 – 1.87 (m, 1H), 1.86 – 1.81 (m, 2H), 1.80 – 1.68 (m, 2H), 1.57 (s, 3H), 1.51 – 1.41 (m, 1H), 1.24 – 1.17 (m, 1H), 1.12 – 1.03 (m, 1H), 0.91 (d, J = 6.8 Hz, 3H) ppm. ^{13}C NMR (101 MHz, CDCl_3) δ = 197.8, 134.5, 132.9, 129.3, 129.2, 128.0, 127.1, 41.5, 39.7, 35.4, 29.2, 29.1, 26.5, 23.5, 20.6 ppm. HRMS: Exact mass calculated for $\text{C}_{17}\text{H}_{22}\text{O}_2$ ($[\text{M}+\text{H}]^+$): 275.1464, Found: 275.1462.

(E)-N'-benzylidene-3-(2,4-dimethylcyclohex-3-en-1-yl)-N-phenylpropanehydrazide (20p):

According to GP I: phenylhydrazine (21 mg, 0.19 mmol), benzaldehyde (20 mg, 0.19 mmol) and 5-((2,4-dimethylcyclohex-3-en-1-yl)methyl)-2,2-dimethyl-1,3-dioxane-4,6-dione **19d** (50 mg,

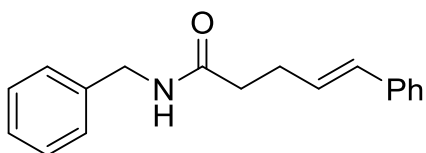


0.19 mmol) was reacted for 12 h and column chromatography (slica gel; EtOAc: hexane, 1:4) gave **20p** as colorless gum (44 mg, 65%).

FTIR: $\tilde{\nu}$ = 2965, 2927, 2872, 1600, 1489, 1448, 1392, 1230, 1178, 1165, 938, 754, 697 cm^{-1} . ^1H NMR (400 MHz, CDCl_3) δ = 7.51 – 7.44 (m, 4H), 7.40 – 7.36 (m, 1H), 7.31 – 7.25 (m, 3H), 7.15 (s, 1H),

7.09 – 7.07 (m, 2H), 5.13 – 5.12 (m, 1H), 3.08 – 2.87 (m, 2H), 2.00 – 1.98 (m, 1H), 1.89 – 1.82 (m, 5H), 1.57 (s, 3H), 1.33 – 1.26 (m, 1H), 1.21 – 1.14 (m, 2H), 0.94 (d, J = 6.8 Hz, 3H) ppm. ^{13}C NMR (101 MHz, CDCl_3) δ = 175.8, 141.4, 136.2, 134.4, 133.0, 130.2, 129.8, 129.29, 129.25, 128.7, 127.4, 127.2, 40.3, 35.6, 32.1, 29.5, 29.0, 26.9, 23.6, 20.8 ppm. Mass calculated for $\text{C}_{24}\text{H}_{28}\text{N}_2\text{O}$ ($[\text{M}+\text{H}]^+$): 361.2274, Found: 361.2593.

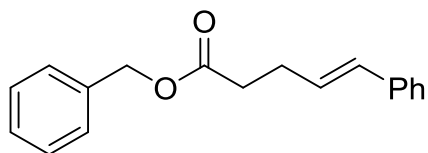
(E)-N-benzyl-5-phenylpent-4-enamide (20q)⁴²: According to GP I: benzylamine (41 mg, 0.38



mmol) and 5-cinnamyl-2,2-dimethyl-1,3-dioxane-4,6-dione **19e** (0.1 g, 0.38 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product **20q** as colorless gum (78 mg, 76%). ^1H NMR (400 MHz, CDCl_3) δ = 7.20 –

7.18 (m, 4H), 7.15 – 7.09 (m, 6H), 6.31 (d, J = 15.6 Hz, 1H), 6.19 – 6.17 (m, 1H), 6.11 – 6.03 (m, 1H), 4.29 (d, J = 5.6 Hz, 2H), 2.47 – 2.41 (m, 2H), 2.25 (t, J = 7.4 Hz, 2H) ppm. ^{13}C NMR (101 MHz, CDCl_3) δ = 172.3, 138.3, 137.3, 131.1, 128.7, 128.6, 127.7, 127.4, 127.2, 126.1, 43.6, 36.3, 29.1 ppm (Less number of ^{13}C observed due to overlap in the aromatic region).

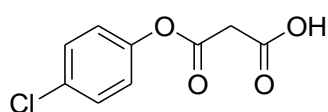
benzyl (E)-5-phenylpent-4-enoate (20r)⁴¹: According to GP III: benzyl alcohol (42 mg, 0.38 mmol), 5-cinnamyl-2,2-dimethyl-1,3-dioxane-4,6-dione **19e**



(0.1 g, 0.38 mmol) and triethylamine (8 mg, 0.08 mmol) was reacted for 12 h and evaporation of the solvent gave analytically pure product **20r** as colorless liquid (83 mg, 81%).

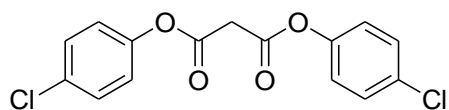
¹H NMR (400 MHz, CDCl₃) δ = 7.29 – 7.24 (m, 5H), 7.22 – 7.18 (m, 4H), 7.14 – 7.10 (m, 1H), 6.33 (d, J = 15.6 Hz, 1H), 6.15 – 6.07 (m, 1H), 5.05 (s, 2H), 2.49 – 2.44 (m, 4H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 172.8, 137.3, 136.0, 131.1, 128.6, 128.5, 128.3, 128.3, 128.3, 127.2, 126.1, 66.3, 34.1, 28.3 ppm.

2-((4-chlorophenoxy)carbonyl)acetic acid (7a)³⁵: 4-Chlorophenol (0.2 g, 1.56 mmol) and Meldrum's acid **2** (0.23 g, 1.56 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:1) gave **7a** as white solid (0.18 g, 55%).



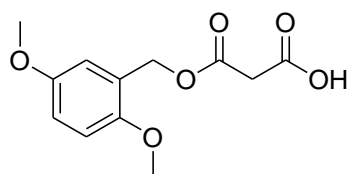
¹H NMR (600 MHz, CDCl₃) δ = 10.70 (br. s, 1H), 7.42 - 7.34 (m, 2H), 7.18 - 7.03 (m, 2H), 3.71 (s, 2H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 172.1, 164.6, 148.7, 131.8, 129.7, 122.7, 41.1 ppm.

bis(4-chlorophenyl) malonate (7b)³⁶: 4-Chlorophenol (0.2 g, 1.56 mmol) and Meldrum's acid **2** (0.23 g, 1.56 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:10) gave **7b** as colourless liquid (0.13 g, 25%).



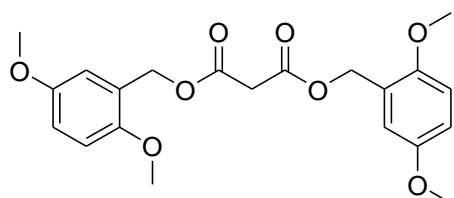
¹H NMR (600 MHz, CDCl₃) δ = 7.37 (d, J = 8.8 Hz, 4H), 7.10 (d, J = 8.8 Hz, 4H), 3.85 (s, 2H) ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 164.5, 148.7, 131.9, 129.7, 122.7, 41.5 ppm.

2-((2,5-dimethoxybenzyloxy)carbonyl)acetic acid (9a): 2,5-Dimethoxy benzyl alcohol (0.2 g, 1.18 mmol) and Meldrum's acid **2** (0.17 g, 1.18 mmol) was reacted



for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:1) gave **9a** as colourless liquid (0.14 g, 48%). ¹H NMR (400 MHz, CDCl₃) δ = 9.45 (br. s, 1H), 6.92 - 6.91 (m, 1H), 6.85 - 6.78 (m, 2H), 5.22 (s, 2H), 3.77 (s, 3H), 3.75 (s, 3H), 3.48 (s, 2H) ppm. ¹³C NMR (101 MHz, CDCl₃) δ = 171.2, 166.7, 153.4, 151.6, 124.3, 115.6, 114.3, 111.7, 63.0, 56.0, 55.8, 40.9 ppm. HRMS: Exact mass calculated for C₁₂H₁₄O₆ ([M+K]⁺): 293.0422, Found: 293.0422.

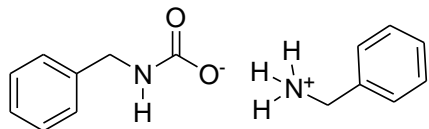
bis(2,5-dimethoxybenzyl) malonate (9b): 2,5-Dimethoxy benzyl alcohol (0.2 g, 1.18 mmol) and



Meldrum's acid **2** (0.17 g, 1.18 mmol) was reacted for 12 h and column chromatography (silica gel; EtOAc: hexane, 1:10) gave **9b** as colourless liquid (0.10 g, 22%). ¹H NMR (600 MHz, CDC₃) δ = 6.93 - 6.90 (m, 2H), 6.83 - 6.78 (m, 4H), 5.21 (s, 4H), 3.77 (s, 6H), 3.75 (s, 6H), 3.50 (s, 2H)

ppm. ¹³C NMR (151 MHz, CDCl₃) δ = 166.4, 153.4, 151.5, 124.6, 115.4, 114.1, 111.6, 62.6, 56.0, 55.8, 41.6 ppm. HRMS: Exact mass calculated for C₂₁H₂₄O₈ ([M+Na]⁺): 427.1363, Found: 427.1369.

Phenylmethanaminium benzylcarbamate (28)³⁷: Meldrum's acid (28 mmol) was added to a solution of benzylamine (28 mmol) in toluene (30 mL) in a 50 mL round bottom flask and round



bottom flask was connected with a condenser. Open mouth of the condenser was closed with septum and one cannula was connected the condenser (through septum) and another closed

round bottom flask containing benzylamine (56 mmol). The reaction mixture for acetylation containing Meldrum's acid (28 mmol), amine (28 mmol) in toluene, was heated at 120 °C and released carbon dioxide was allowed to pass through cannula to the benzylamine at room temperature. Carbamate salt was formed as the white solid with 76% yield (5.5 g, yield was calculated w.r.t carbon dioxide (28 mmol) released). ¹H NMR (400 MHz, MeOH-d₄) δ = 7.40 – 7.27 (m, 10H), 4.29 (s, 1H), 3.88 (s, 4H) ppm. ¹³C NMR (101 MHz, MeOH-d₄) δ = 164.0, 160.1, 141.1, 136.7, 128.5, 128.0, 127.9, 126.7, 126.2, 45.1, 43.8 ppm.

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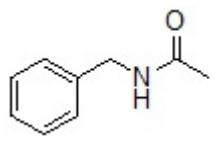
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7.184
7.168
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7.147
7.129
7.110
7.107
7.093
7.085
7.066

4.163
4.148

1.754



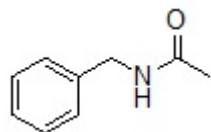
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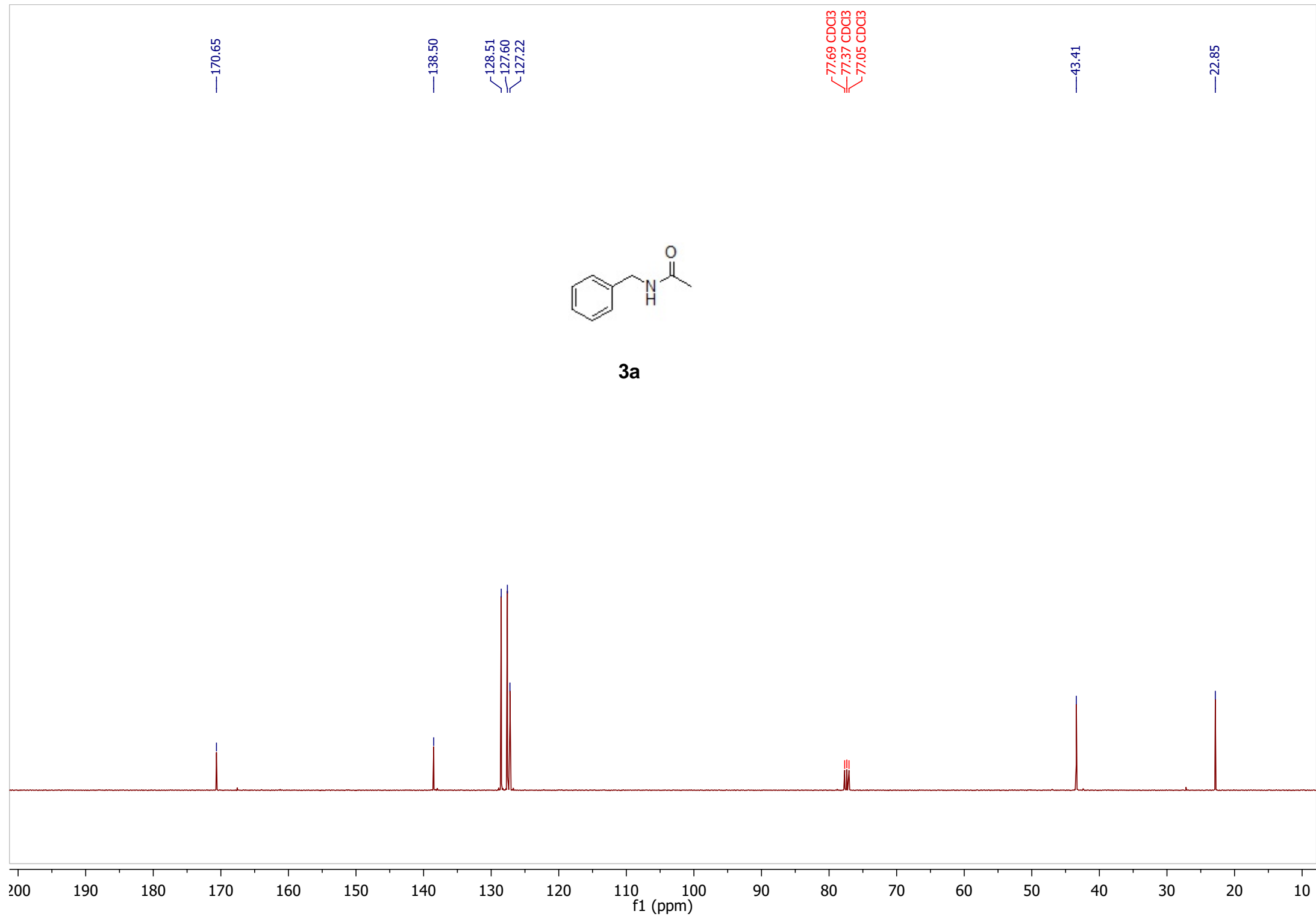
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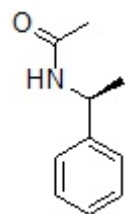
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f1 (ppm)

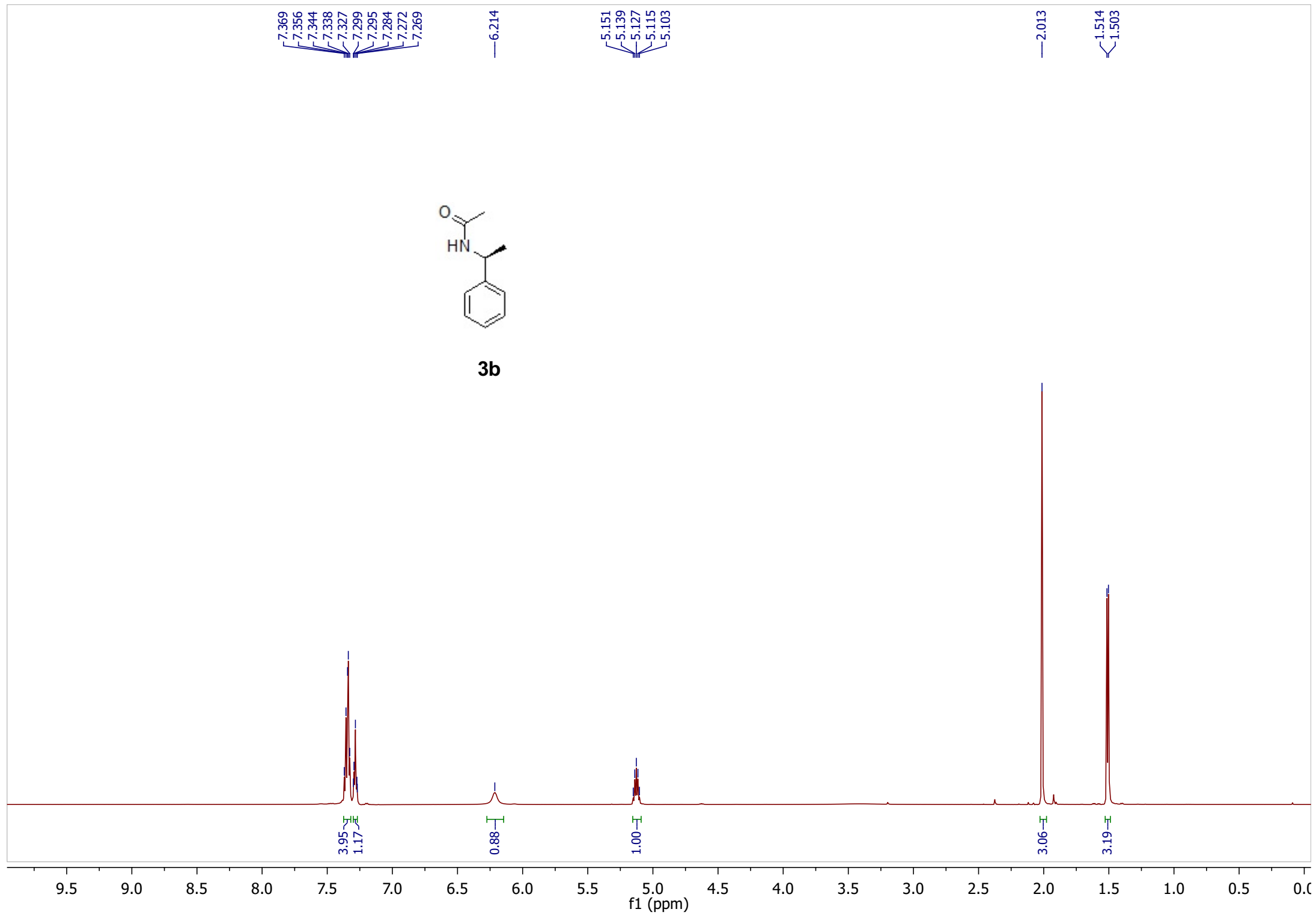


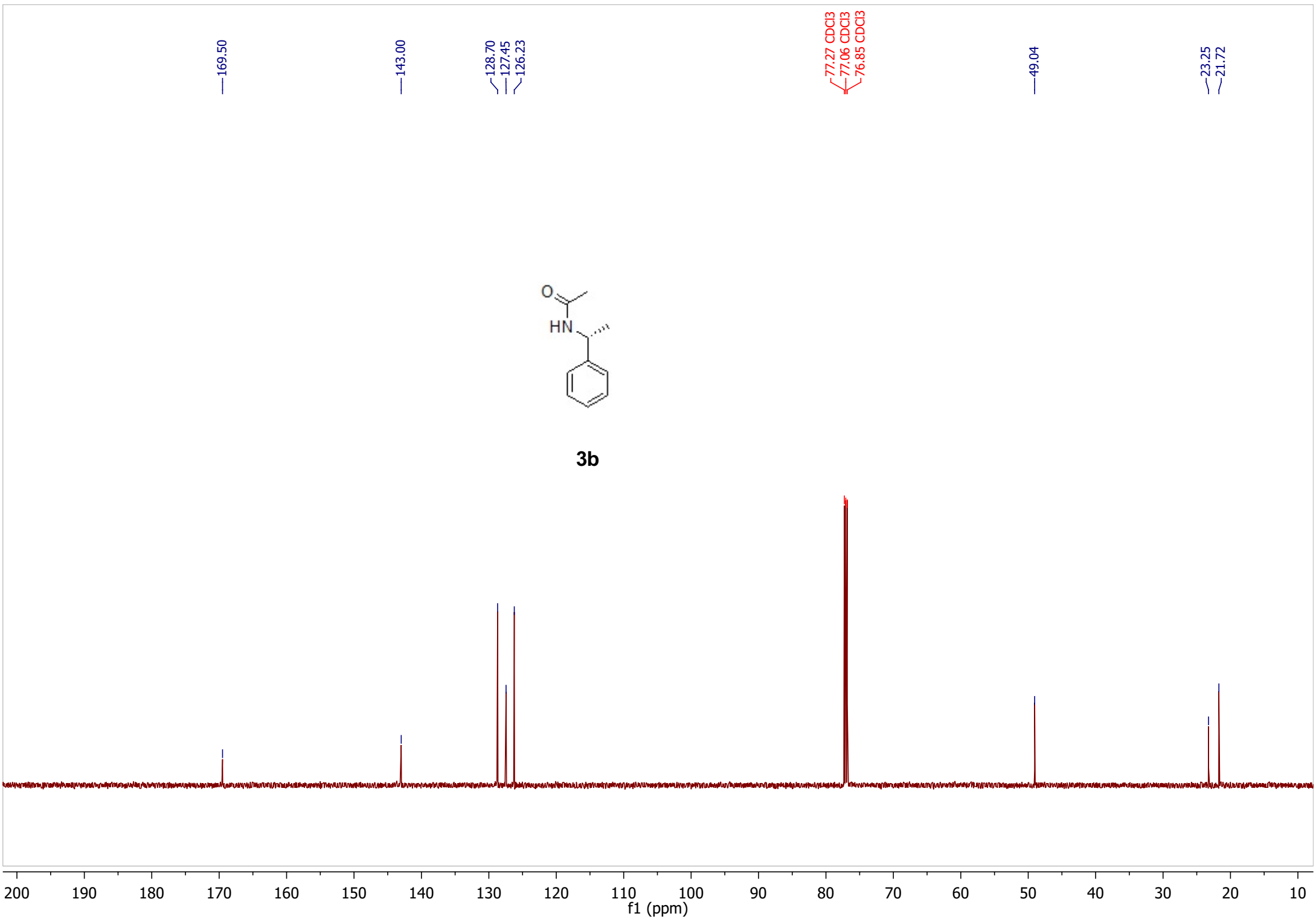
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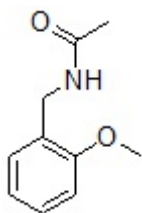




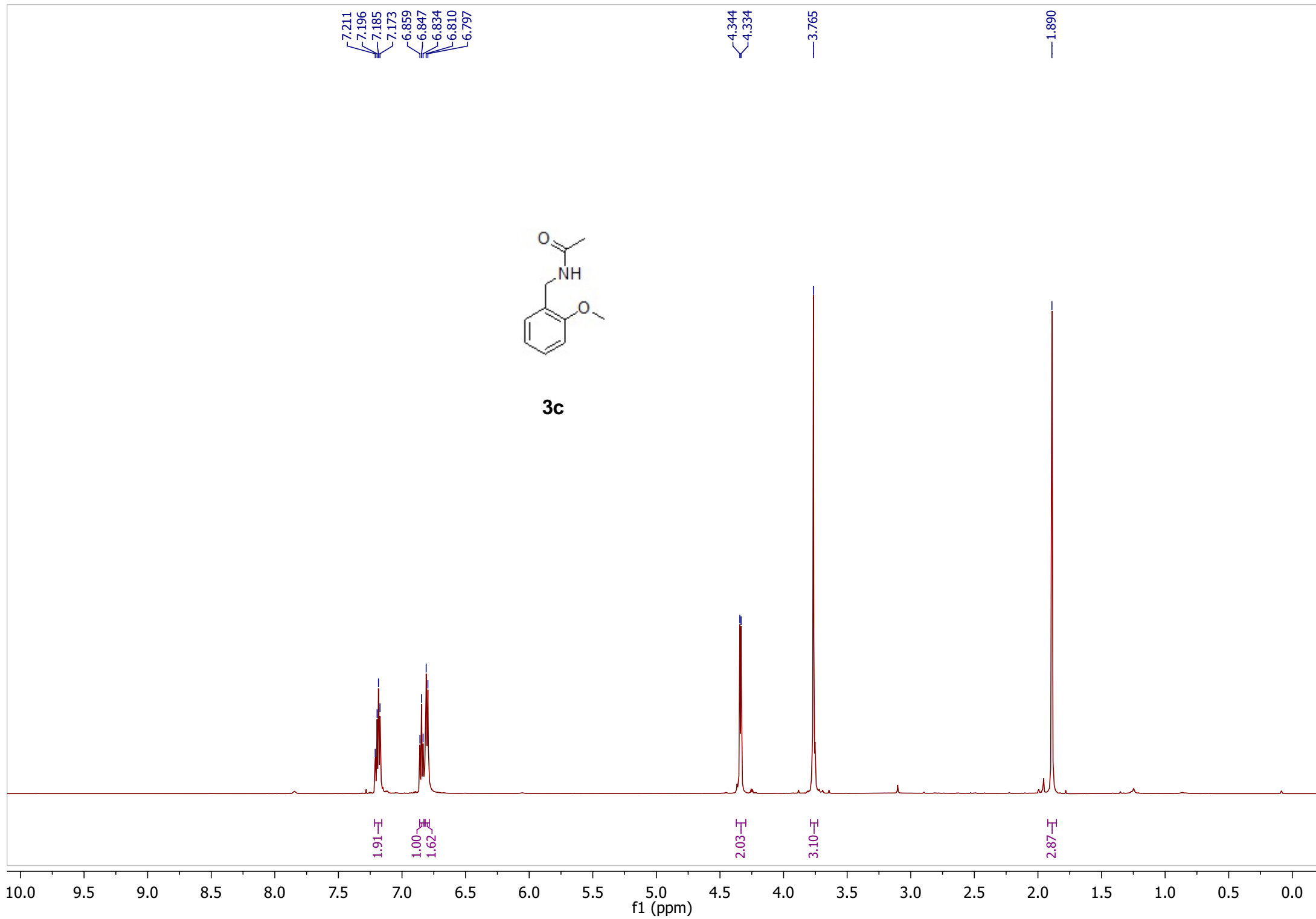
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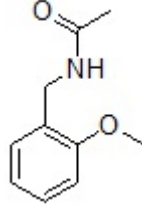




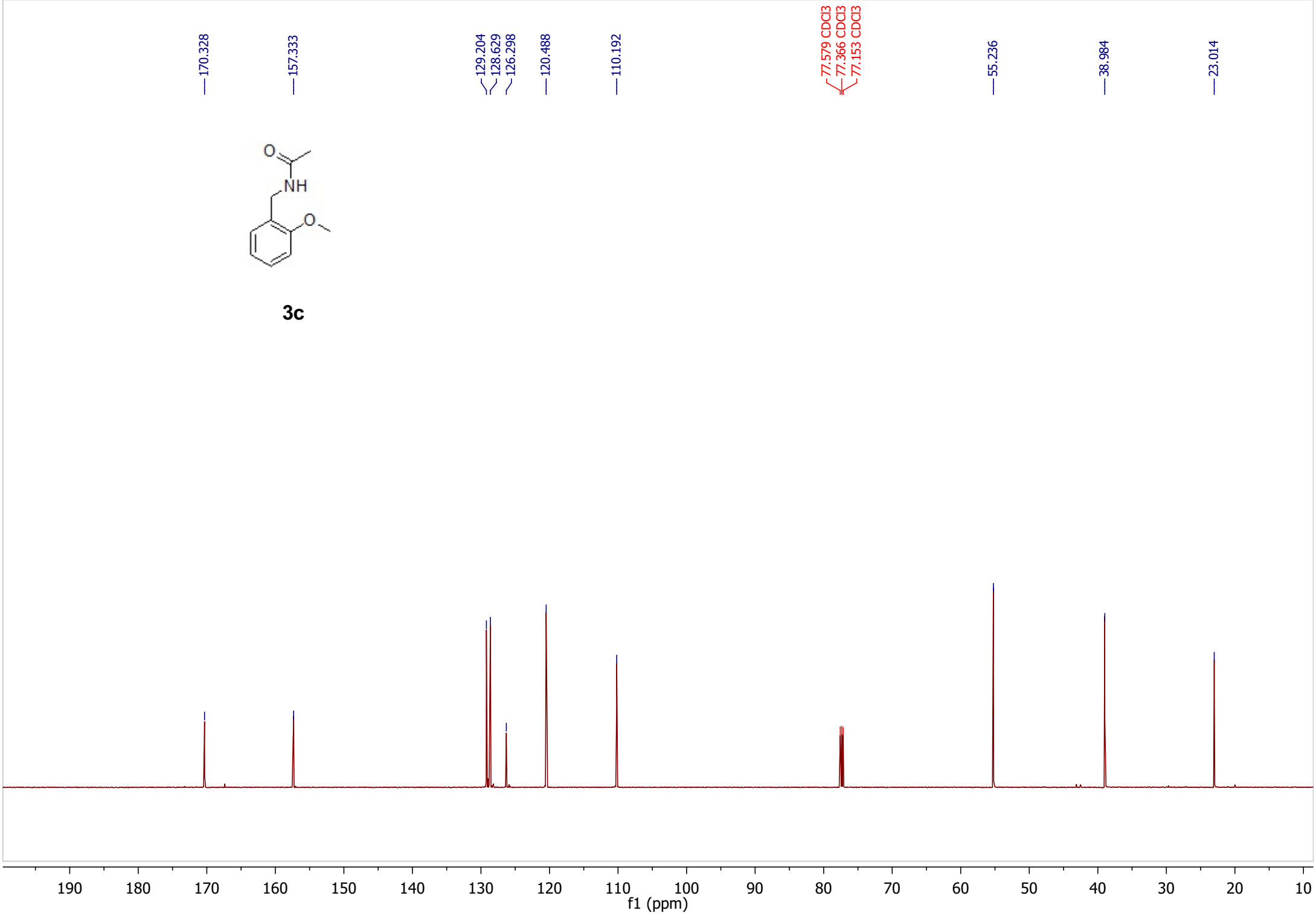


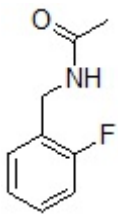
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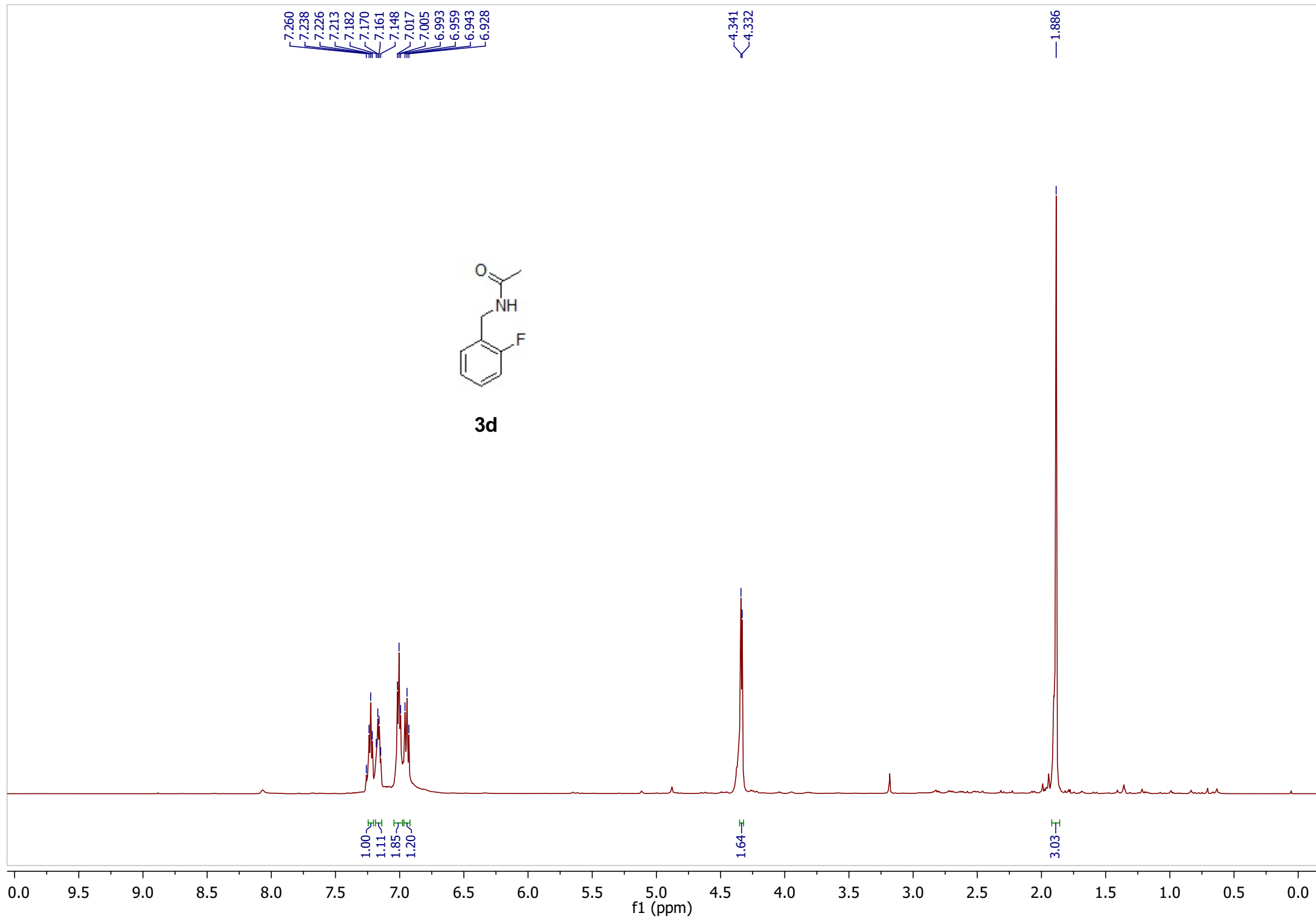


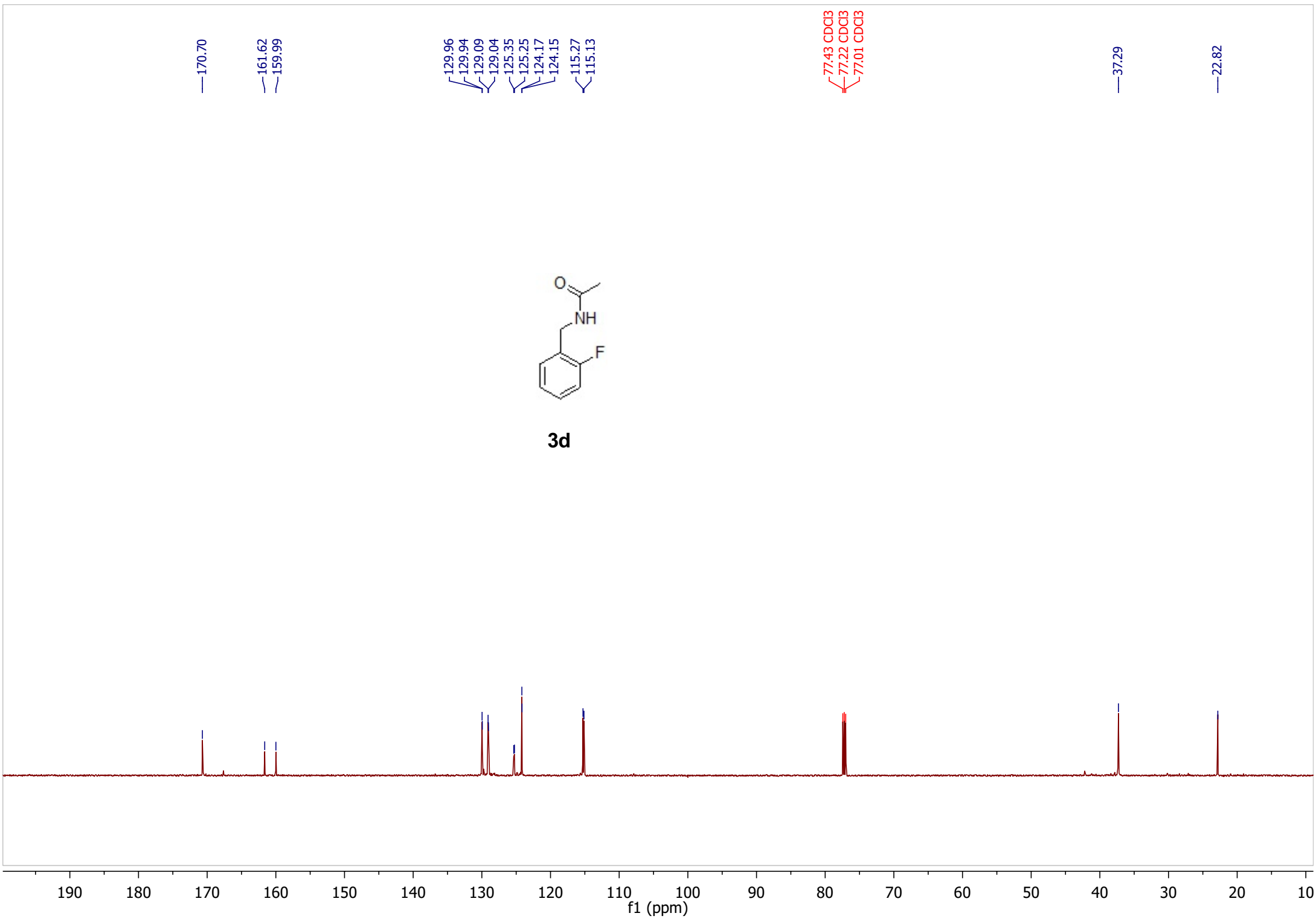
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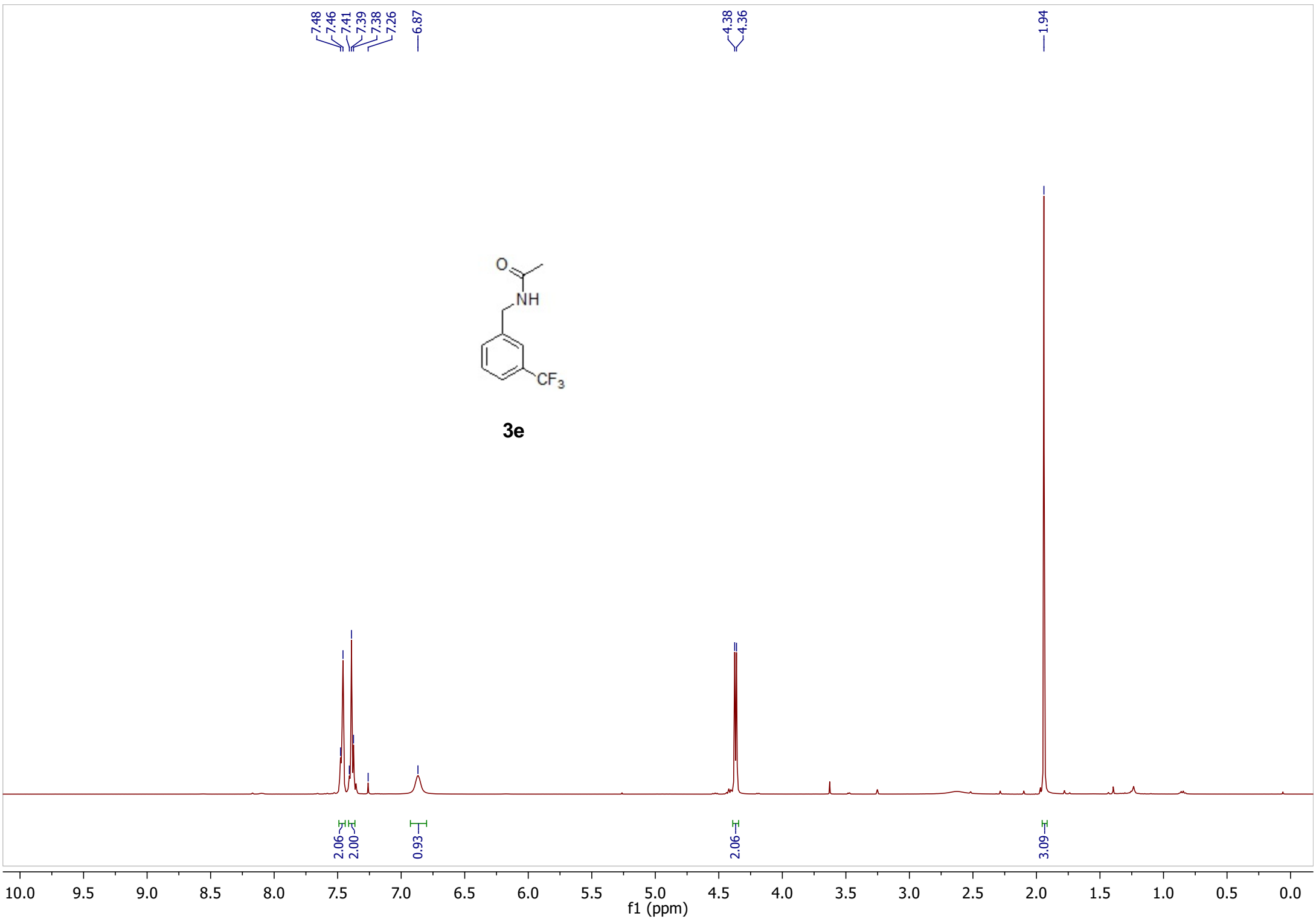


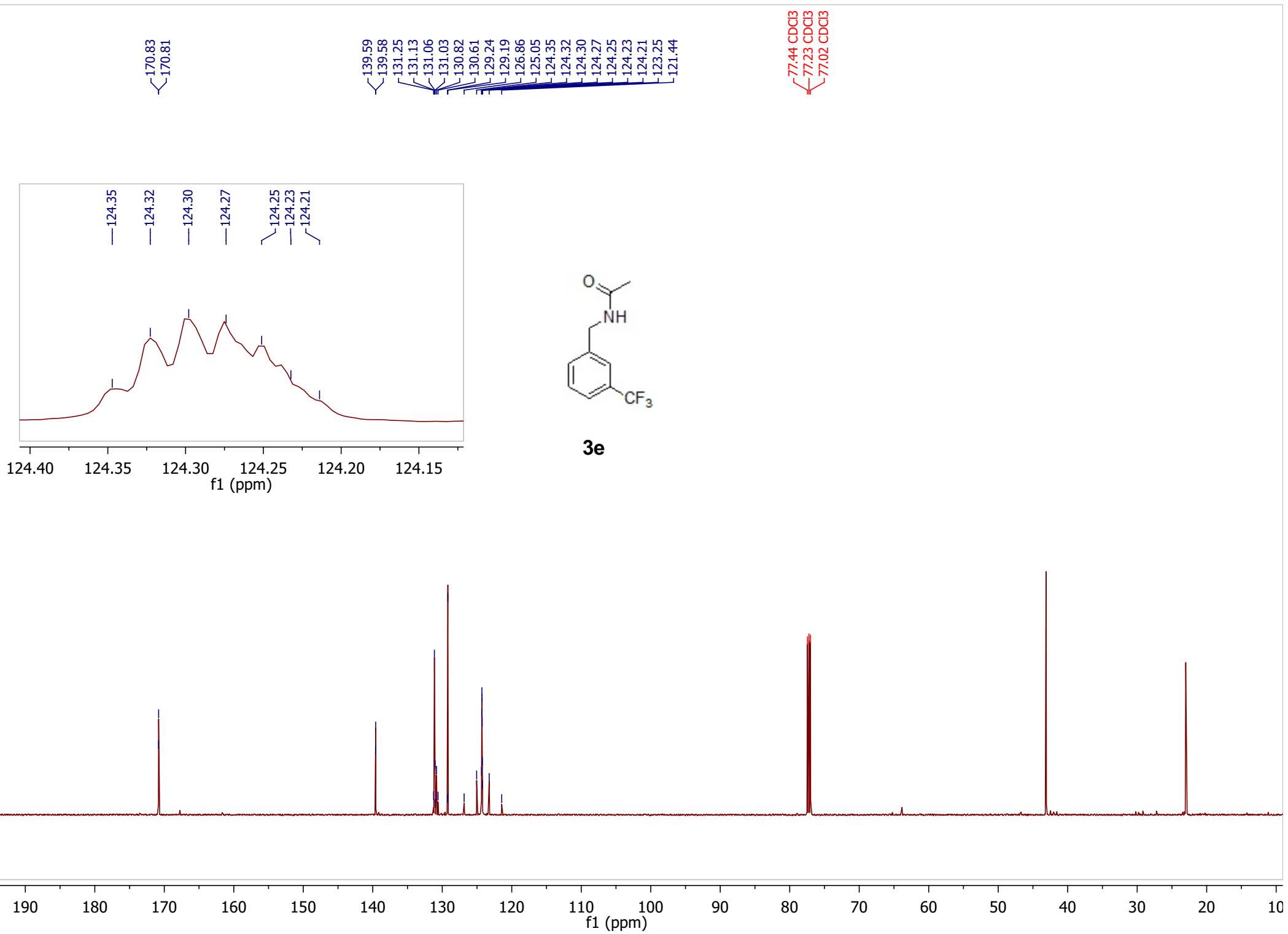


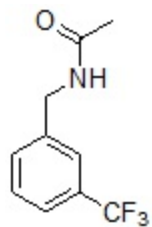
3d









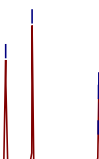


3e, DEPT-135

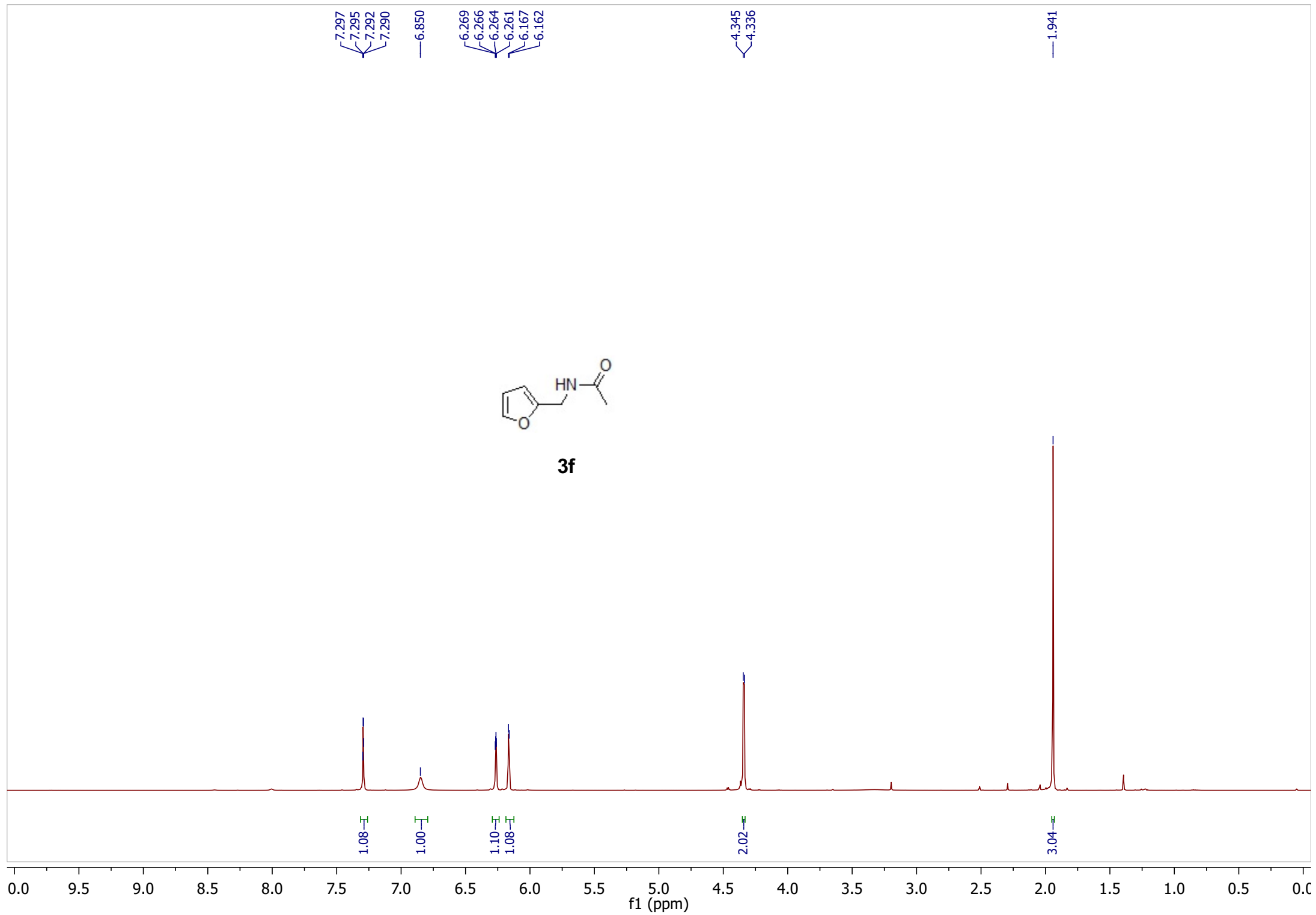
— 131.00
— 129.06
— 124.21
— 124.19
— 124.17
— 124.14
— 124.11
— 124.09

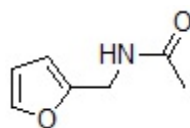
— 42.98

— 22.87



150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)





3f

—170.47

—151.36

—142.04

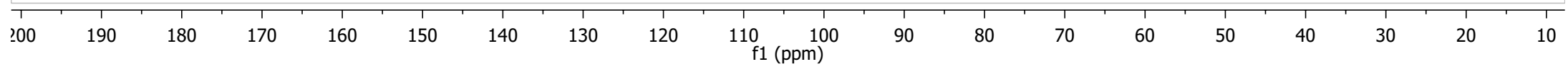
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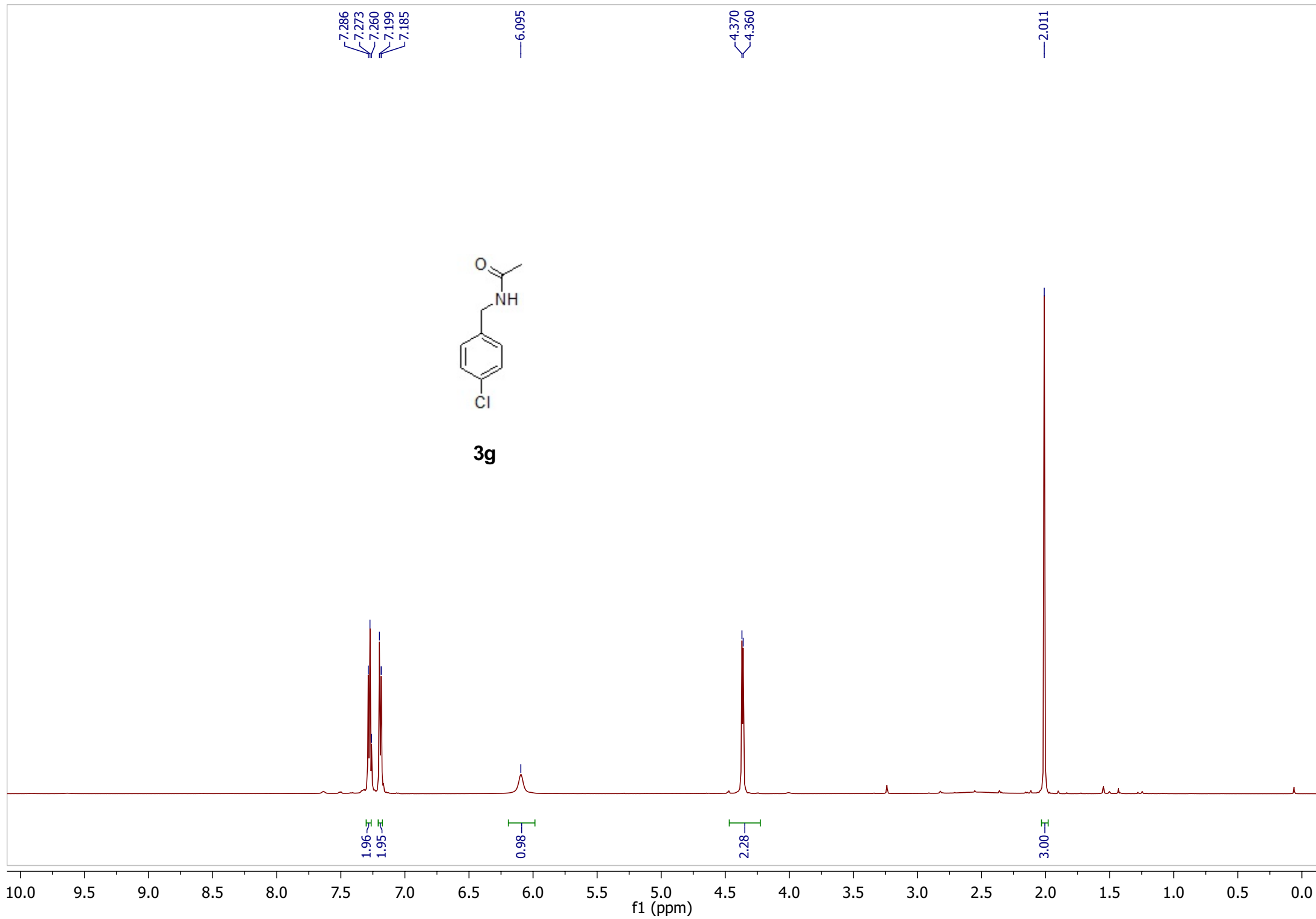
—107.31

77.39 CDCl₃
77.17 CDCl₃
76.96 CDCl₃

—36.47

—22.85





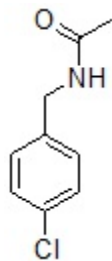
—170.19

136.74
133.31
129.15
128.81

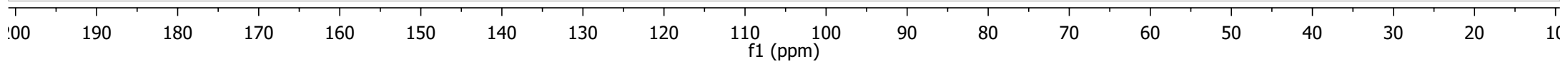
77.27 CDCl3
77.06 CDCl3
76.85 CDCl3

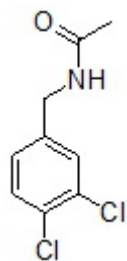
—43.01

—23.19

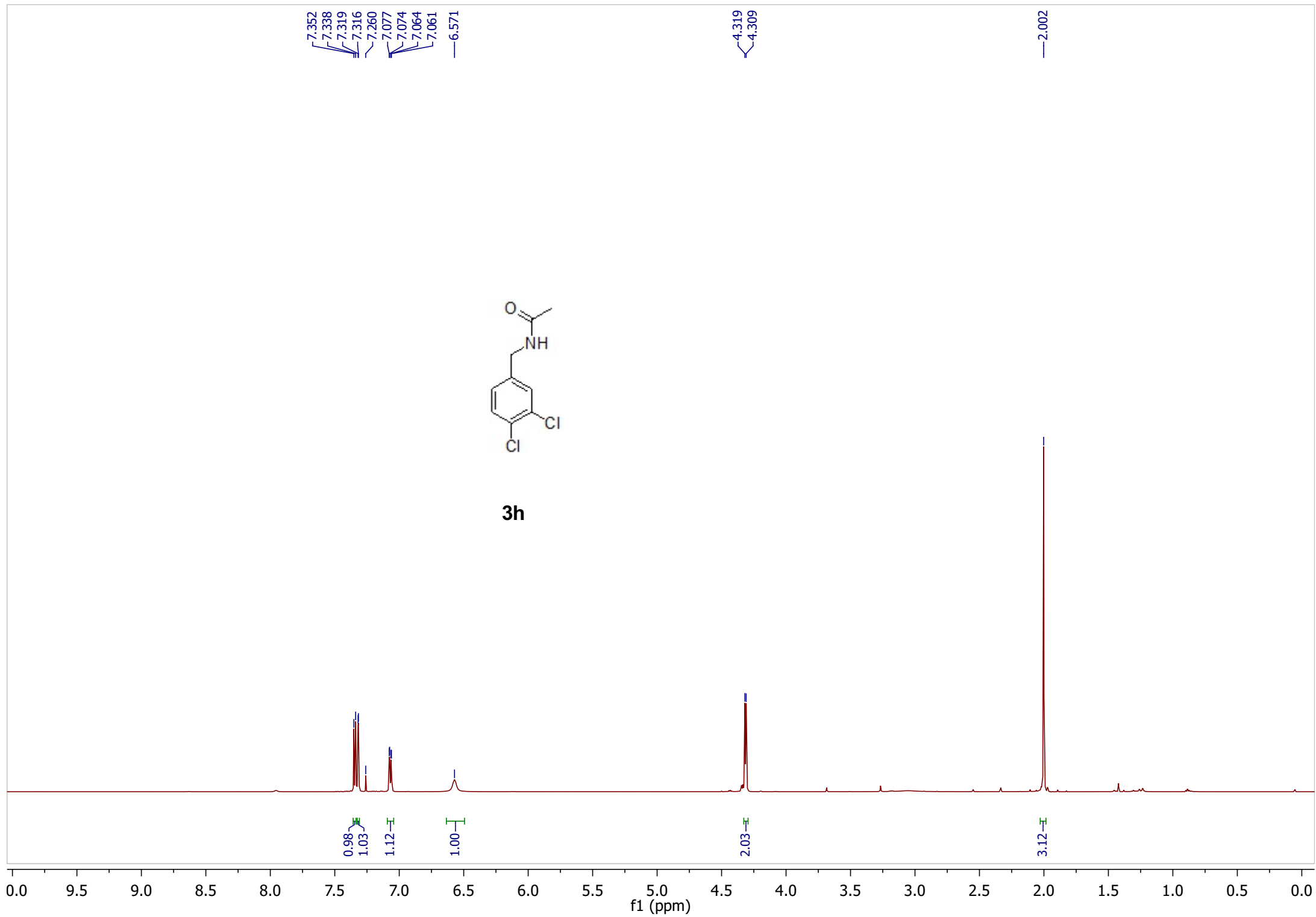


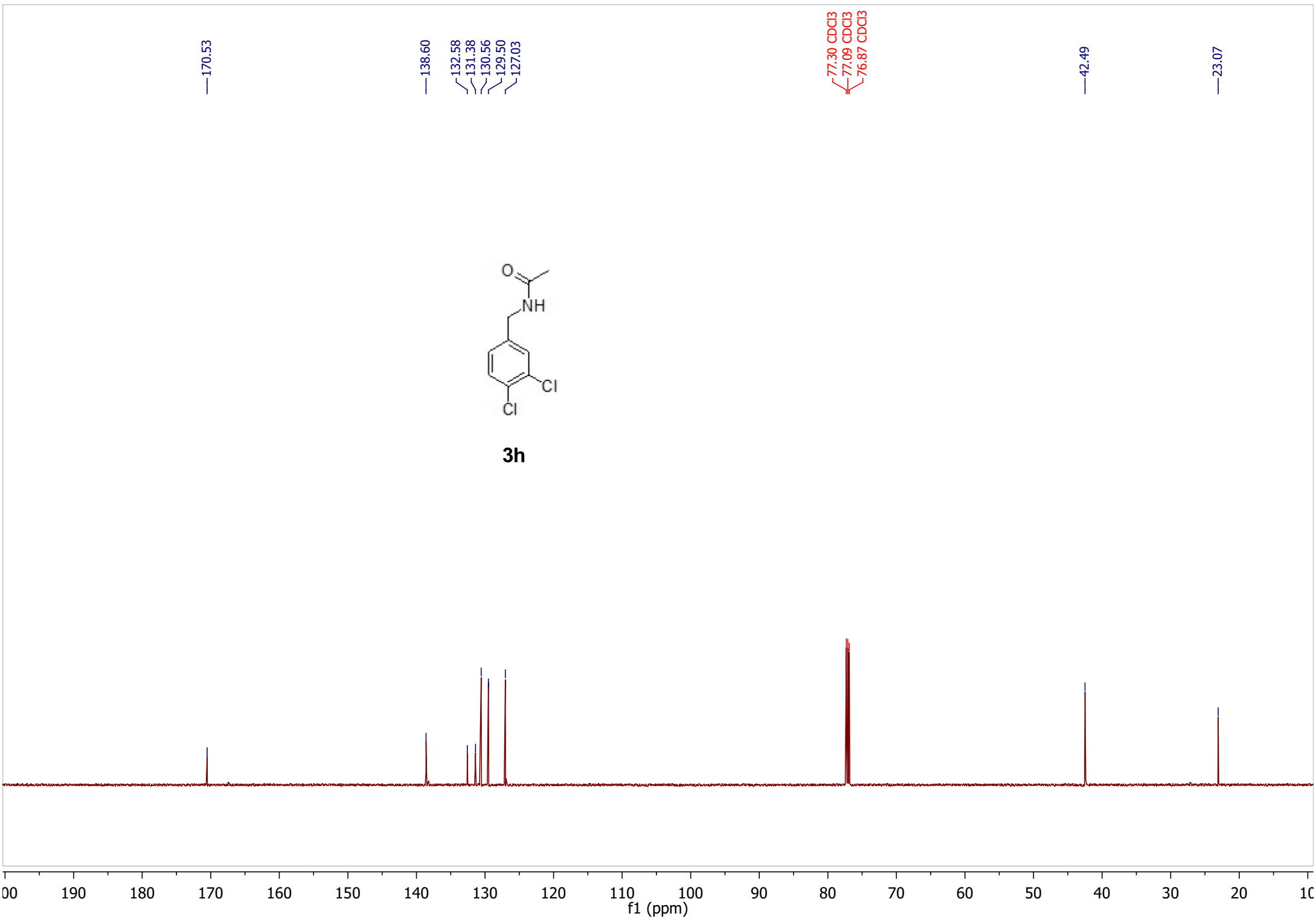
3g

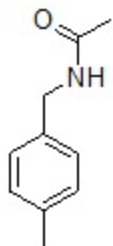




3h







3i

7.260
7.153
7.140
7.124
7.111
7.105

6.209

4.336
4.327

2.319

1.970

3.89

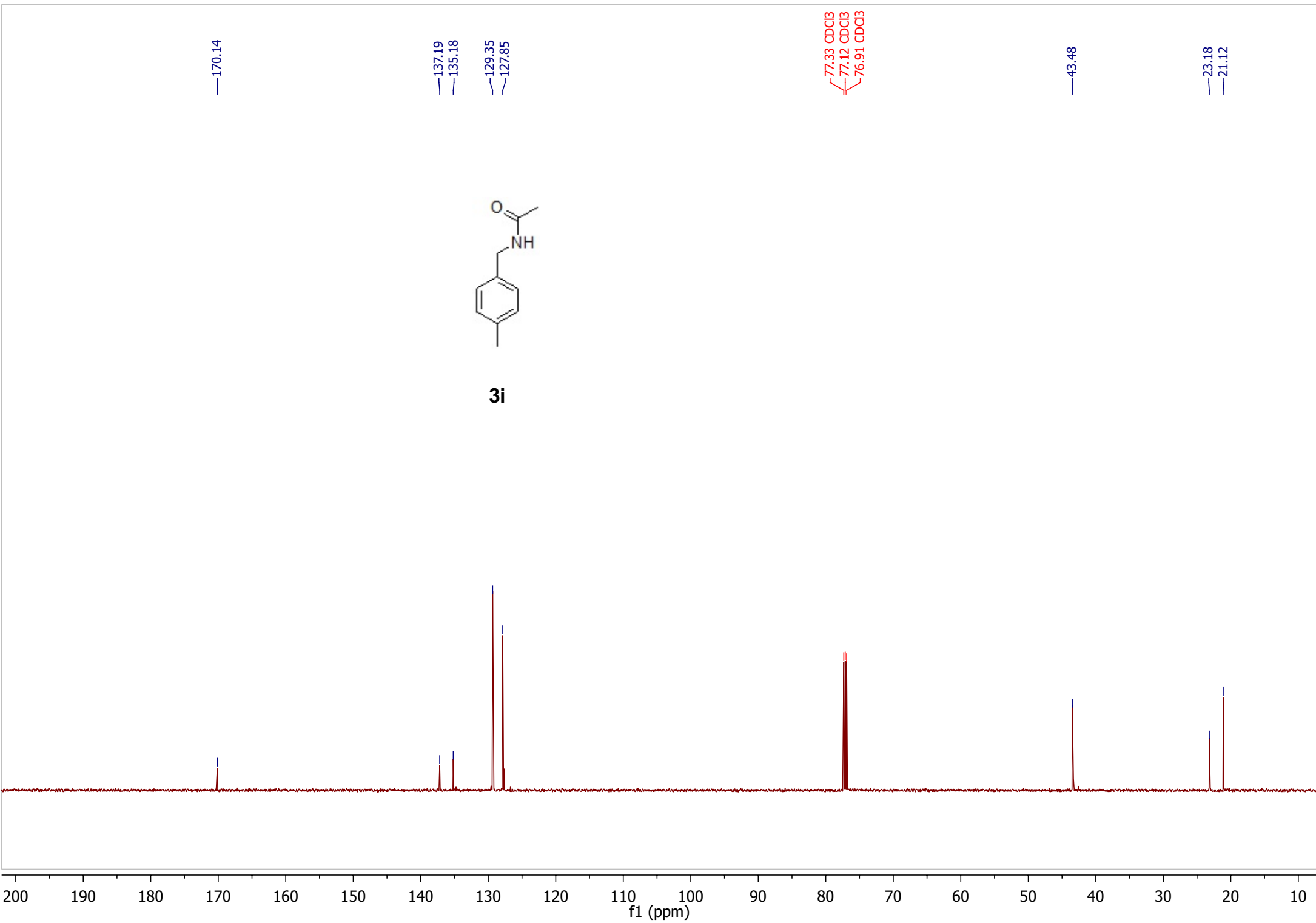
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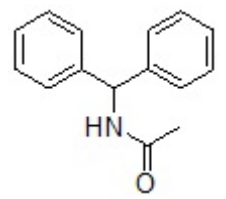
2.00

3.06

2.64

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f1 (ppm)





3j

7.356
7.344
7.331
7.302
7.290
7.284
7.277
7.256
7.250
7.238
7.233
6.492
6.258
6.244

2.058

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

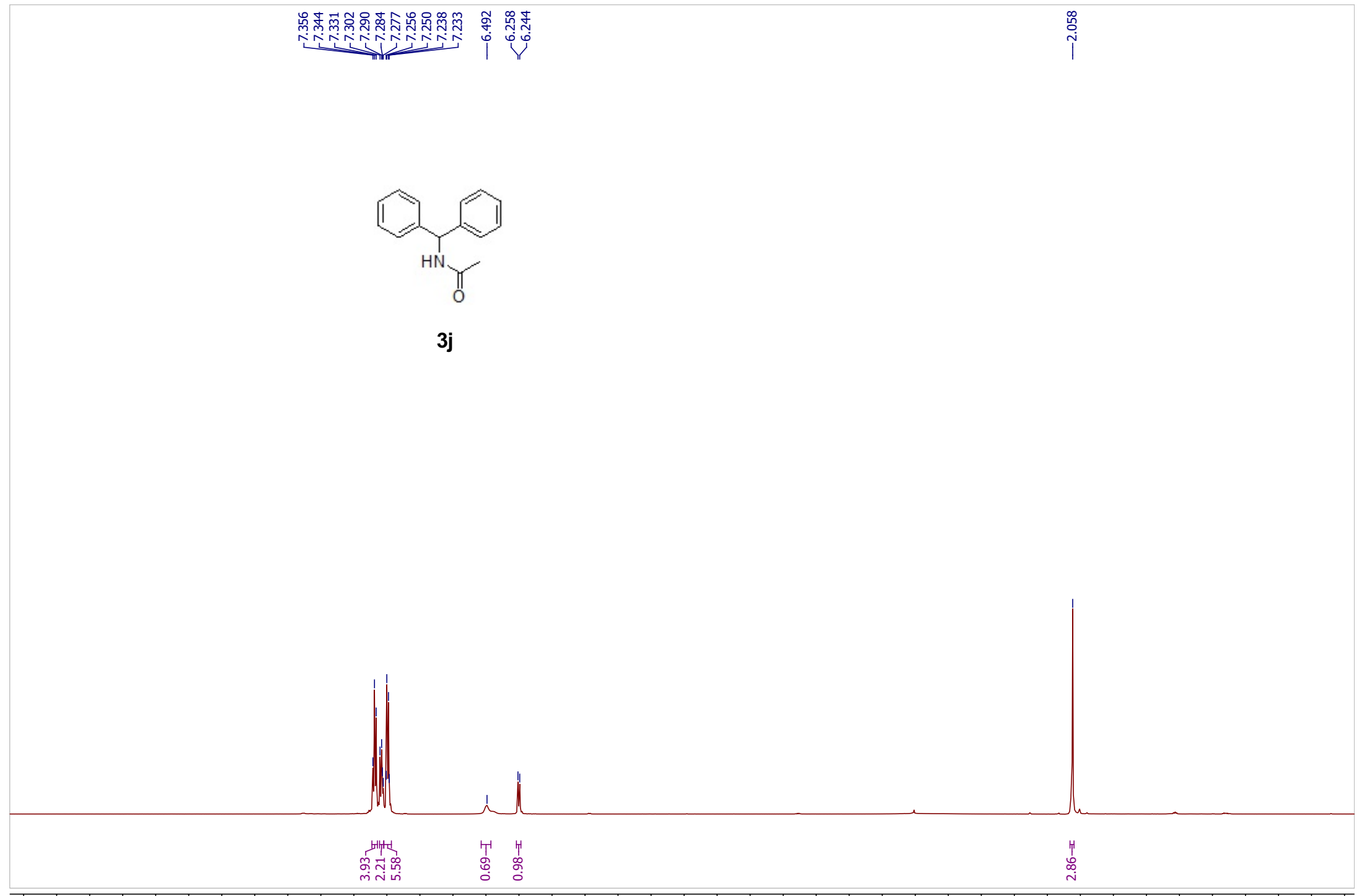
f1 (ppm)

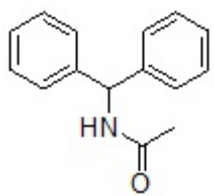
3.93
2.21
5.58

0.69

0.98

2.86





3j

—169.49

—141.36

128.69

127.53

127.44

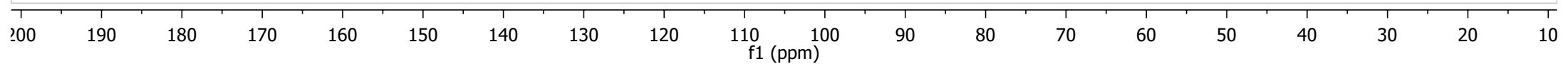
77.28 CDCl₃

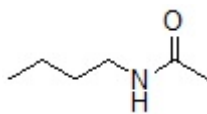
77.07 CDCl₃

76.85 CDCl₃

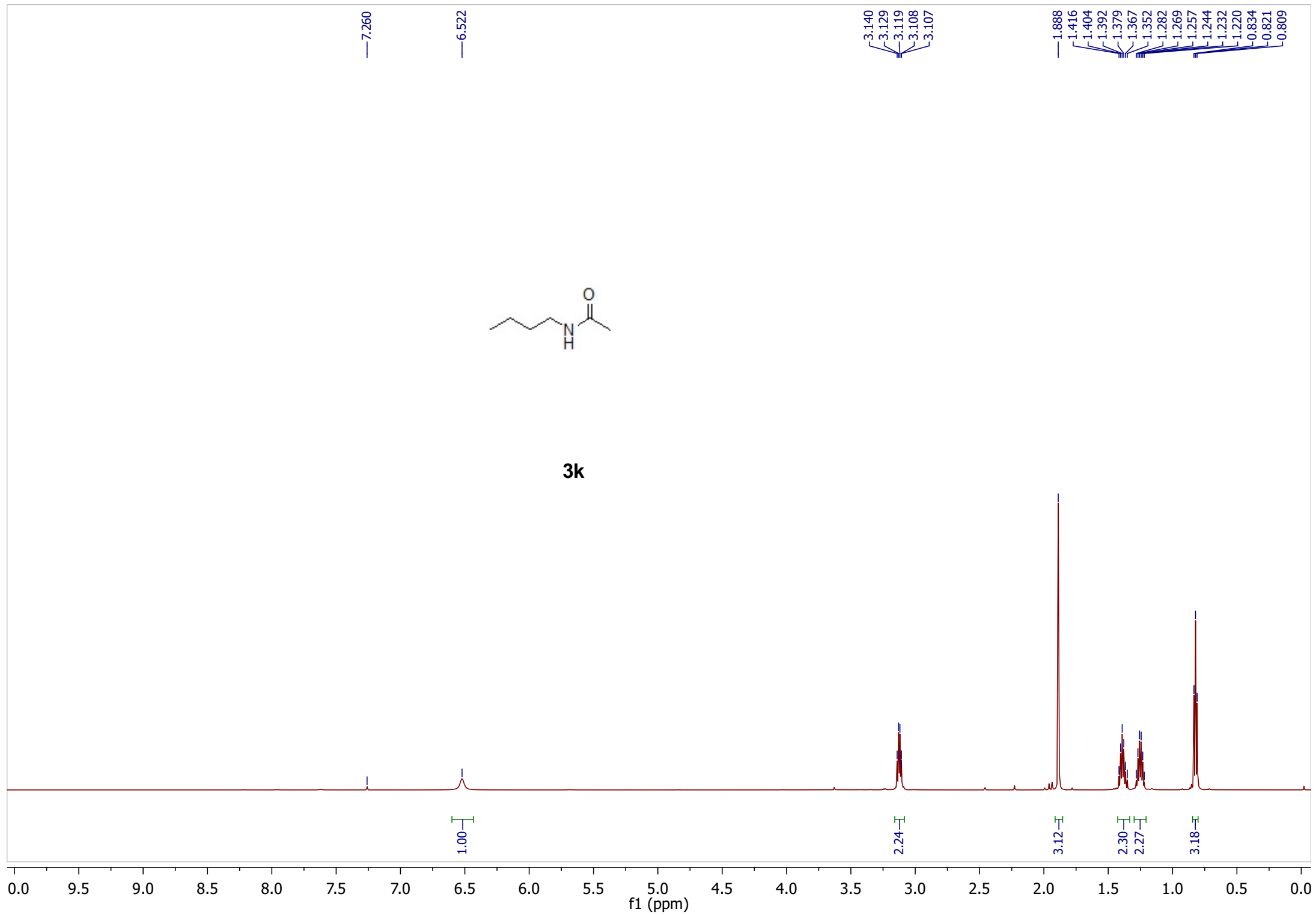
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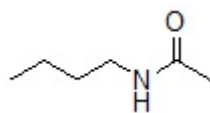
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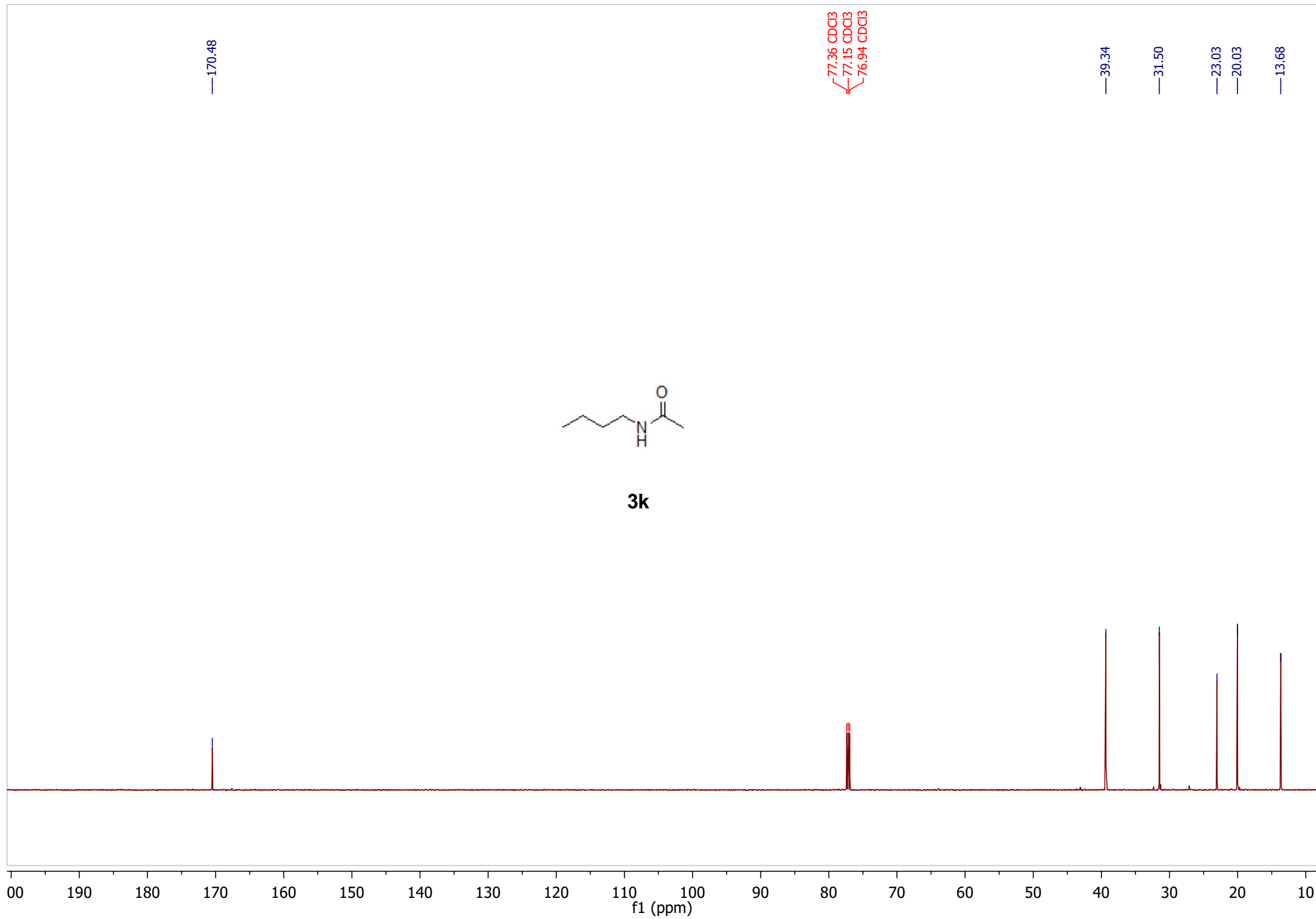


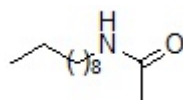
3k



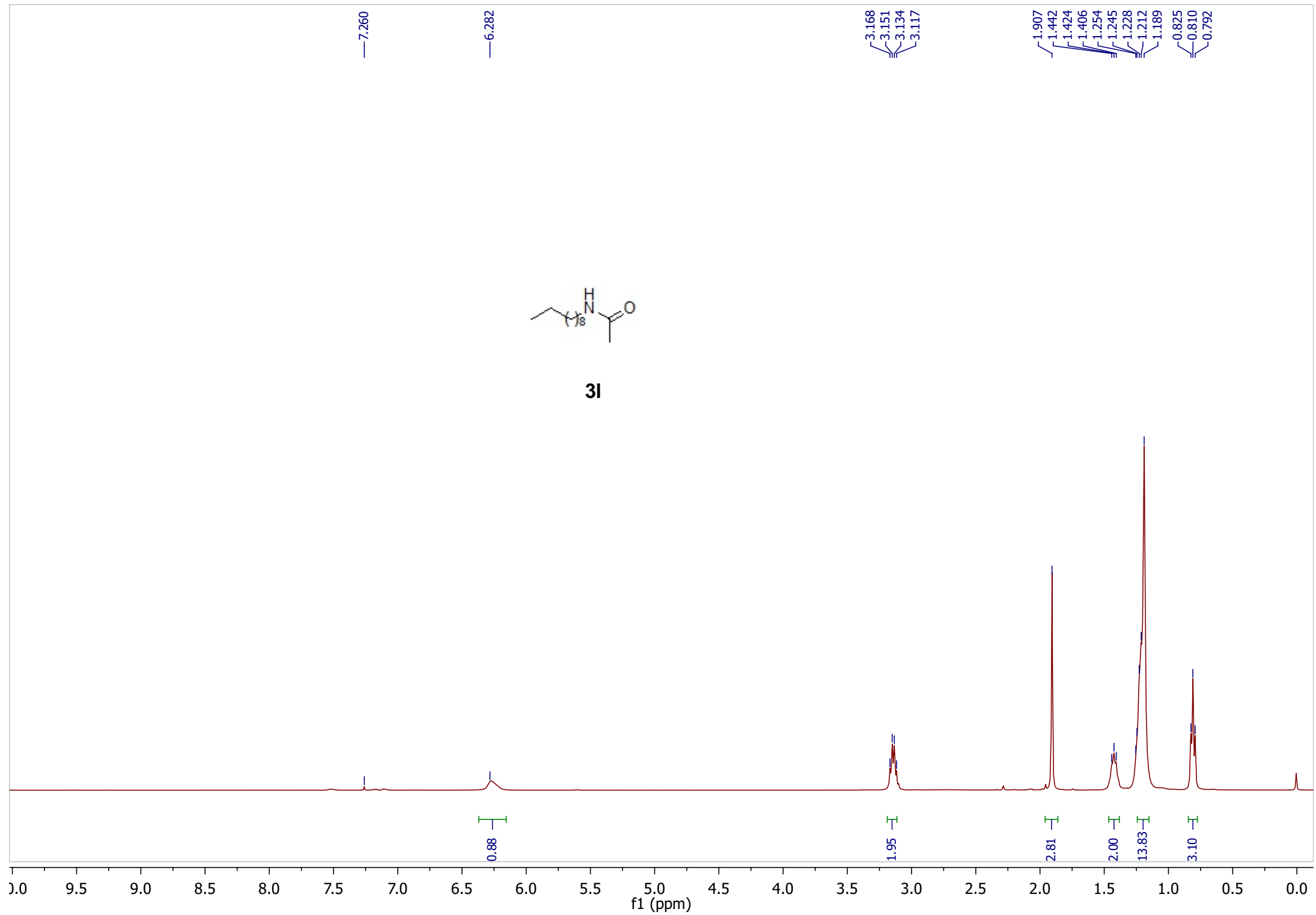


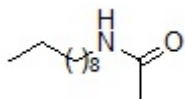
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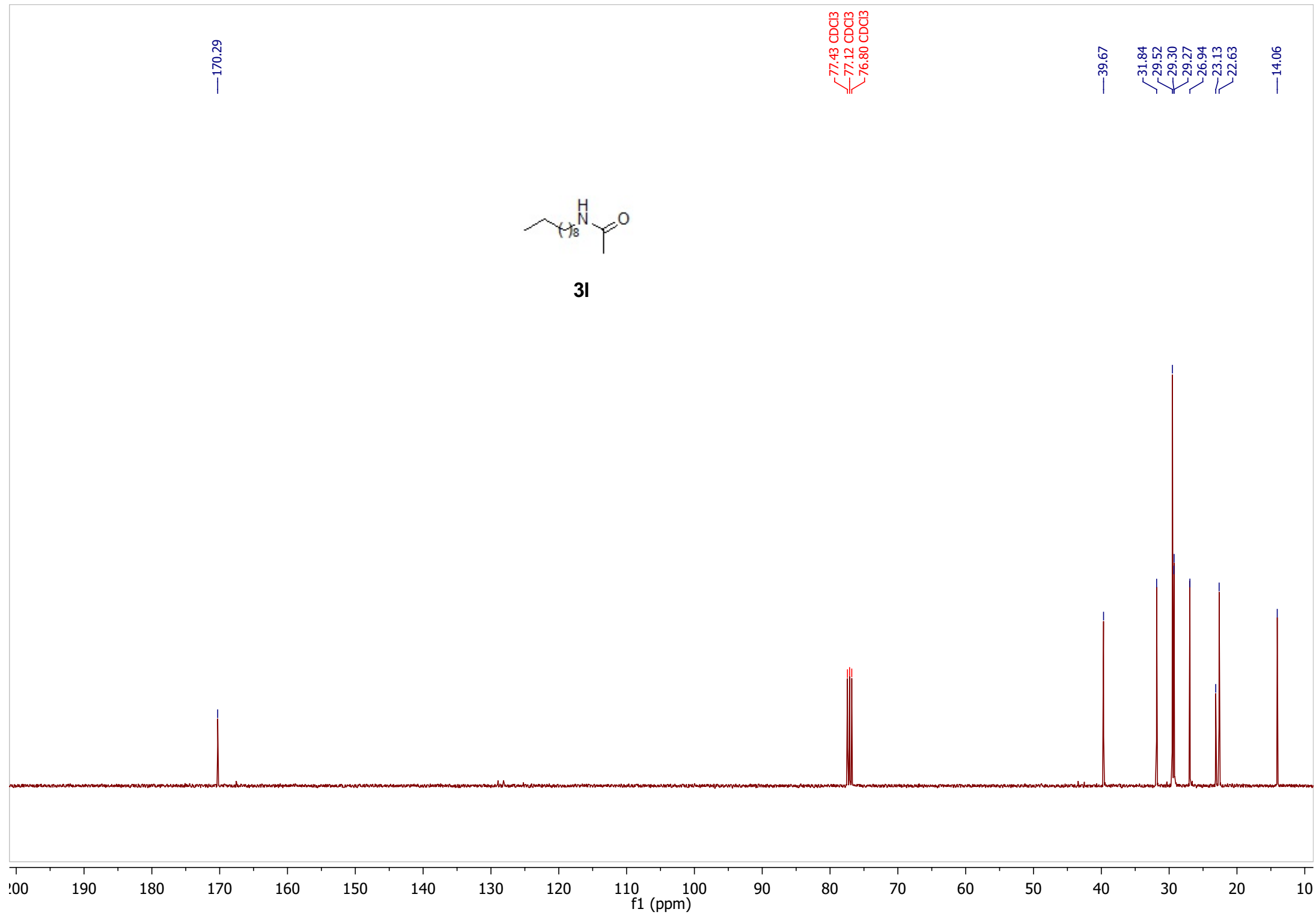


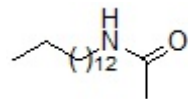
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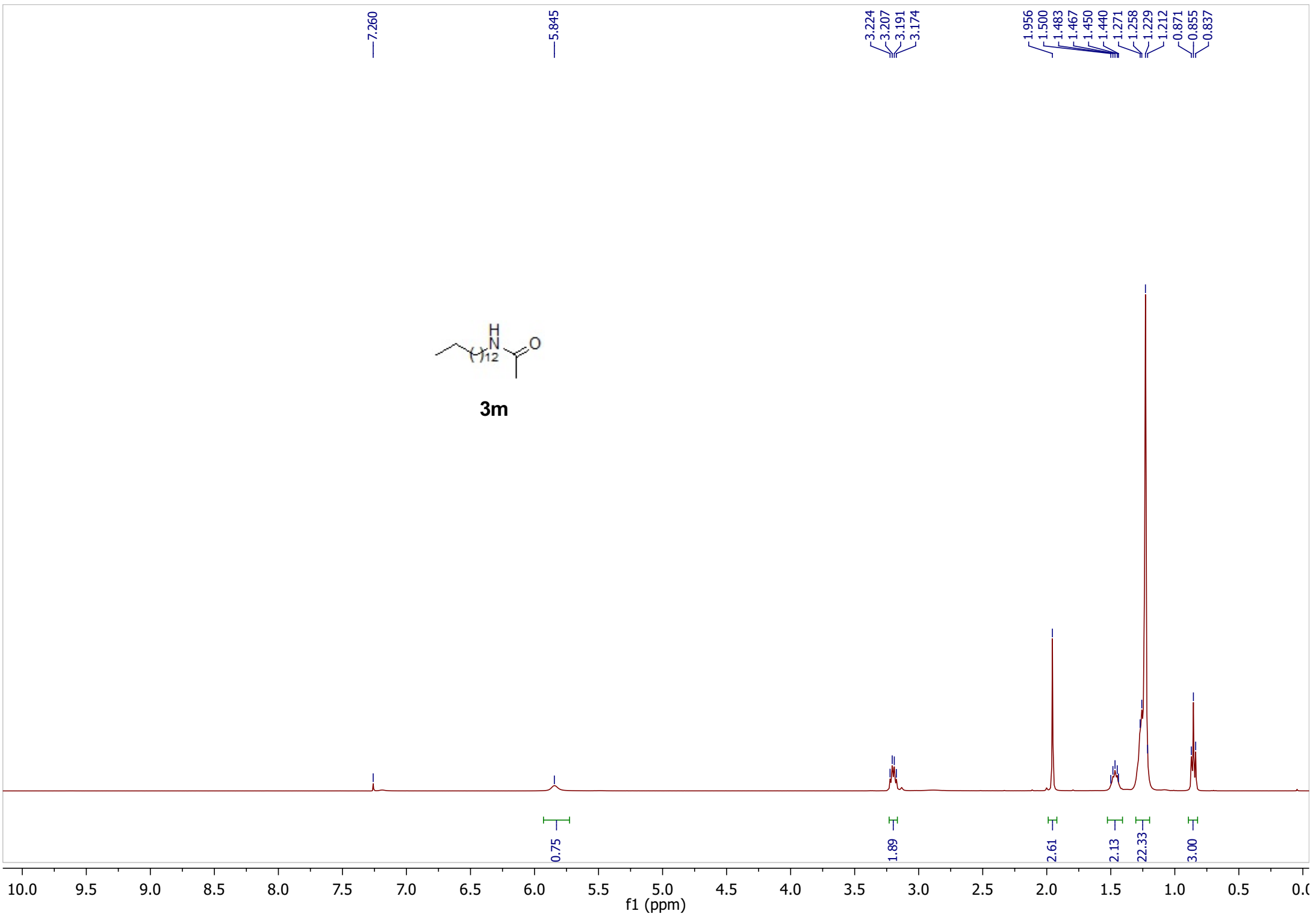


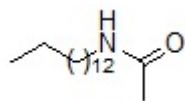
3I



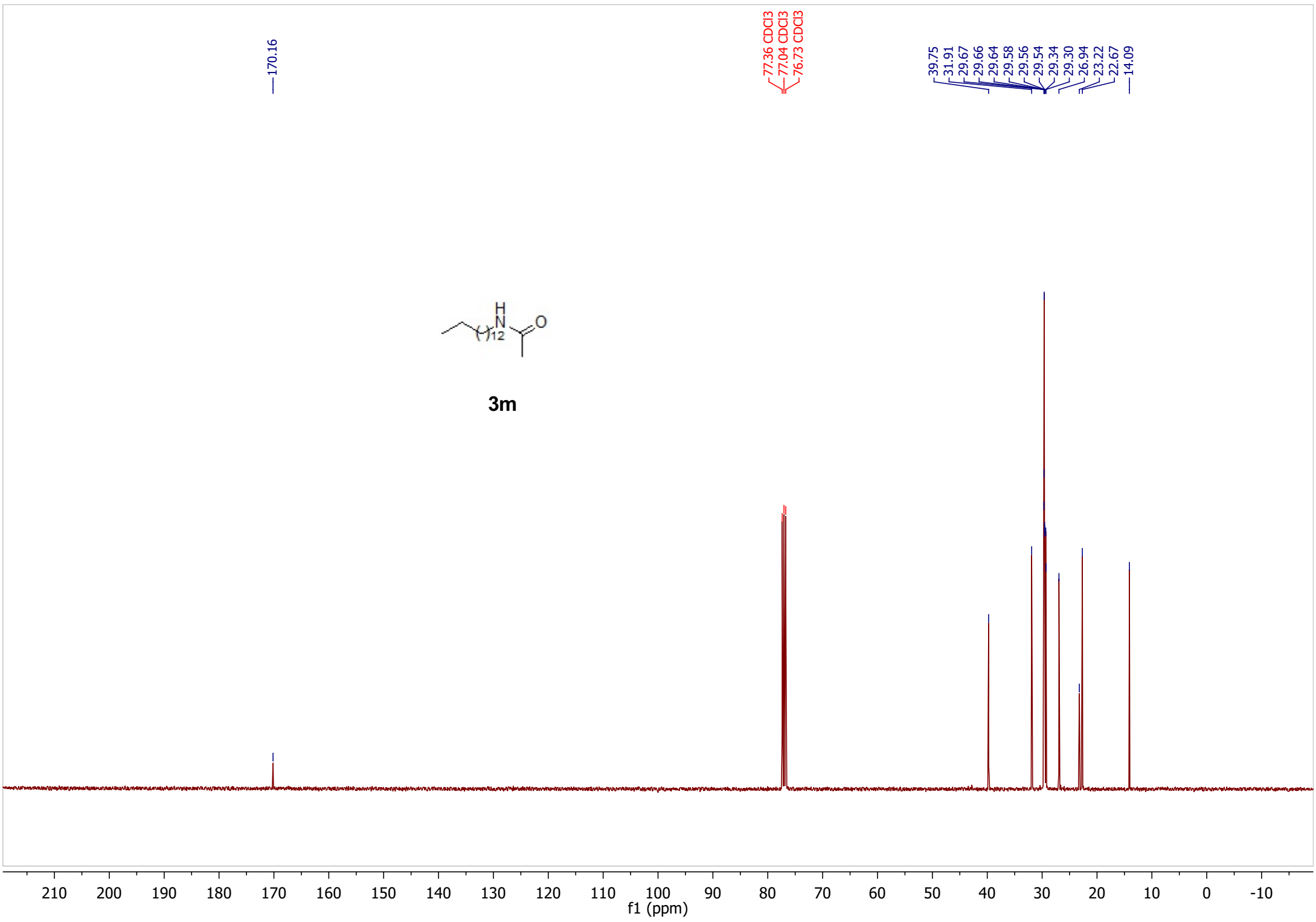


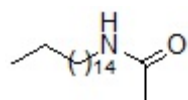
3m



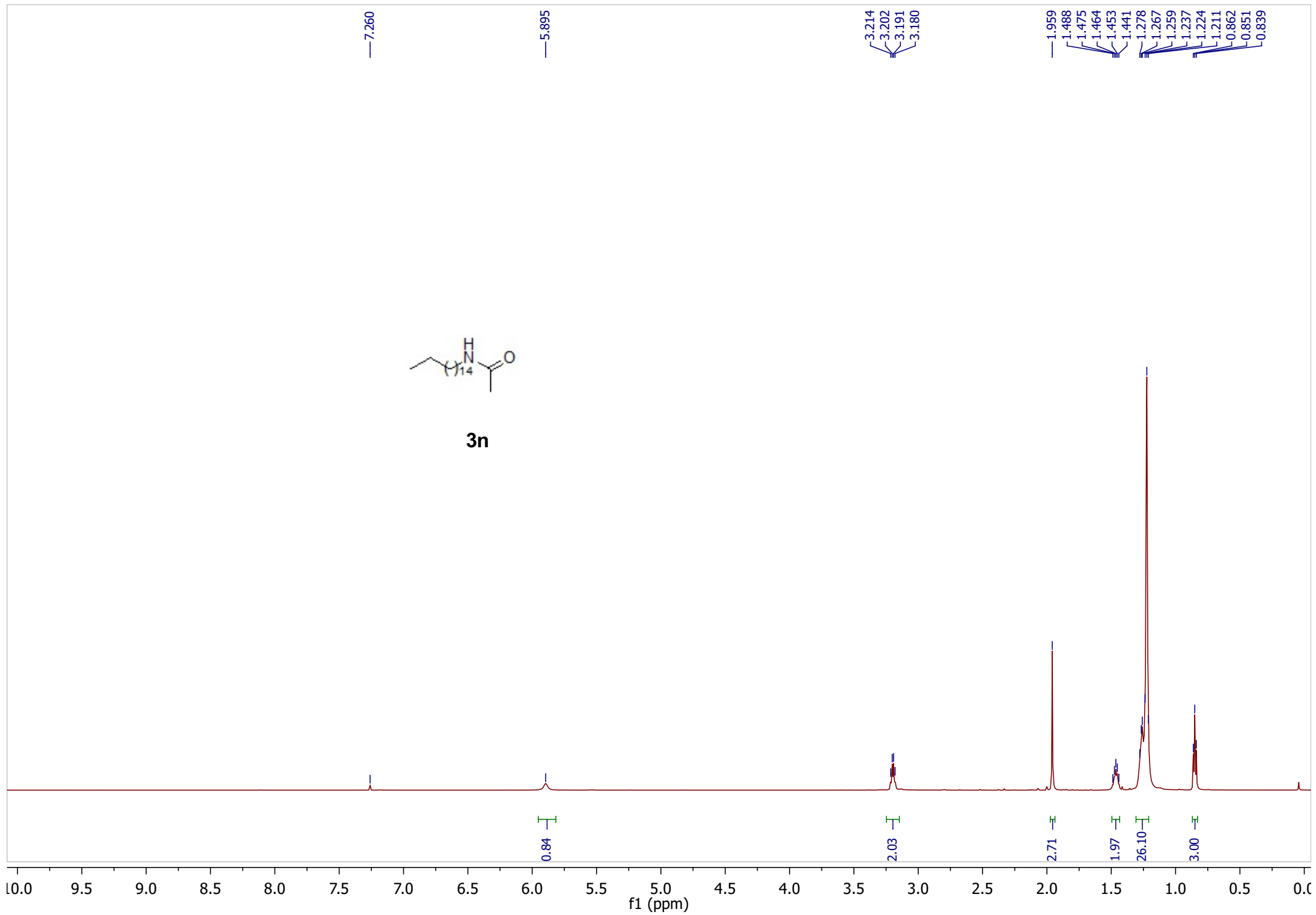


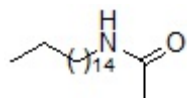
3m



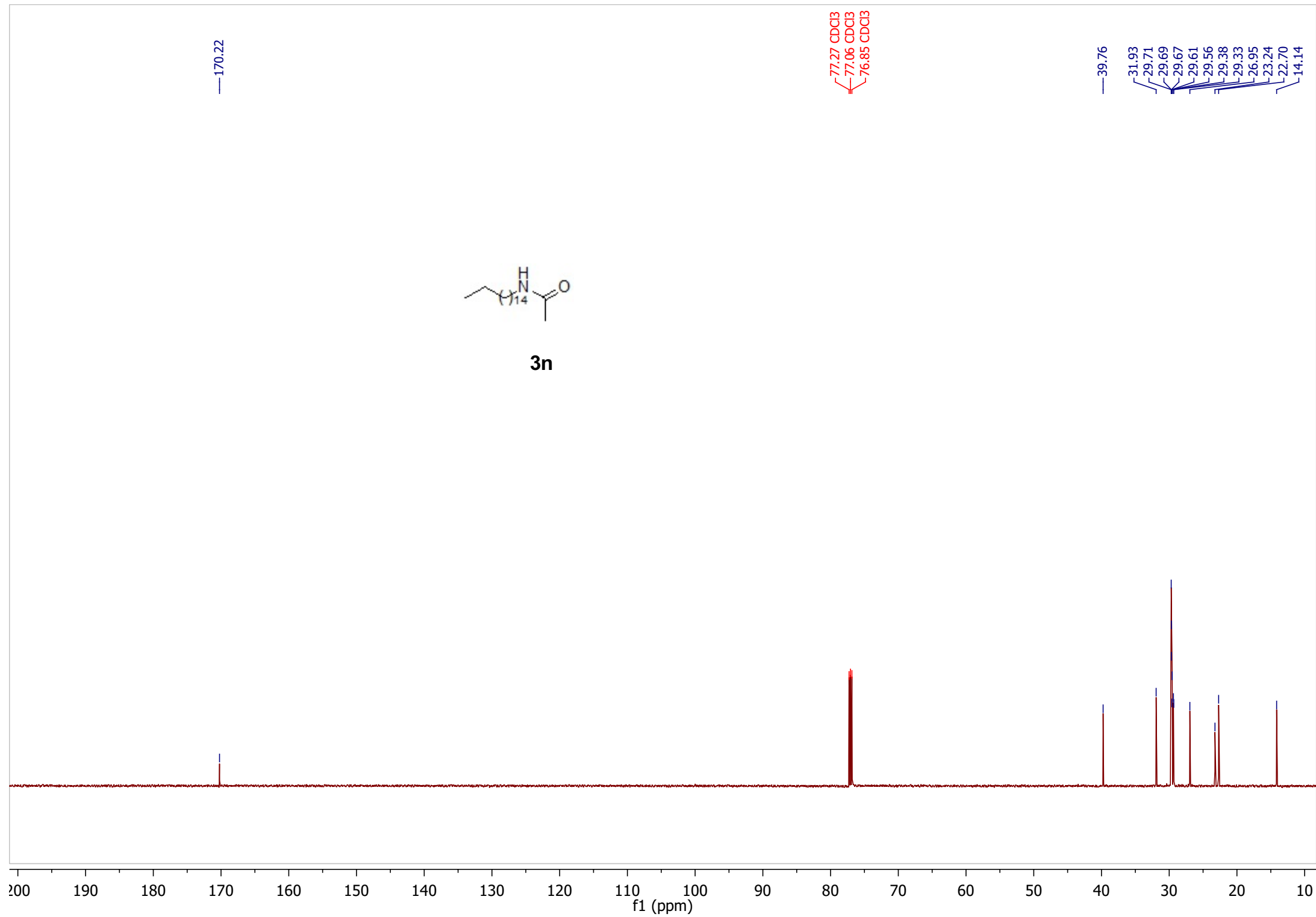


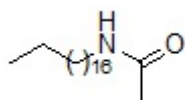
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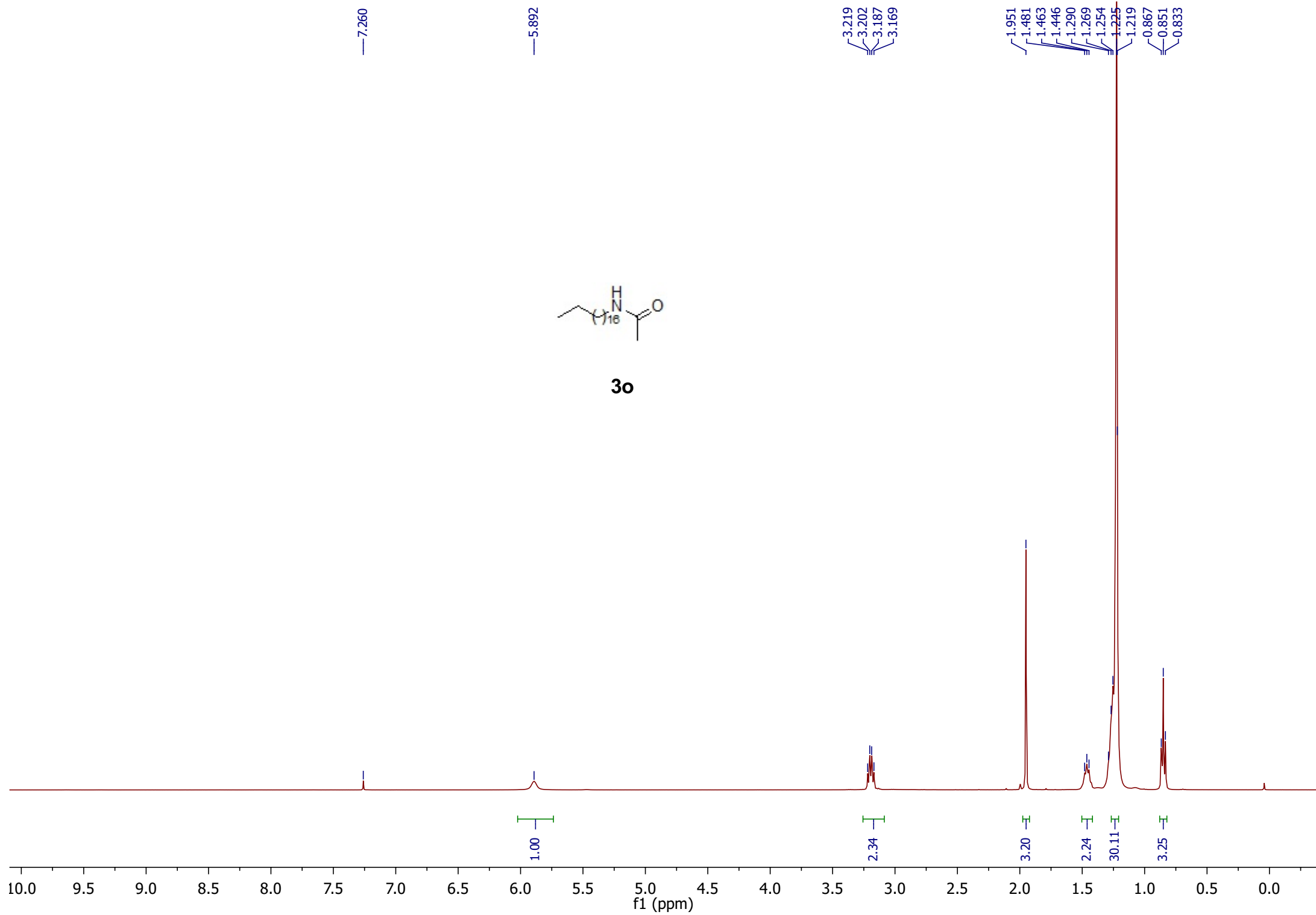


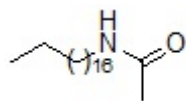
3n



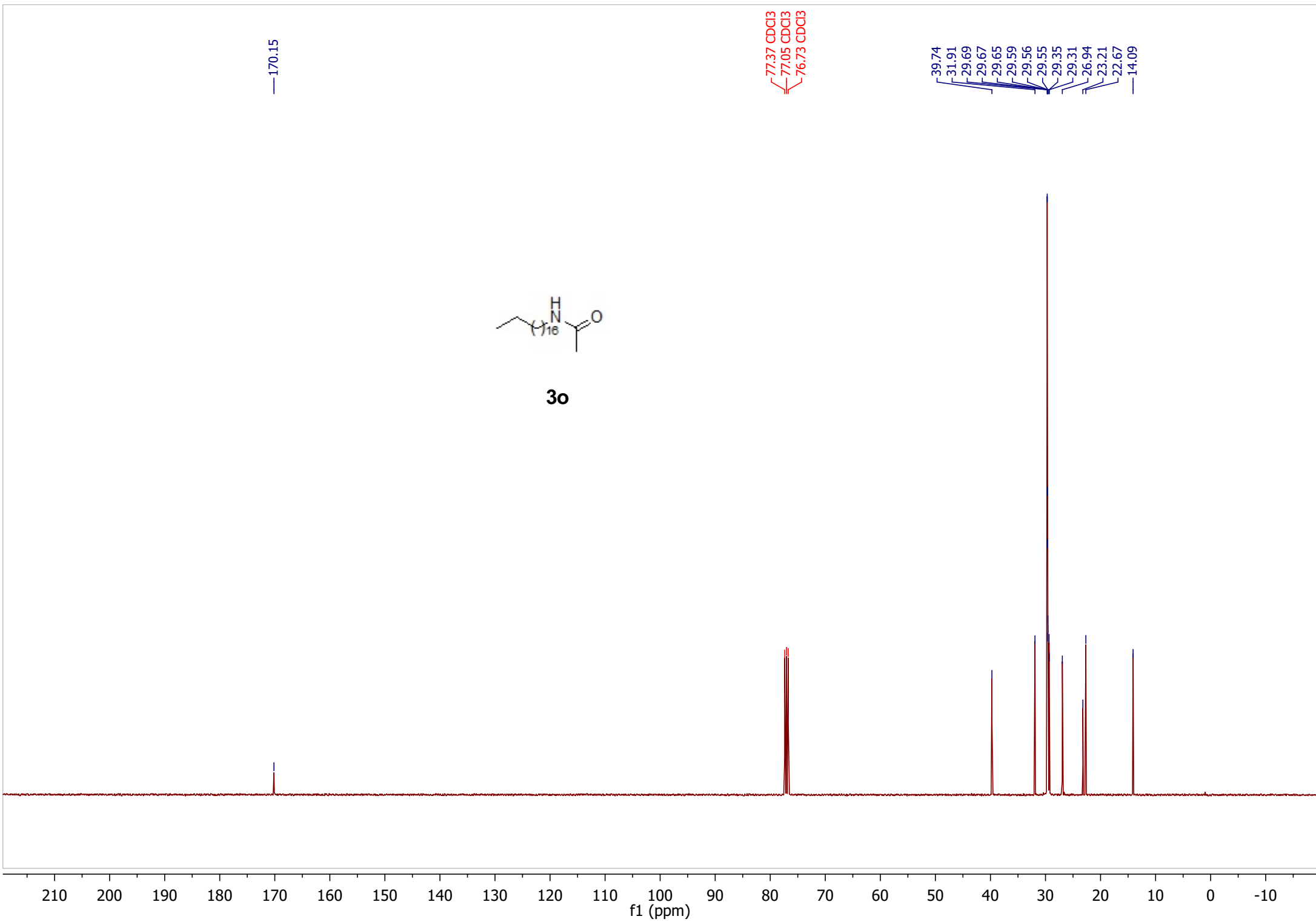


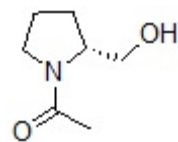
3o



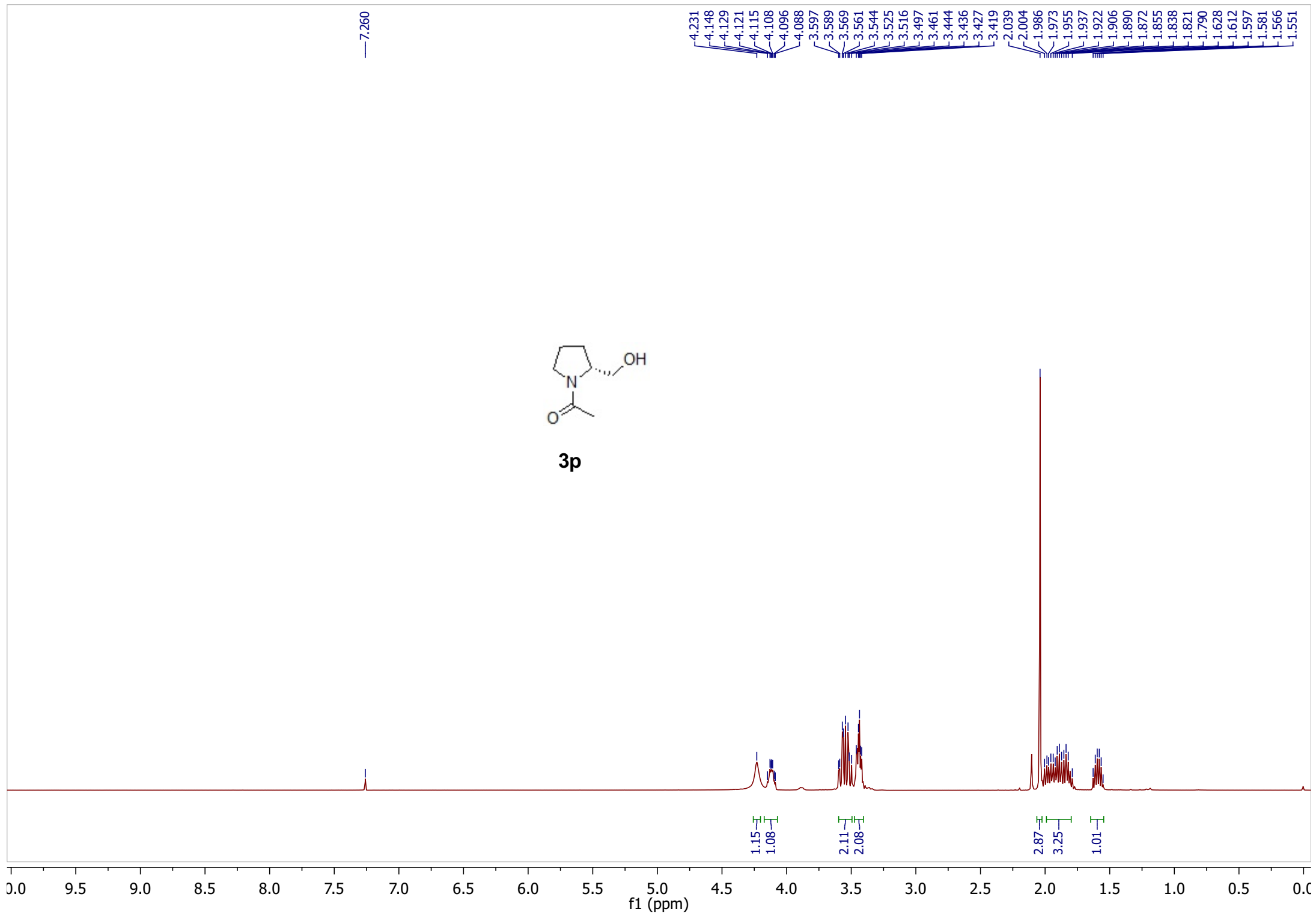


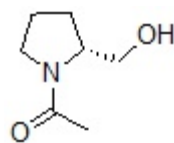
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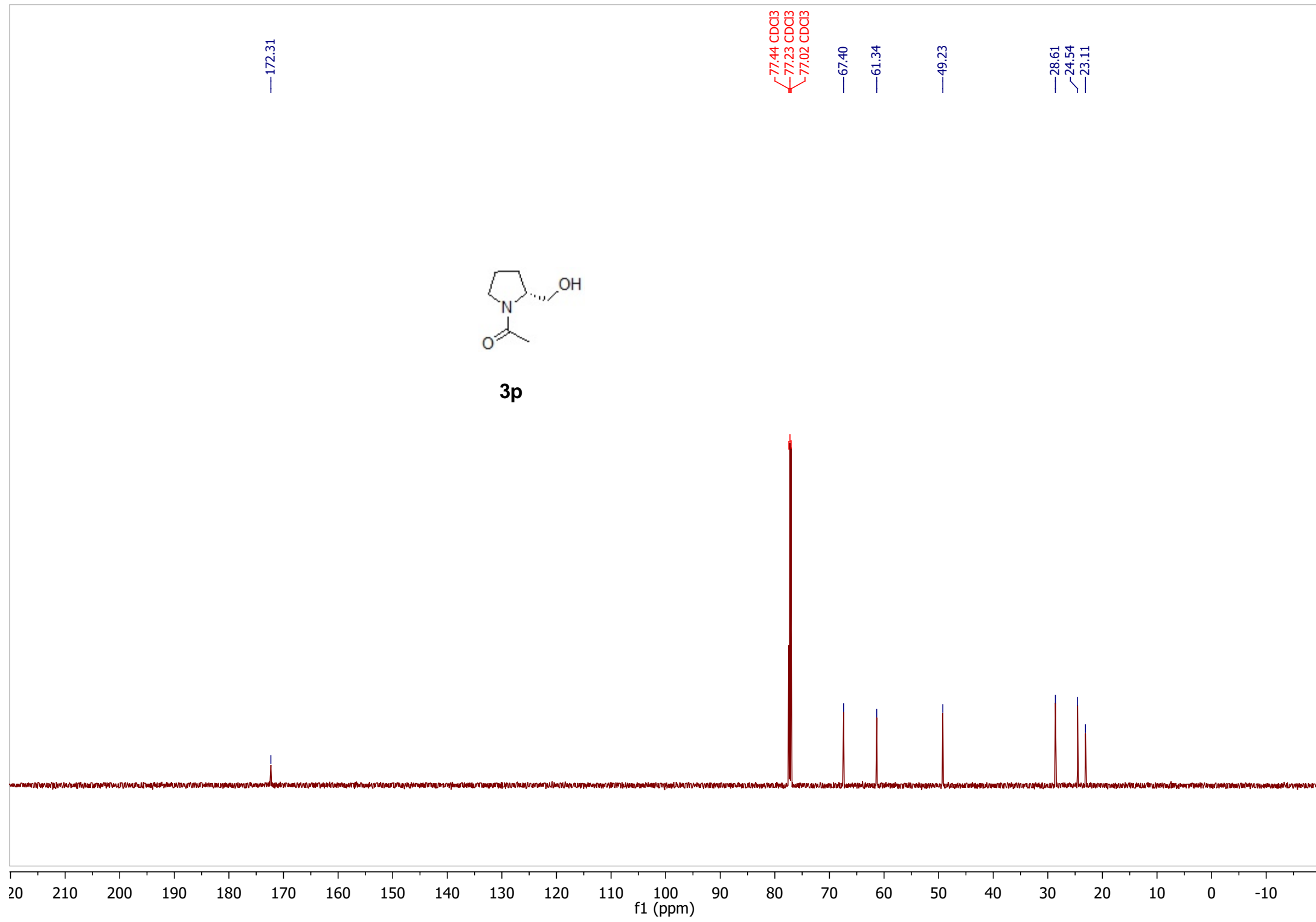


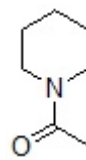
3p



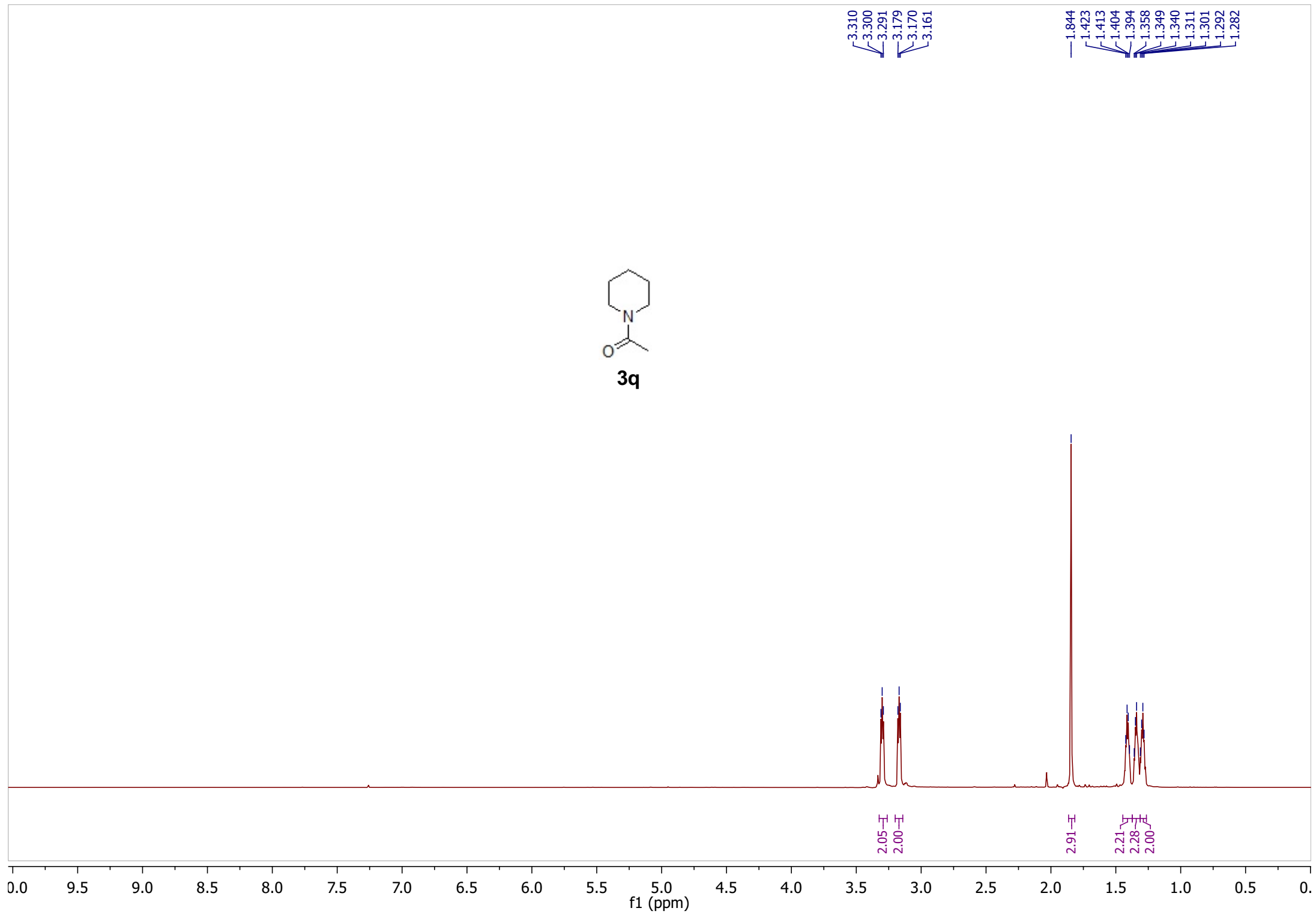


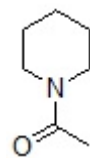
3p



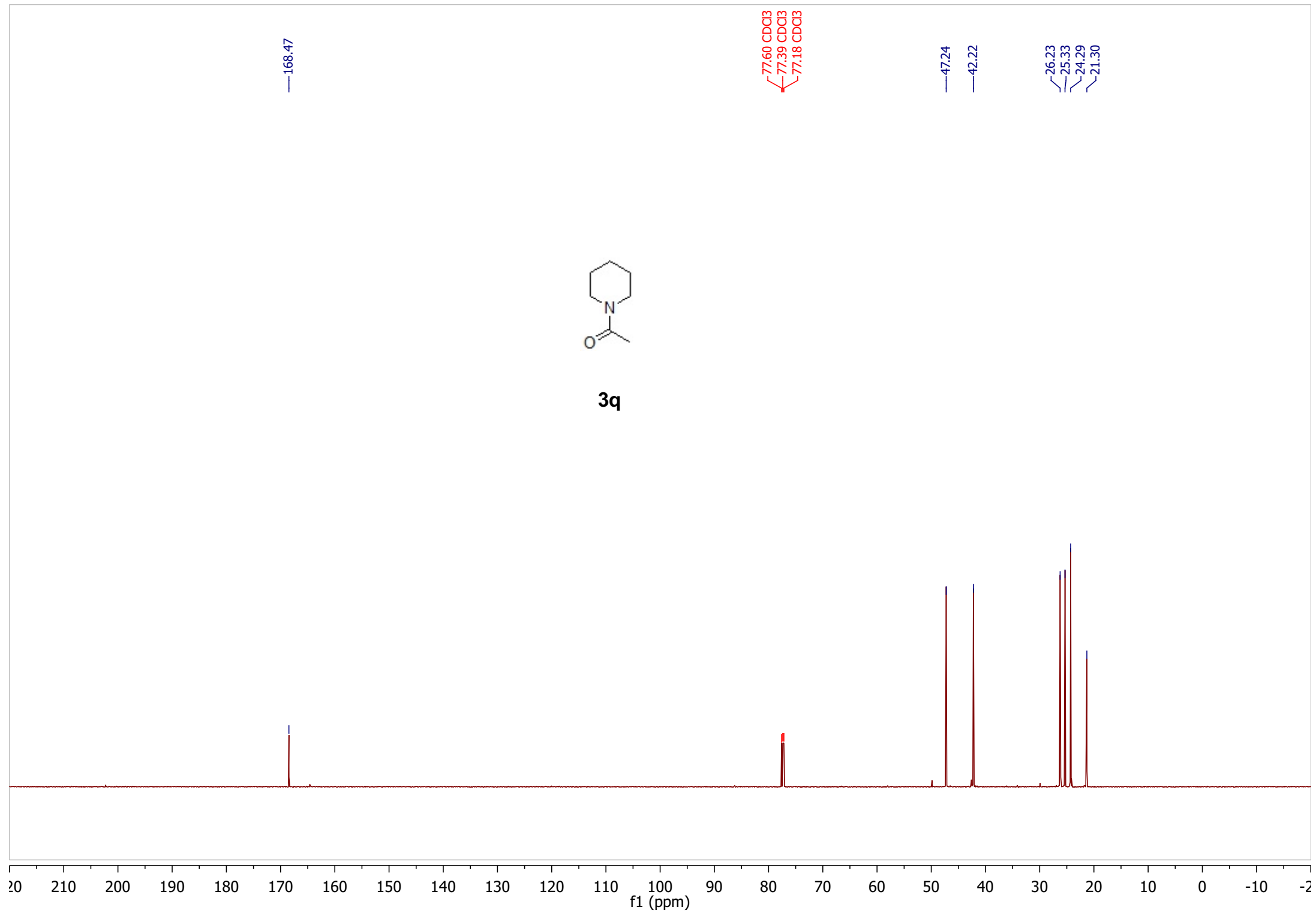


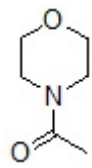
3q



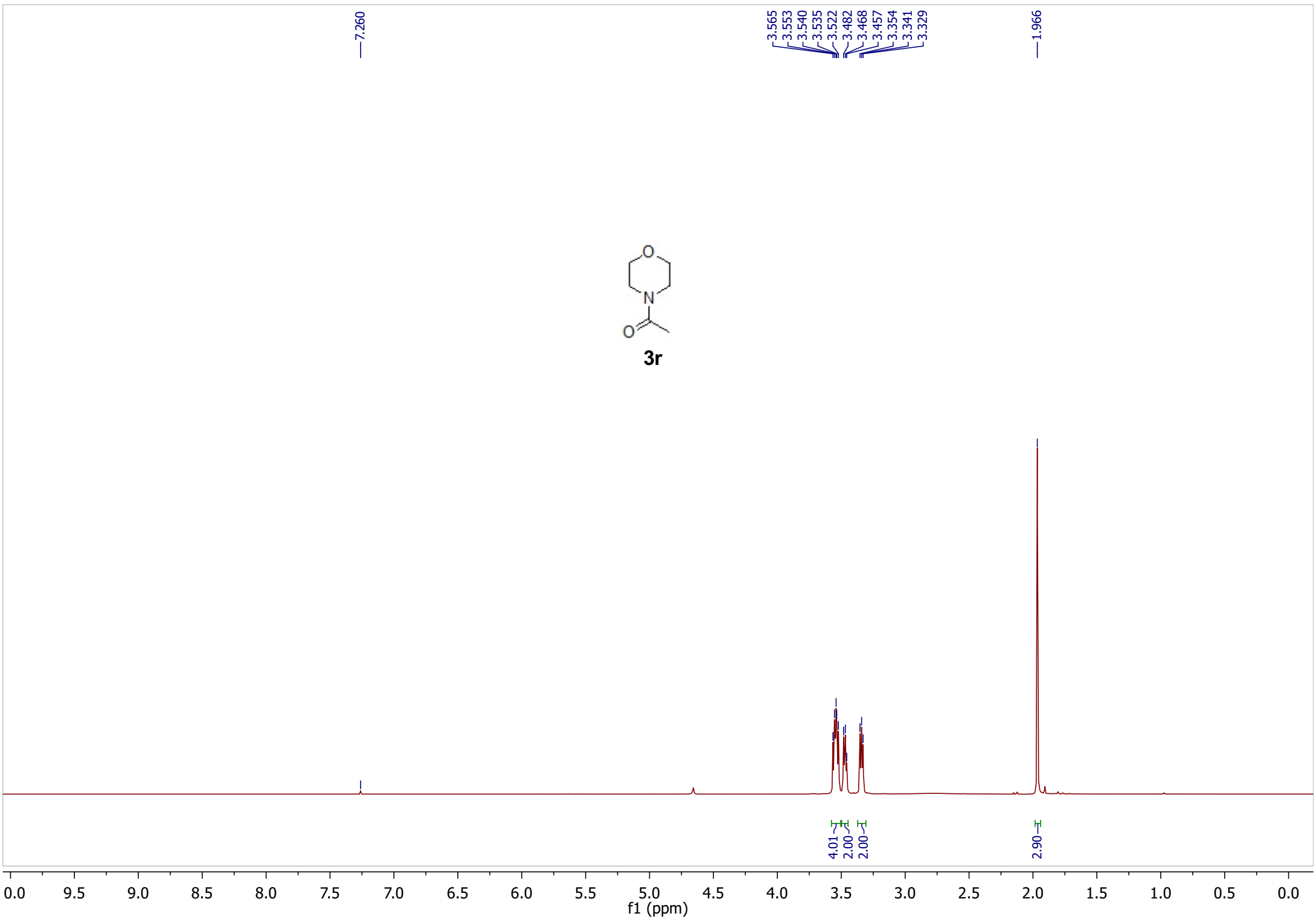


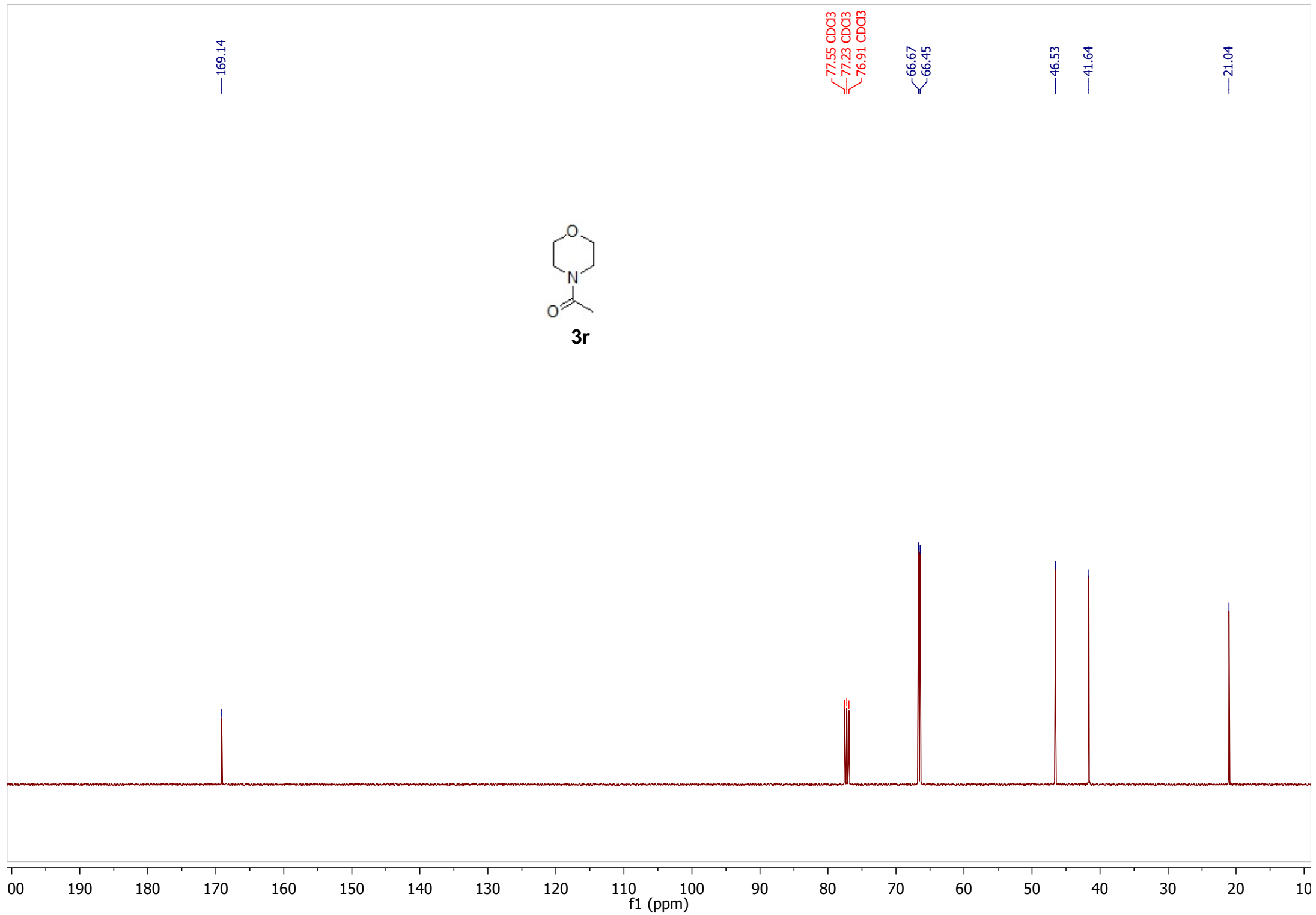
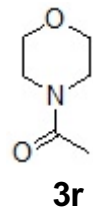
3q

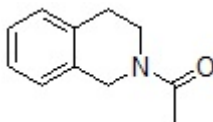




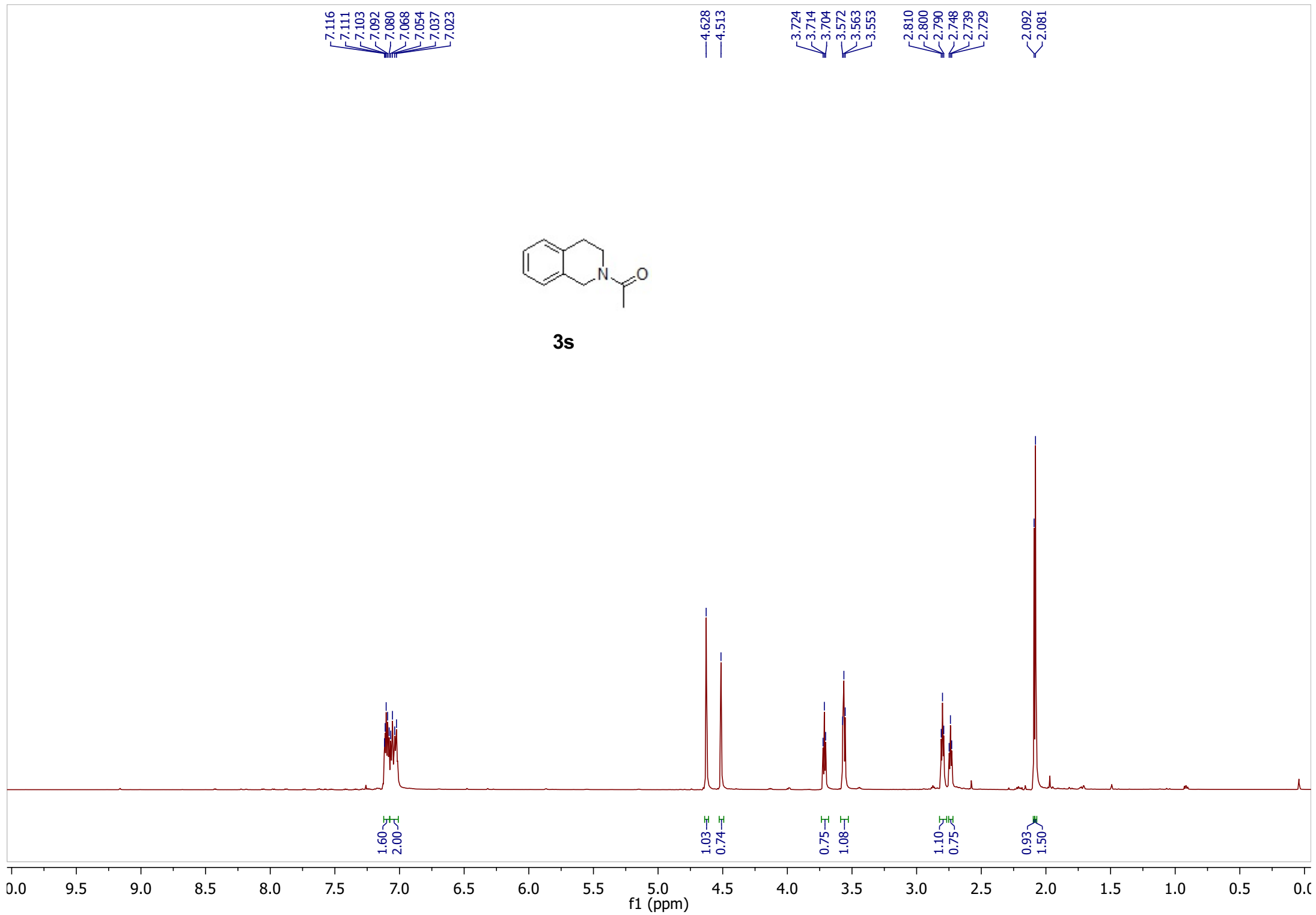
3r







3s



169.46
169.37

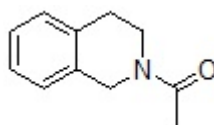
134.95
134.04
133.41
132.55
128.86
128.31
126.85
126.56
126.53
126.49
126.32
126.04

77.65 CDCl3
77.44 CDCl3
77.23 CDCl3

47.98
44.00
43.94
39.42

29.36
28.47

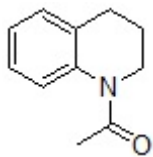
21.95
21.67



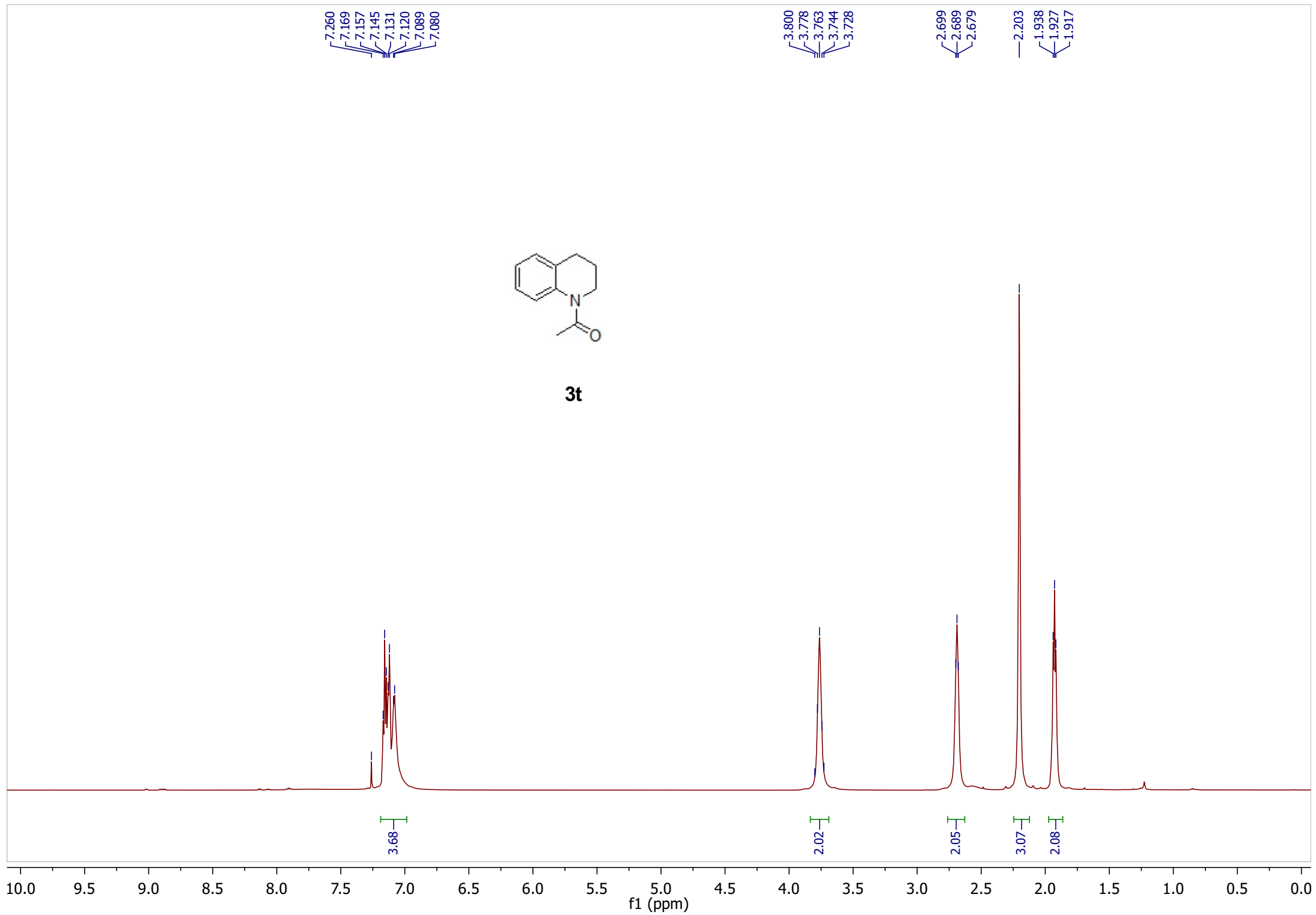
3s

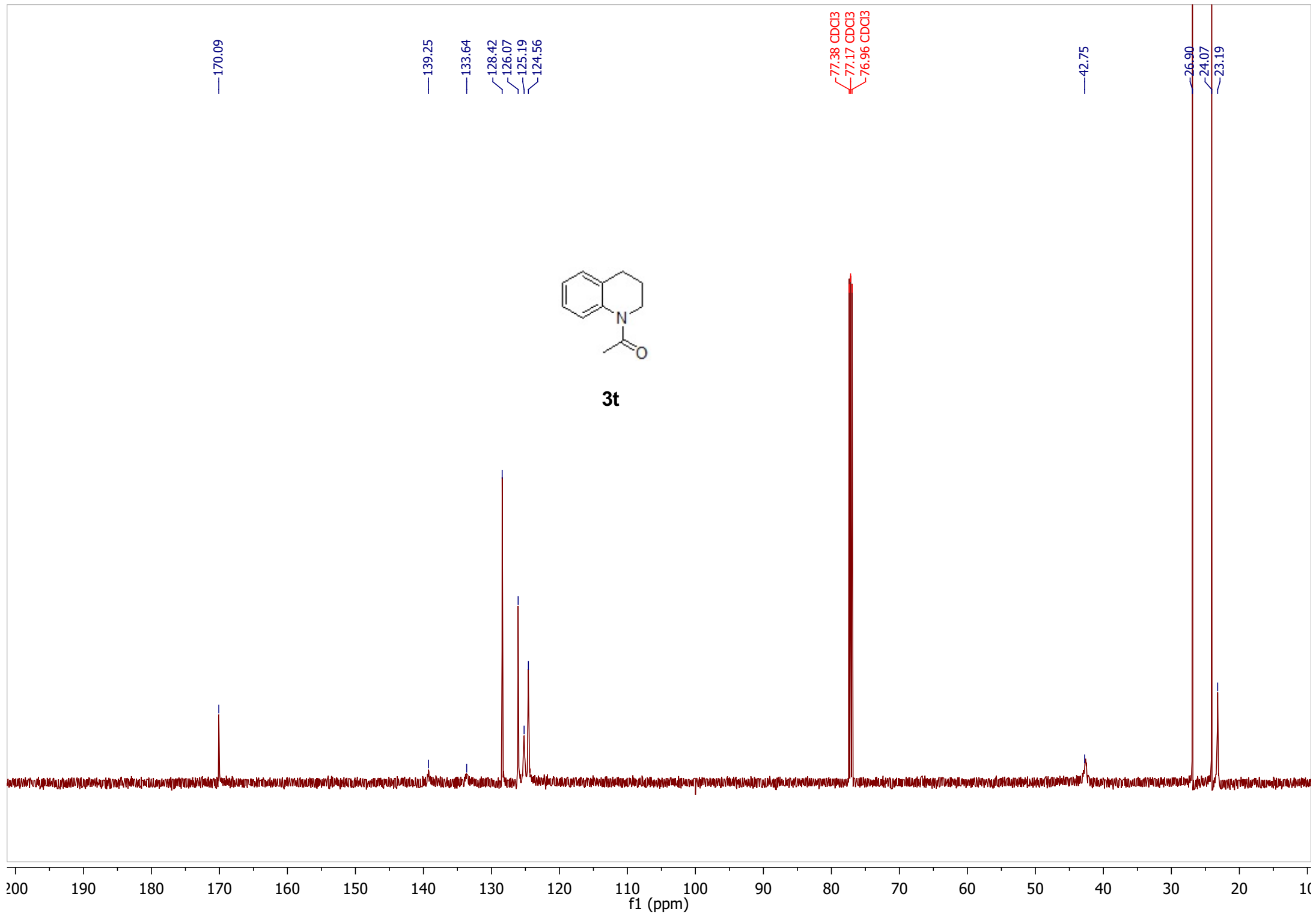
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10

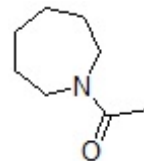
f1 (ppm)



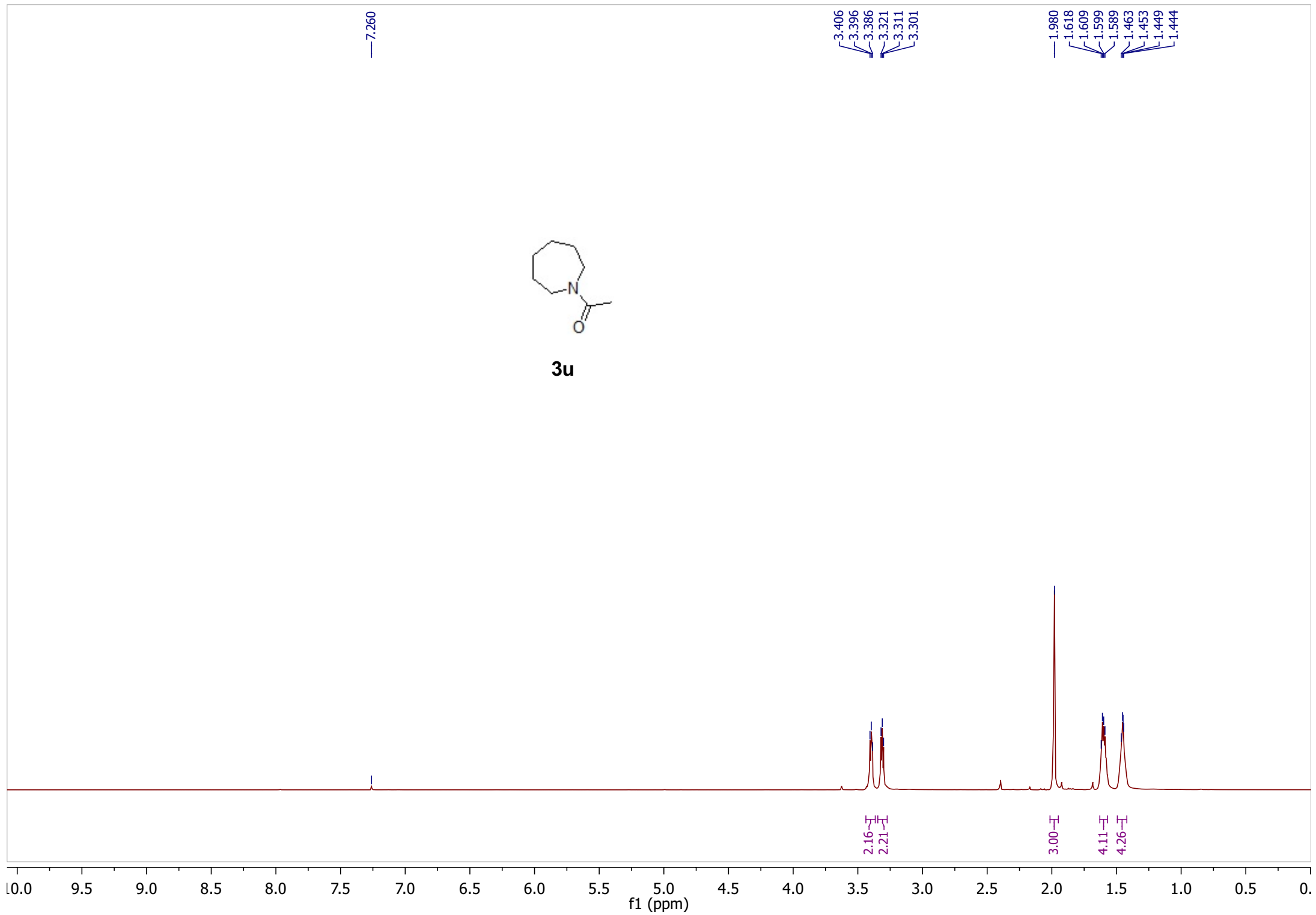
3t

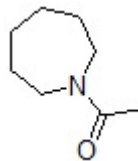






3u





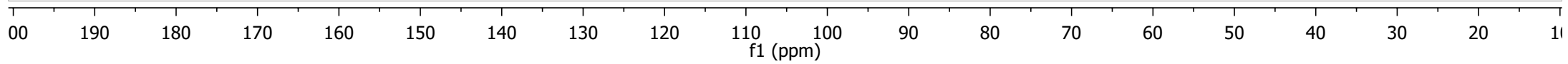
3u

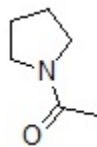
—170.26

77.42 CDCl₃
77.21 CDCl₃
77.00 CDCl₃

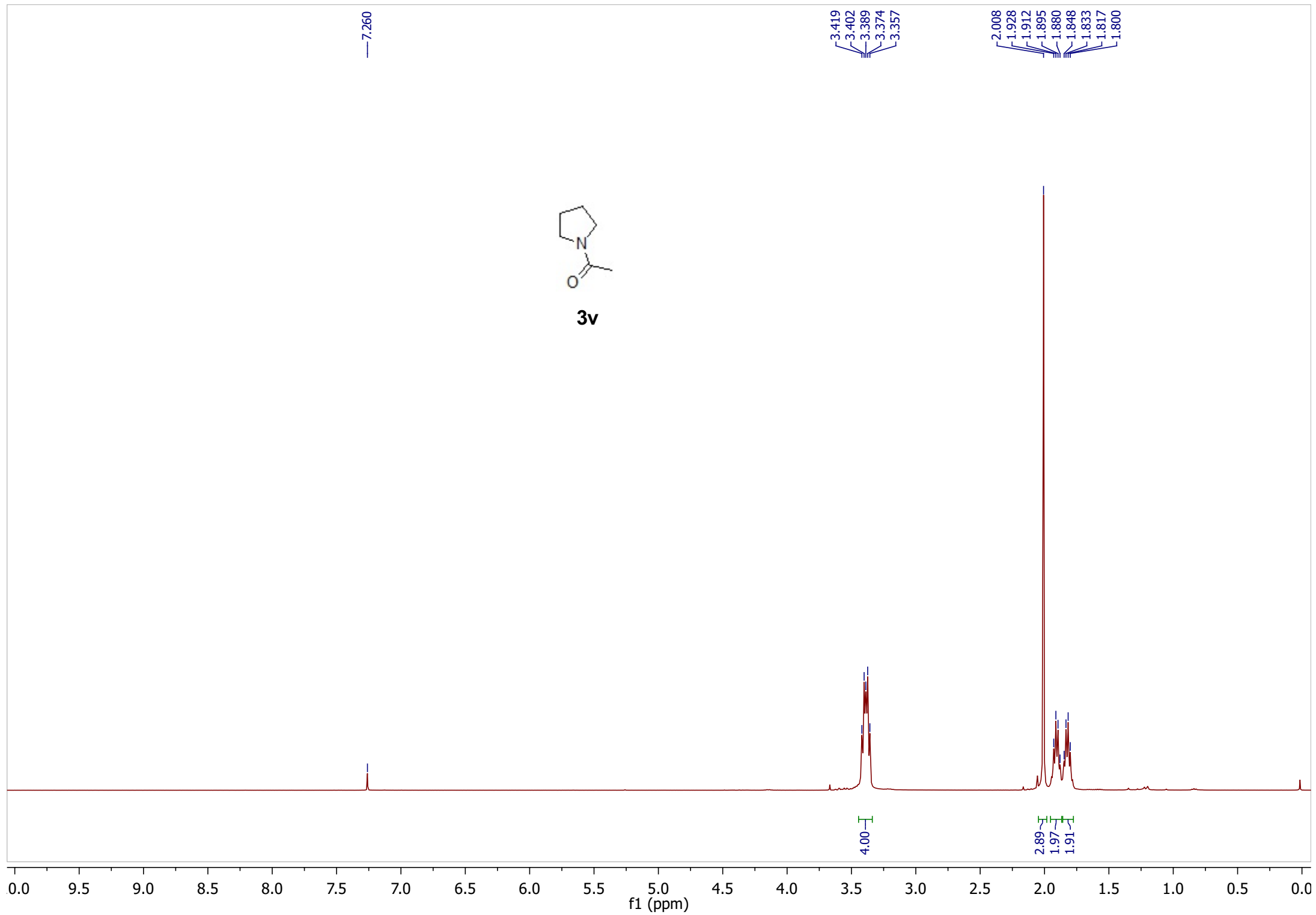
—48.62
—45.67

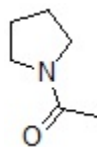
28.90
27.47
26.97
26.81
—21.52



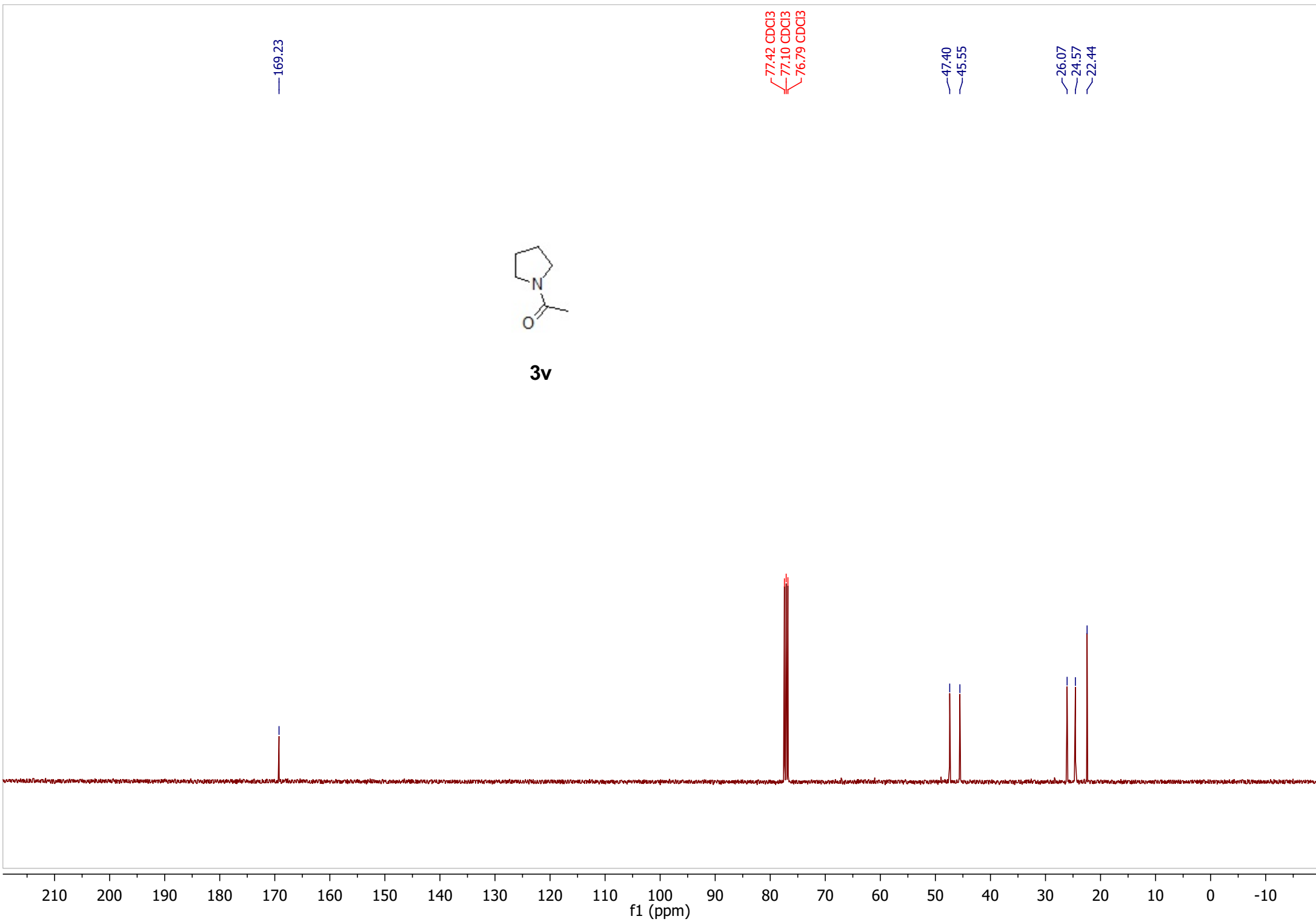


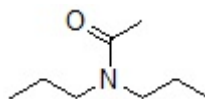
3v



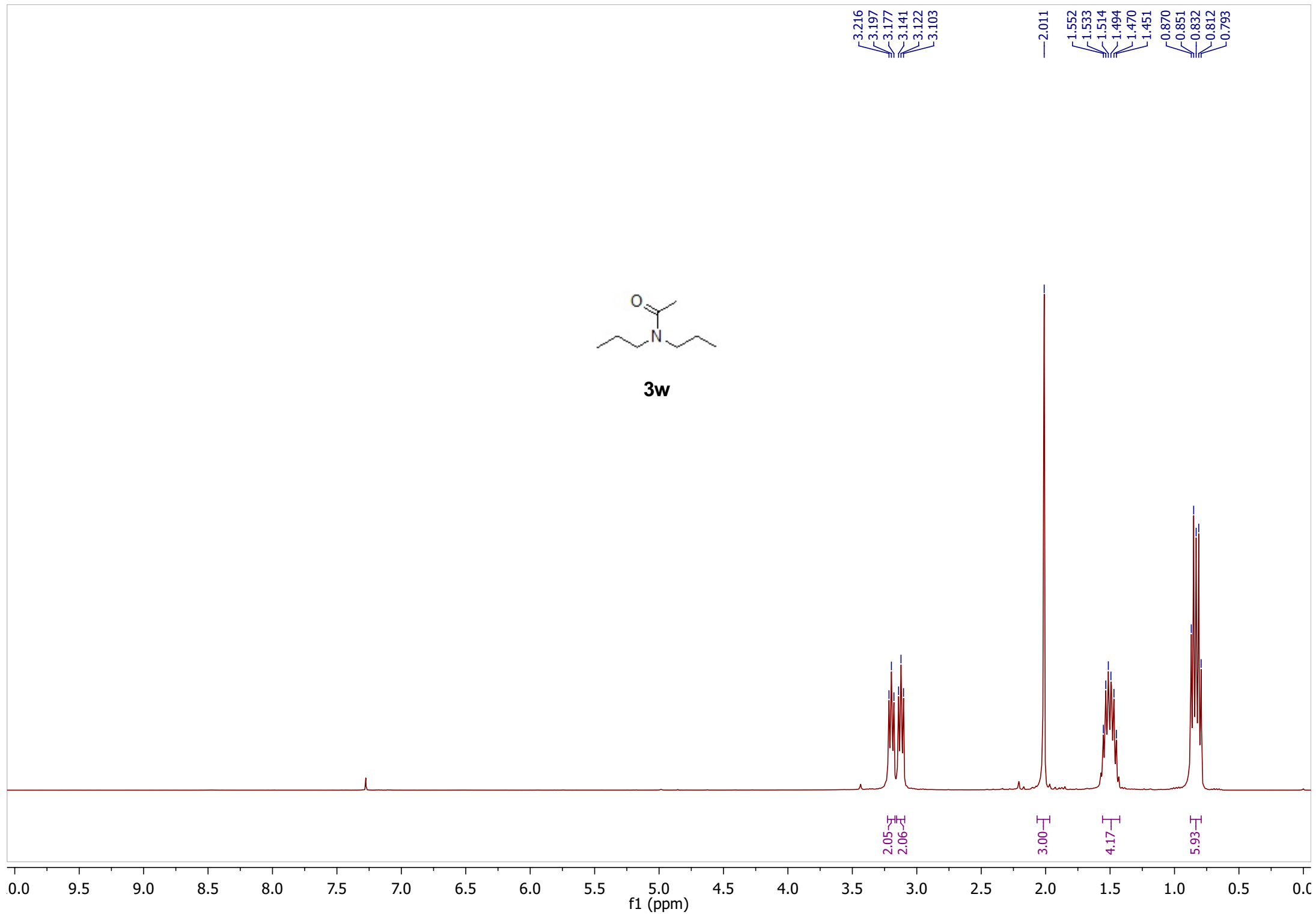


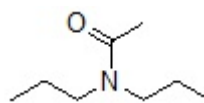
3v



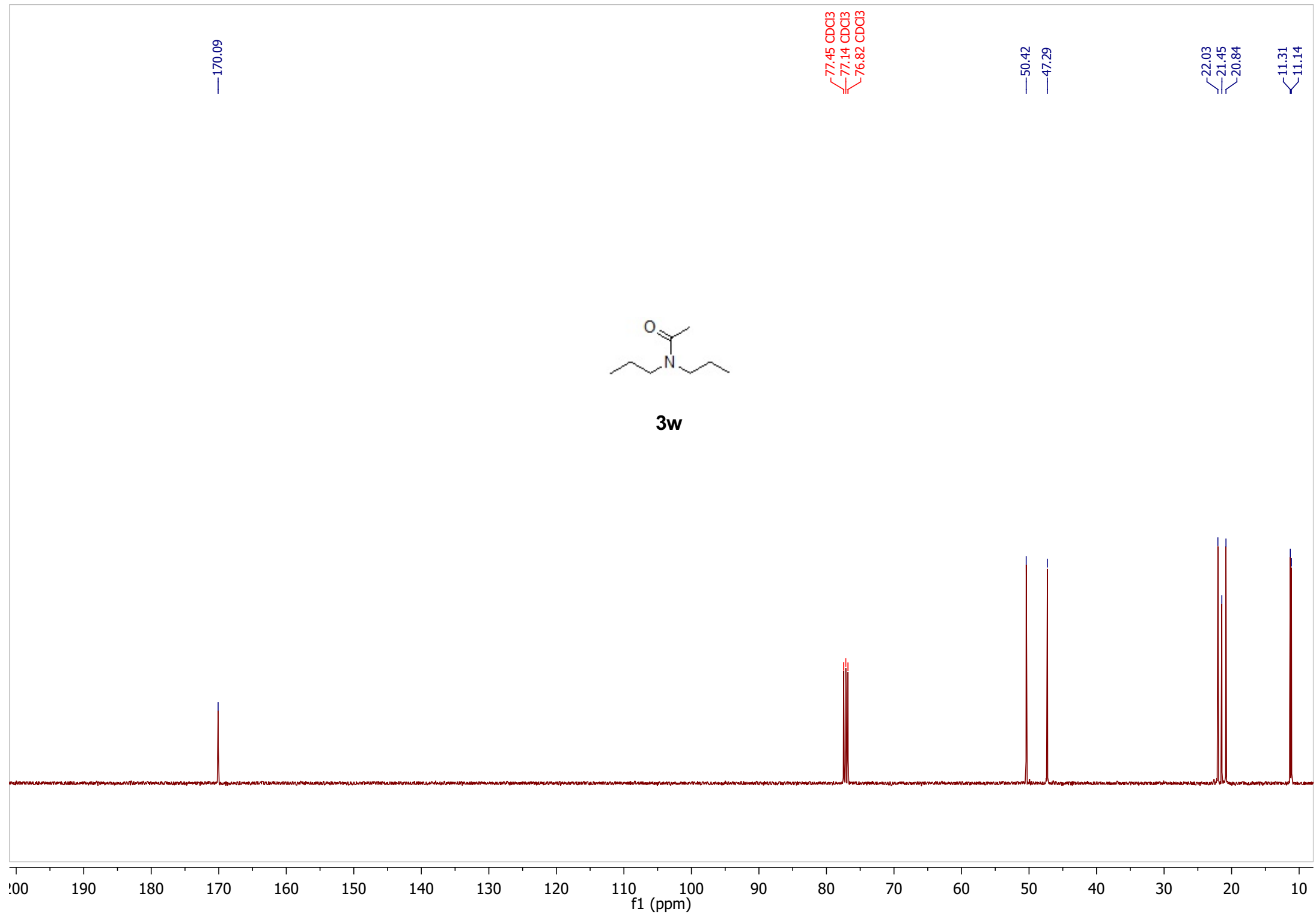


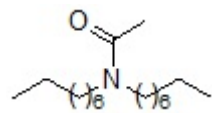
3w



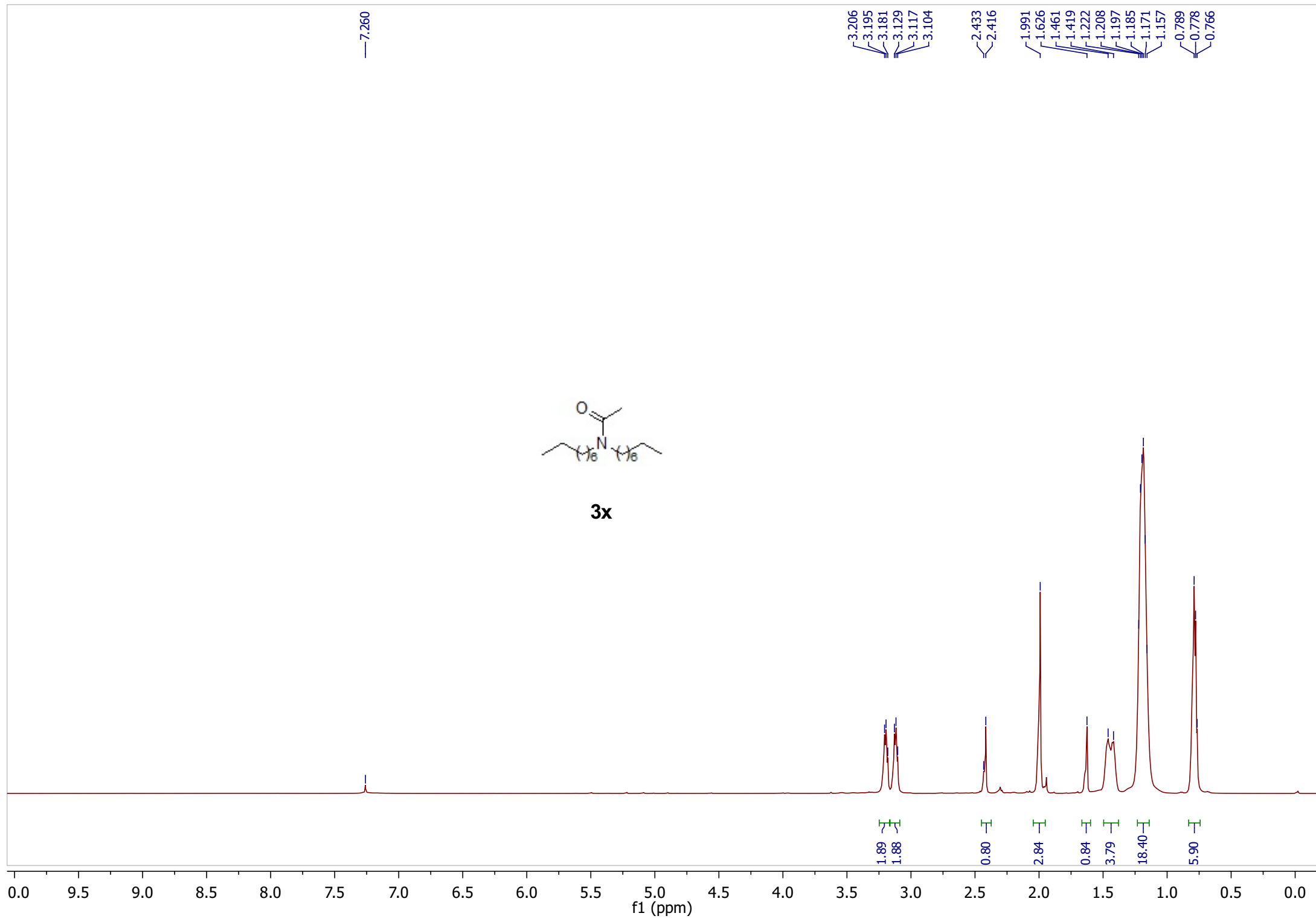


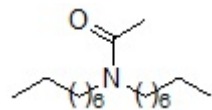
3w



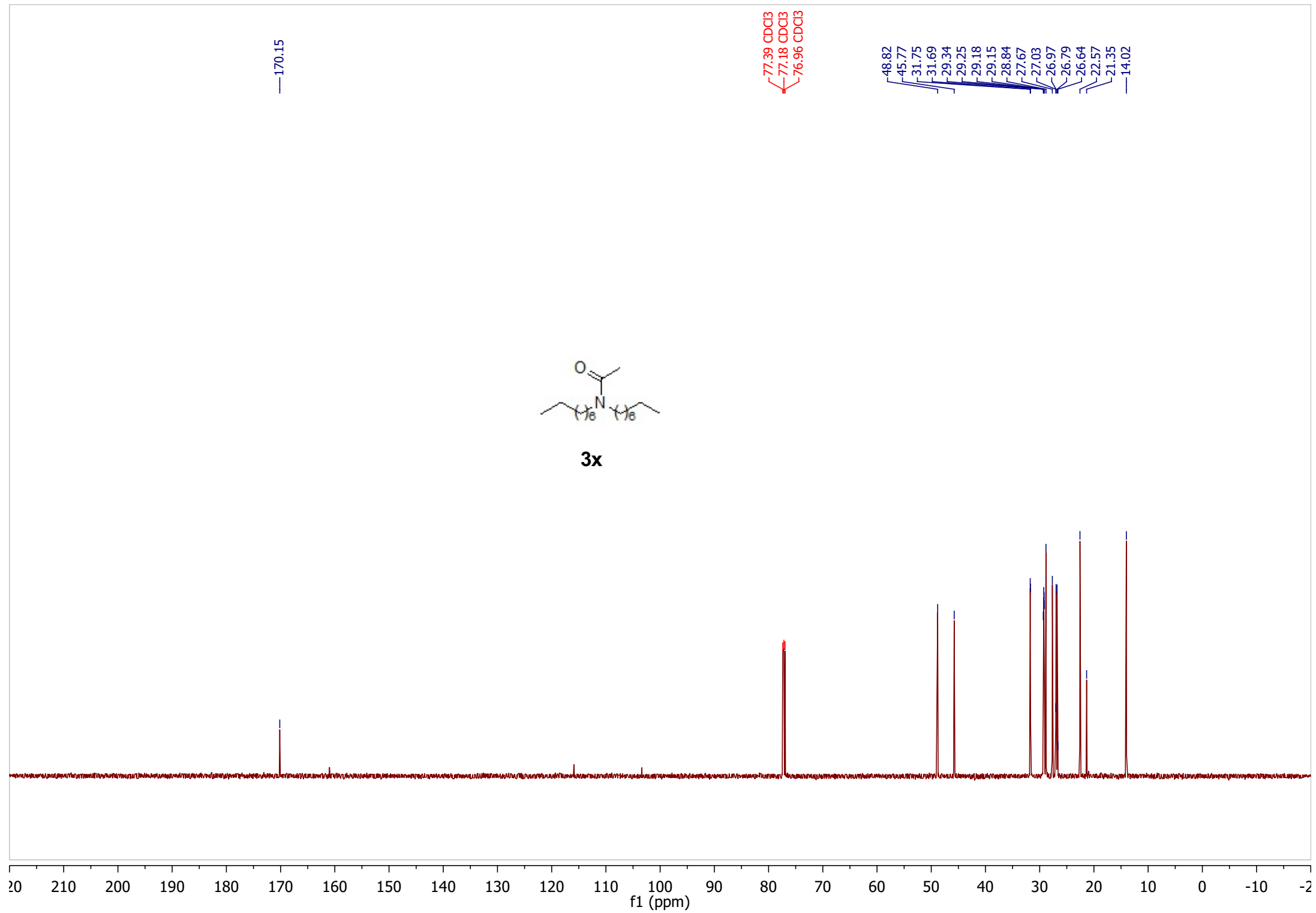


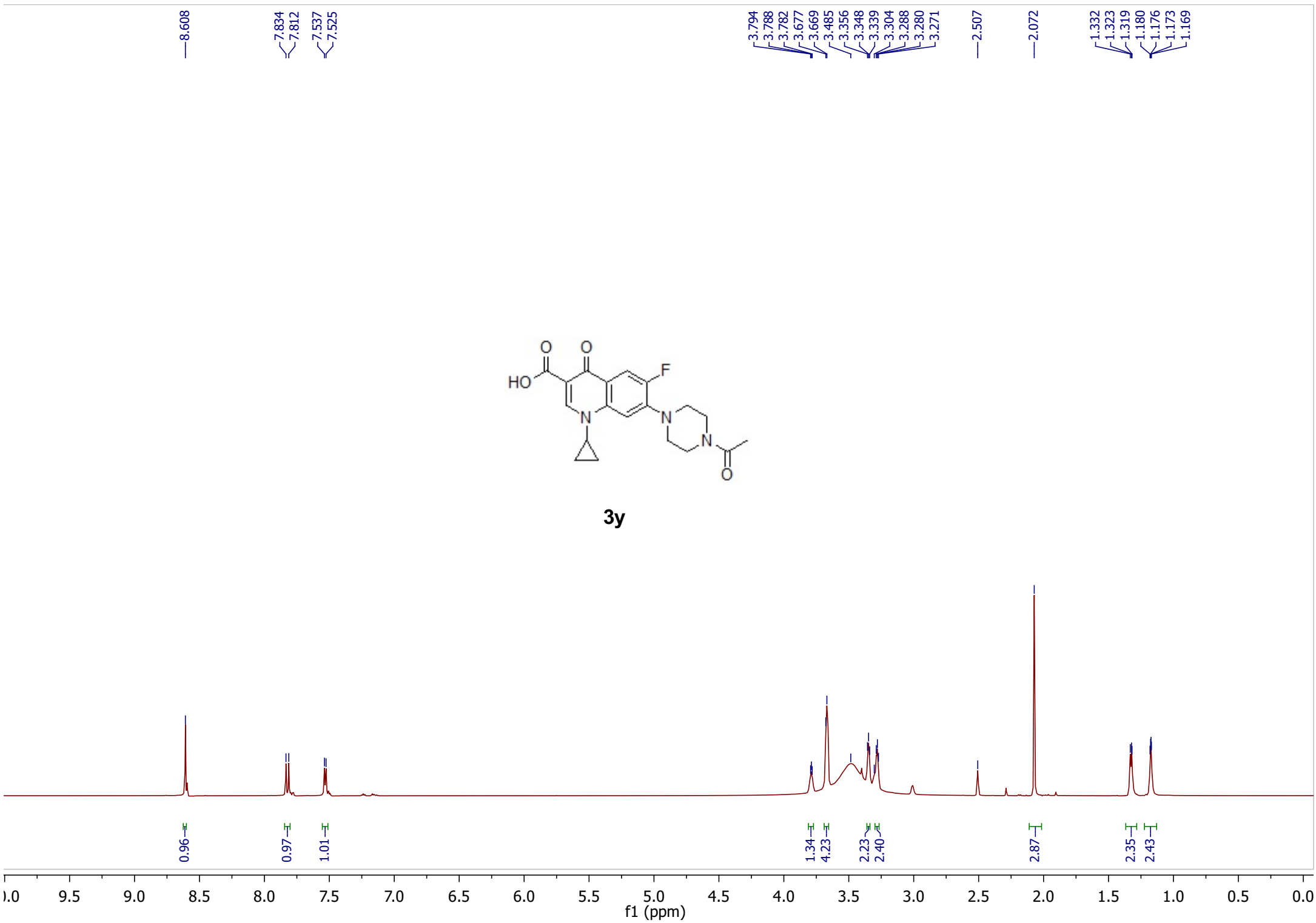
3x

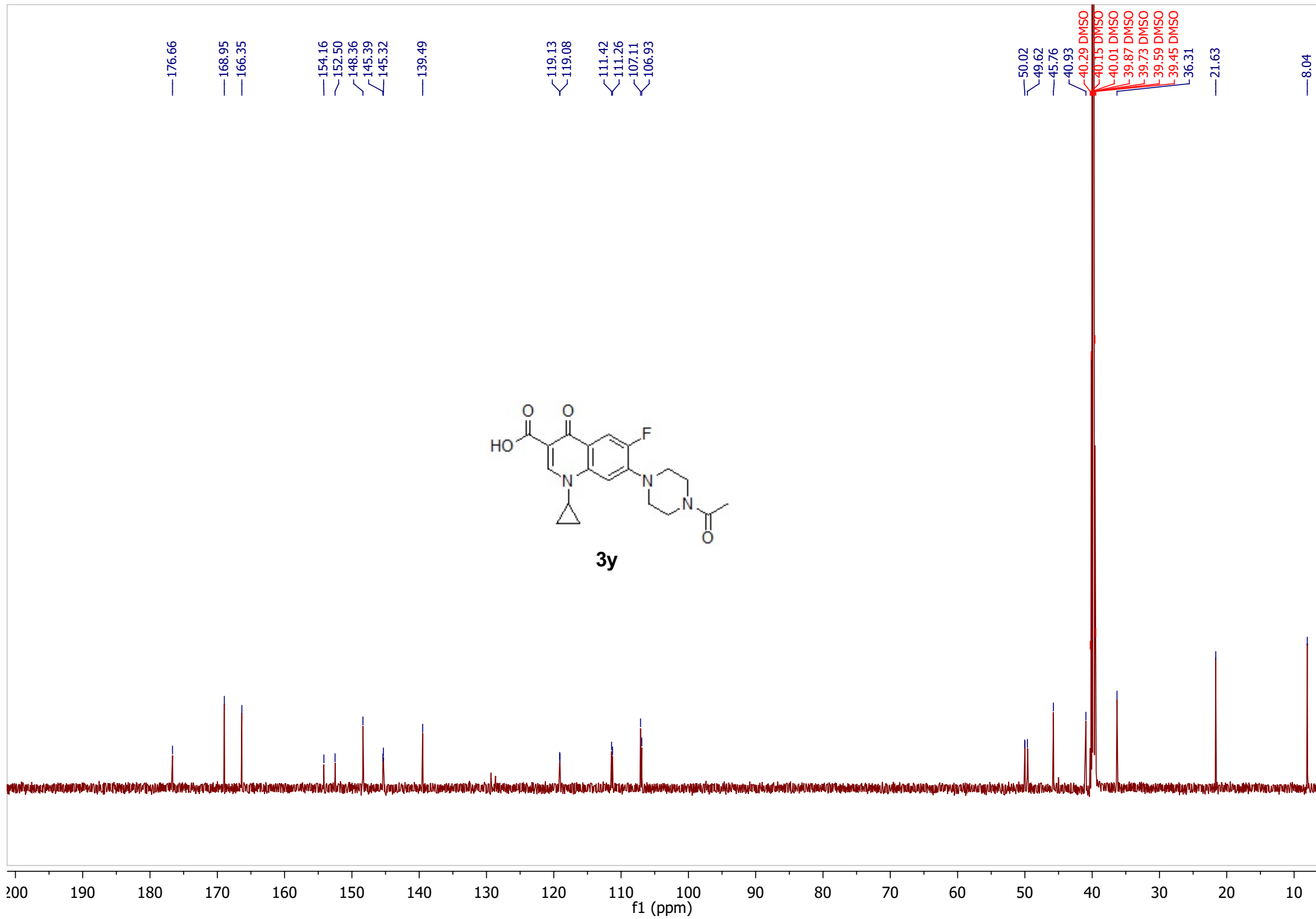


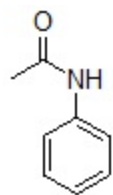


3x

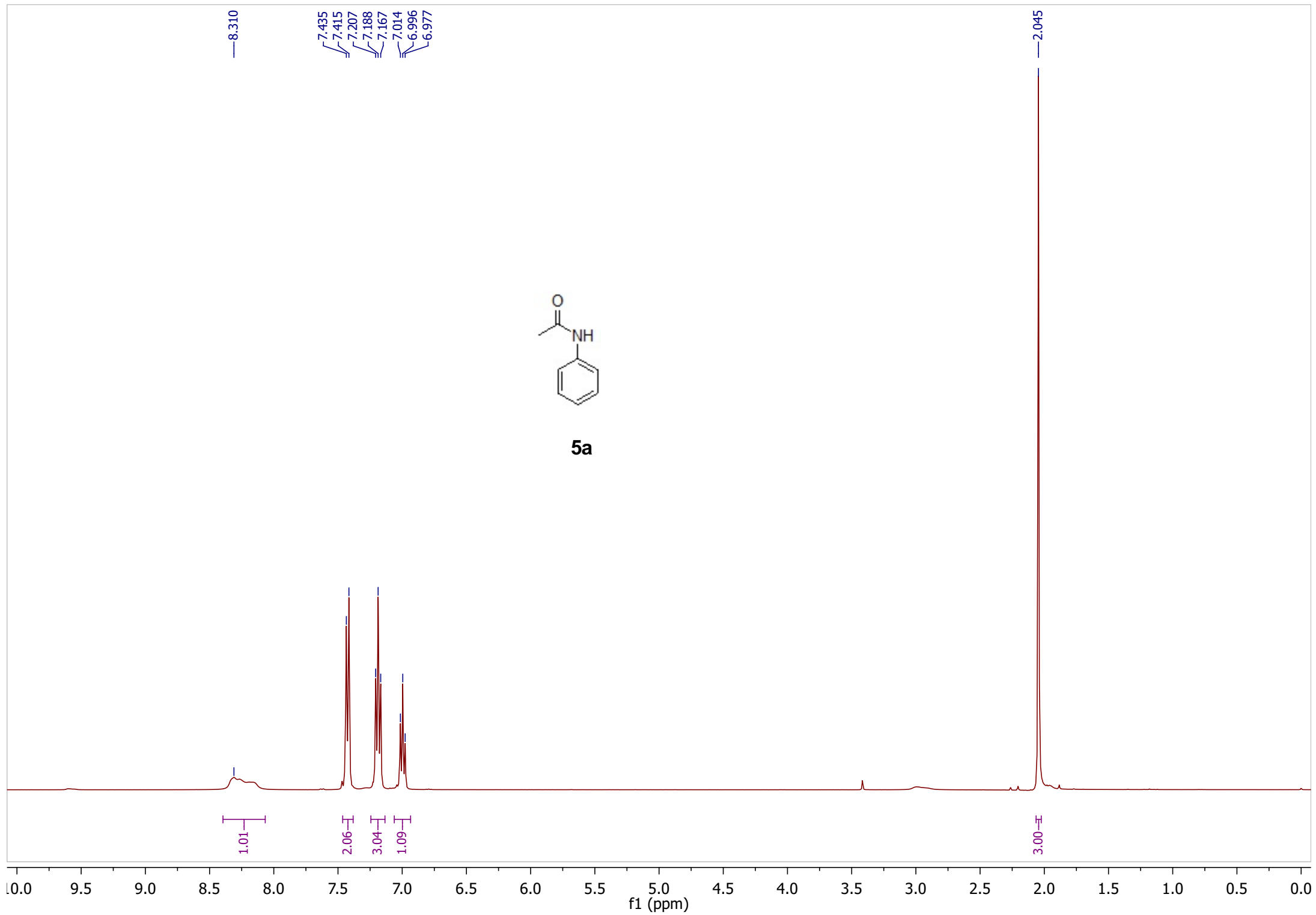


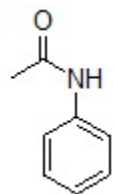




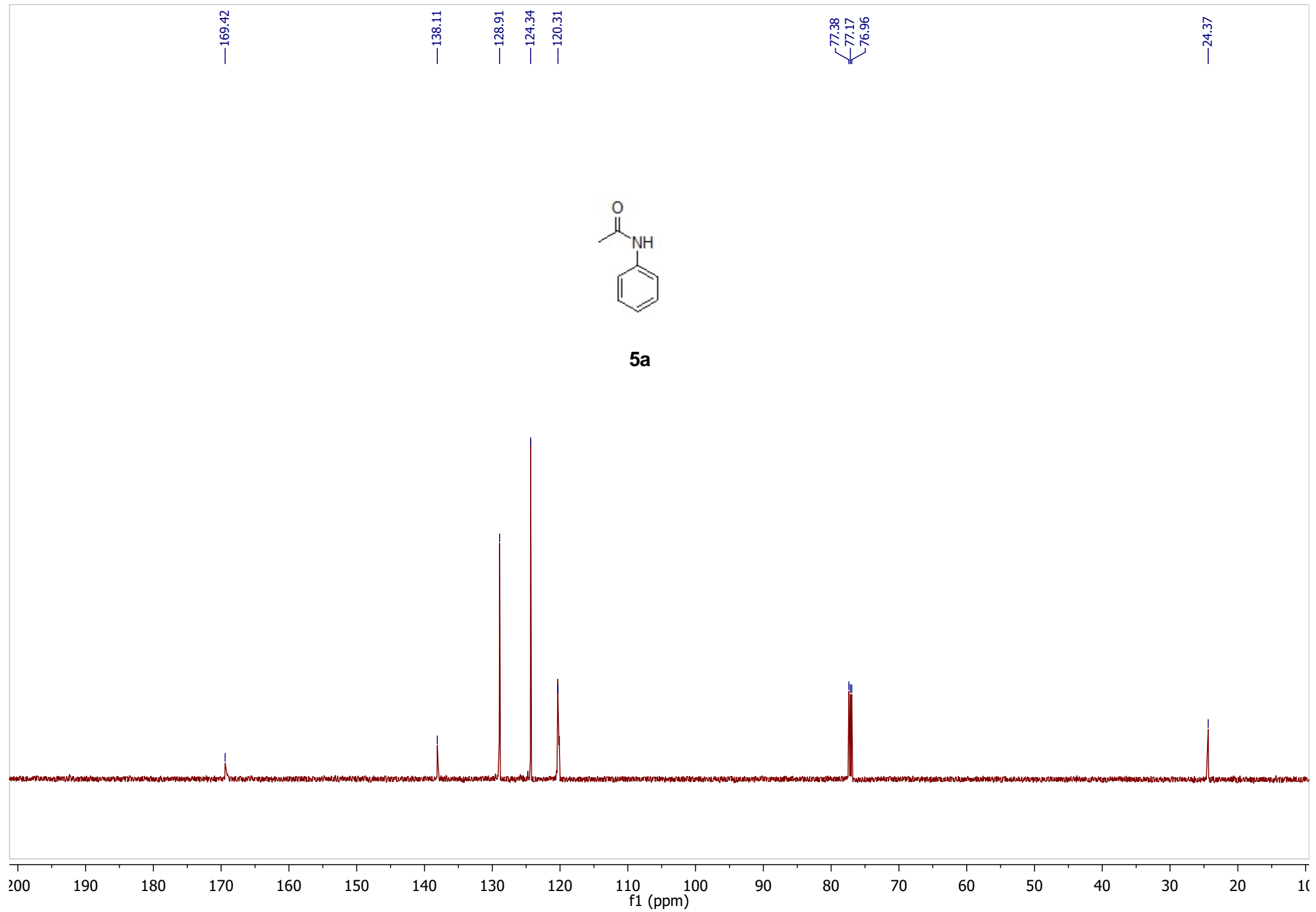


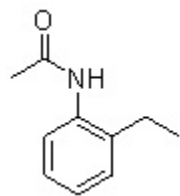
5a



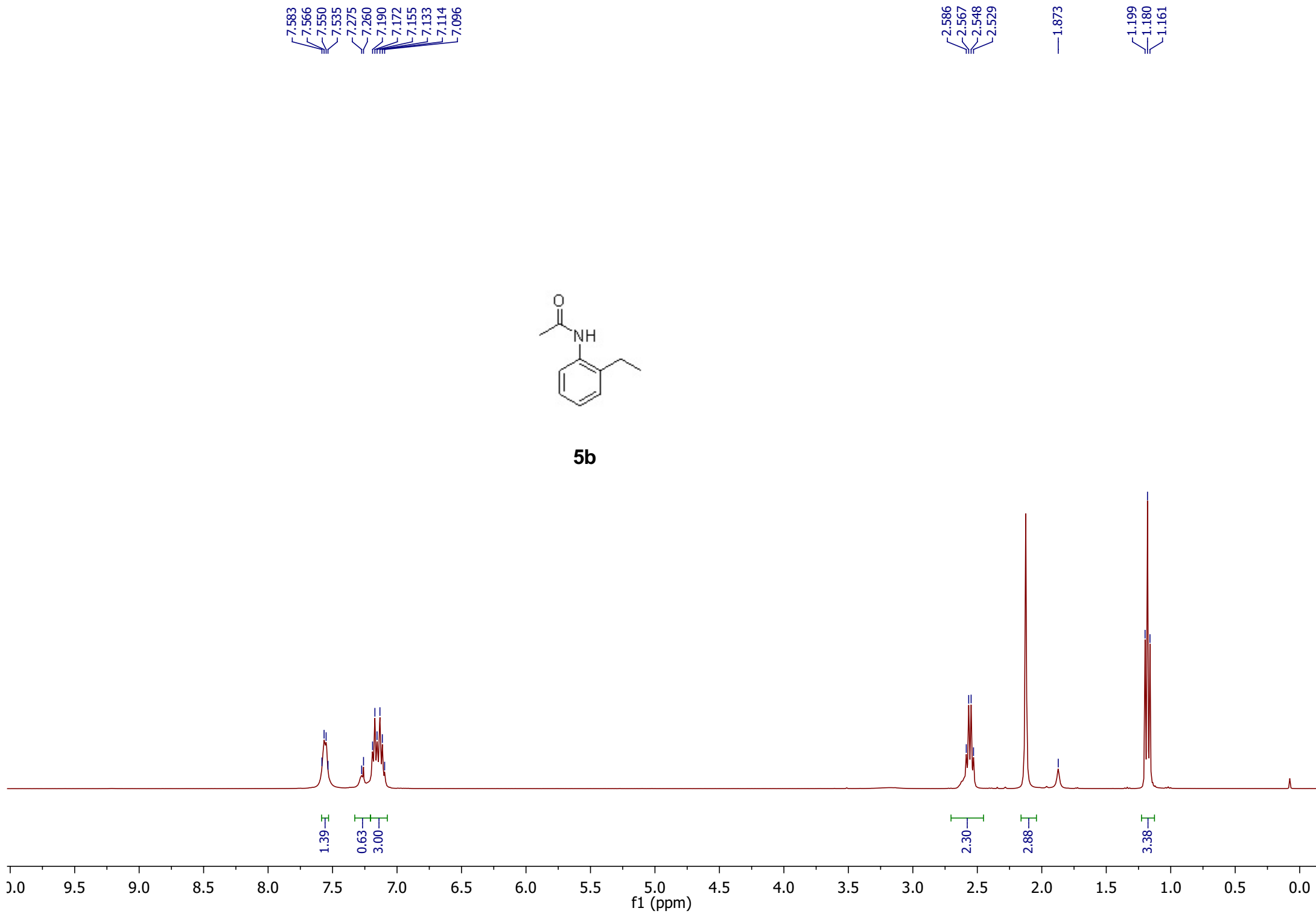


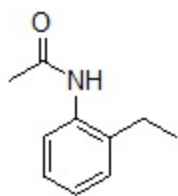
5a





5b





5b

— 169.22

— 136.33

— 134.86

— 128.52

— 126.45

— 125.97

— 124.88

77.35 CDCl₃

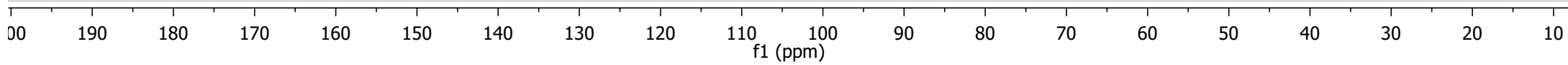
77.14 CDCl₃

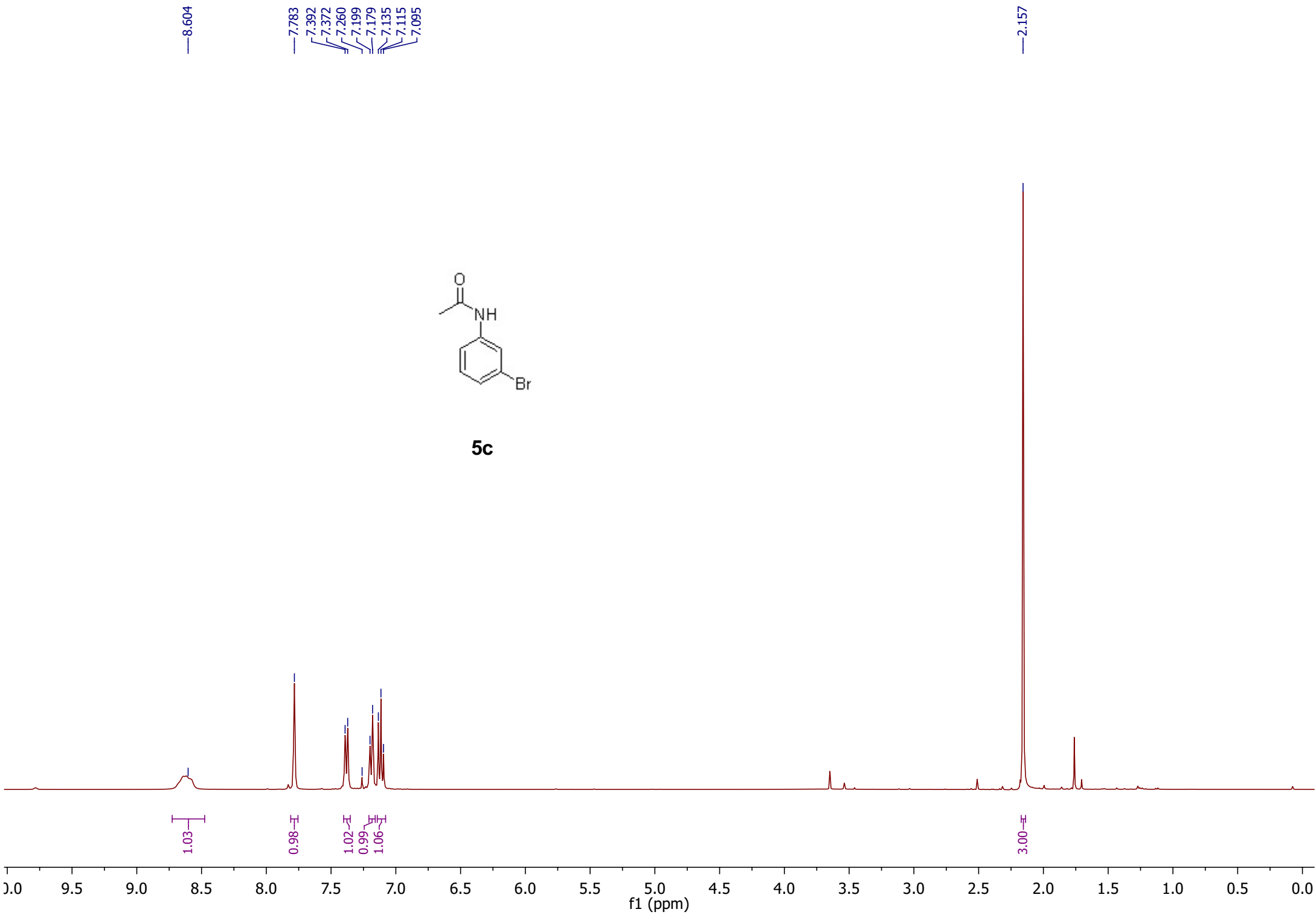
76.92 CDCl₃

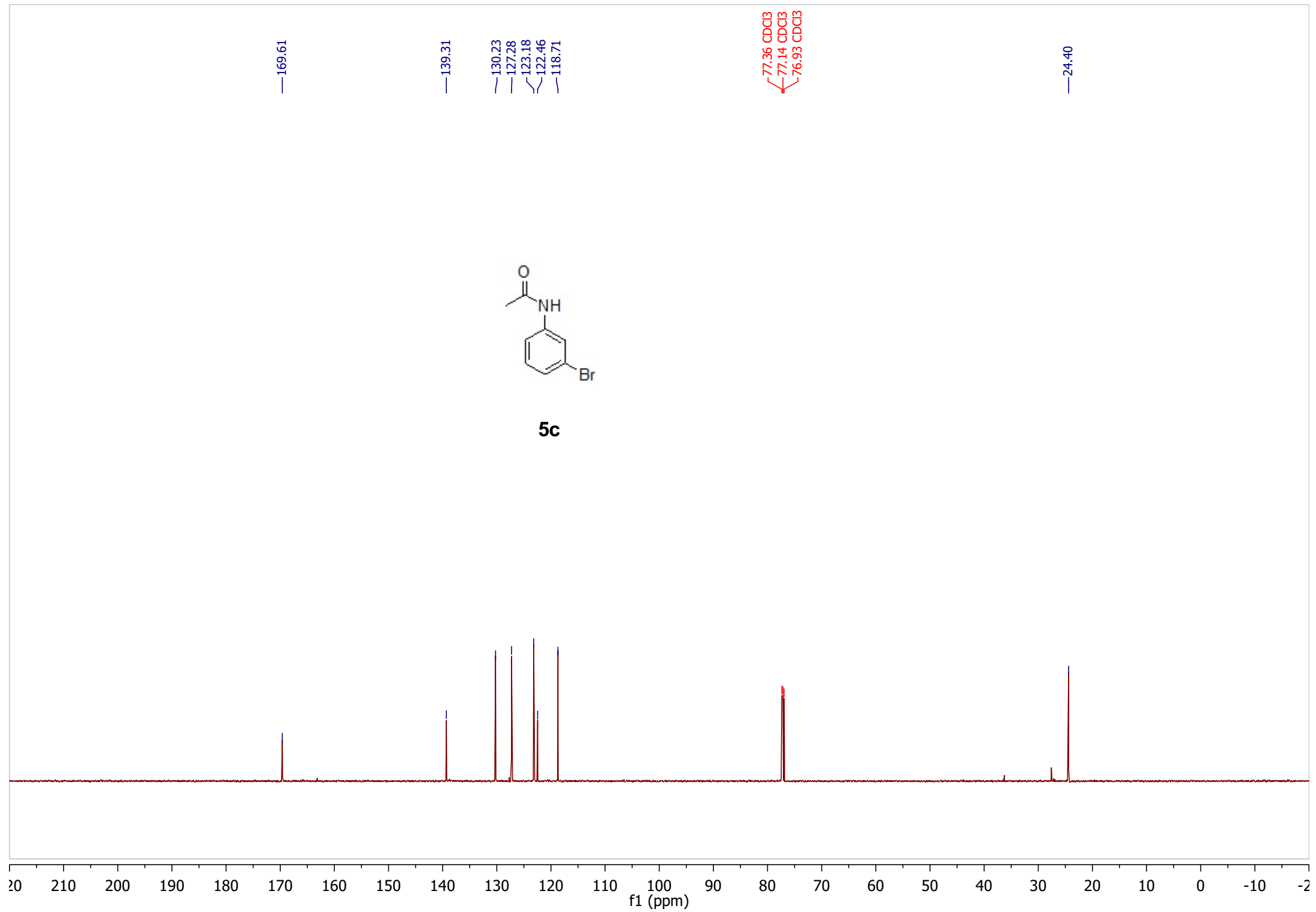
— 24.20

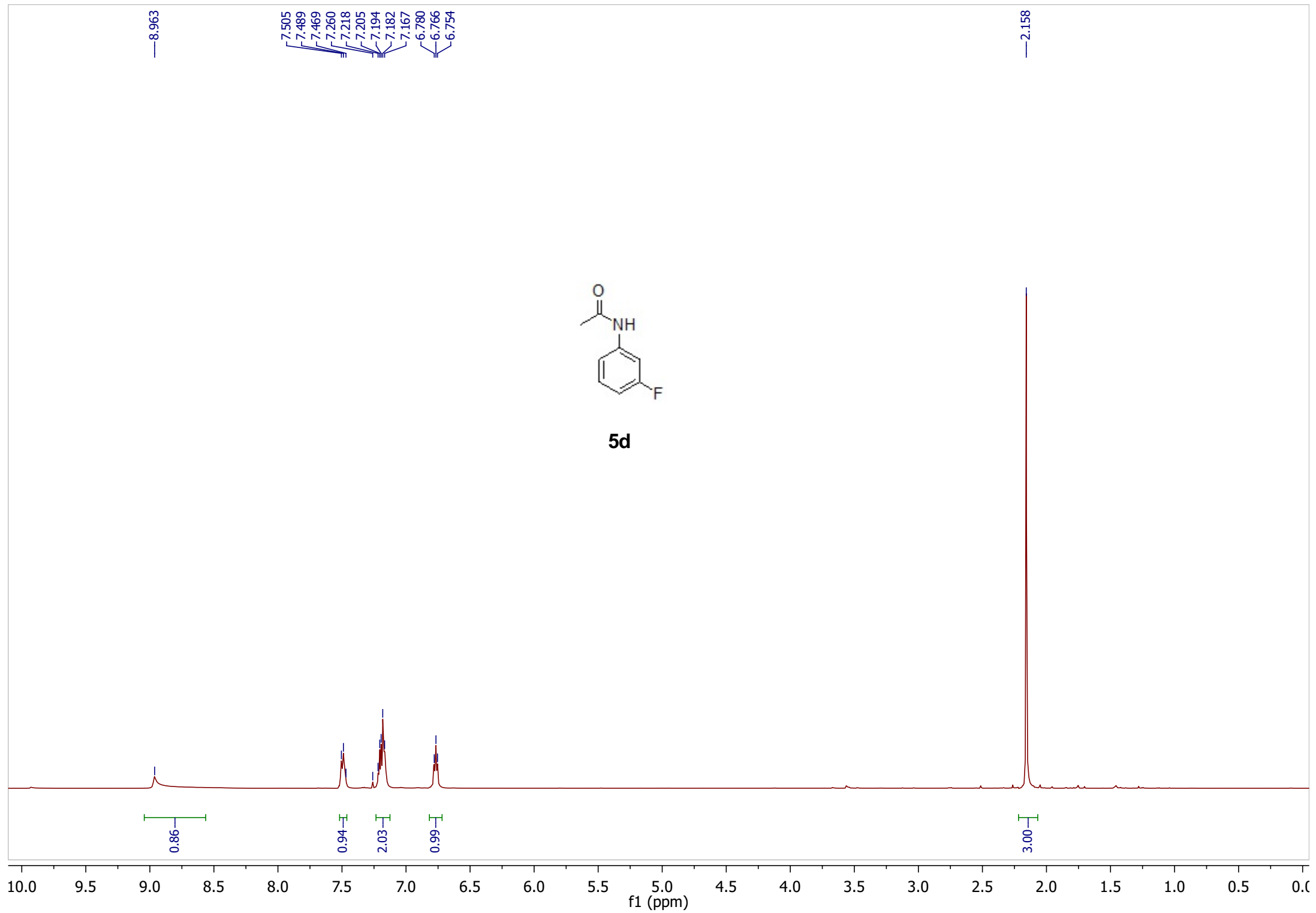
— 23.93

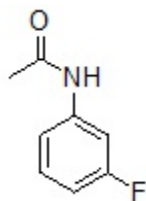
— 14.01



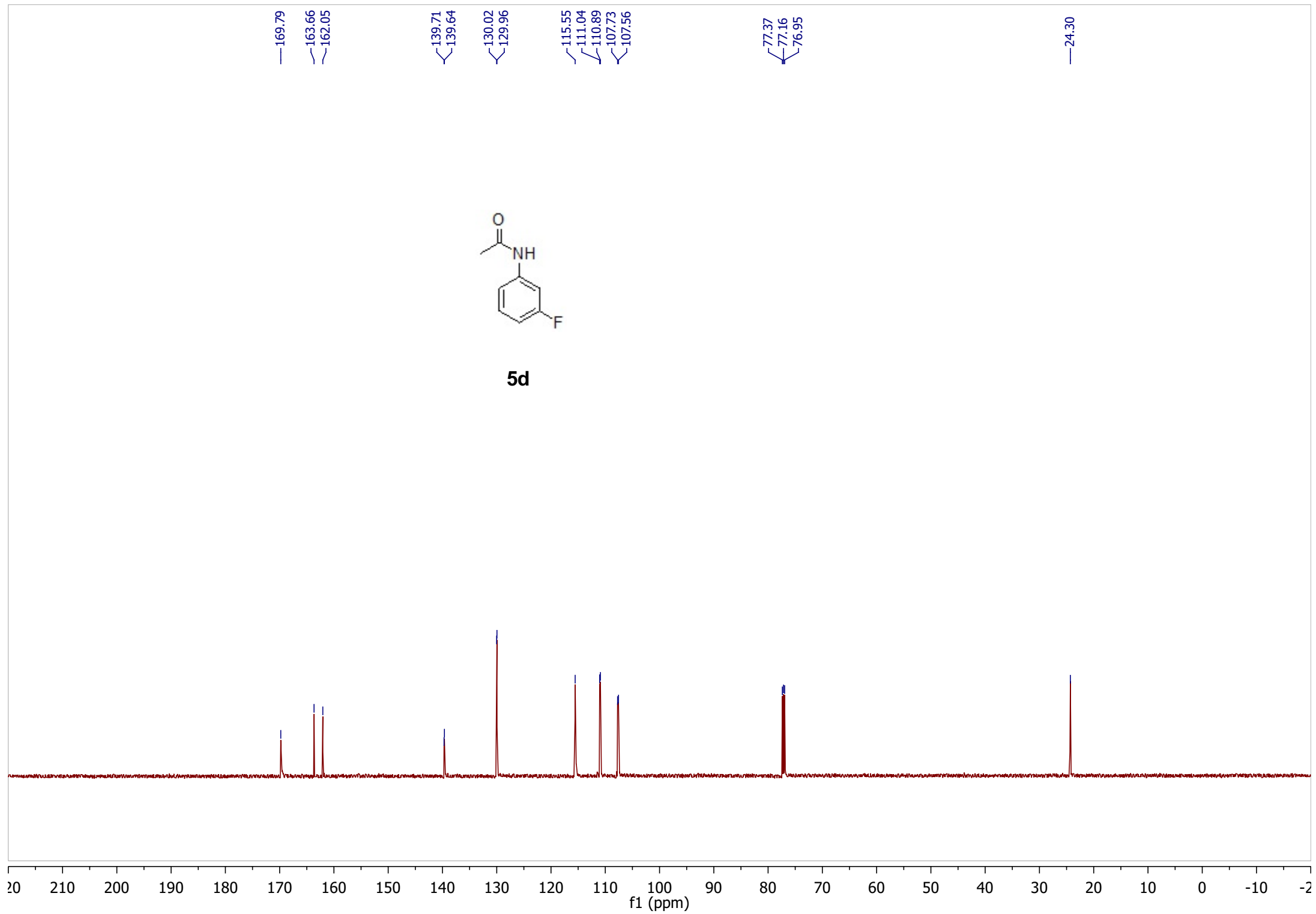


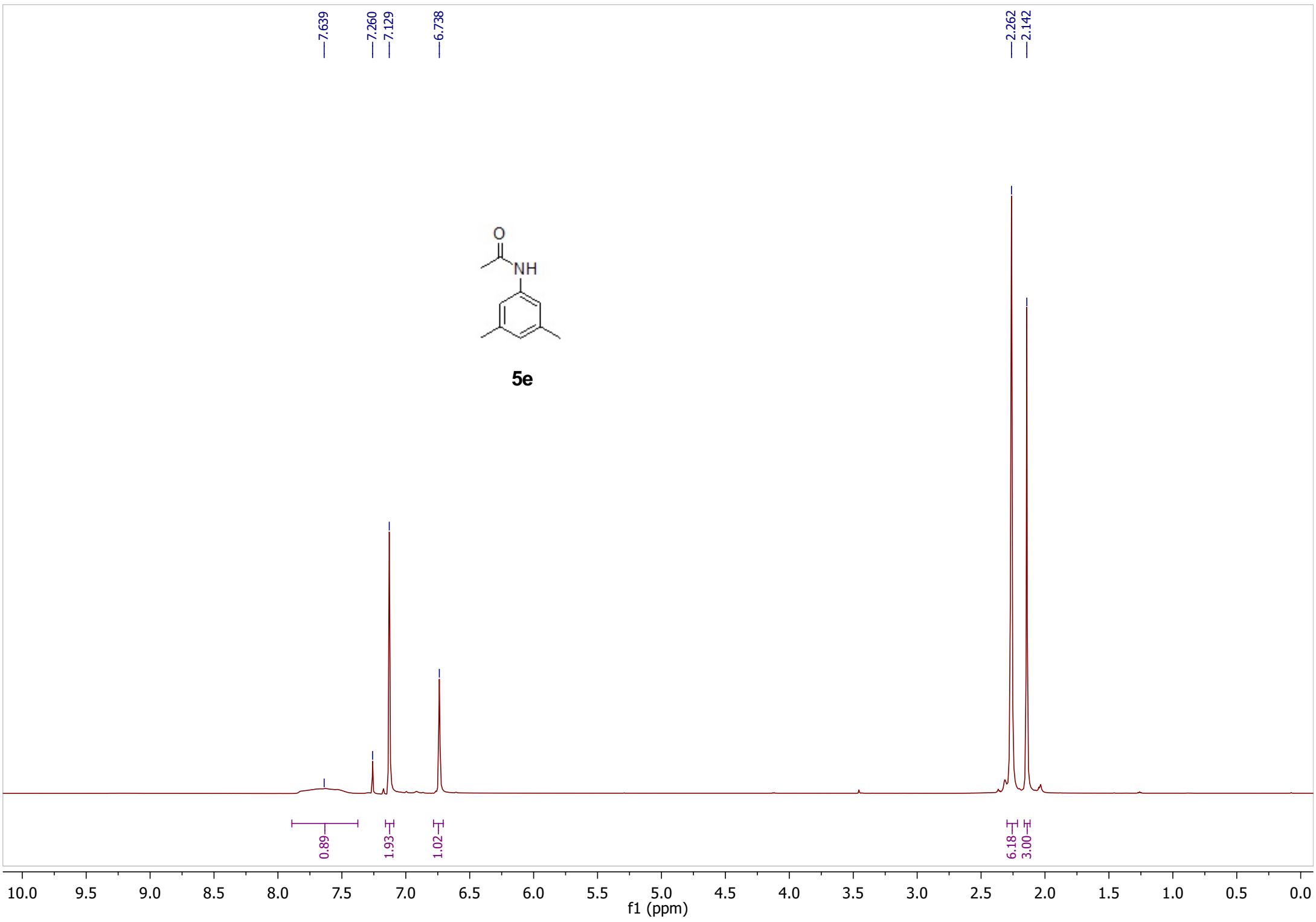


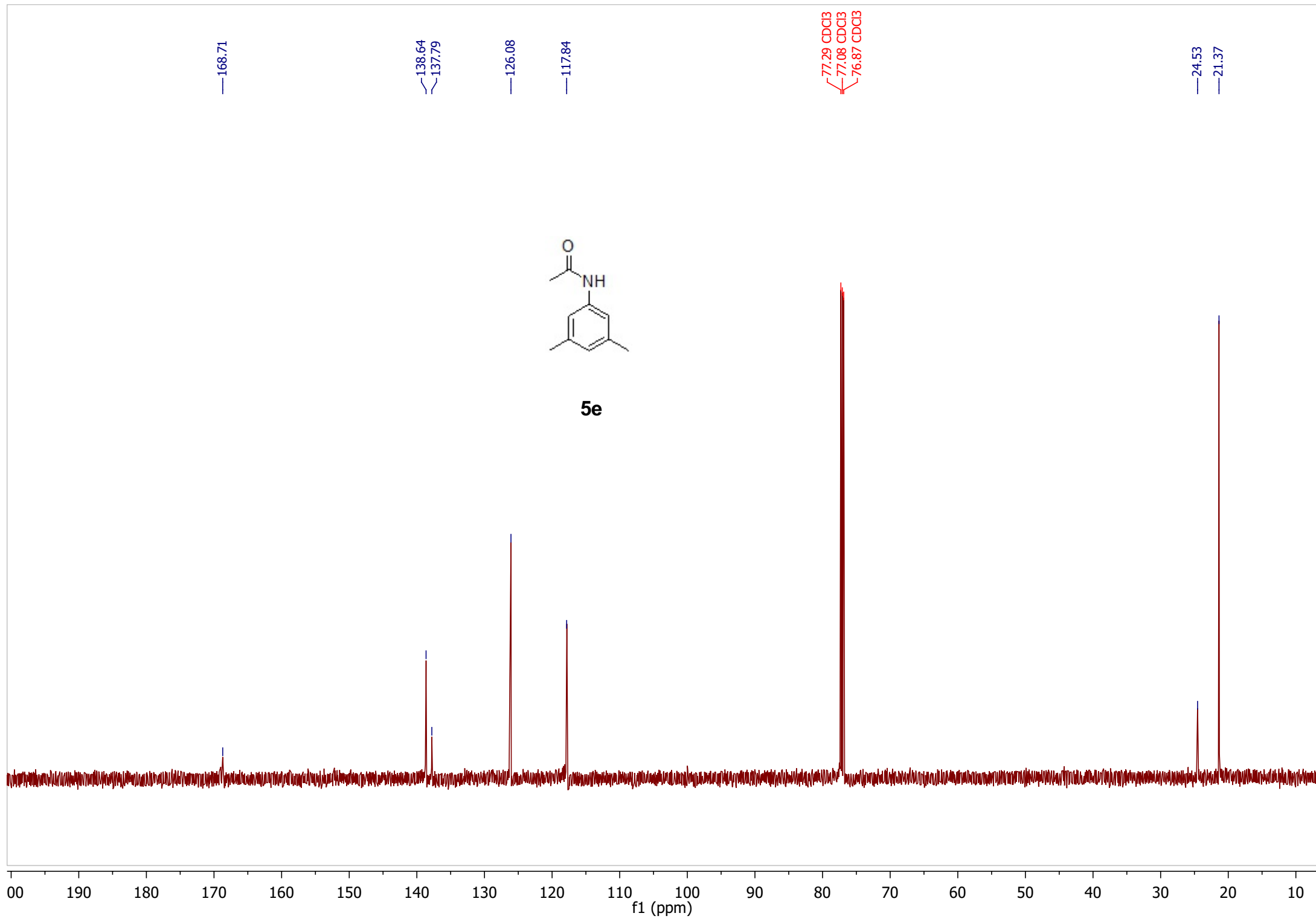


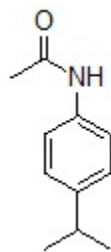


5d

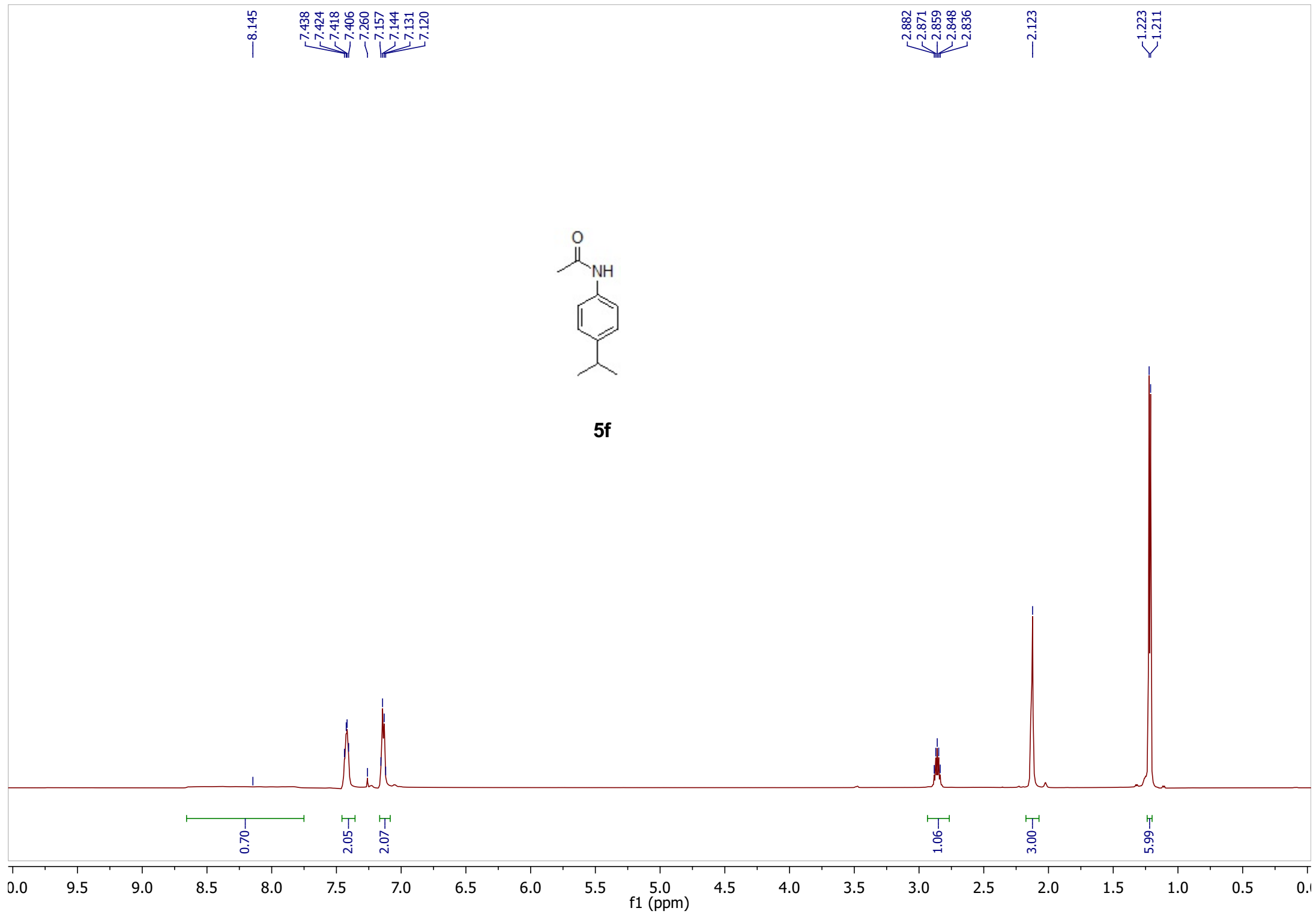


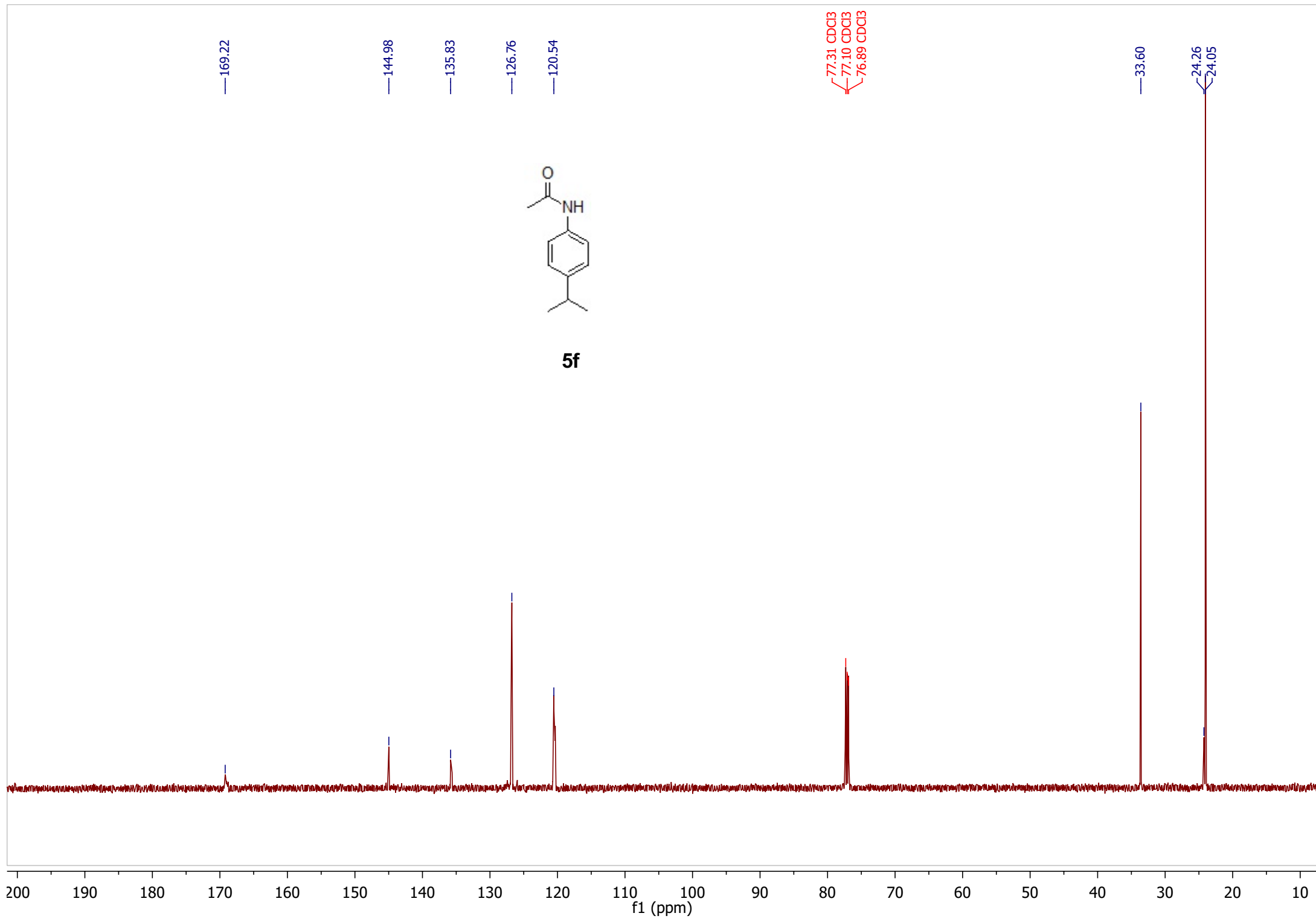


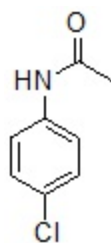




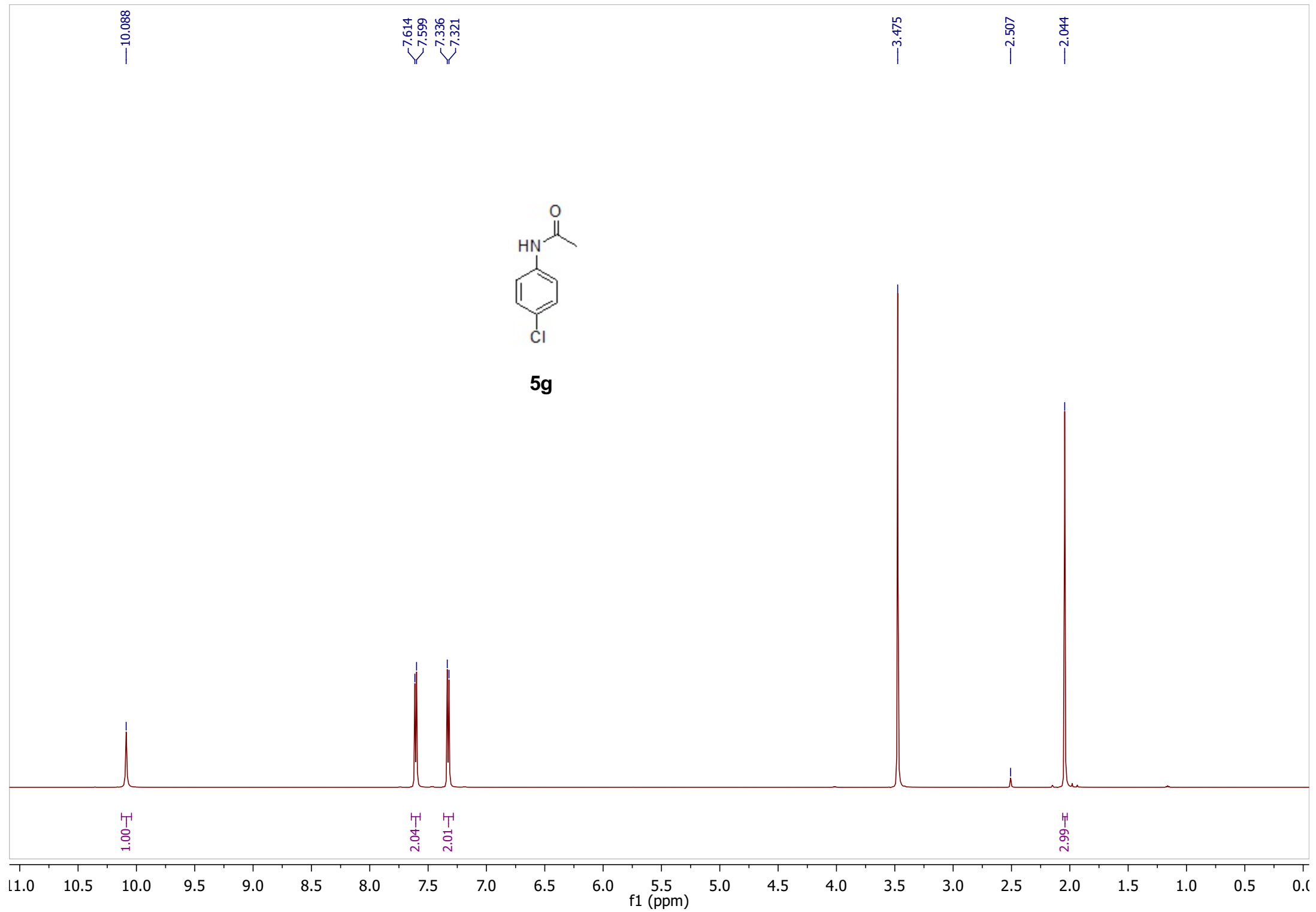
5f

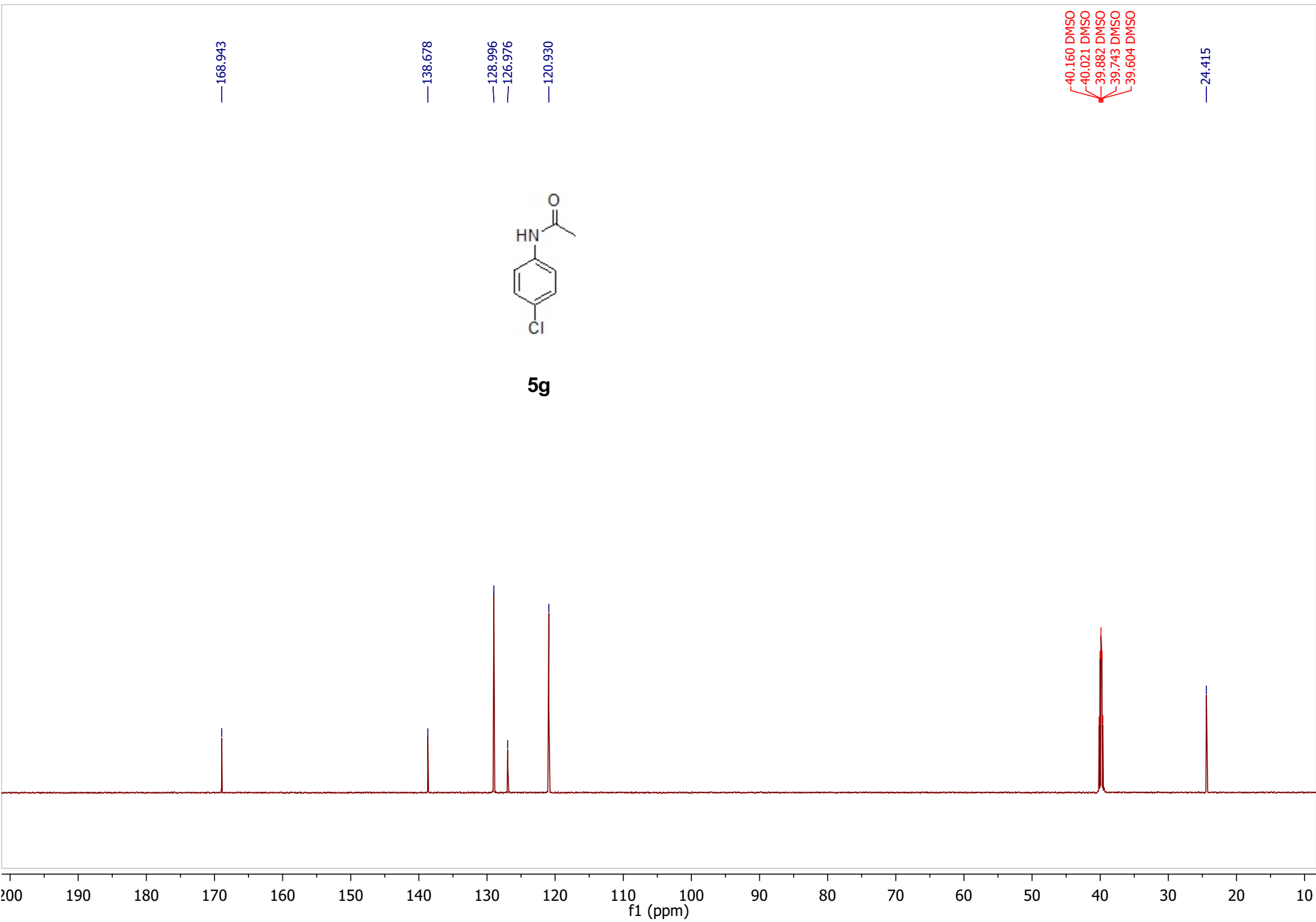


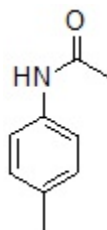




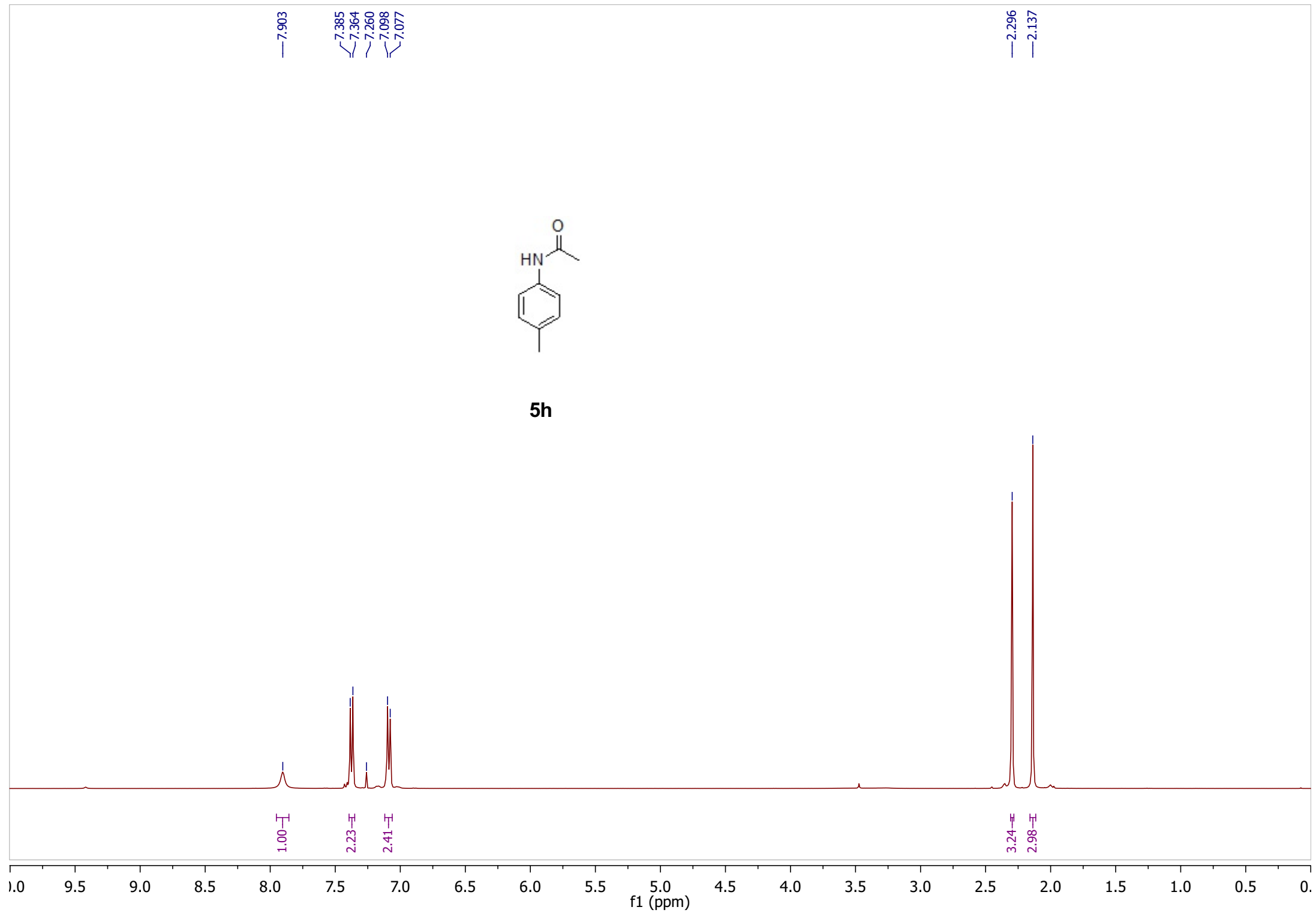
5g







5h



— 168.82

— 135.35

— 134.00

— 129.42

— 120.27

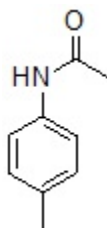
77.40 CDCl₃

77.08 CDCl₃

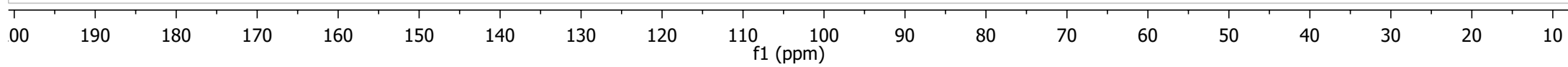
76.76 CDCl₃

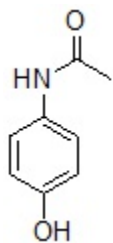
— 24.33

— 20.88

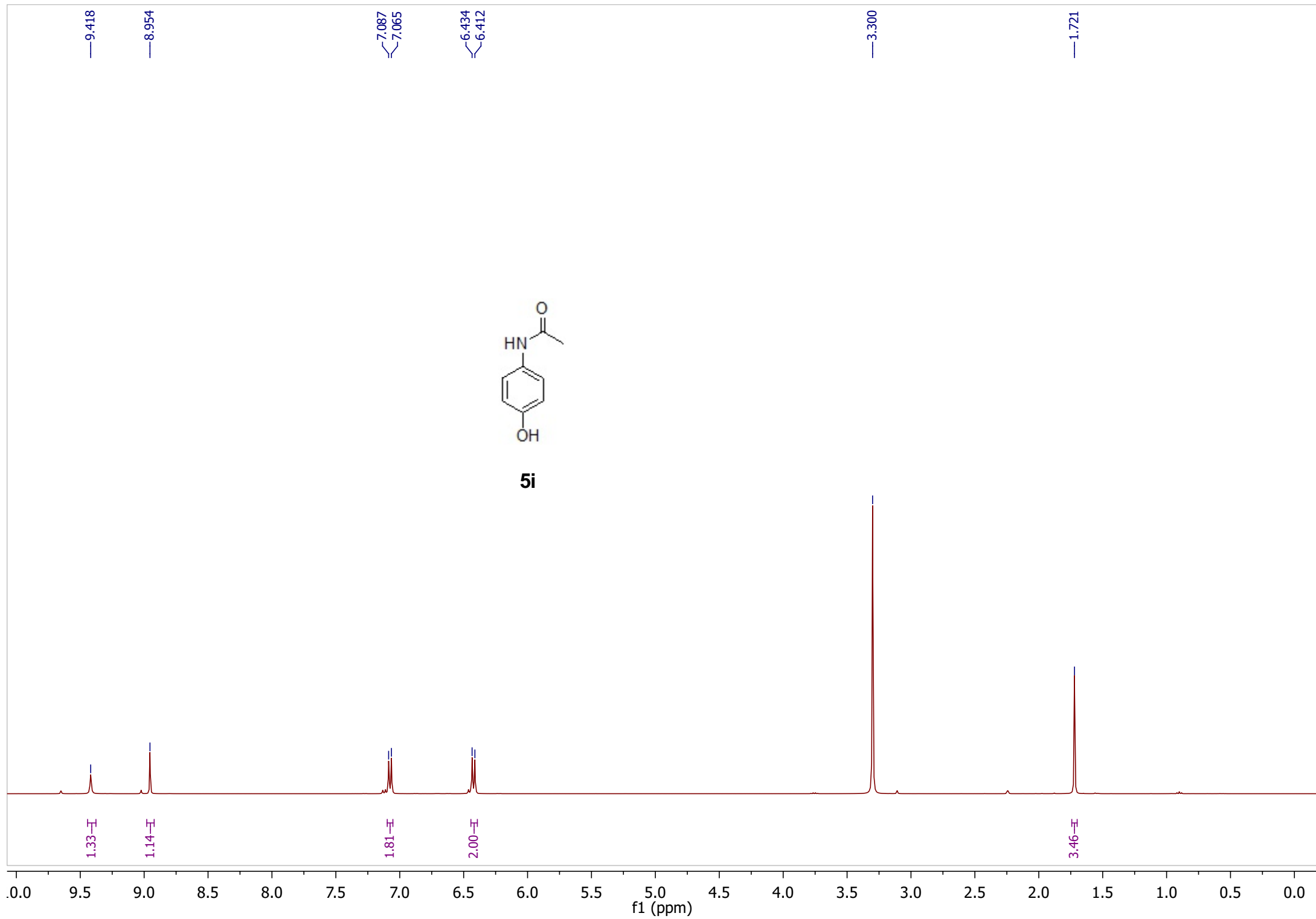


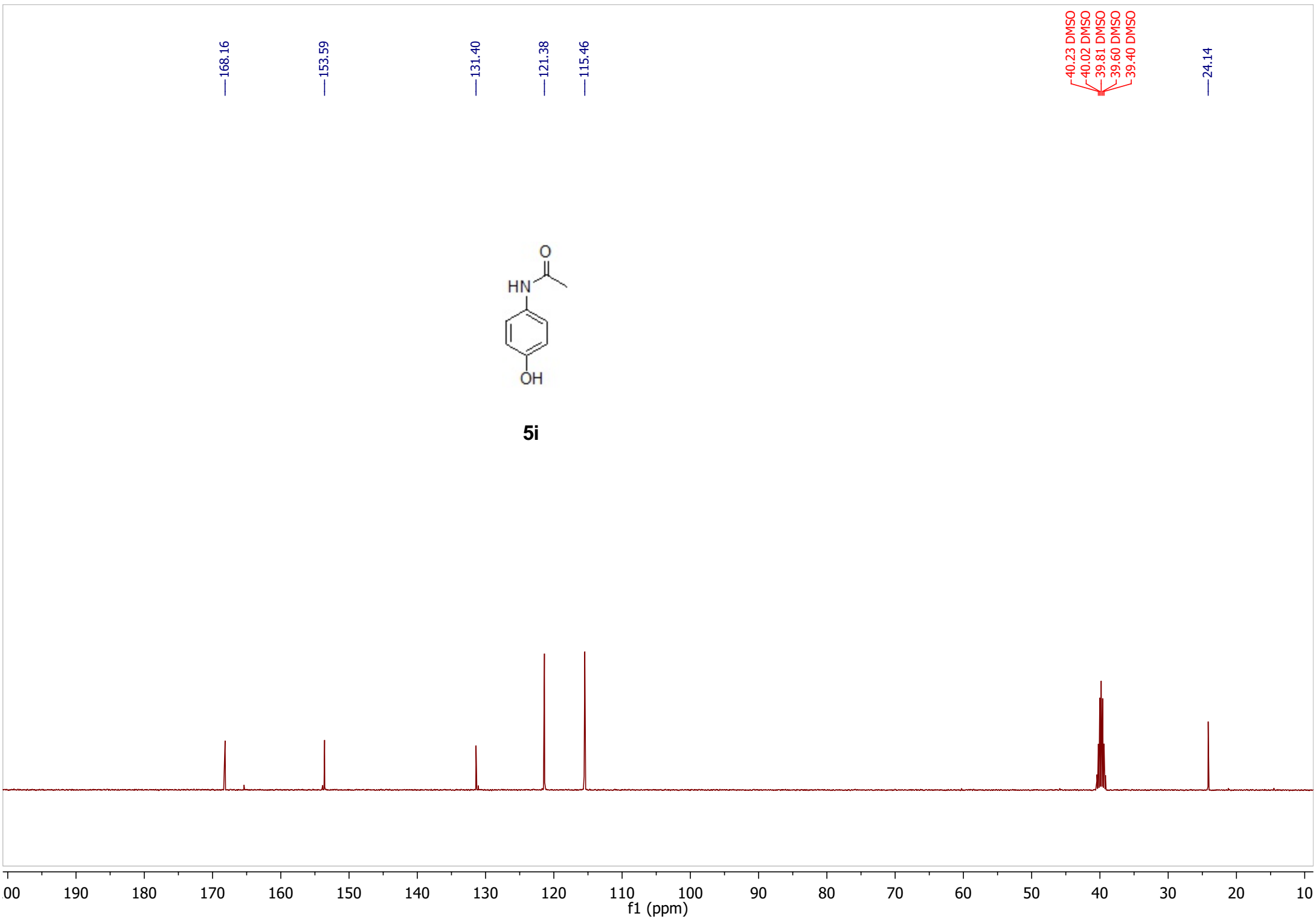
5h

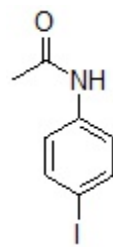




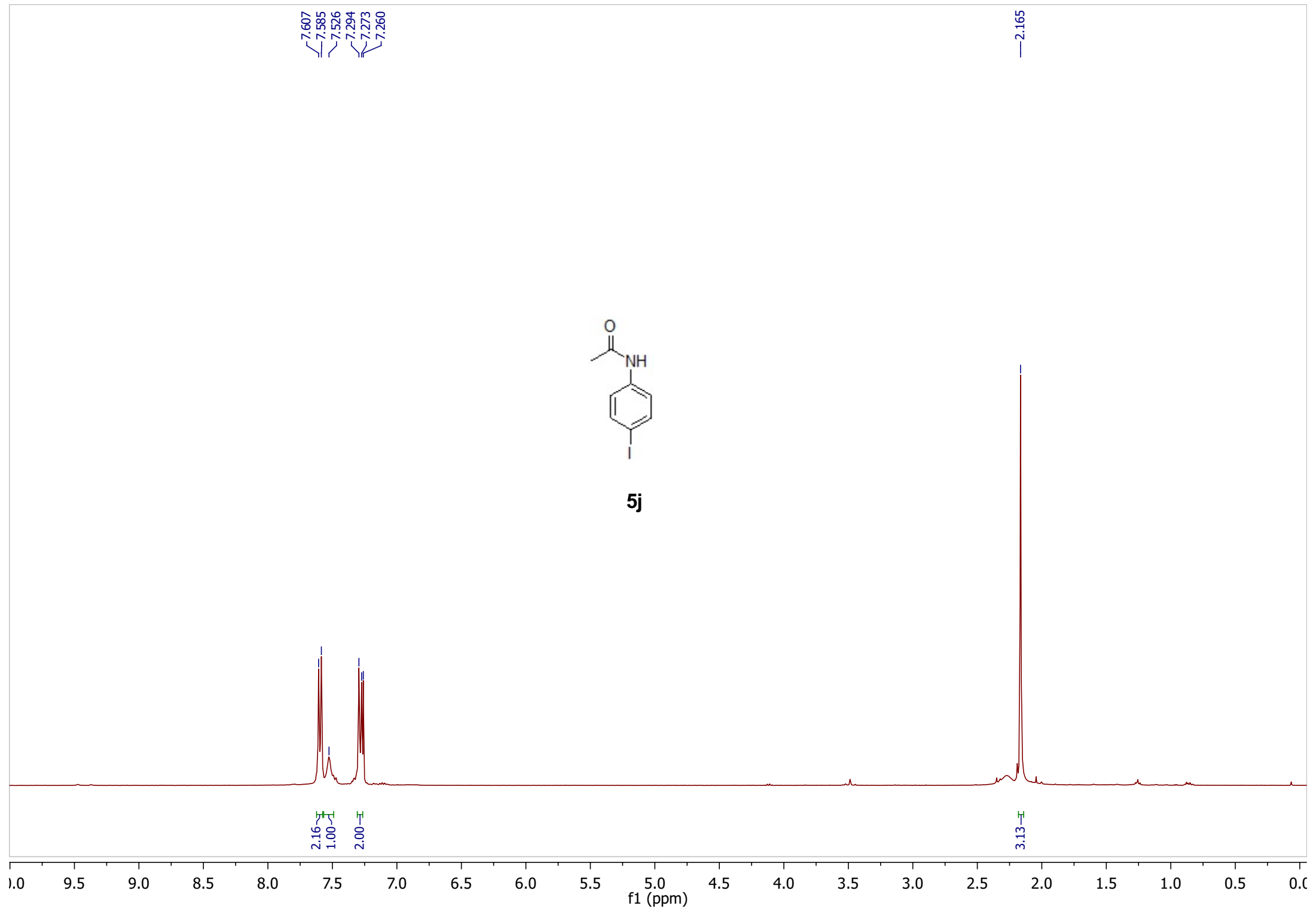
5i

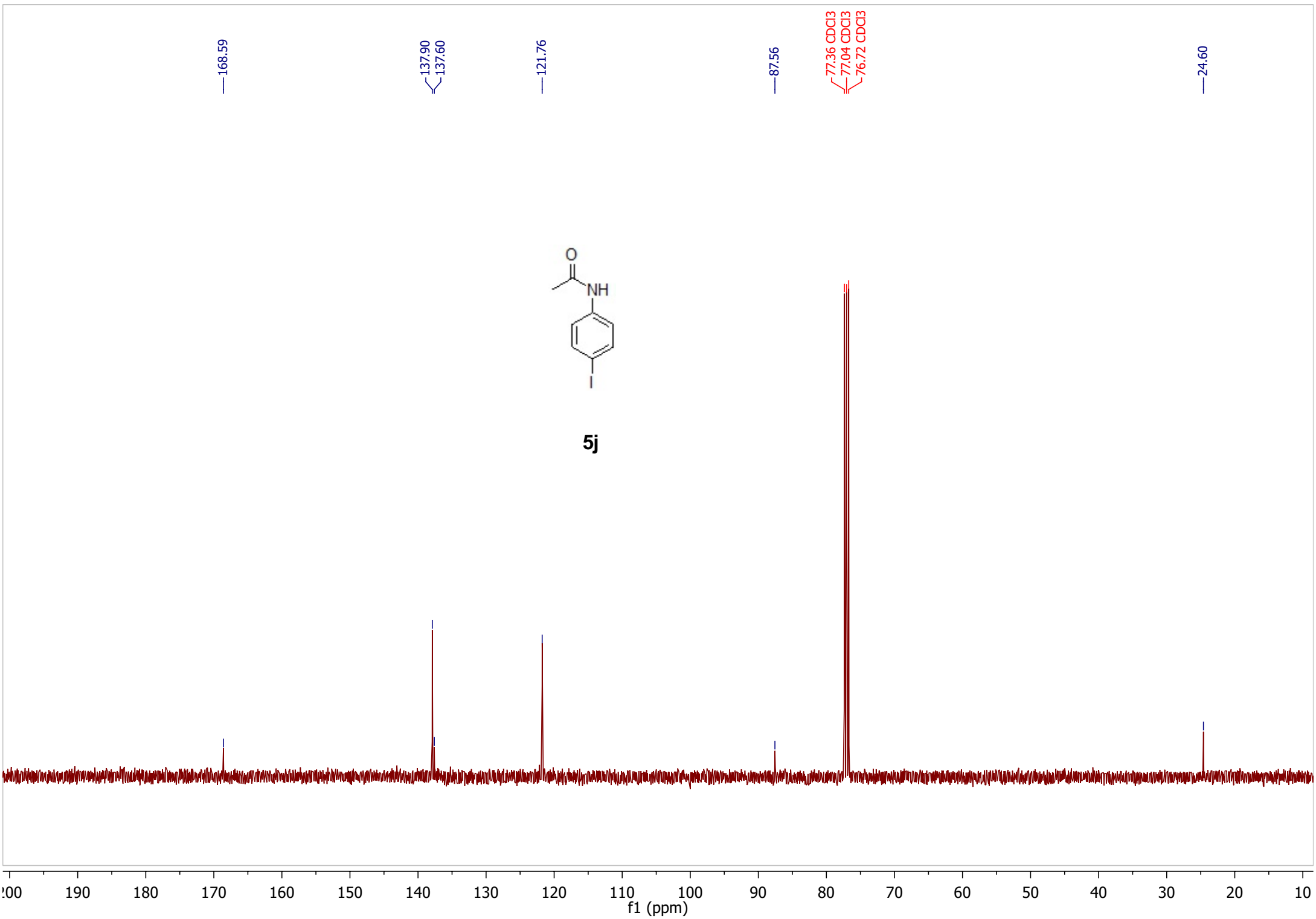


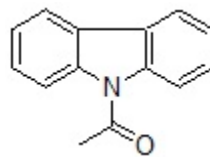




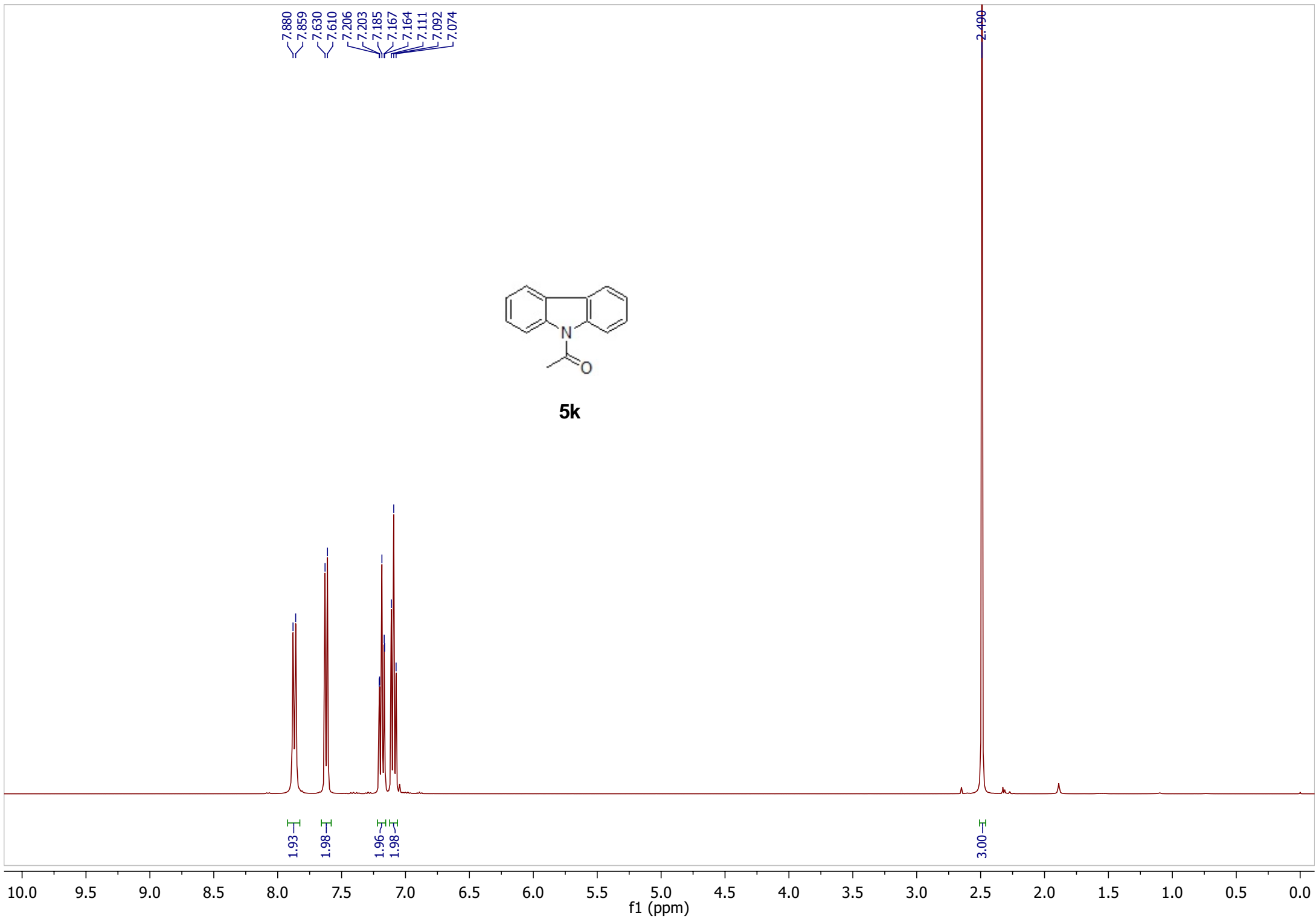
5j







5k



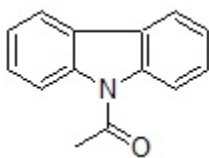
—169.97

—138.47

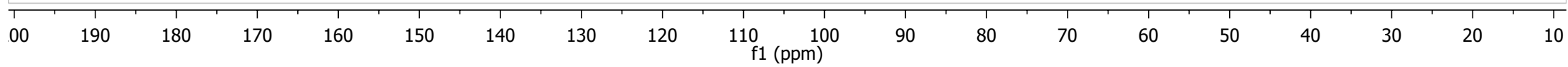
127.22
126.26
123.56
119.71
116.17

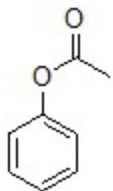
77.62 CDCl₃
77.30 CDCl₃
76.98 CDCl₃

—27.66

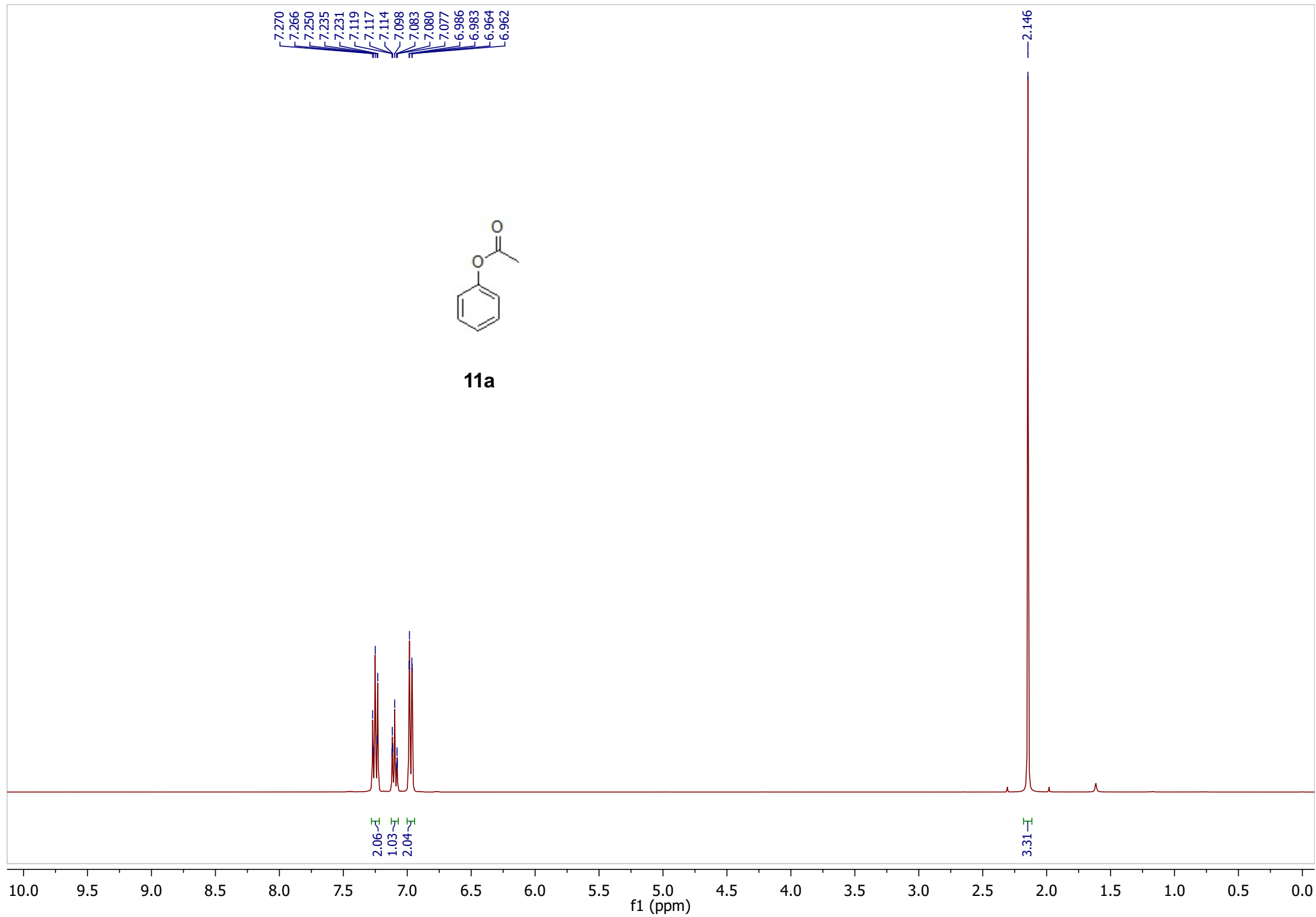


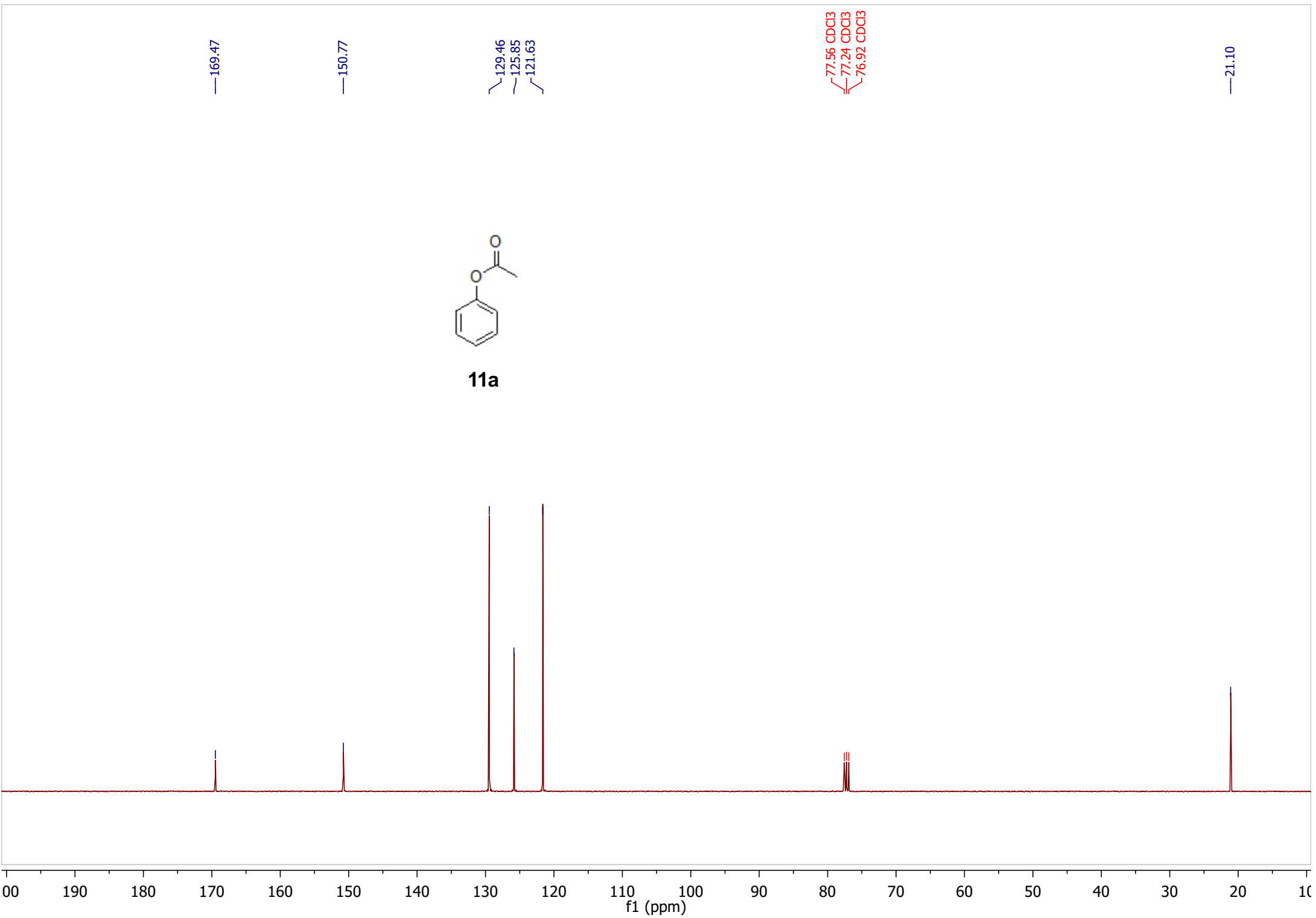
5k

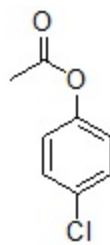




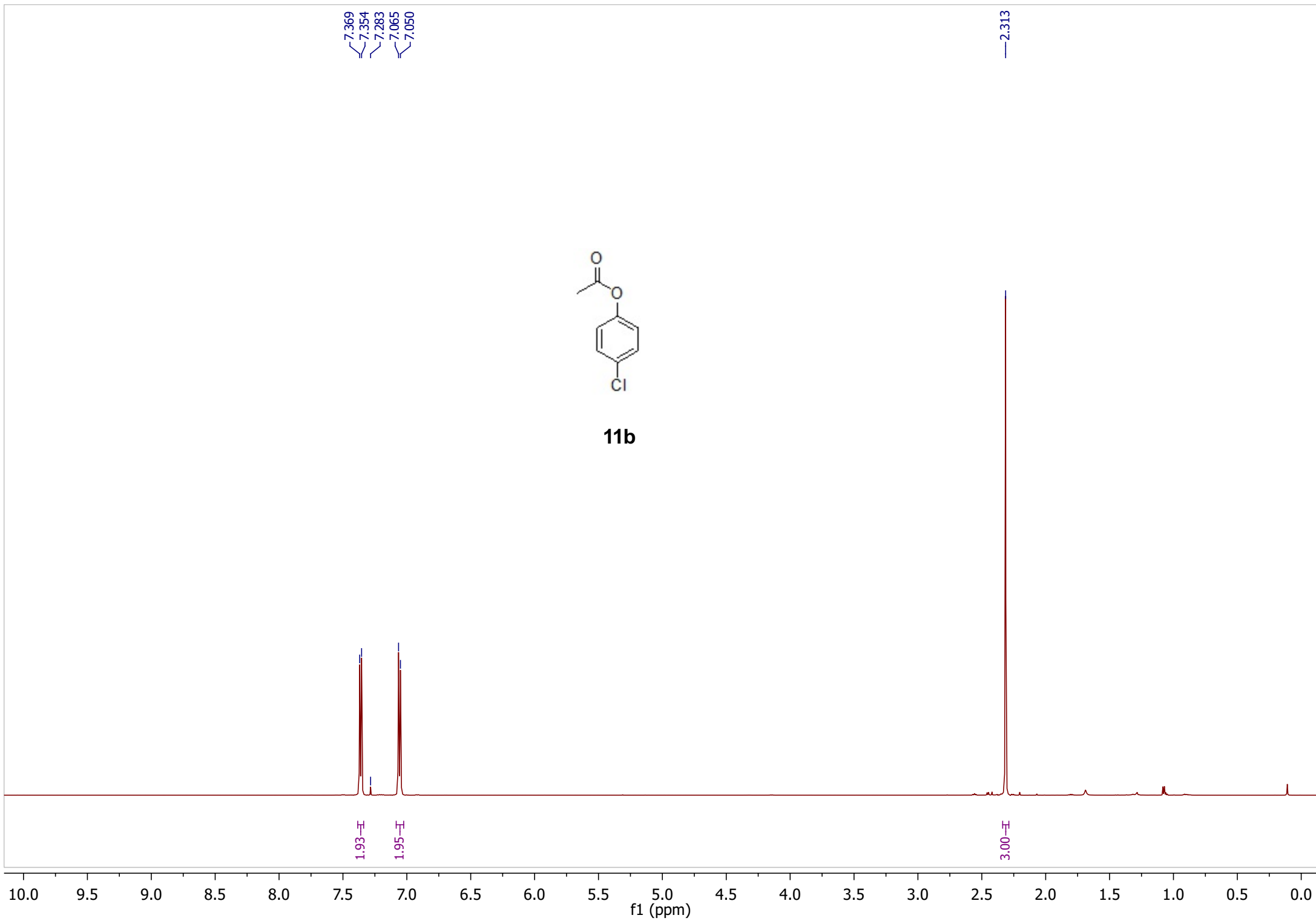
11a

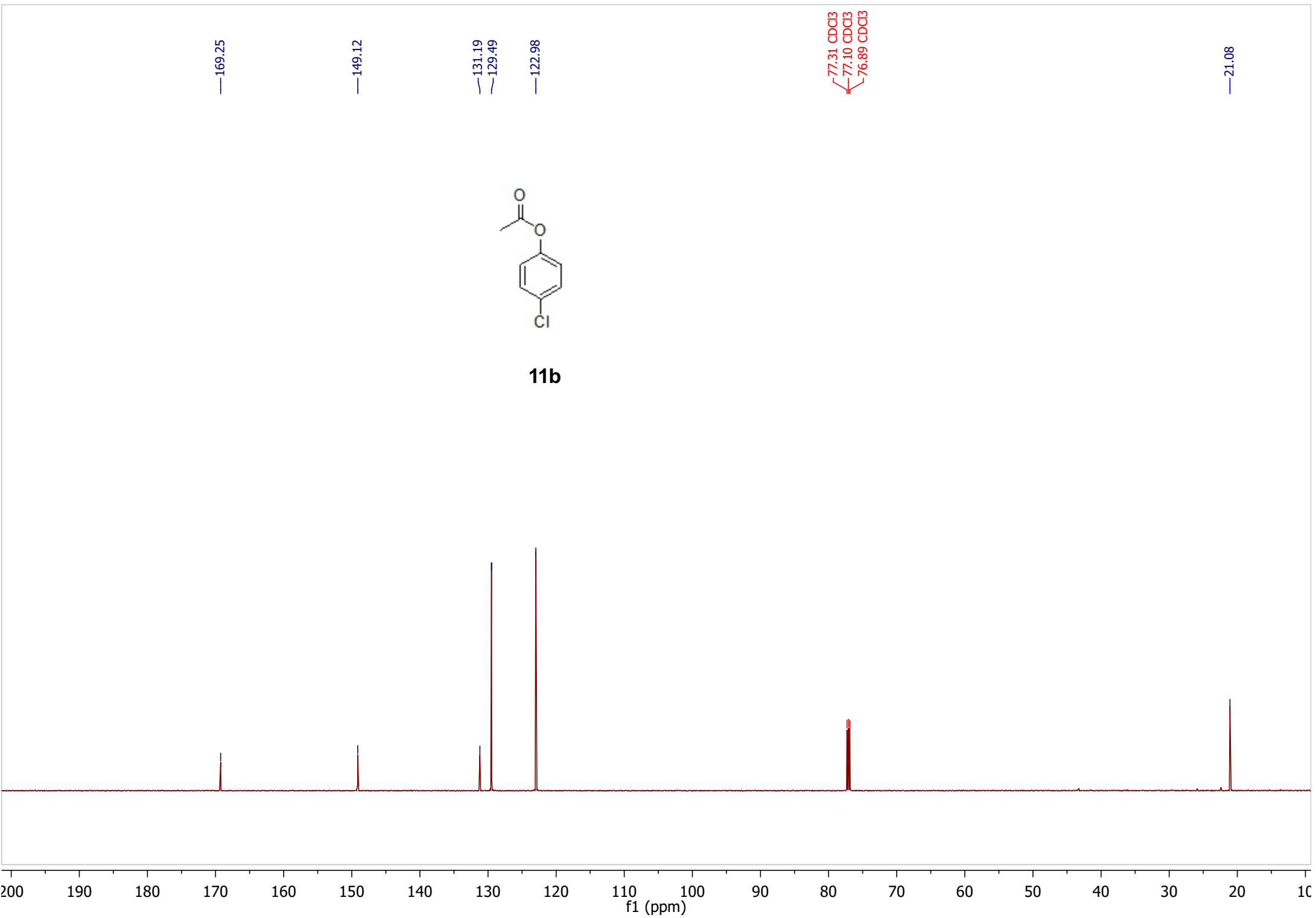


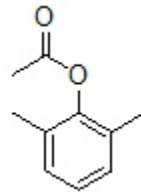




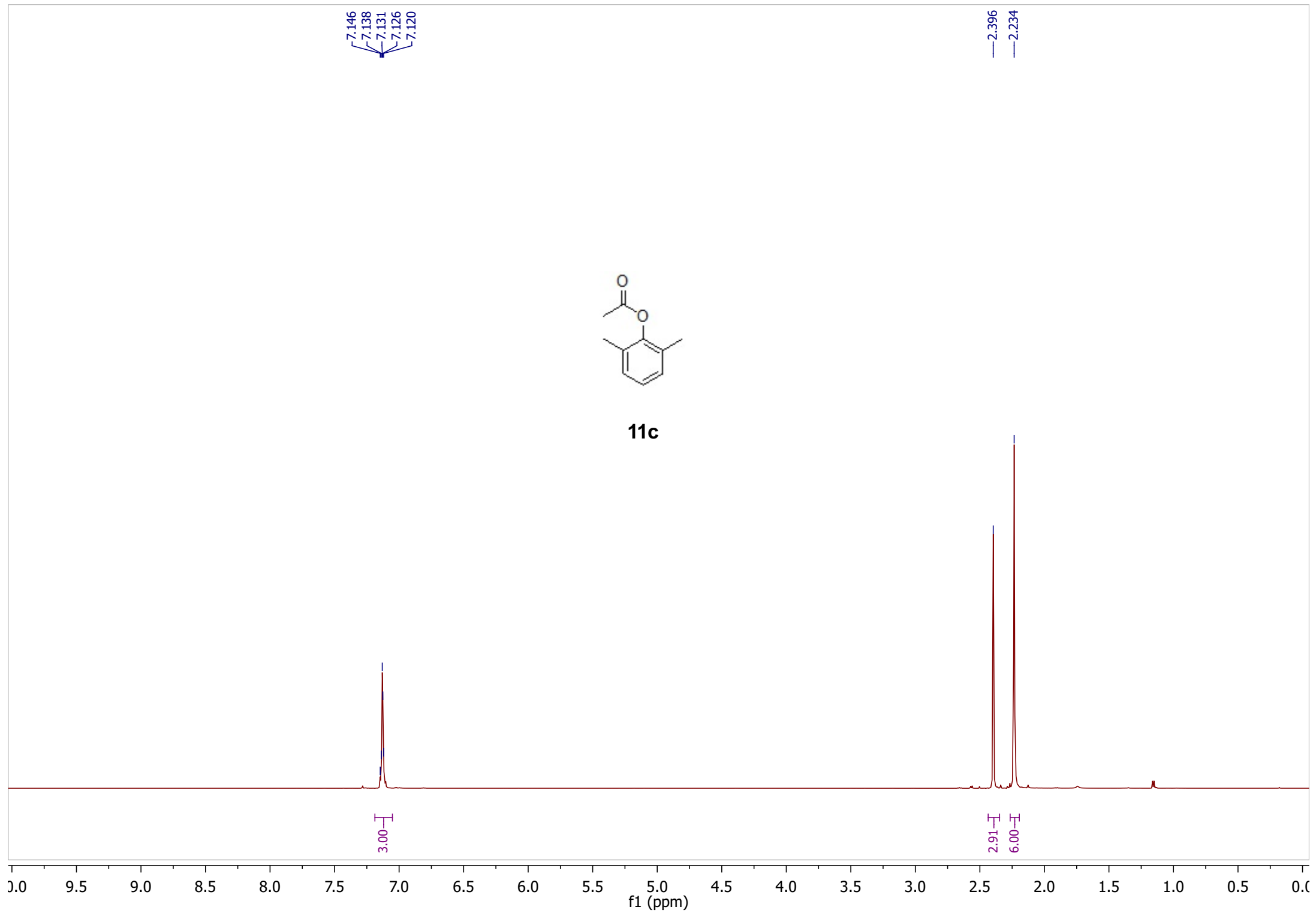
11b

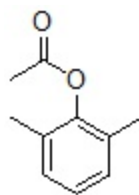




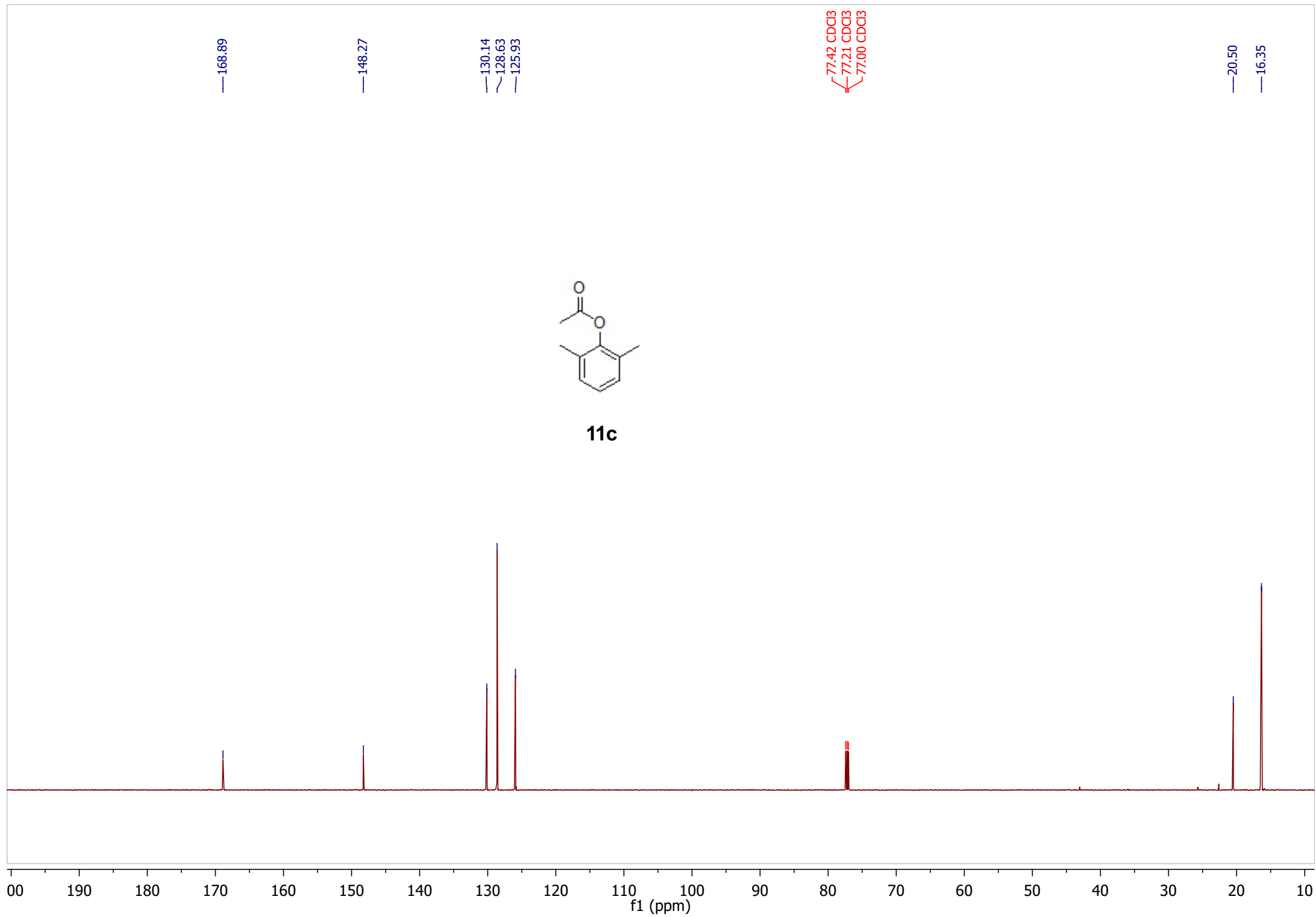


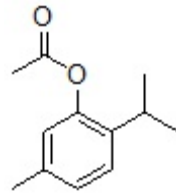
11c



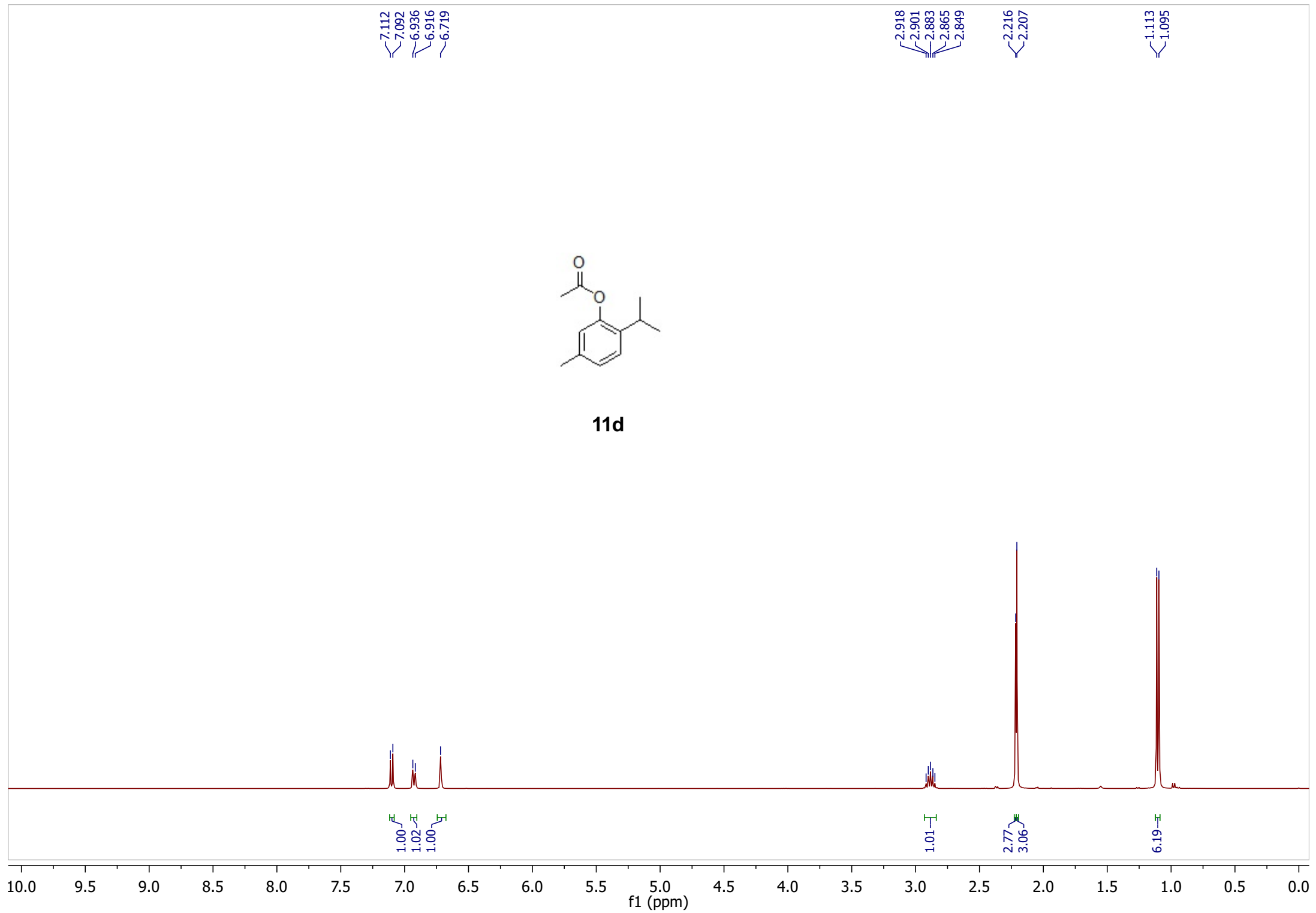


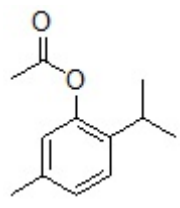
11c



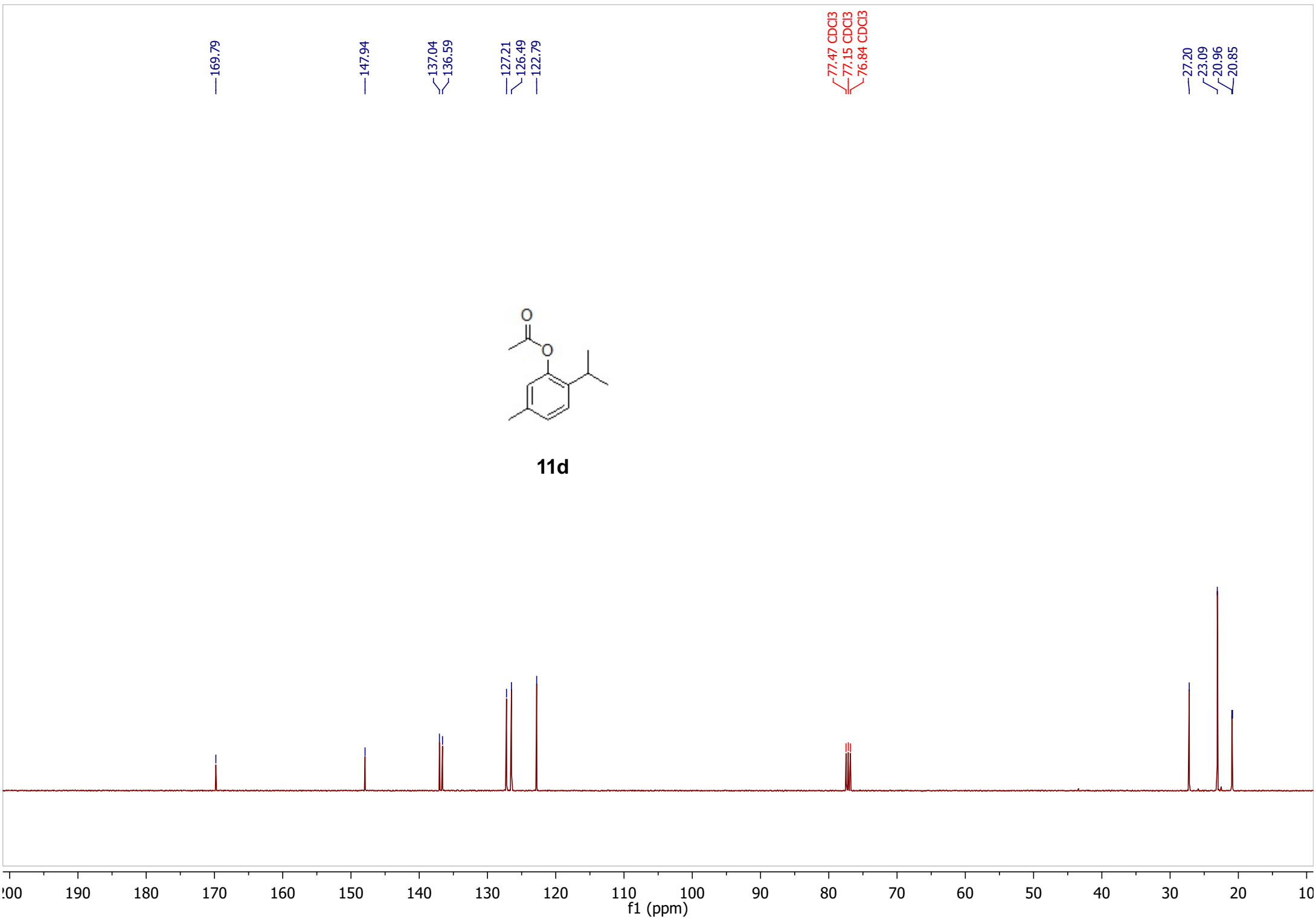


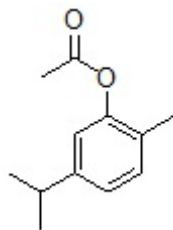
11d



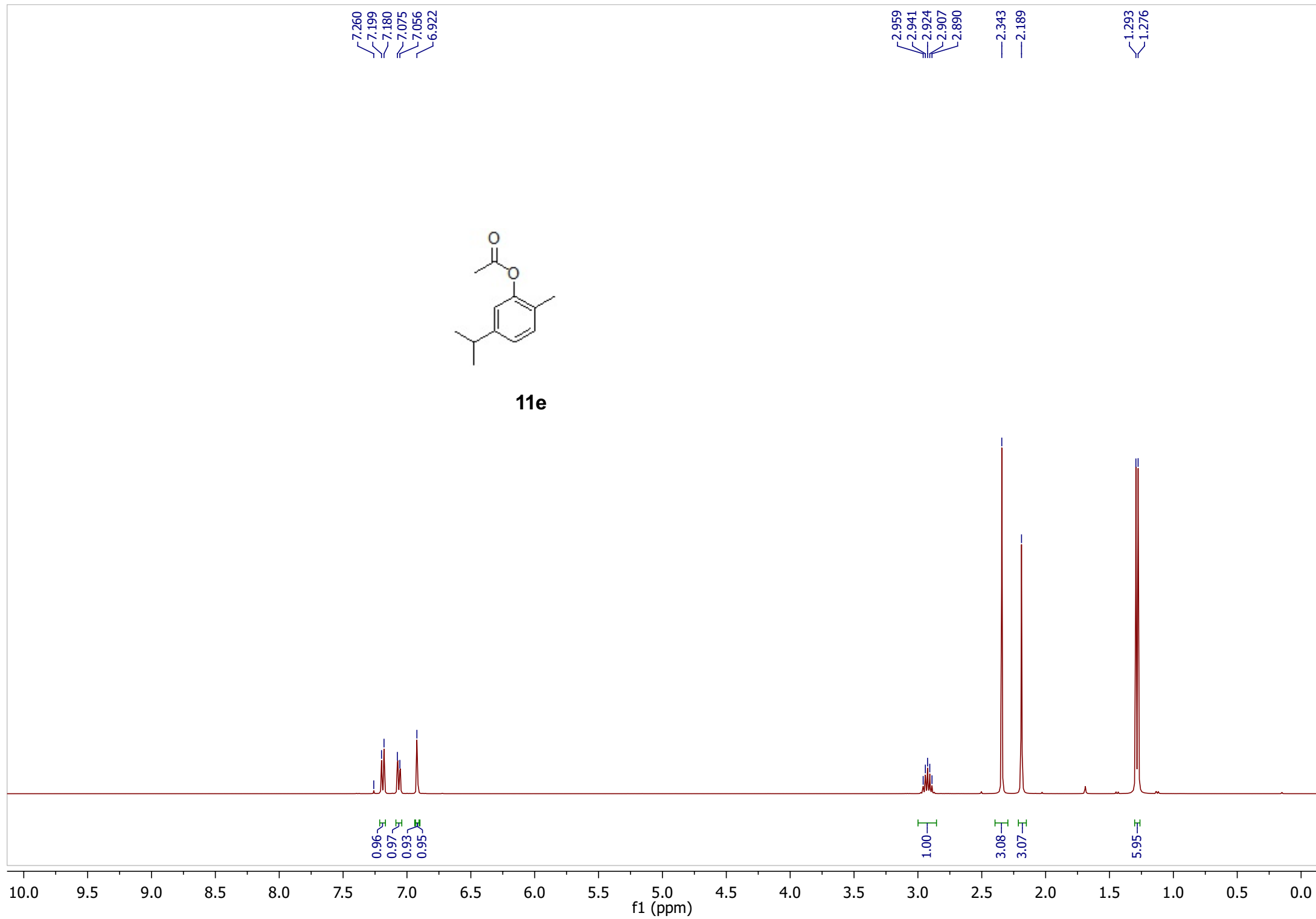


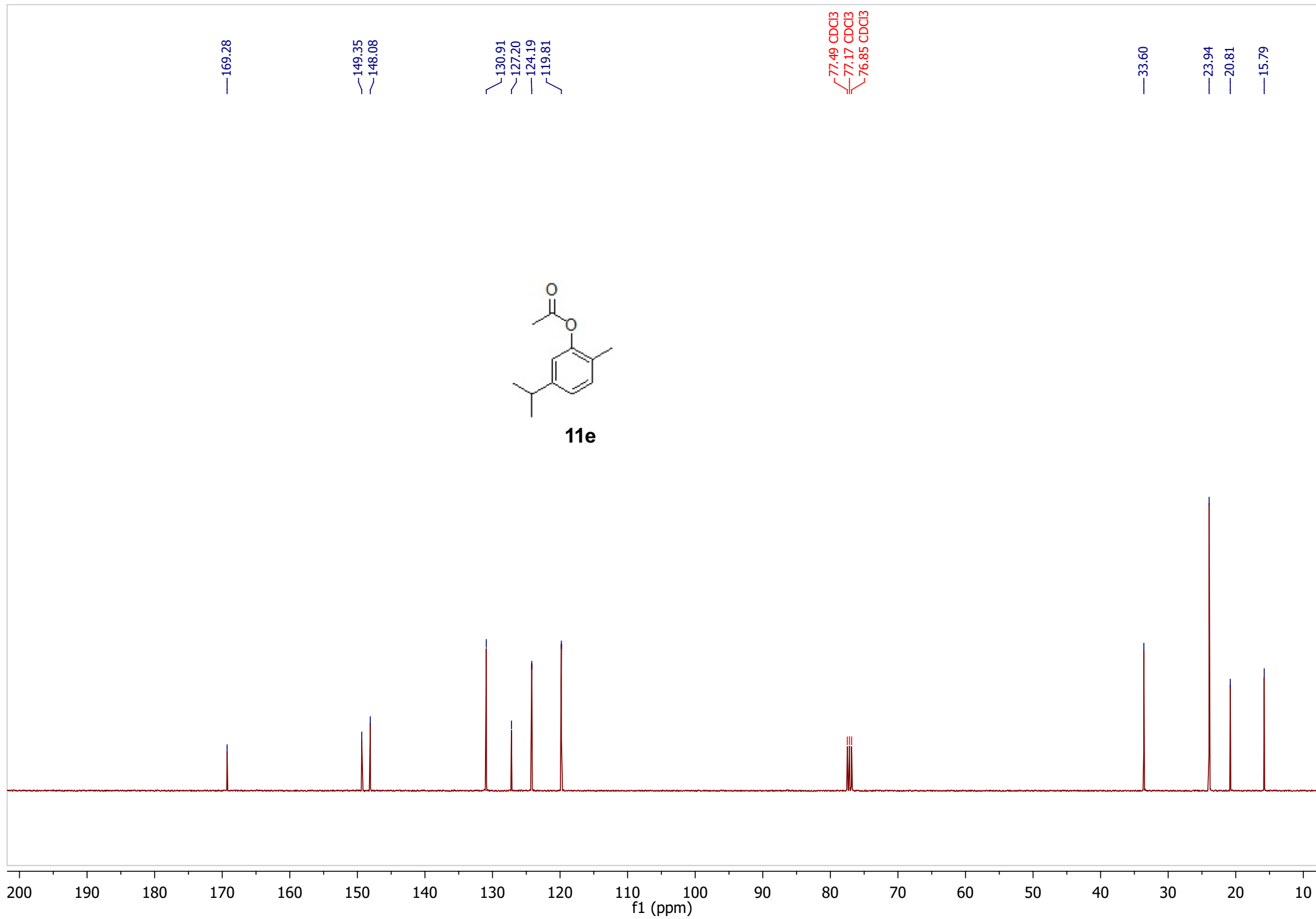
11d



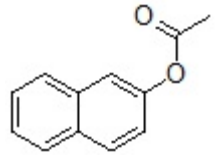


11e



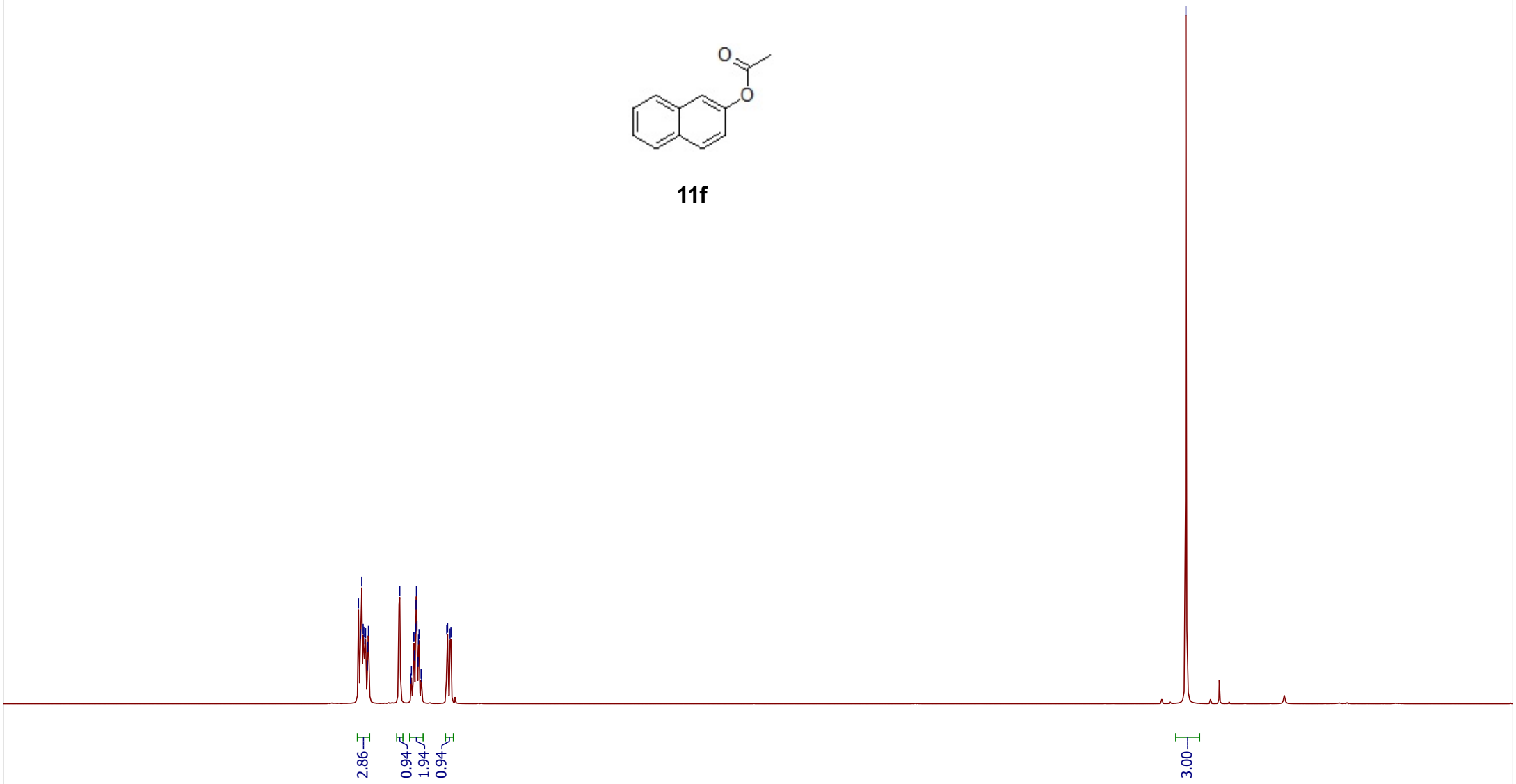


7.690
7.677
7.668
7.659
7.654
7.647
7.642
7.631
7.628
7.623
7.414
7.341
7.336
7.323
7.319
7.317
7.309
7.306
7.303
7.299
7.292
7.289
7.285
7.272
7.268
7.101
7.095
7.079
7.073



11f

2.167



2.86

0.94

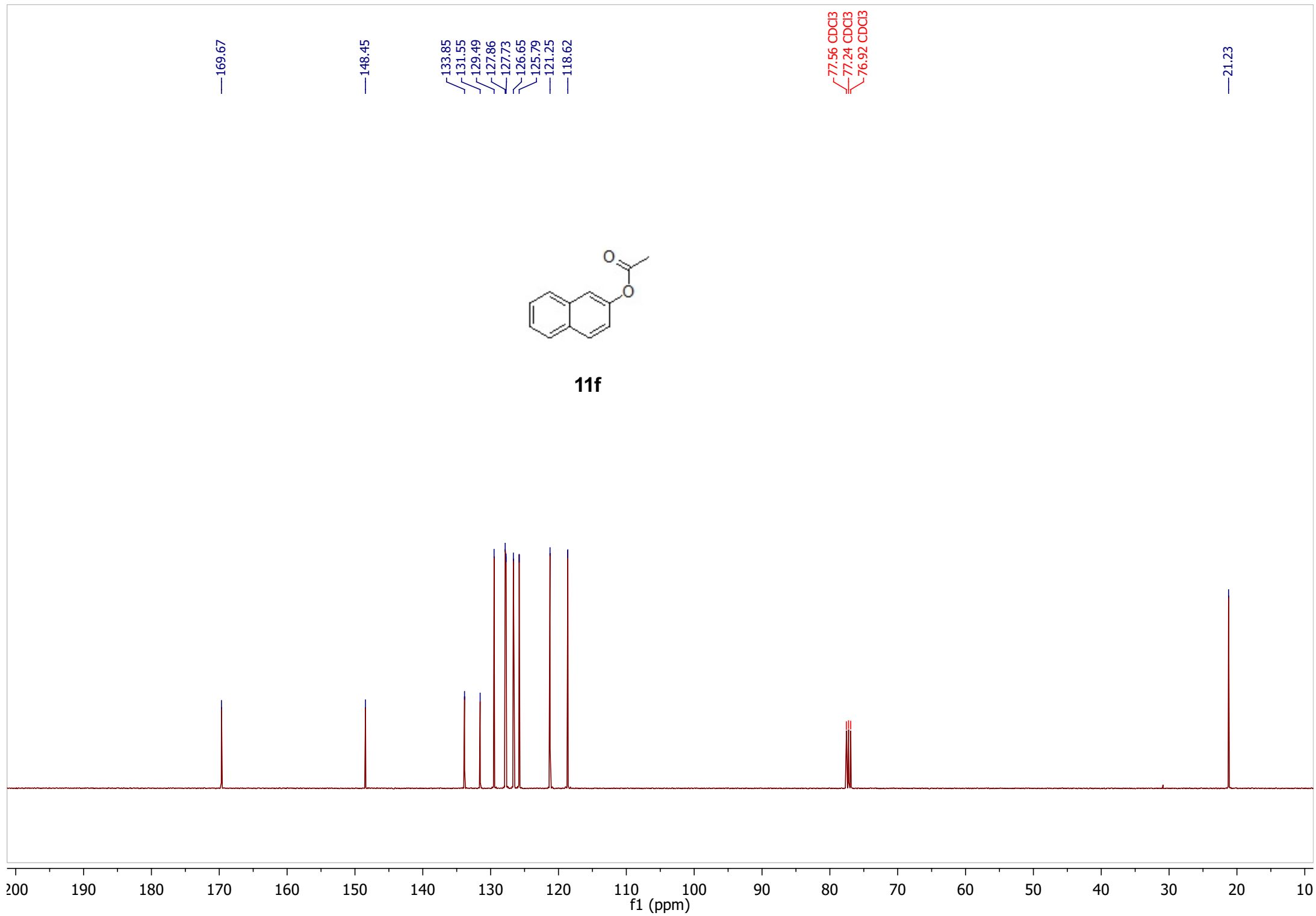
1.94

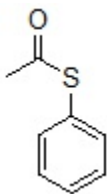
0.94

3.00

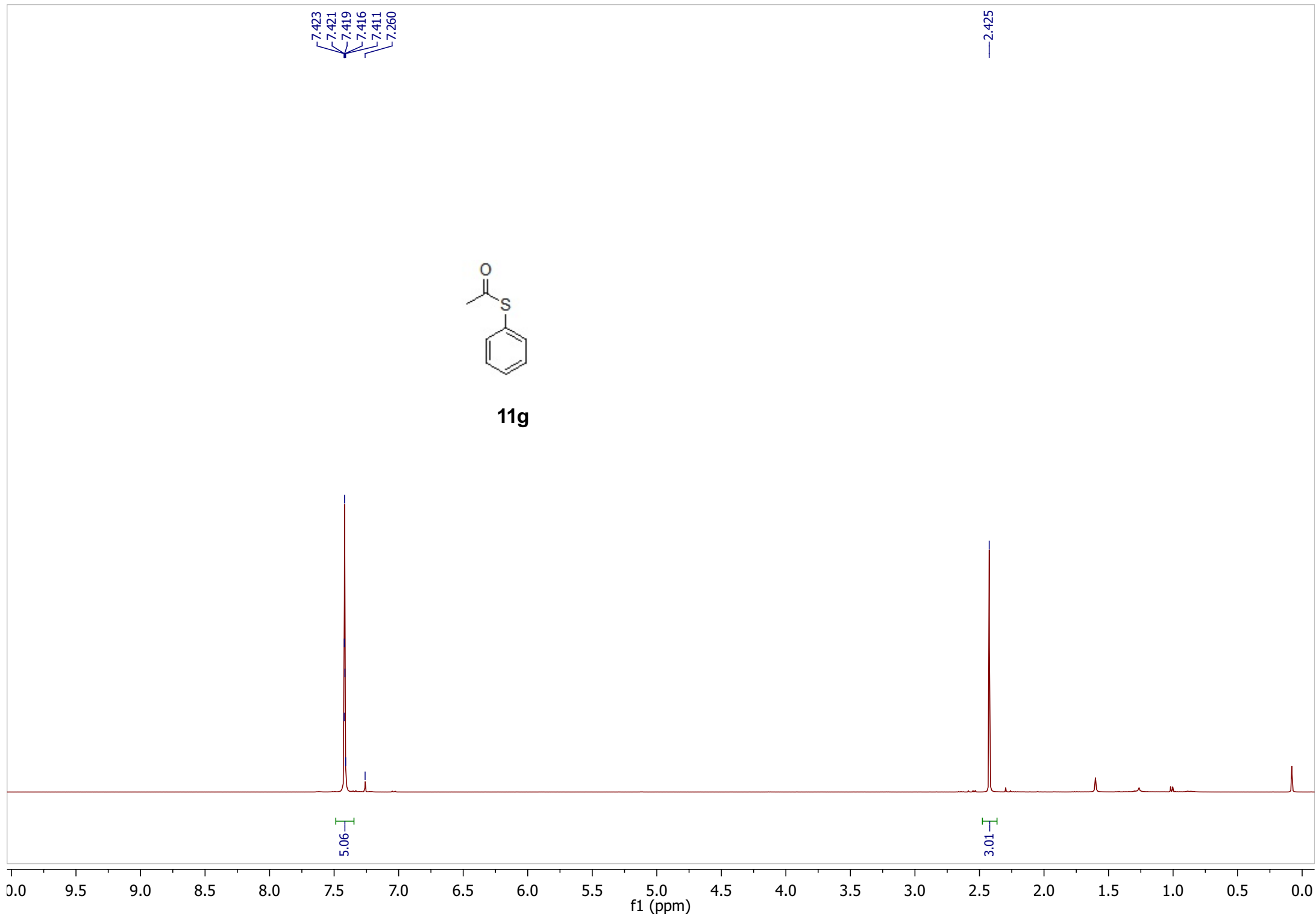
0.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.

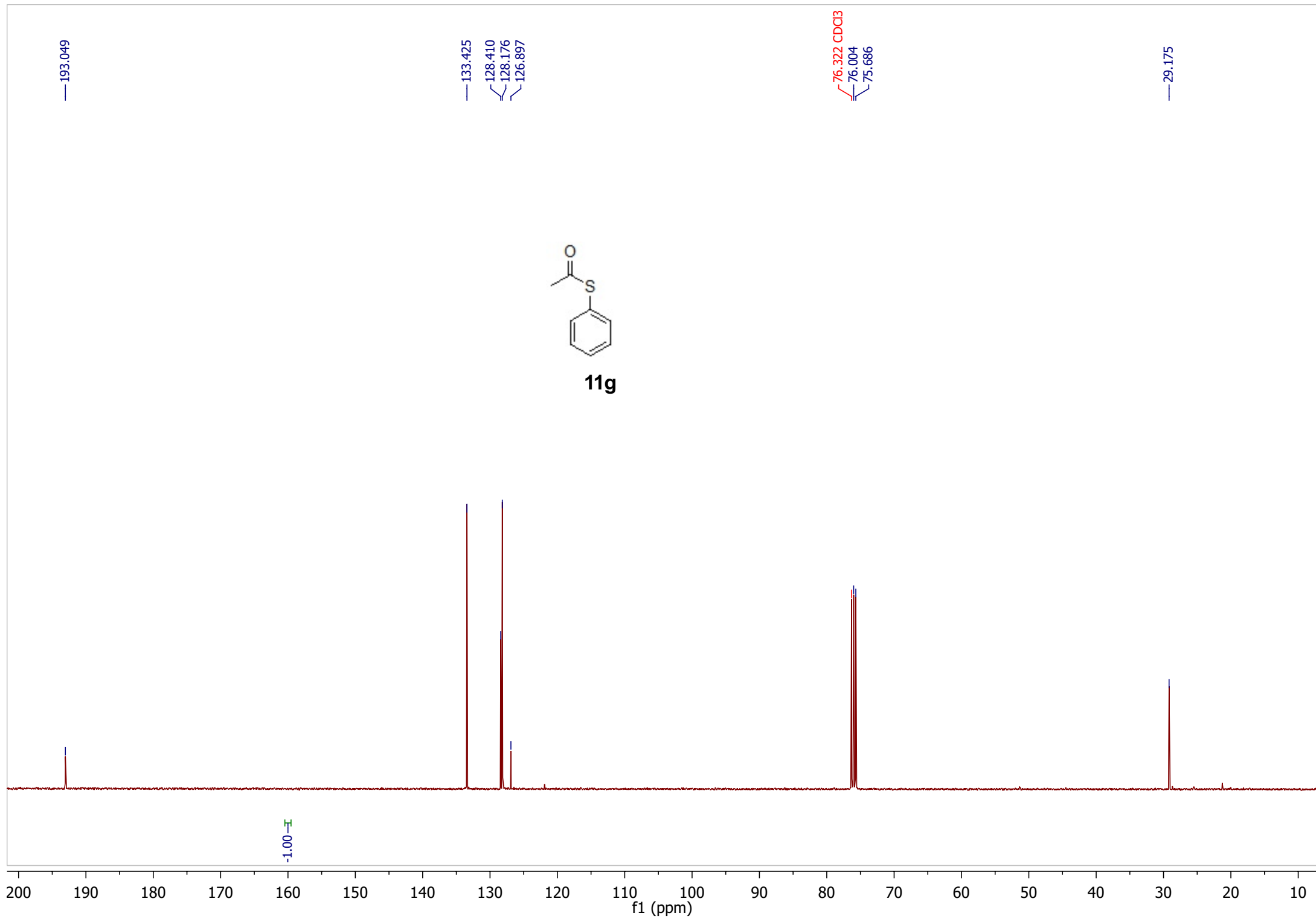
f1 (ppm)

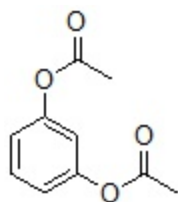




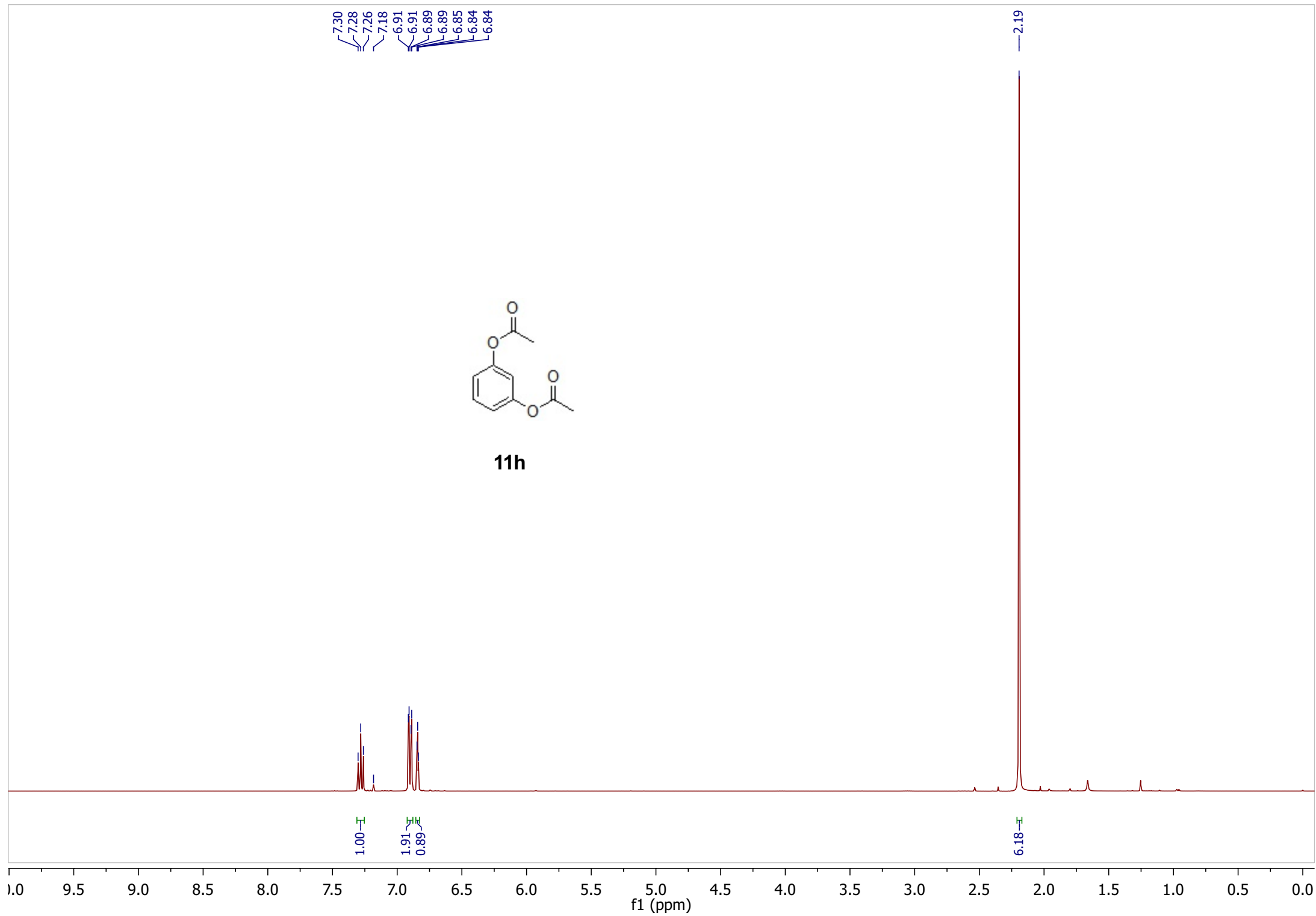
11g

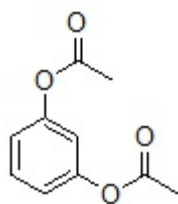






11h





11h

—169.02

—151.10

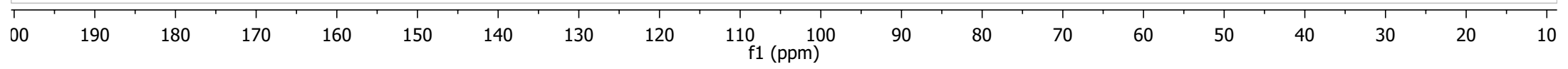
—129.70

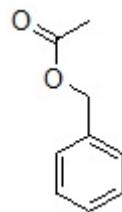
—118.99

—115.48

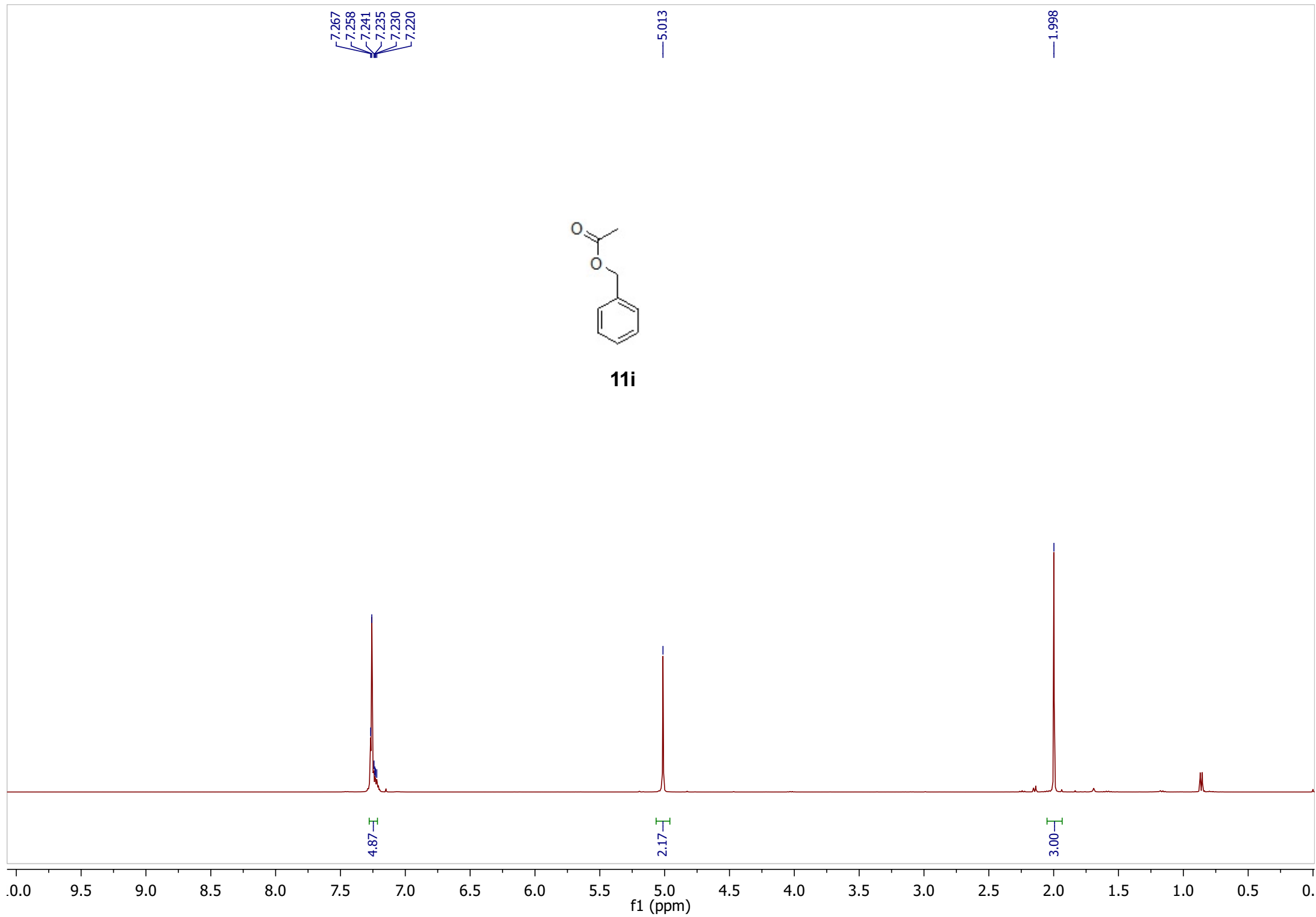
77.44 CDCl₃
77.13 CDCl₃
76.81 CDCl₃

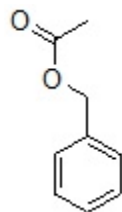
—21.08



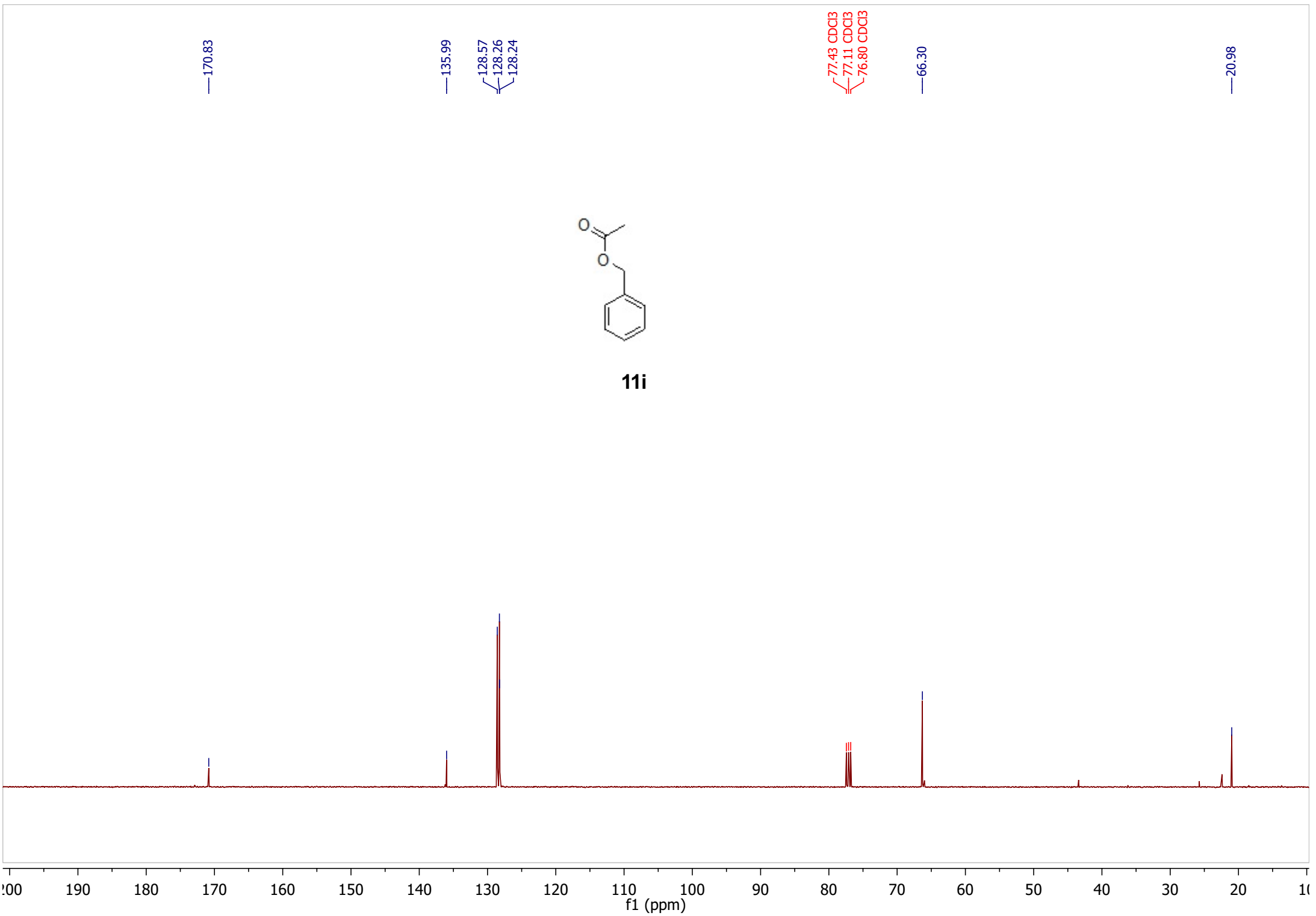


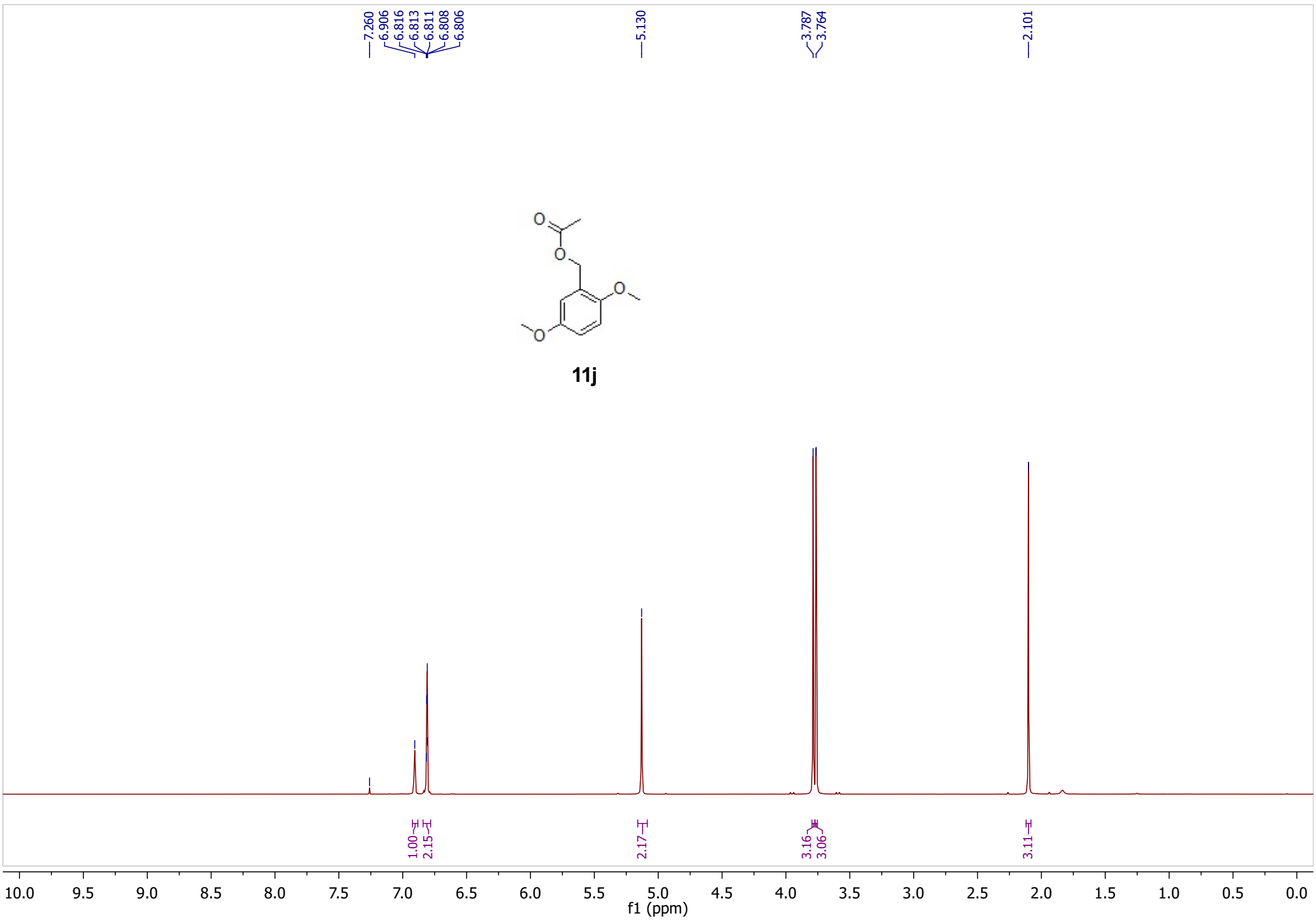
11i

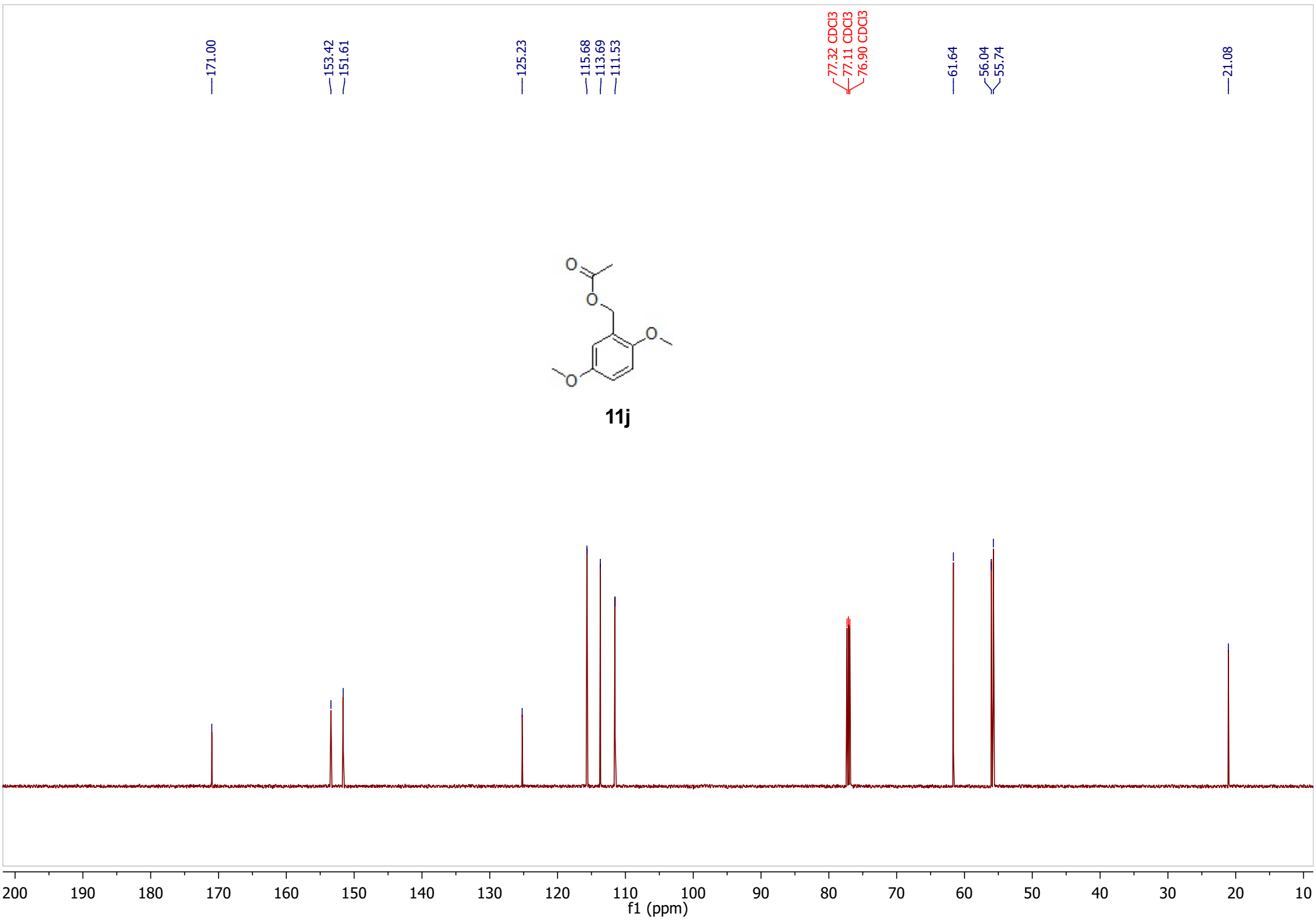


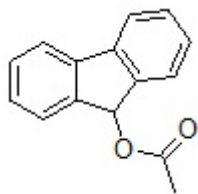


11i

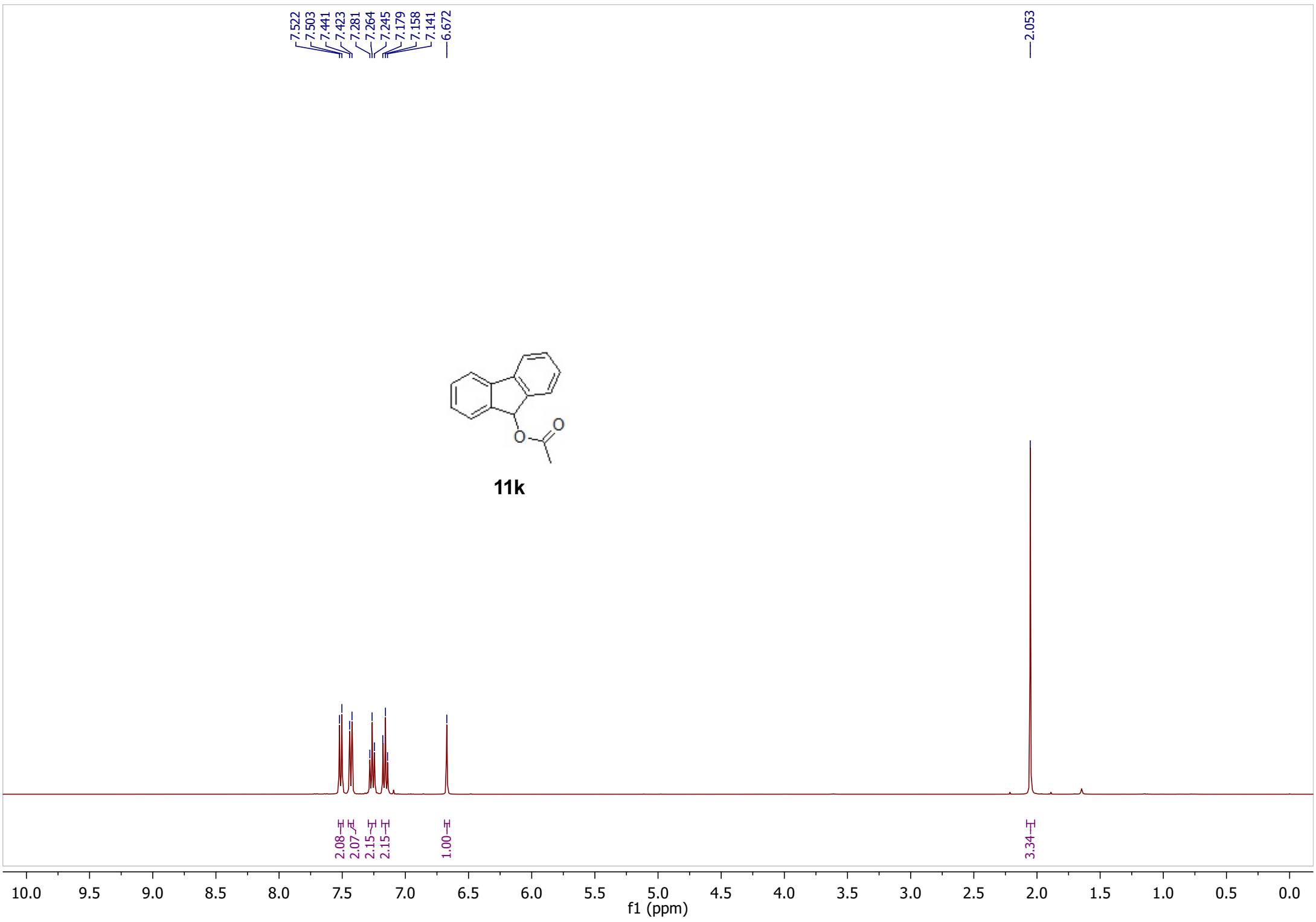


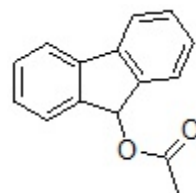






11k





11k

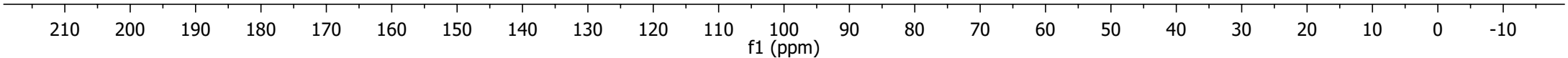
—171.80

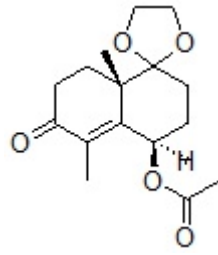
—142.09
—141.07

—129.54
—127.90
—125.94
—120.09

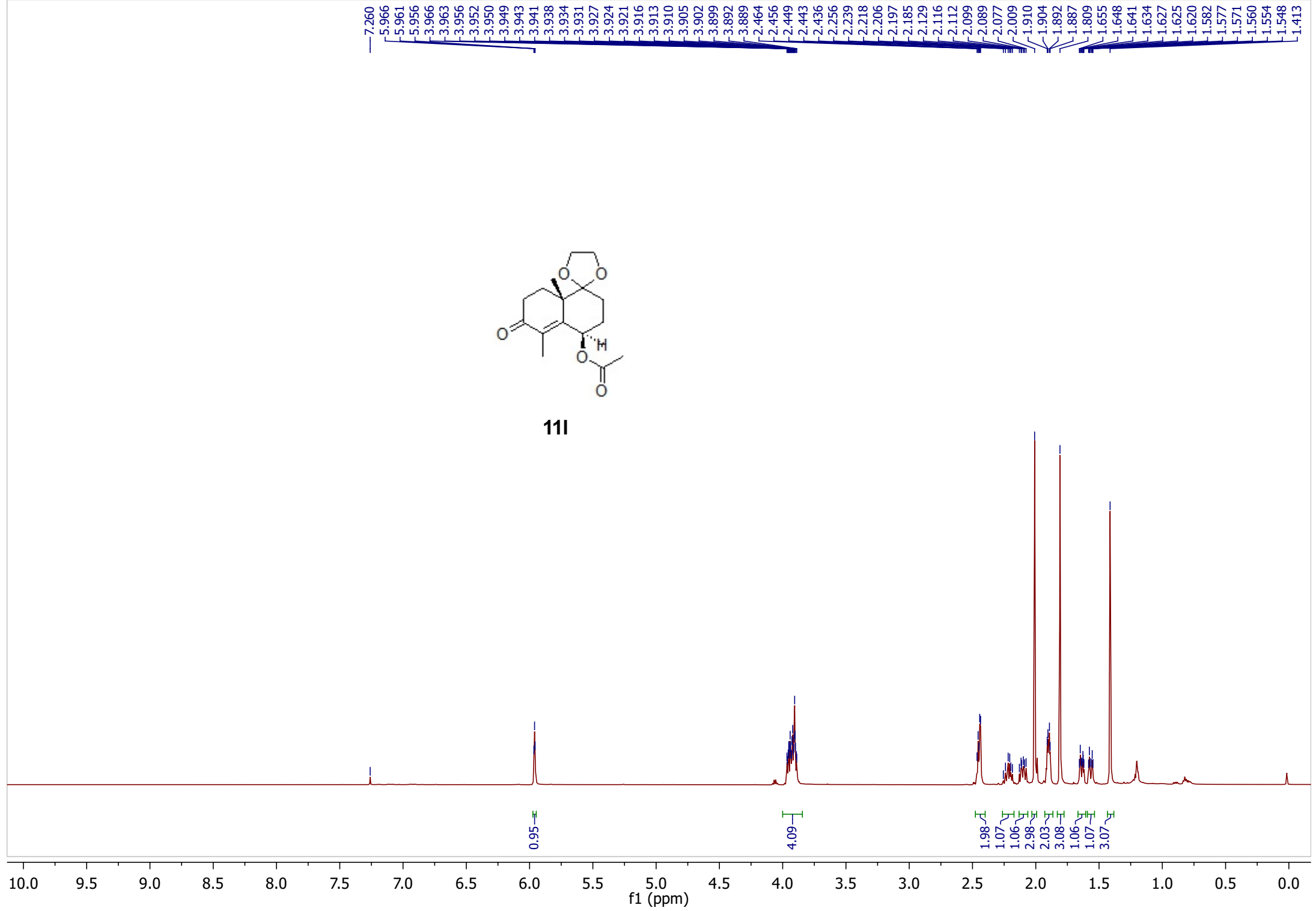
77.49 CDCl₃
77.18 CDCl₃
76.86 CDCl₃
75.21

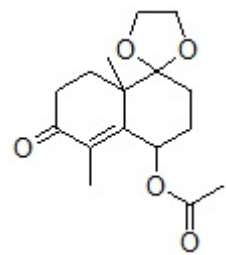
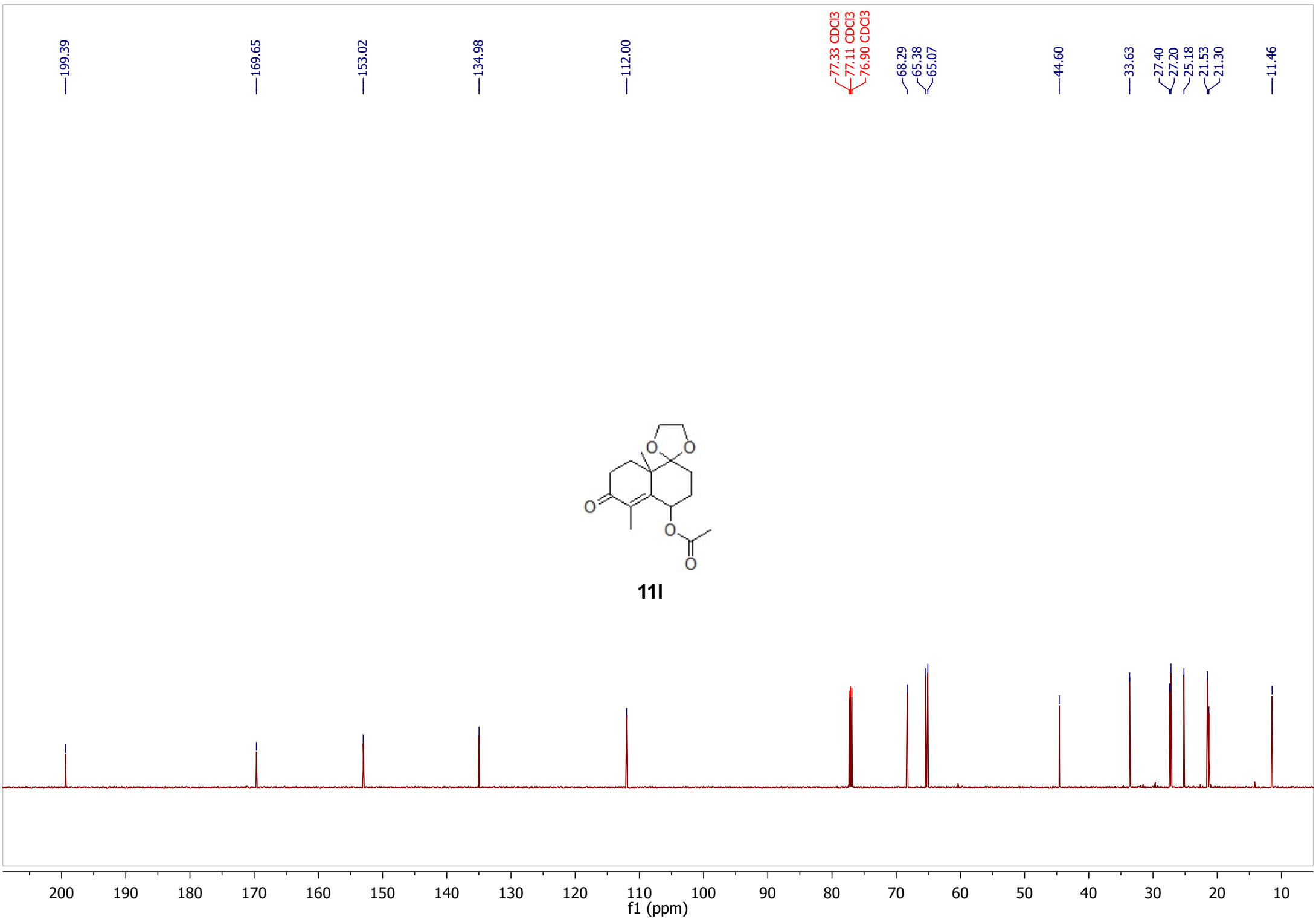
—21.29





111





11I

199.39

169.65

153.02

134.98

112.00

77.33 CDCl3
77.11 CDCl3
76.90 CDCl3

68.29
65.38
65.07

44.60

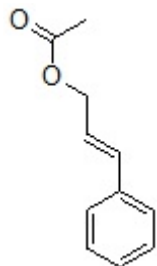
33.63

27.40
27.20
25.18
21.53
21.30

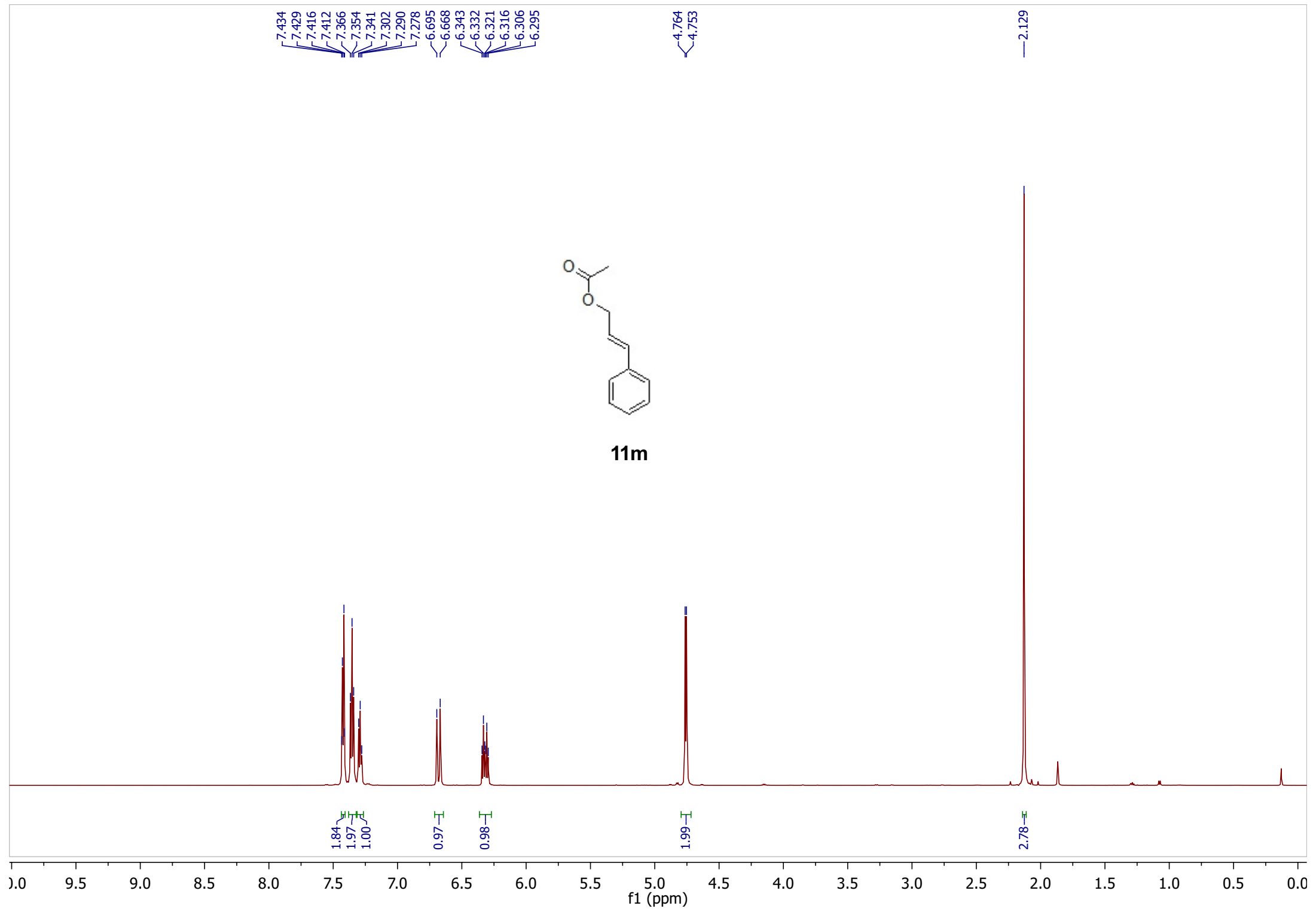
11.46

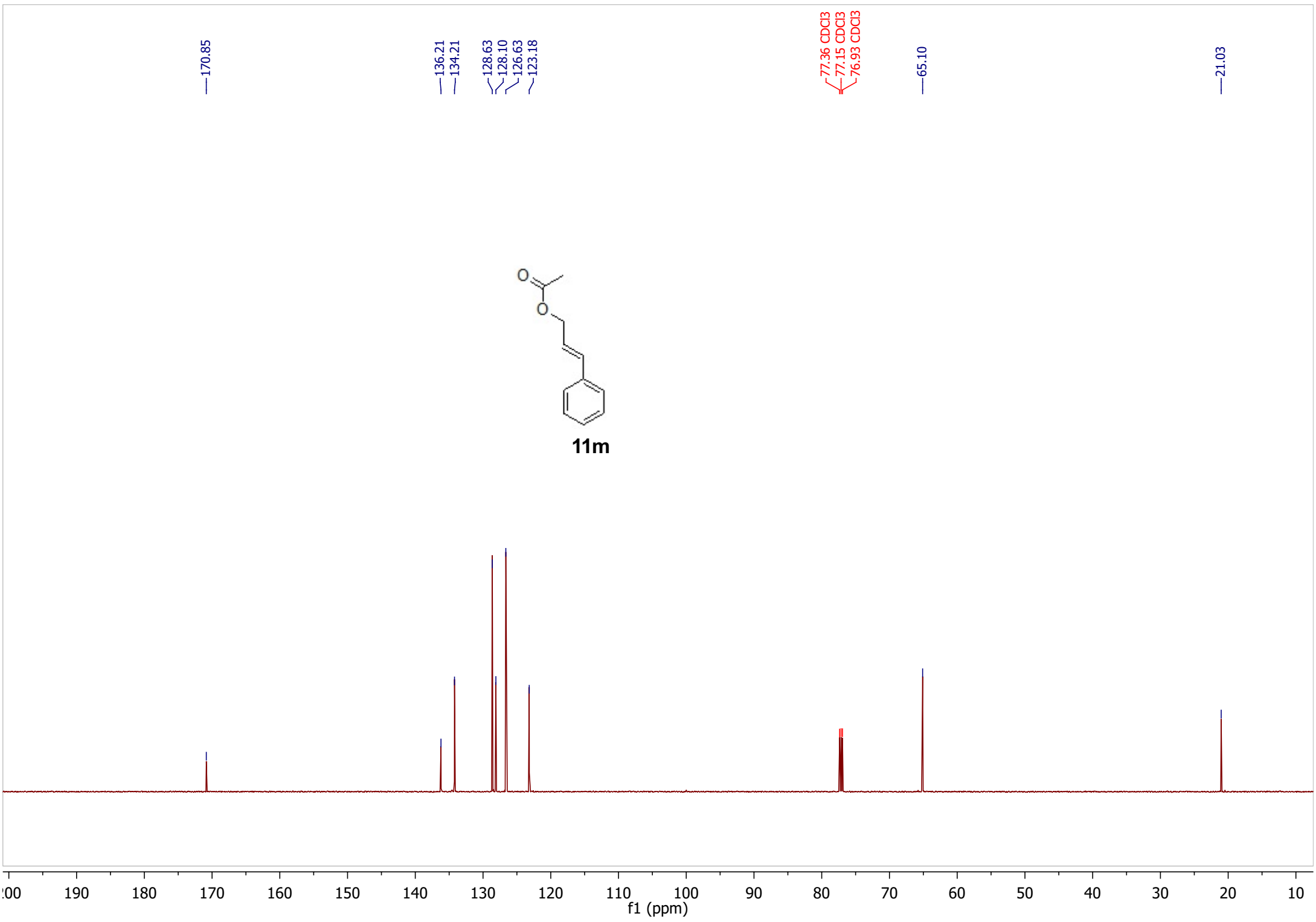
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10

f1 (ppm)



11m

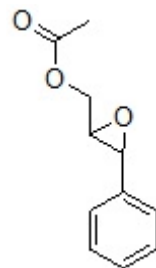




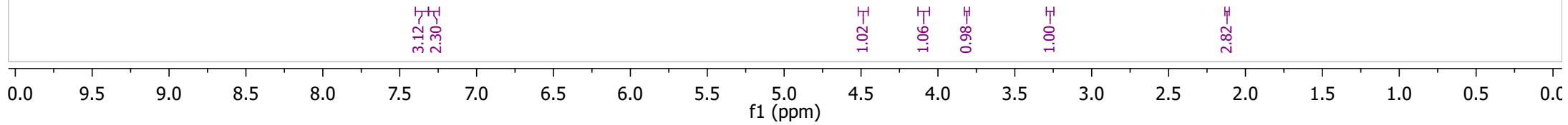
7.384
7.378
7.373
7.368
7.362
7.357
7.355
7.348
7.344
7.338
7.334
7.330
7.323
7.294
7.289
7.283
7.278
7.274
7.269

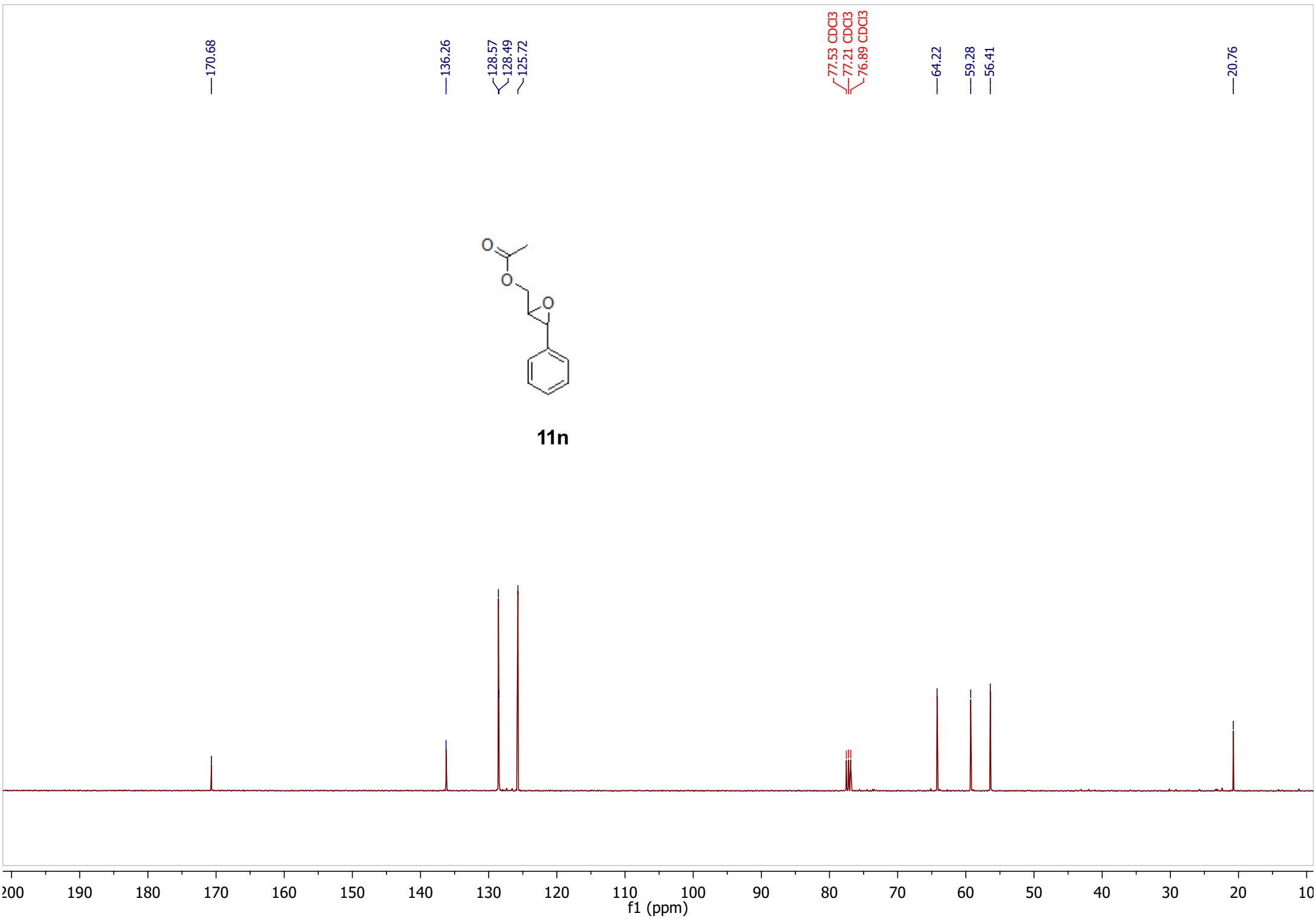
4.509
4.501
4.478
4.470
4.115
4.100
4.084
4.069
3.818
3.813
3.287
3.282
3.279
3.273
3.267
3.263
3.259

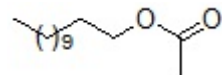
2.121



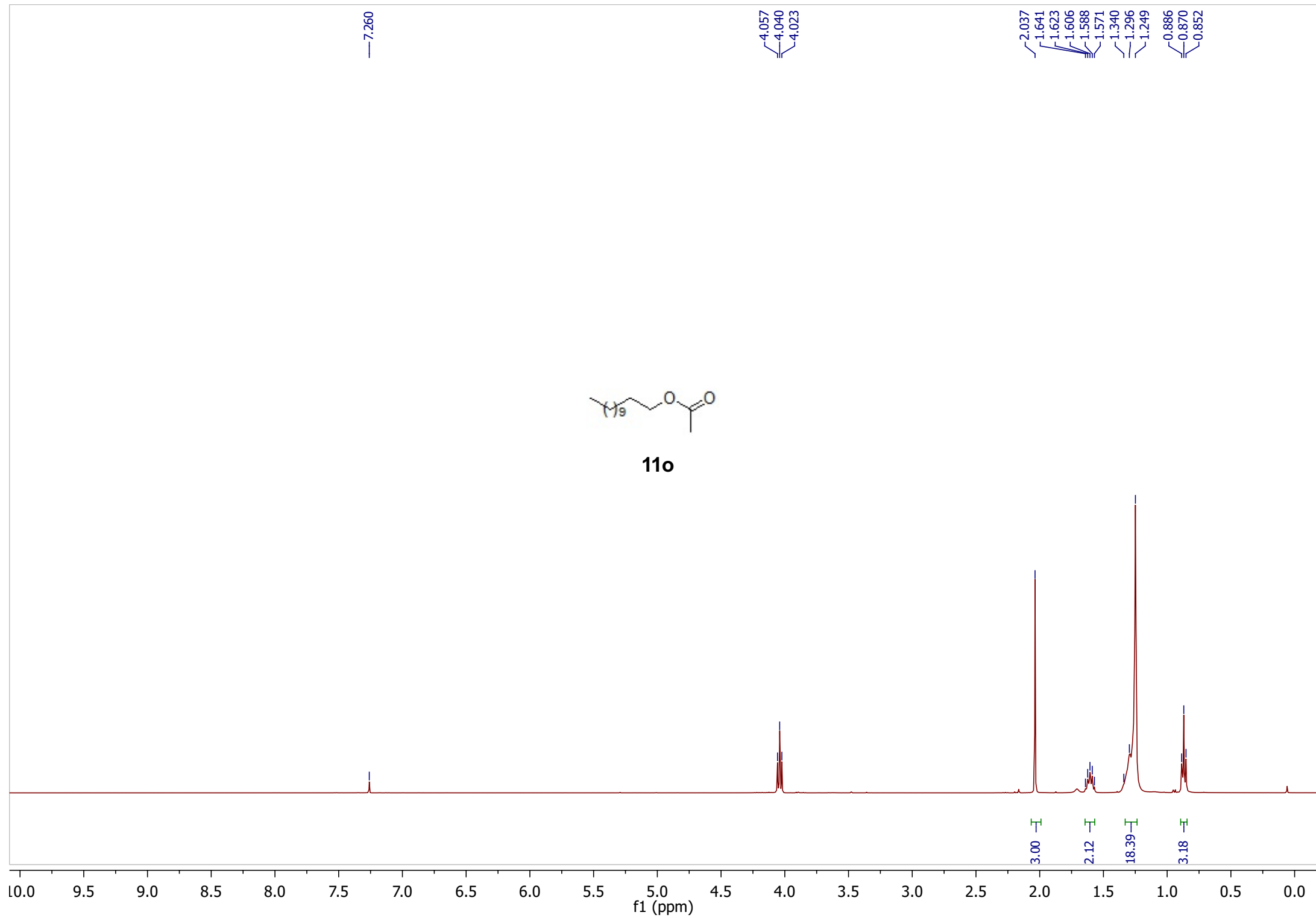
11n

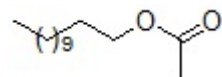




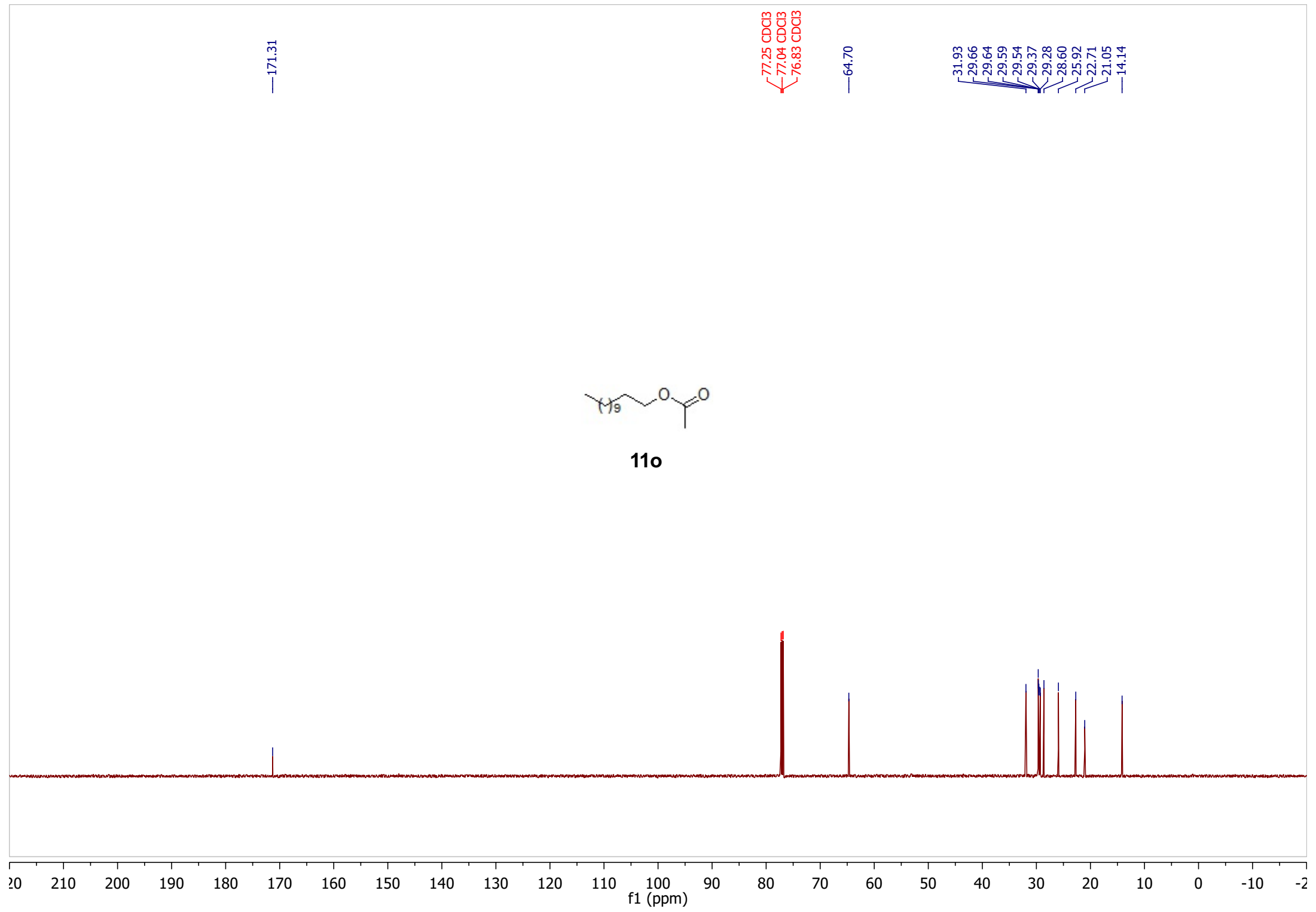


11o



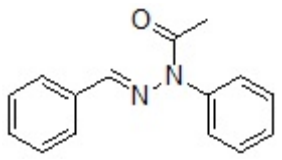


11o

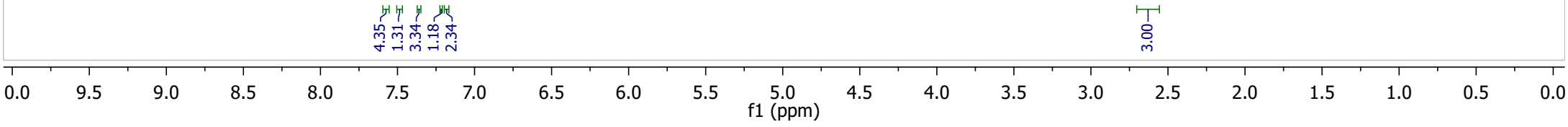


7.595
7.588
7.579
7.567
7.553
7.501
7.489
7.476
7.376
7.370
7.367
7.361
7.358
7.260
7.217
7.188
7.176

2.636

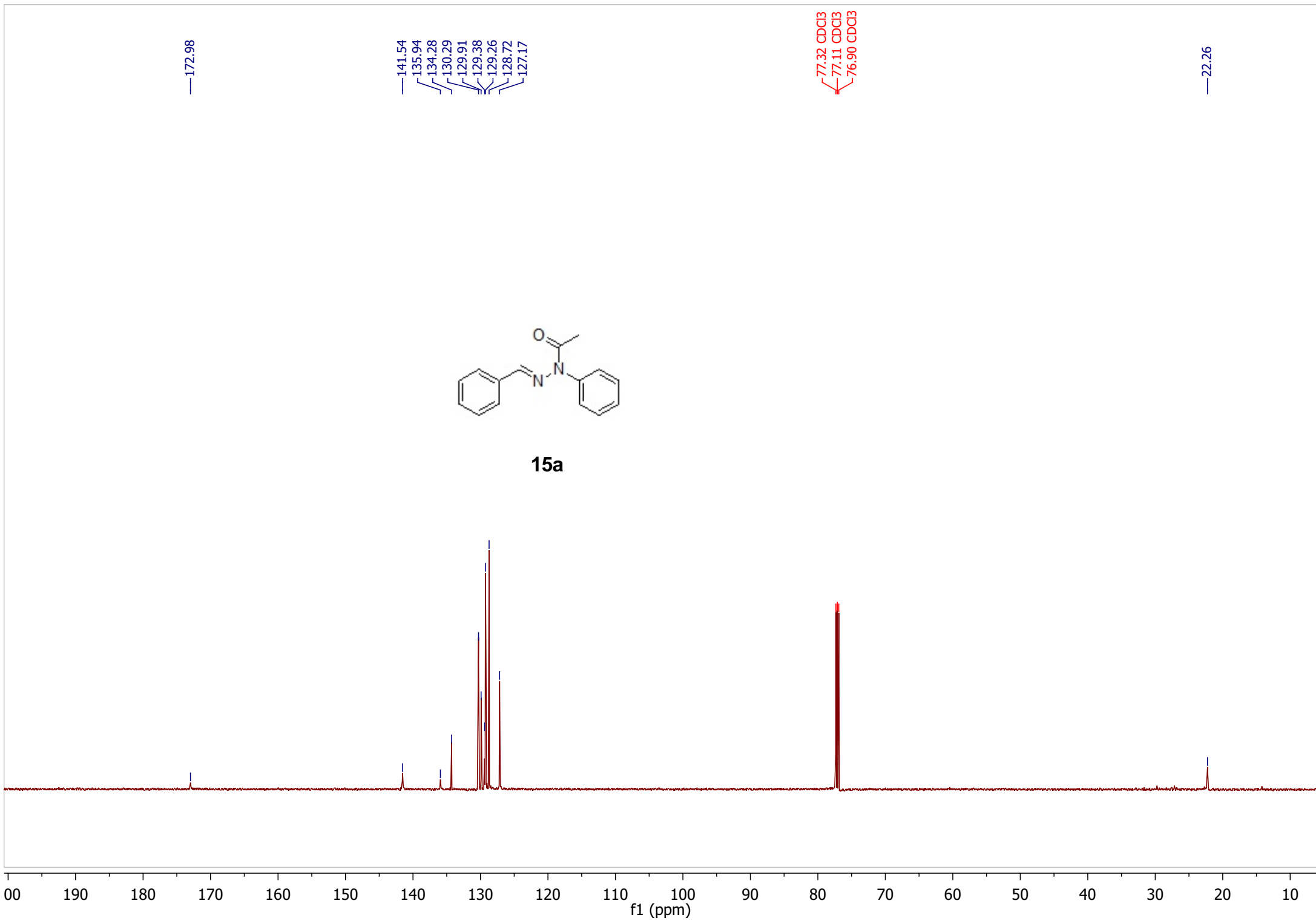


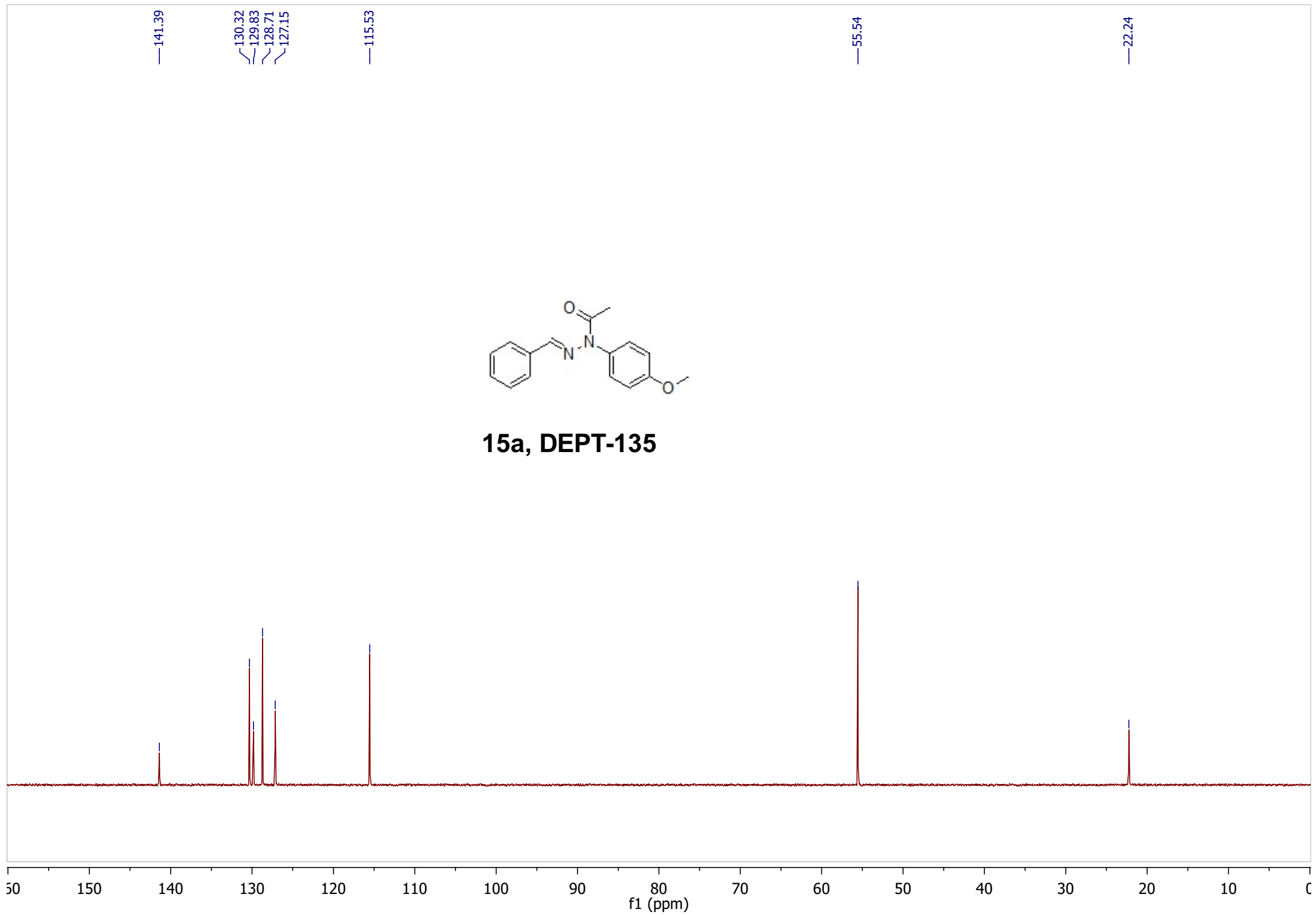
15a



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

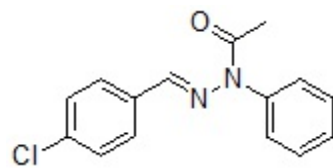
f1 (ppm)



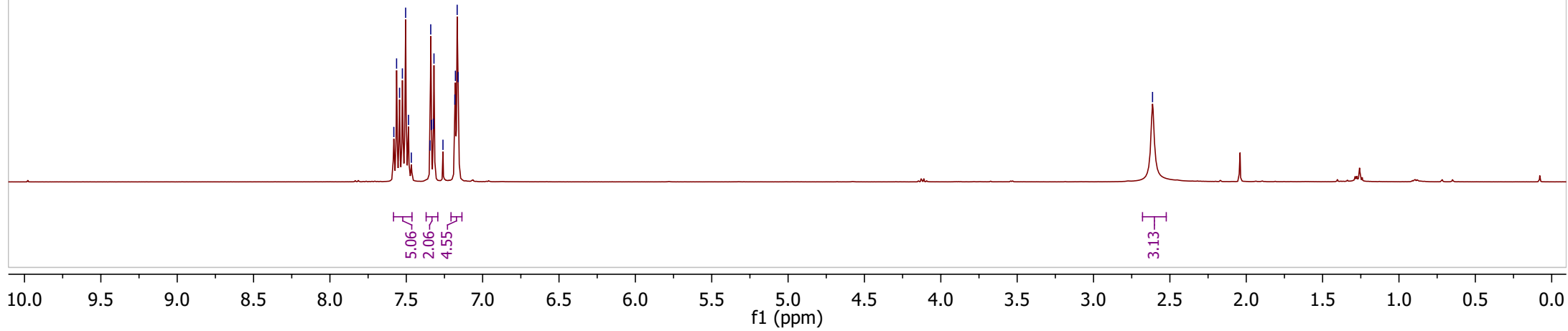


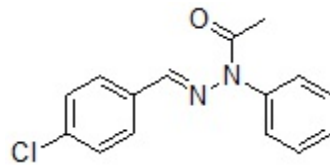
7.582
7.564
7.544
7.526
7.504
7.486
7.467
7.345
7.340
7.335
7.323
7.319
7.260
7.182
7.178
7.167
7.161

—2.614



15b





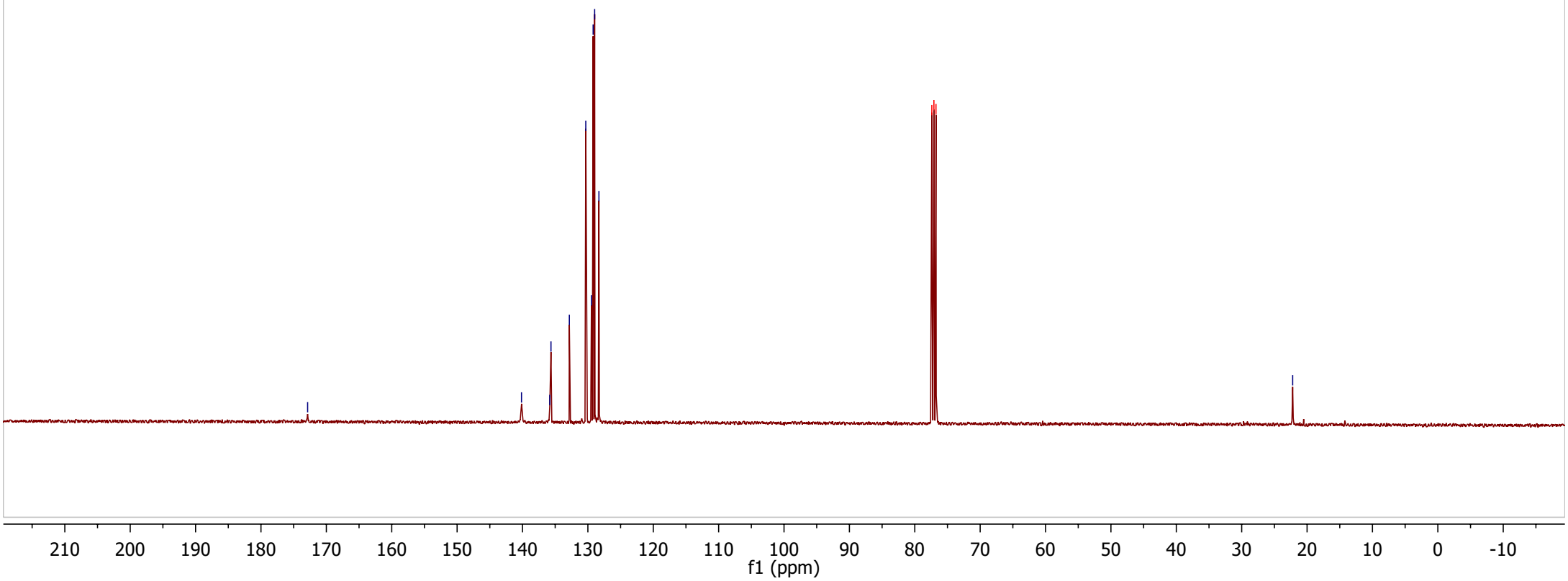
15b

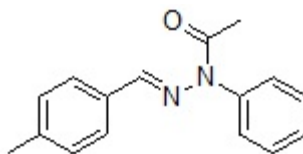
—172.86

140.15
135.83
135.64
132.84
130.32
129.46
129.19
128.96
128.31

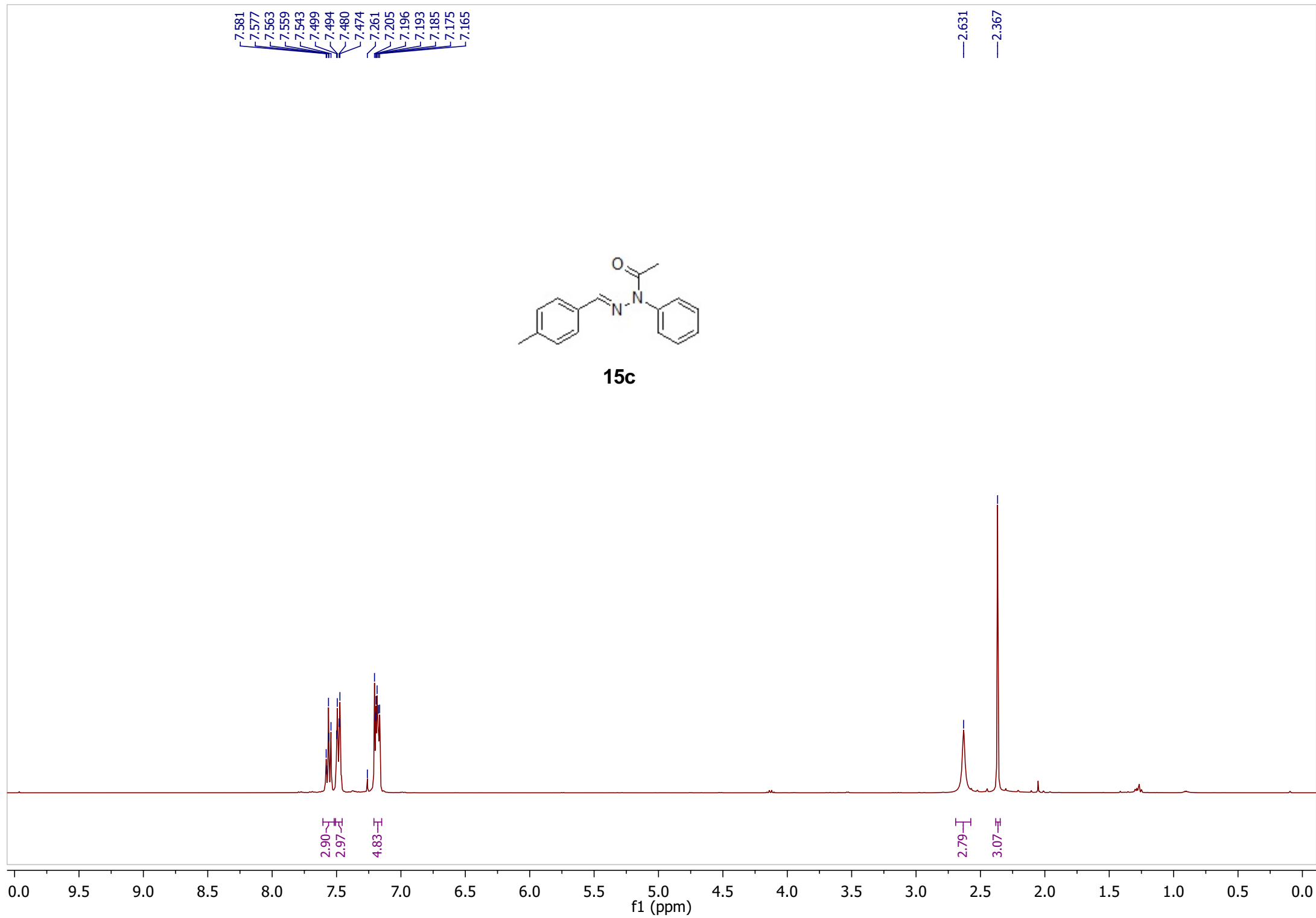
77.39 CDCl3
77.07 CDCl3
76.76 CDCl3

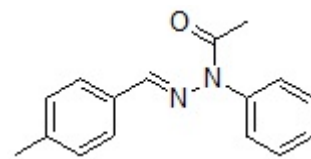
—22.20



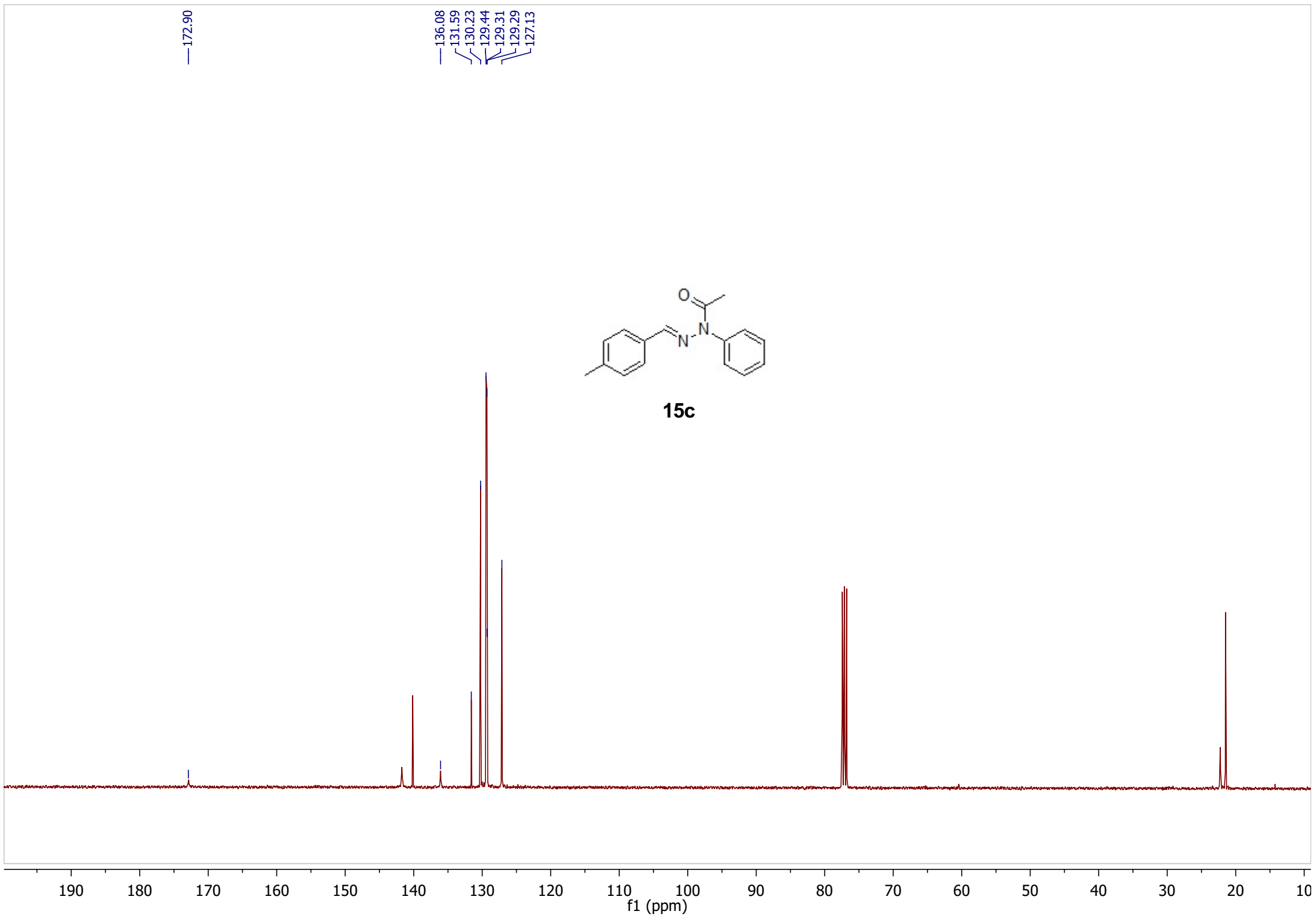


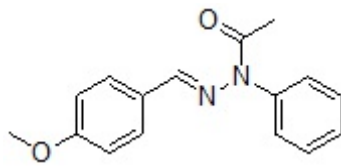
15c



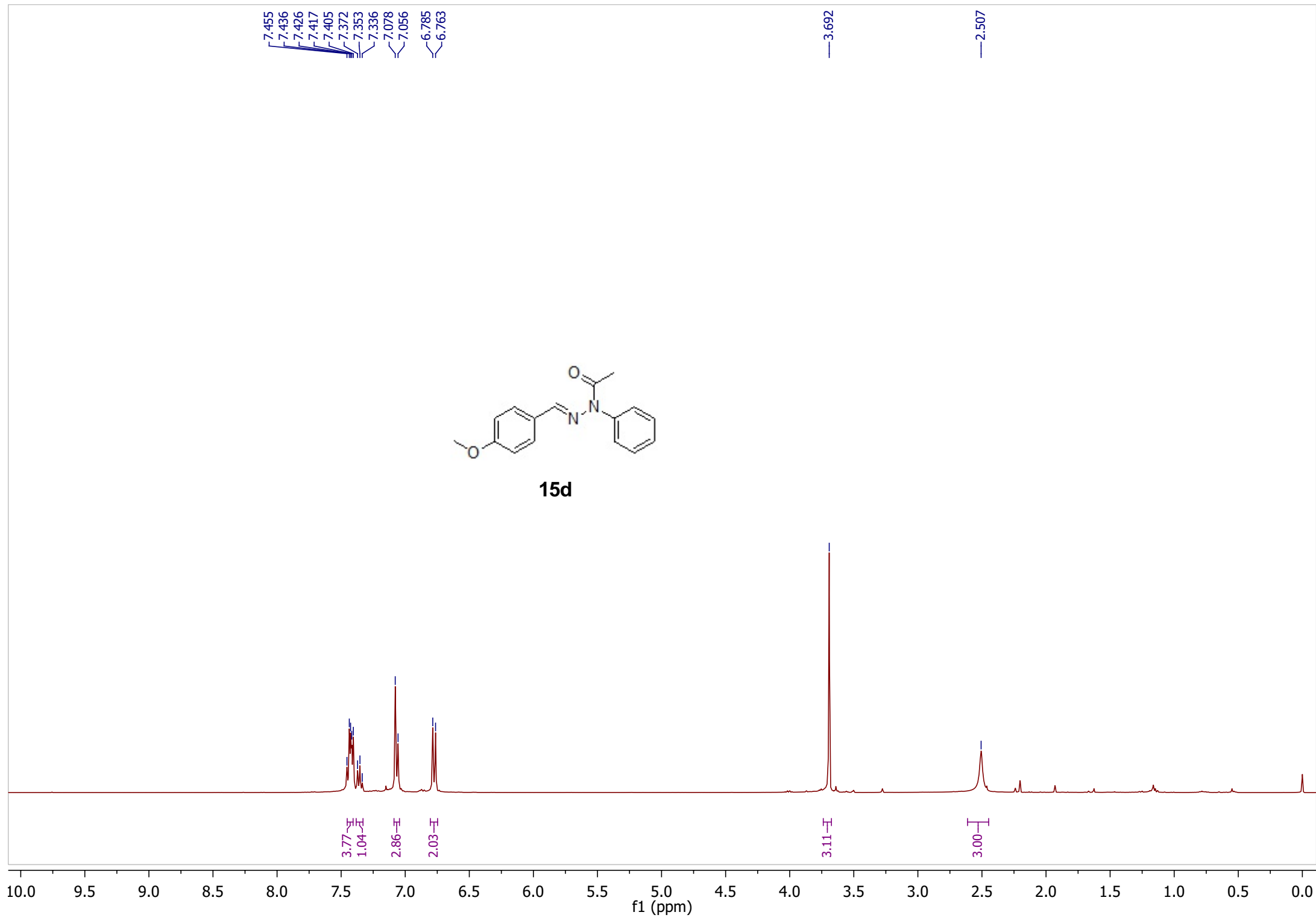


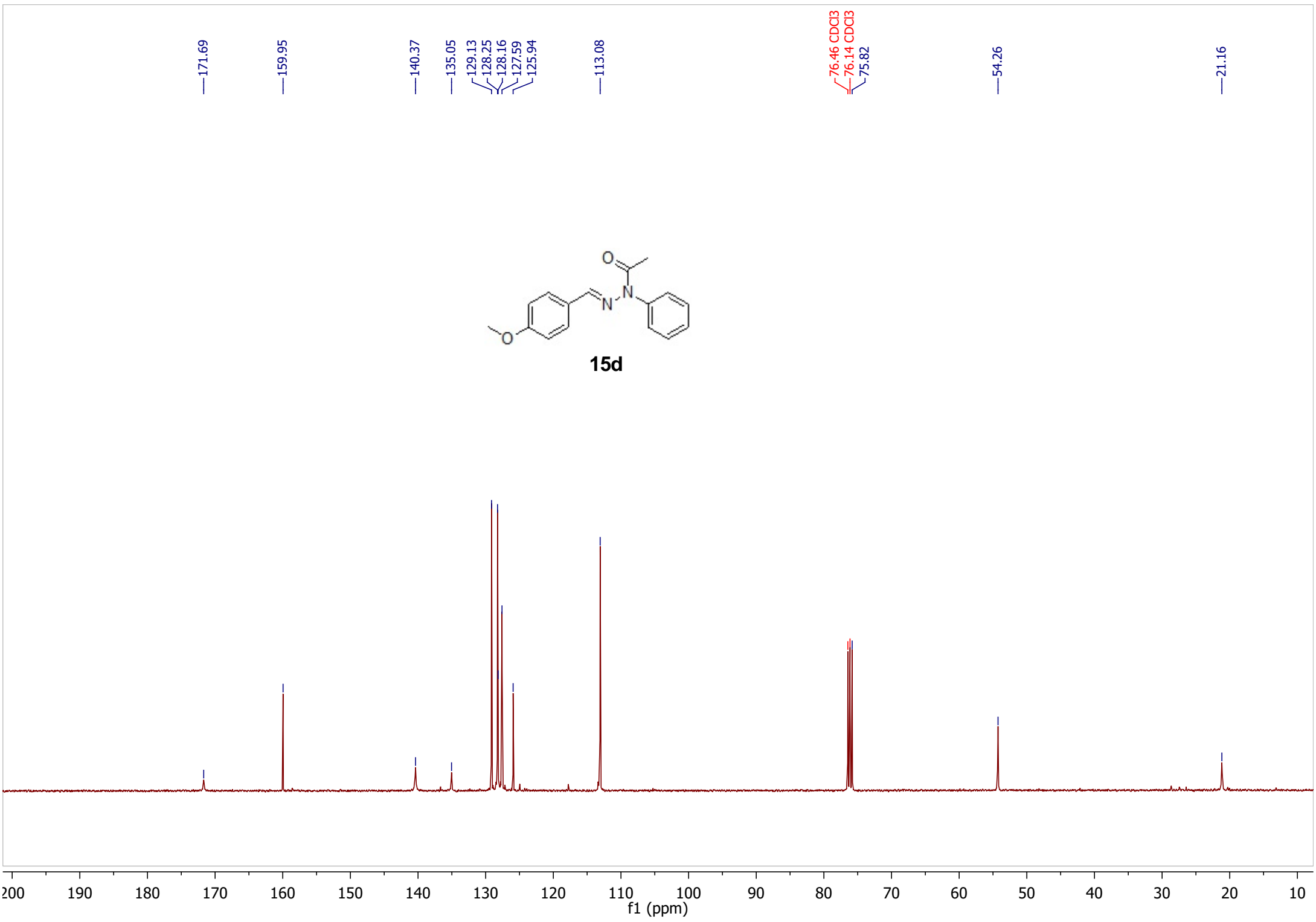
15c





15d

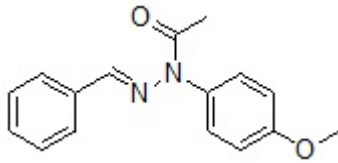




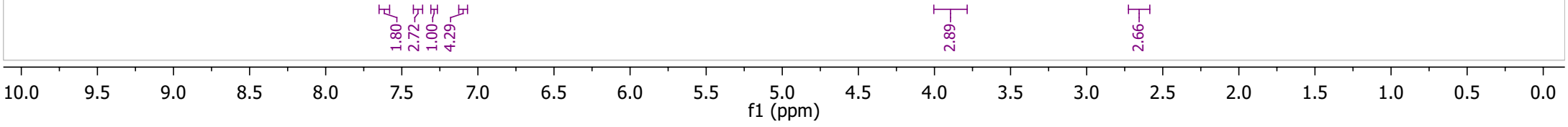
7.629
7.620
7.610
7.417
7.405
7.395
7.390
7.382
7.377
7.289
7.130
7.125
7.108
7.094
7.078
7.071

3.889

2.651



15e



10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0

f1 (ppm)

—173.18

—159.97

—141.39

134.36

130.32

129.82

128.71

128.21

127.15

—115.53

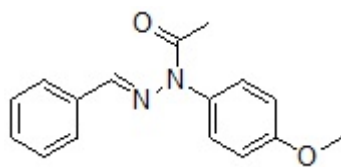
77.43 CDCl3

77.11 CDCl3

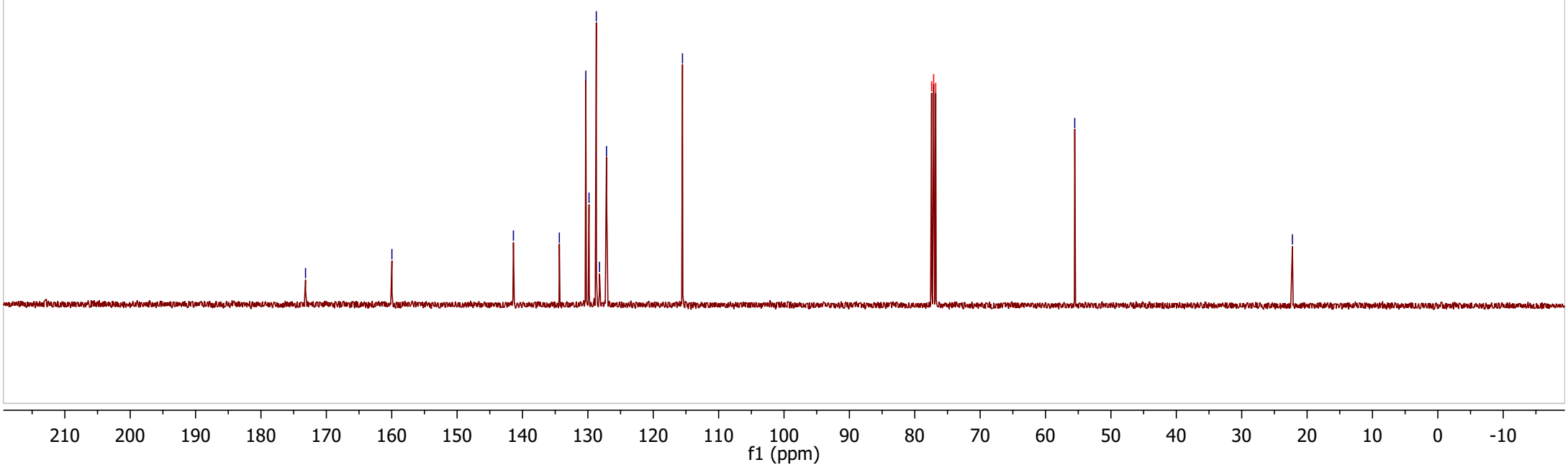
76.79 CDCl3

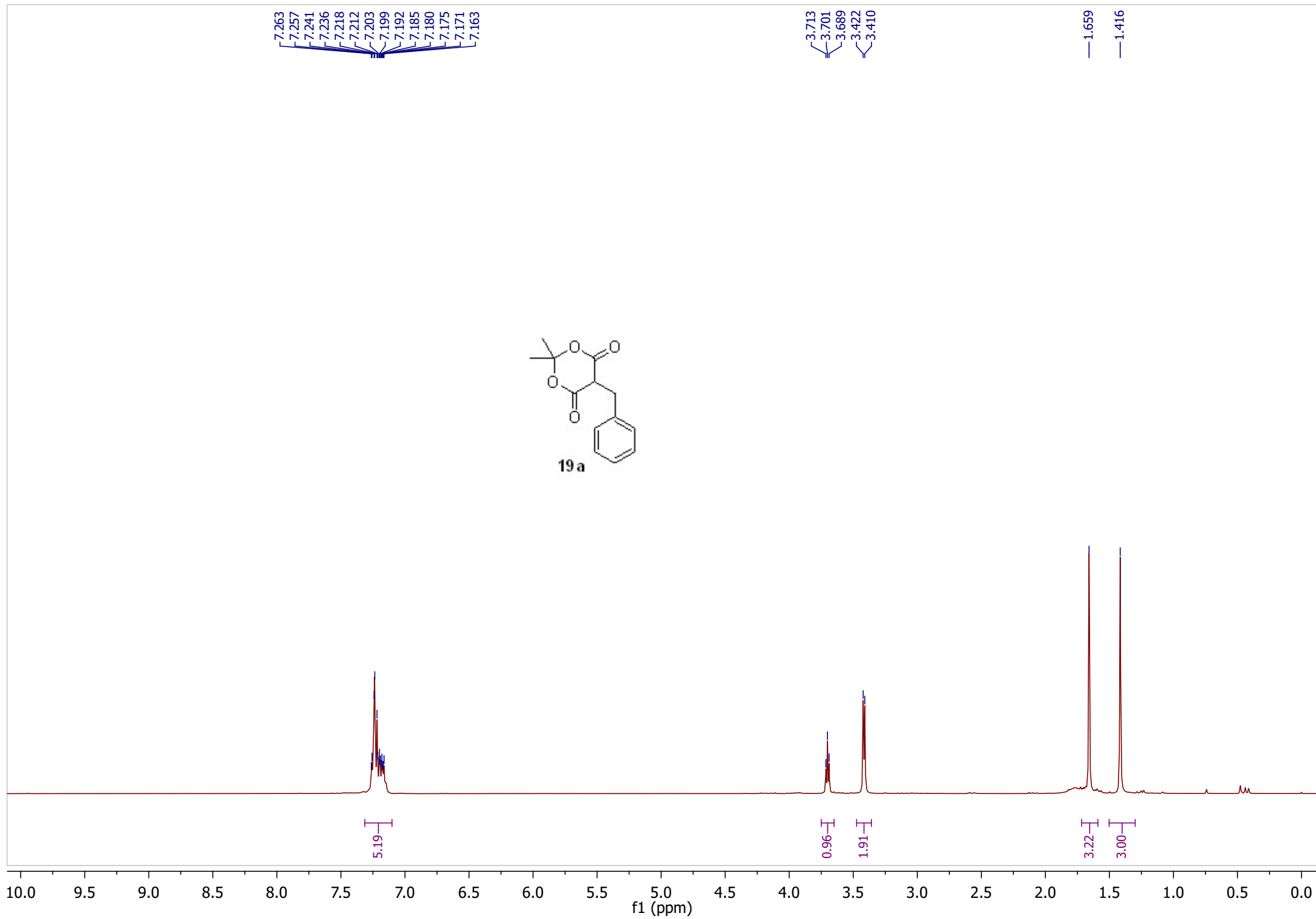
—55.54

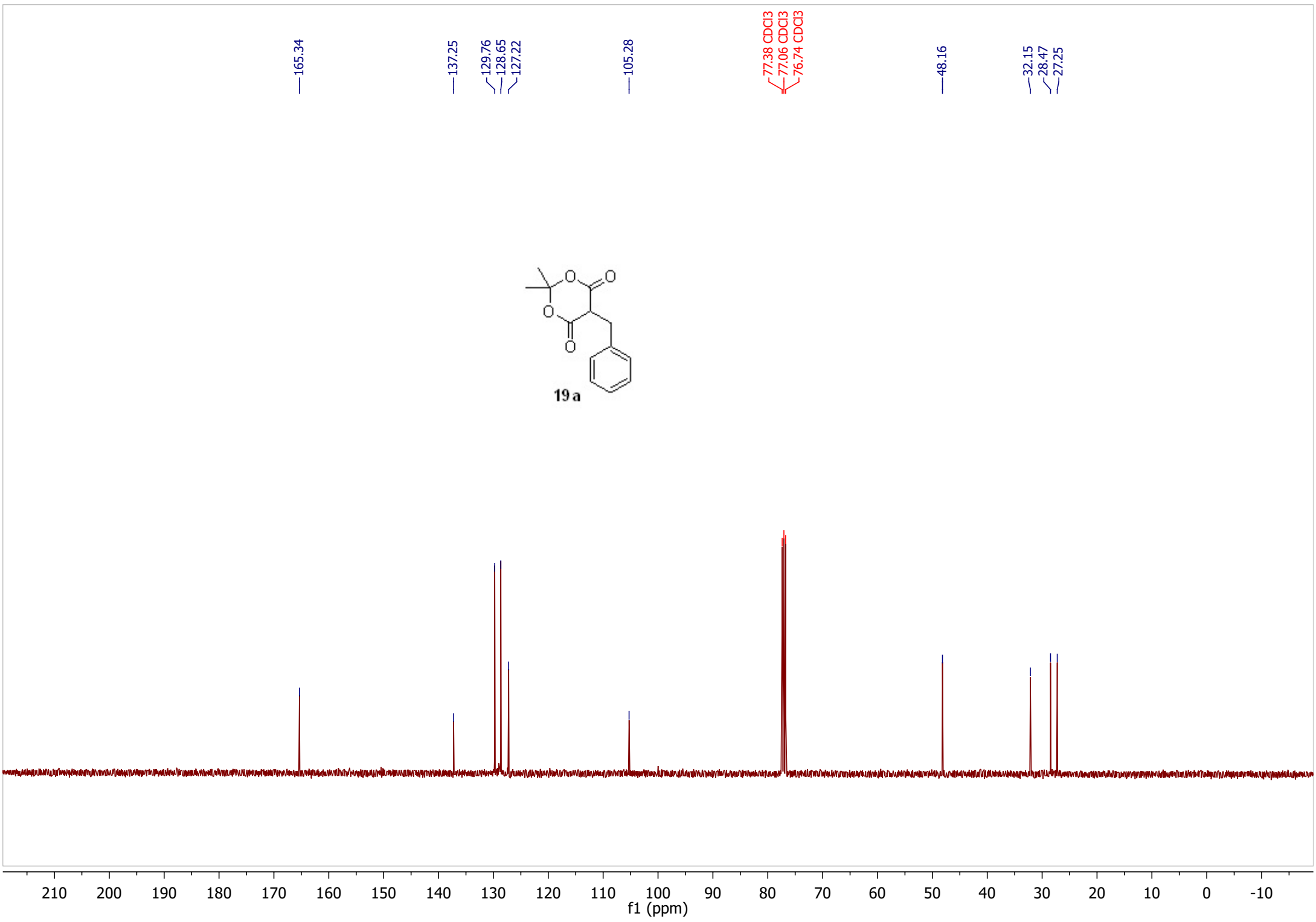
—22.24

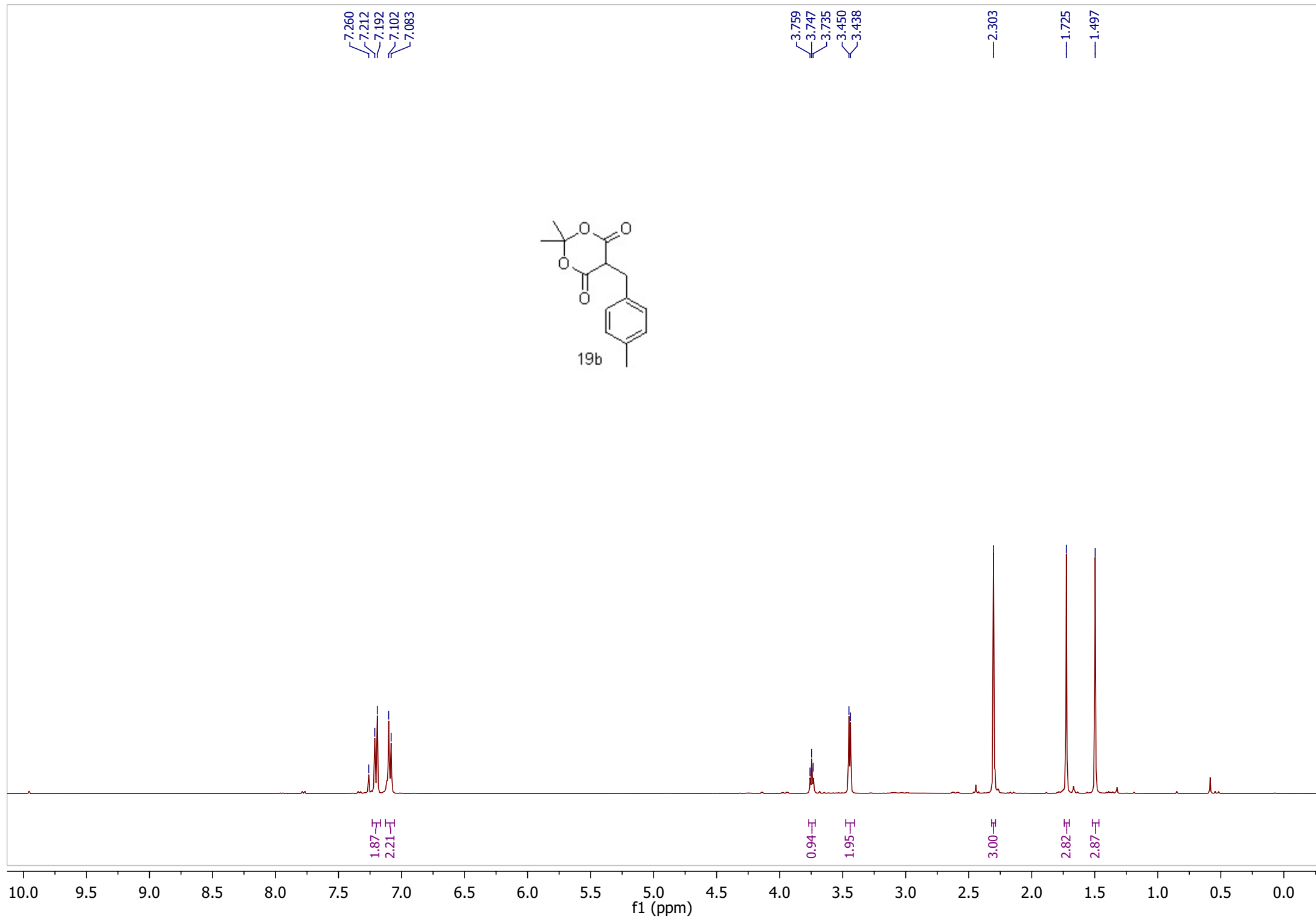
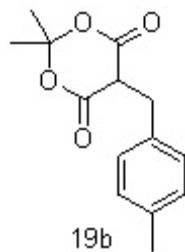


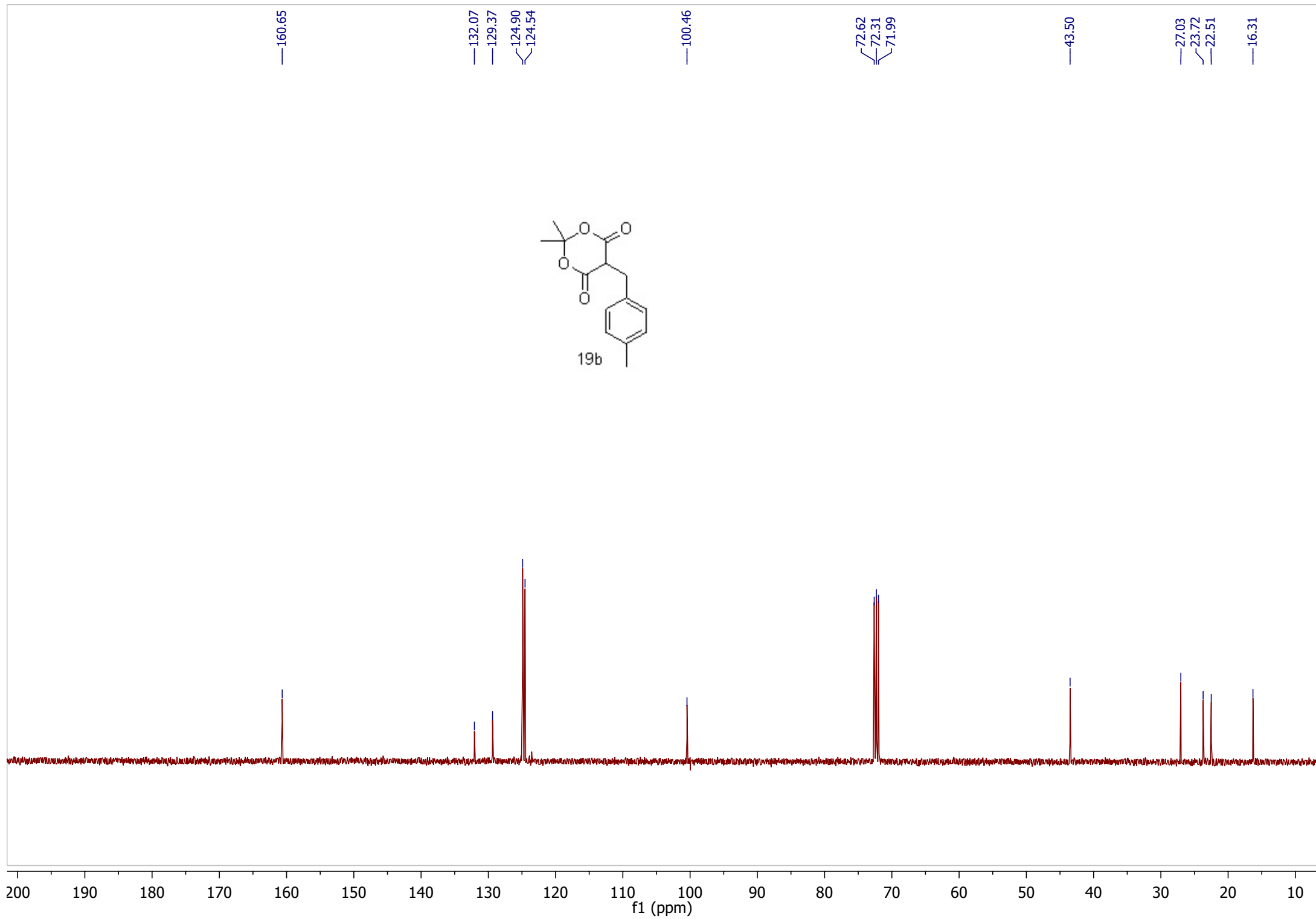
15e

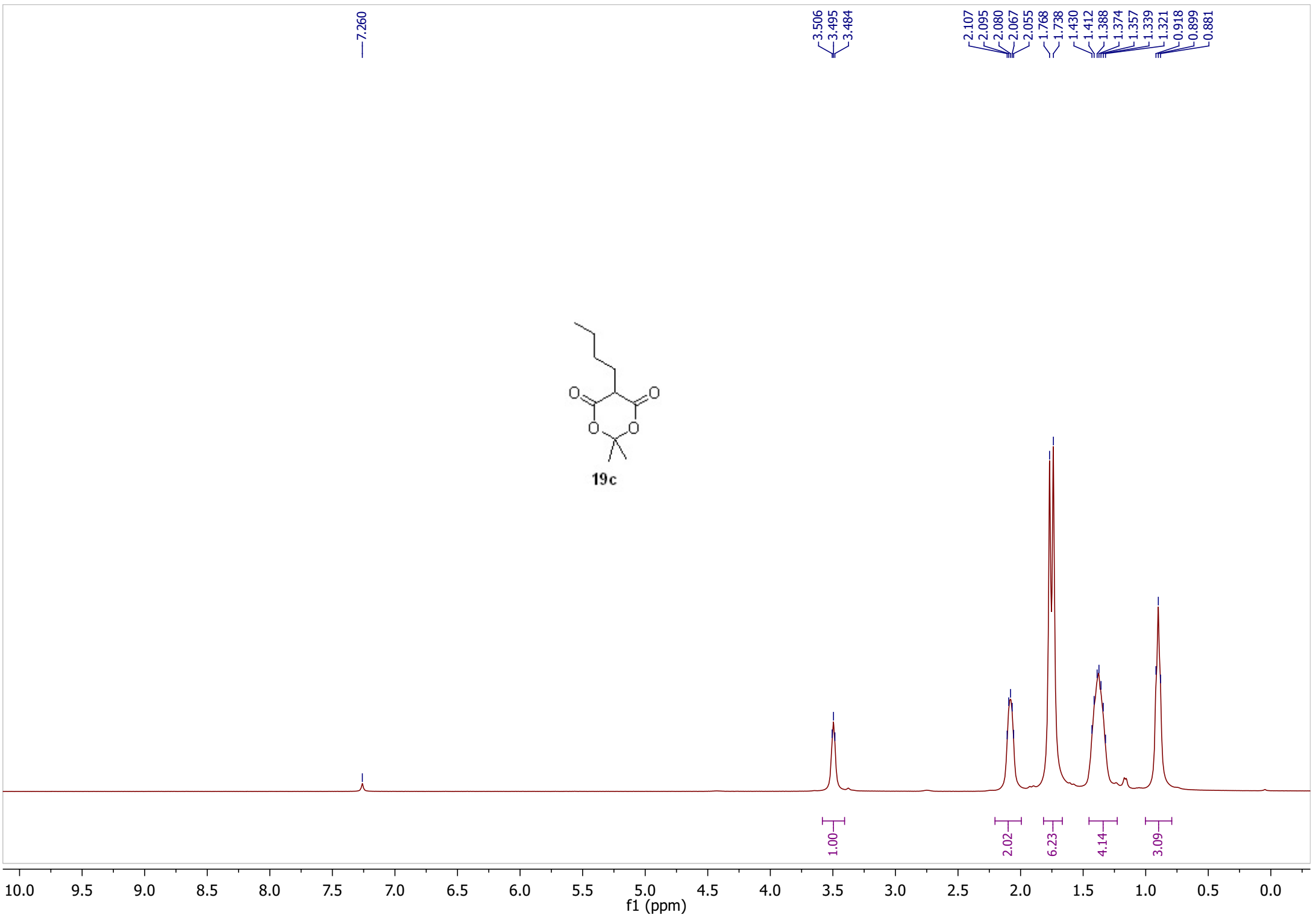
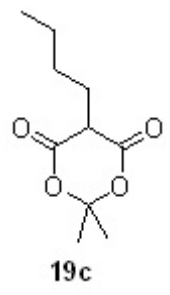


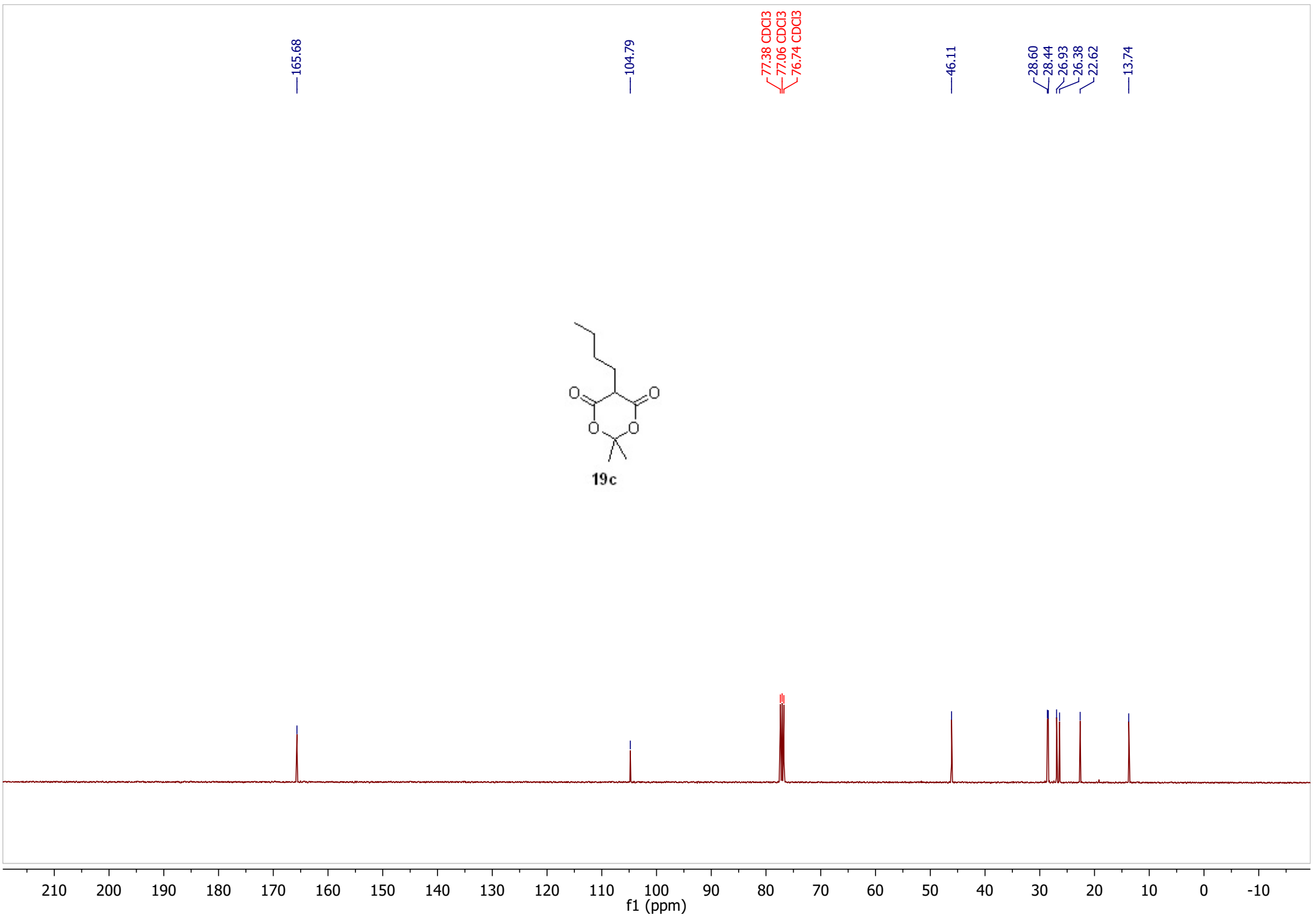
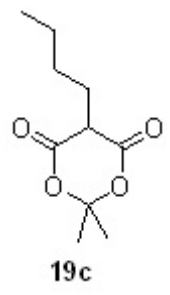




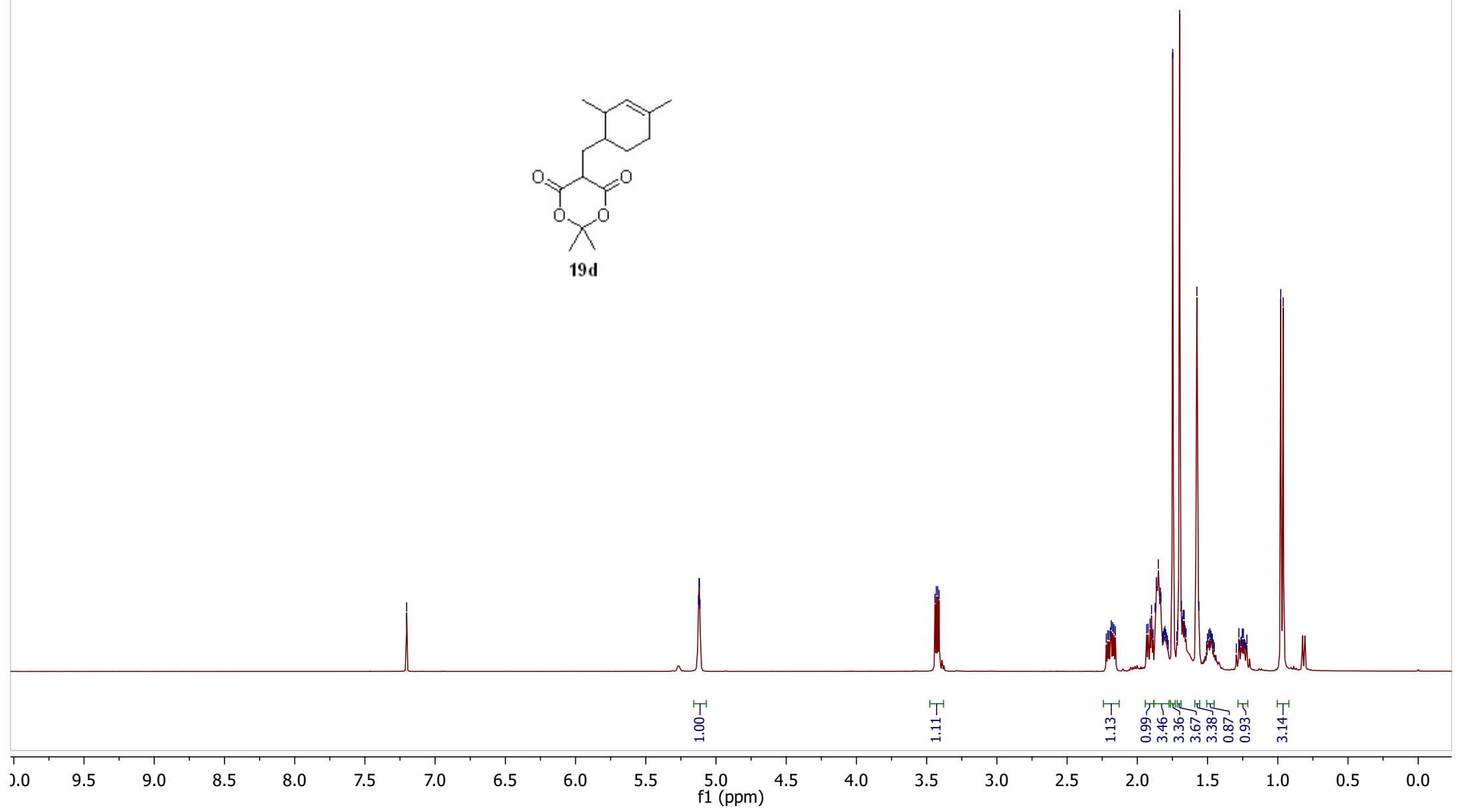
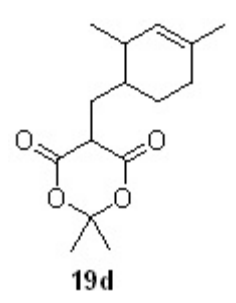




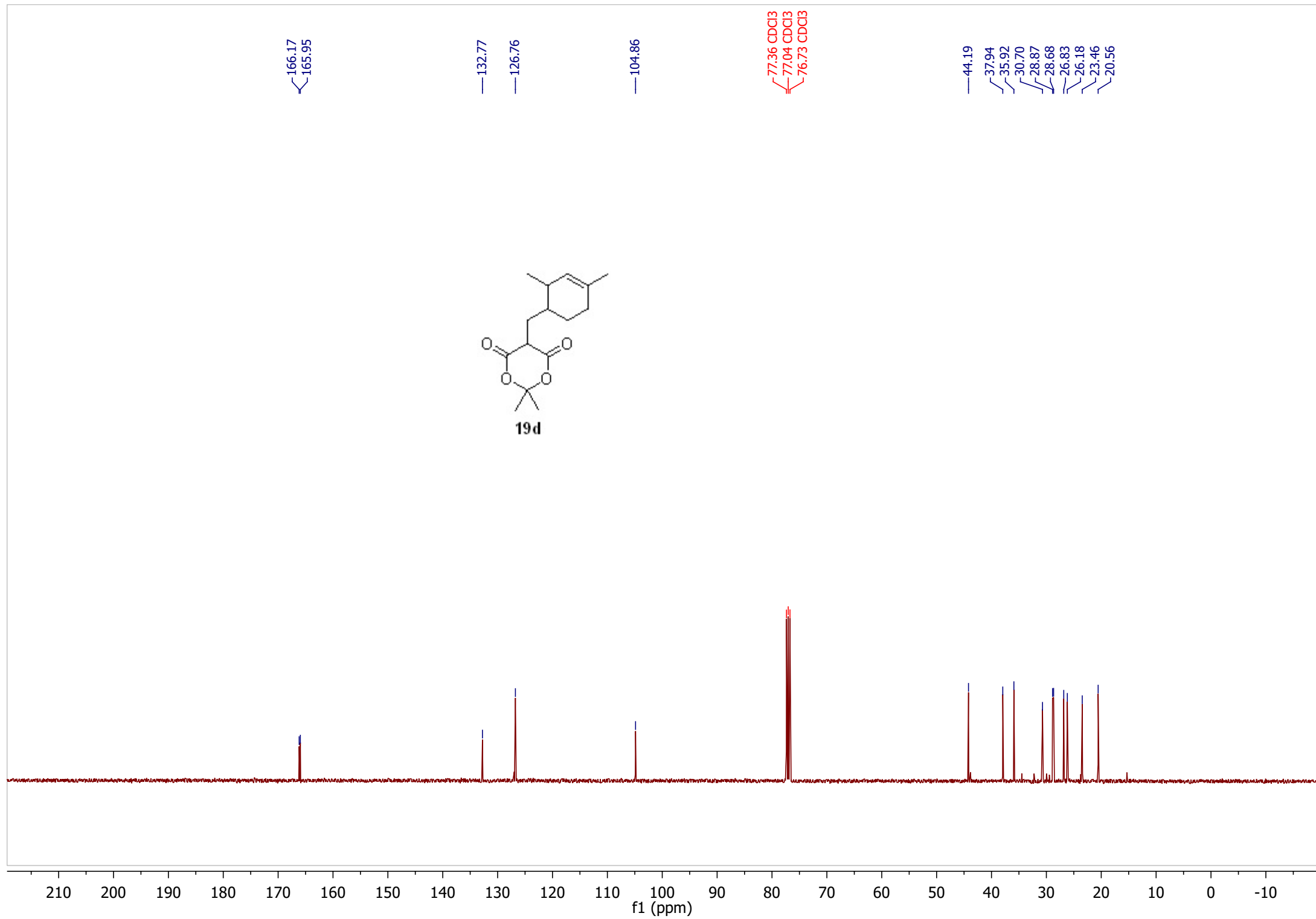
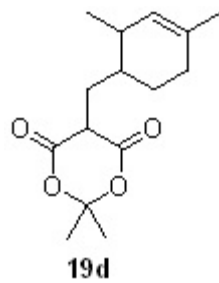


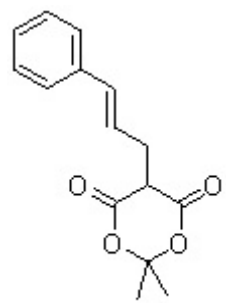


7.203
5.125
5.121
5.118
5.114
3.441
3.431
3.422
3.412
2.221
2.211
2.202
2.192
2.186
2.175
2.166
2.156
2.156
1.933
1.923
1.909
1.899
1.888
1.873
1.865
1.851
1.836
1.832
1.821
1.816
1.810
1.804
1.798
1.792
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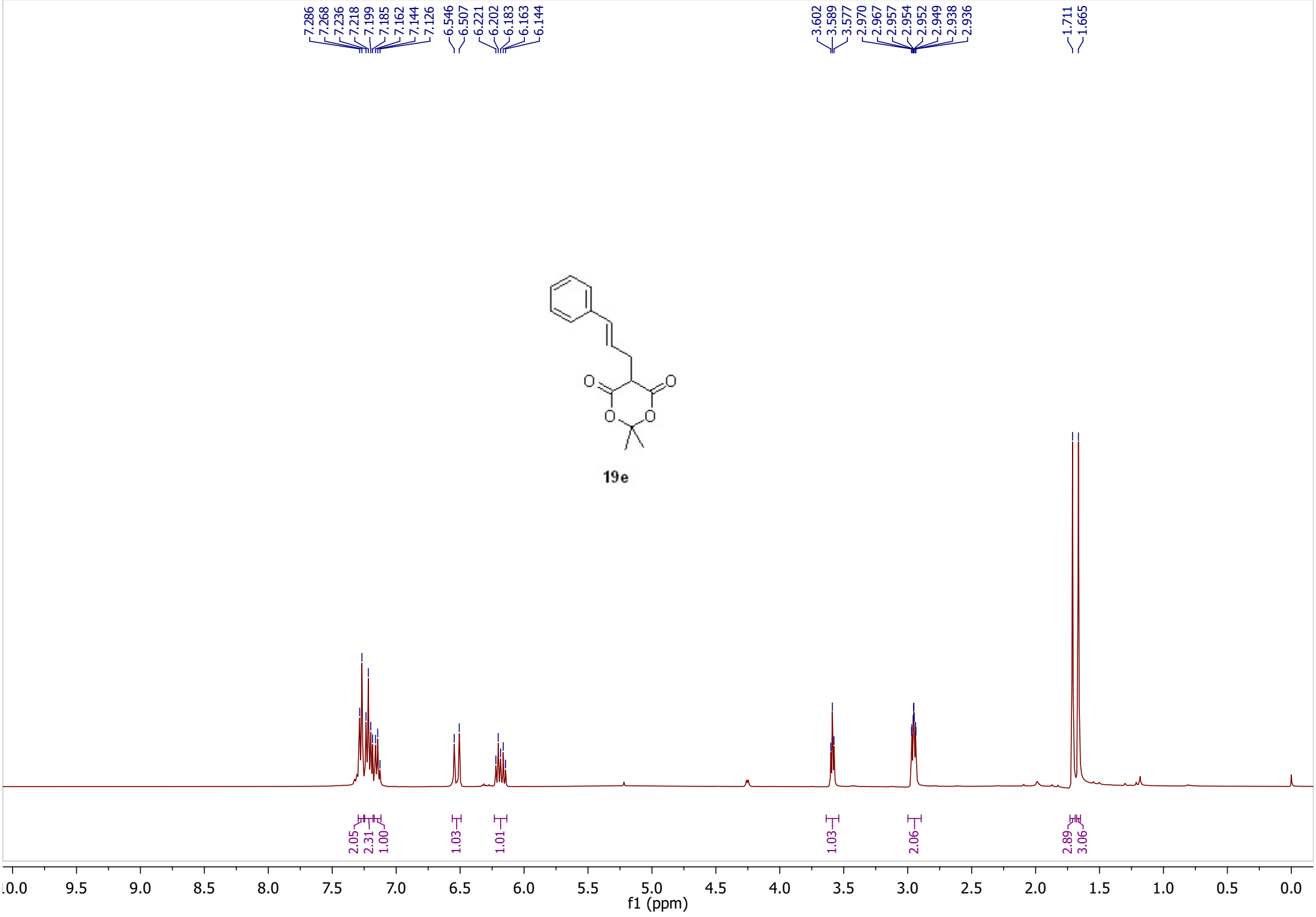


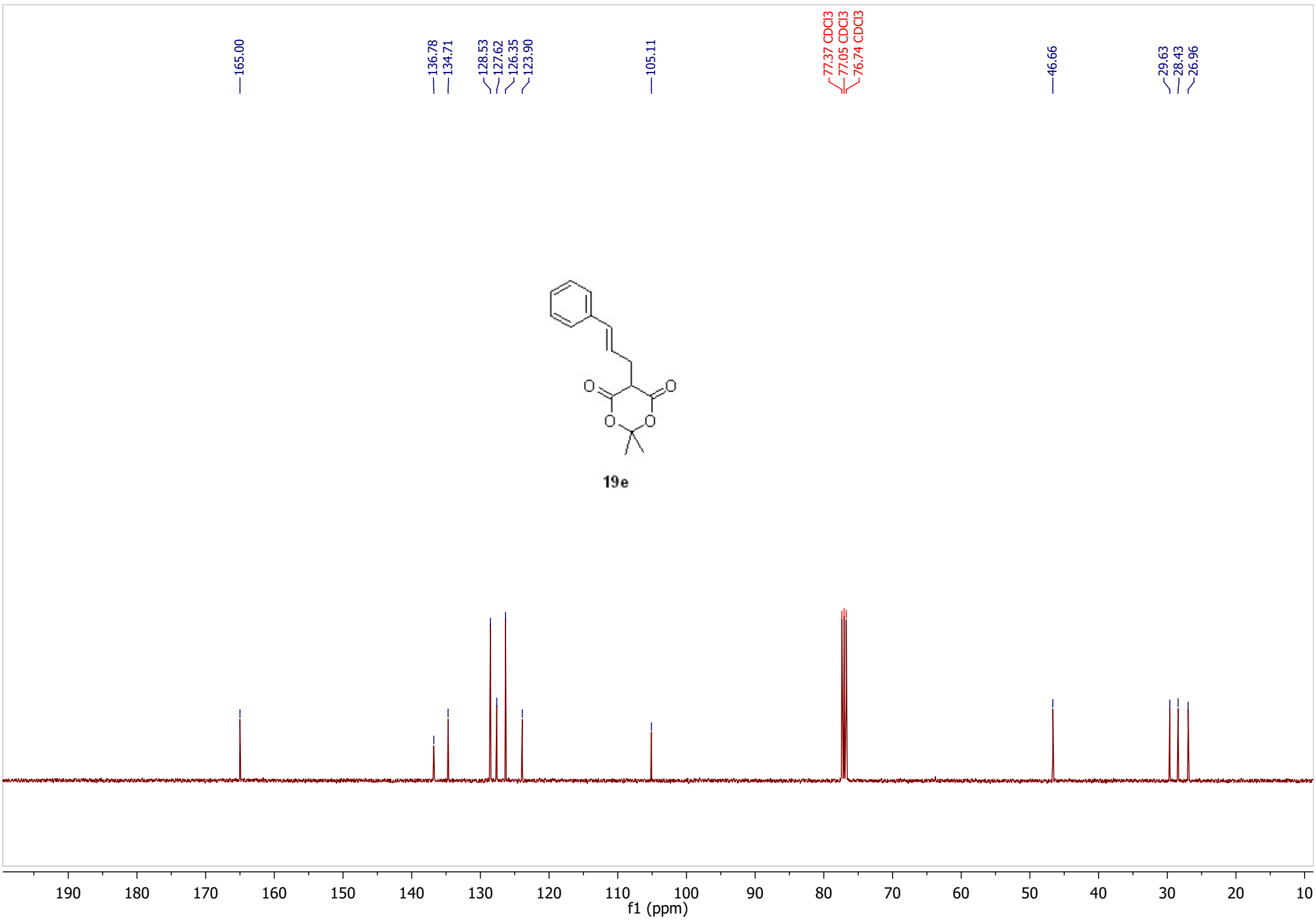
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0

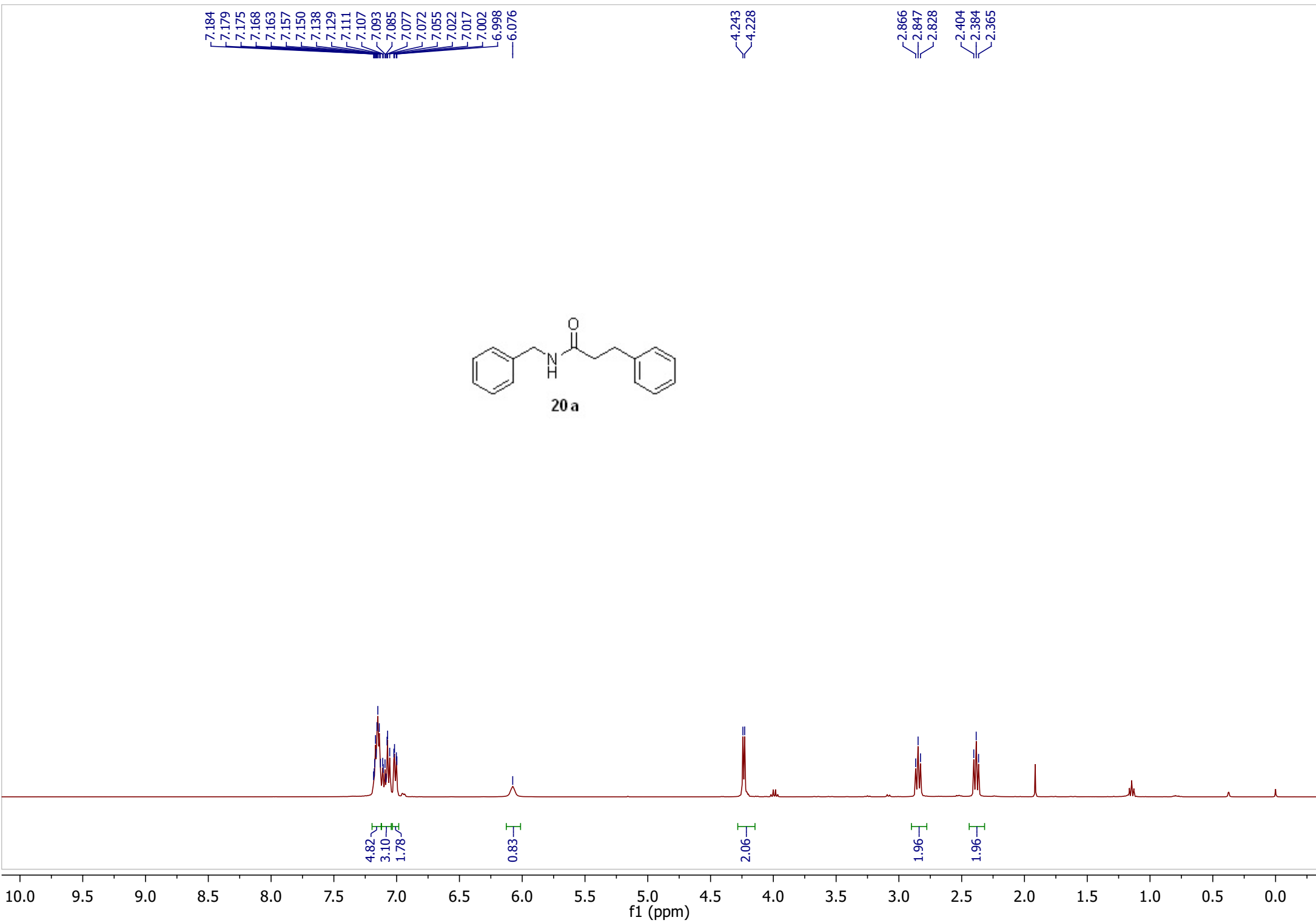


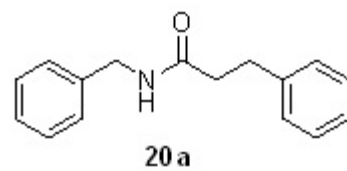


19e









—172.23

—140.80

—138.22

—128.63

—128.56

—128.42

—127.68

—127.39

—126.25

77.50 CDCl₃

77.18 CDCl₃

76.86 CDCl₃

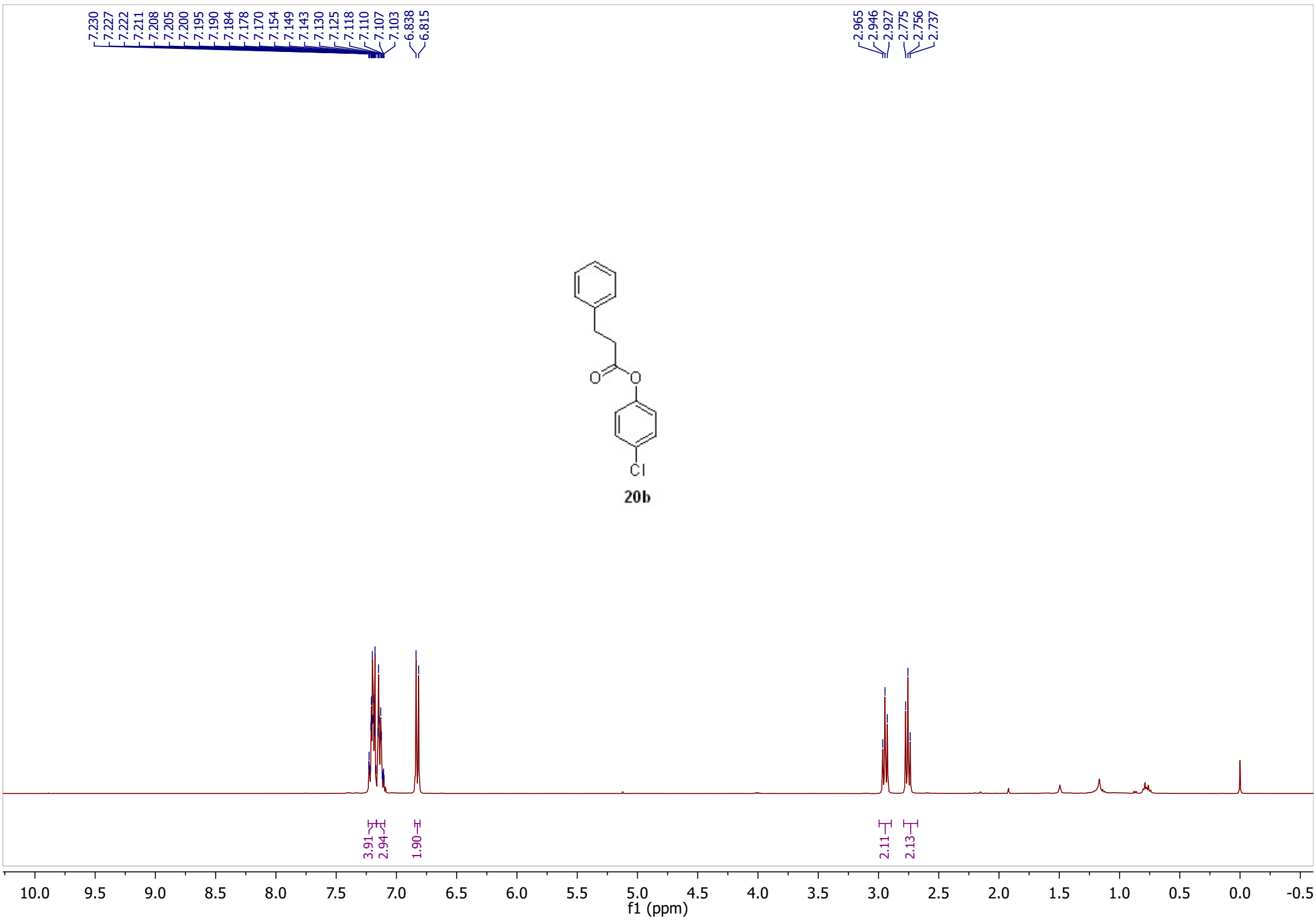
—43.50

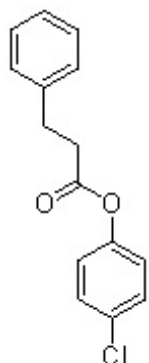
—38.35

—31.75

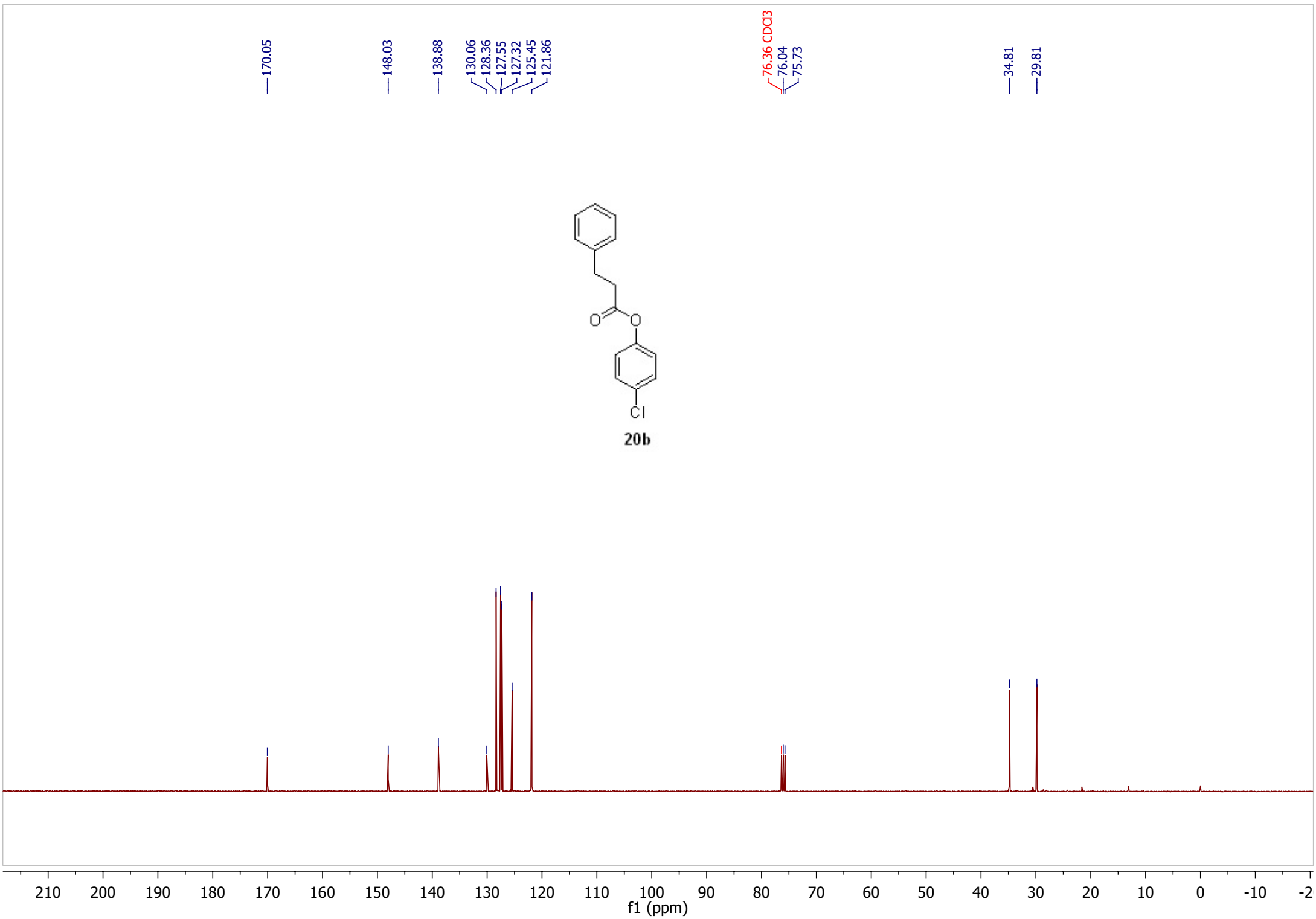
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10

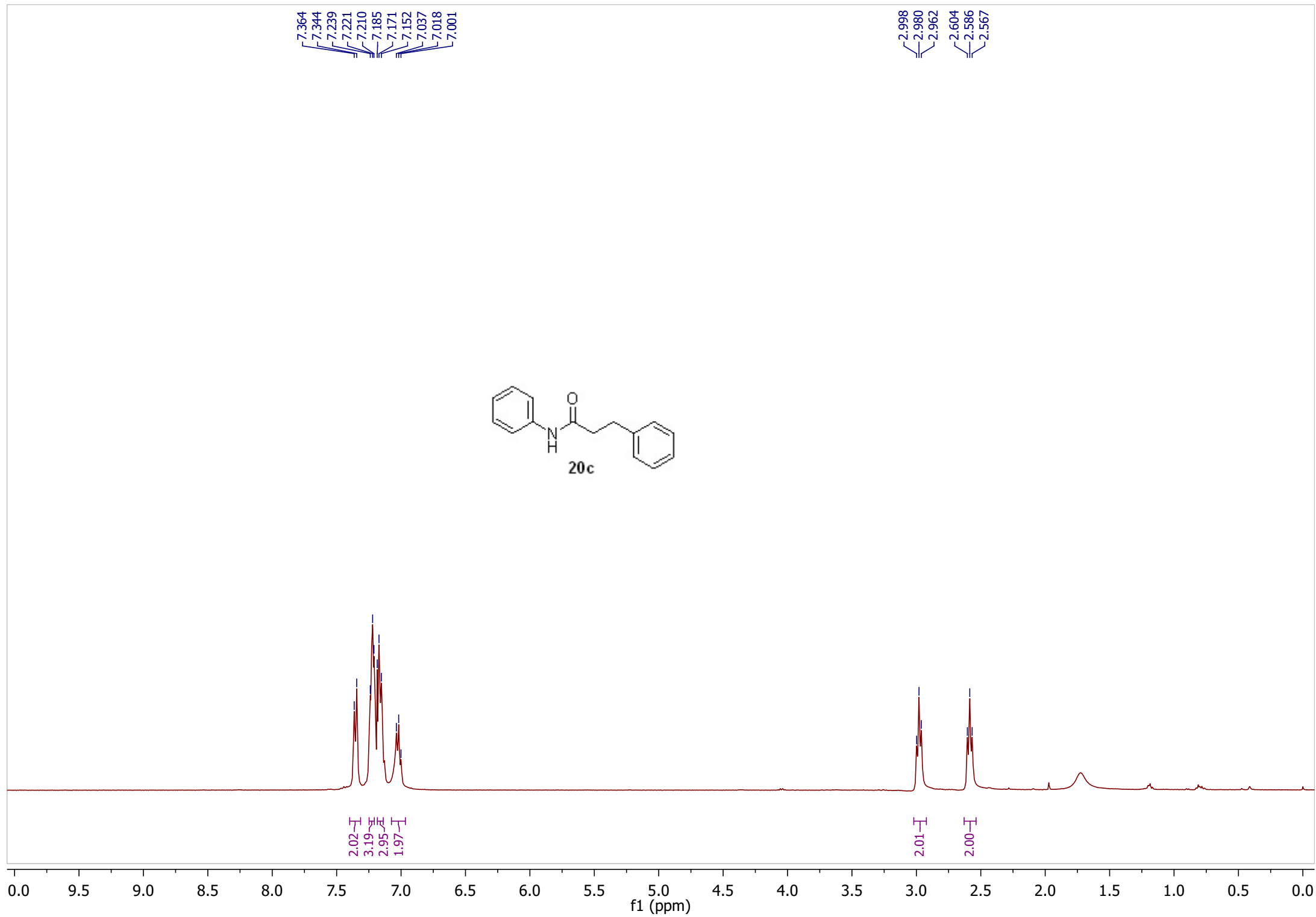
f1 (ppm)

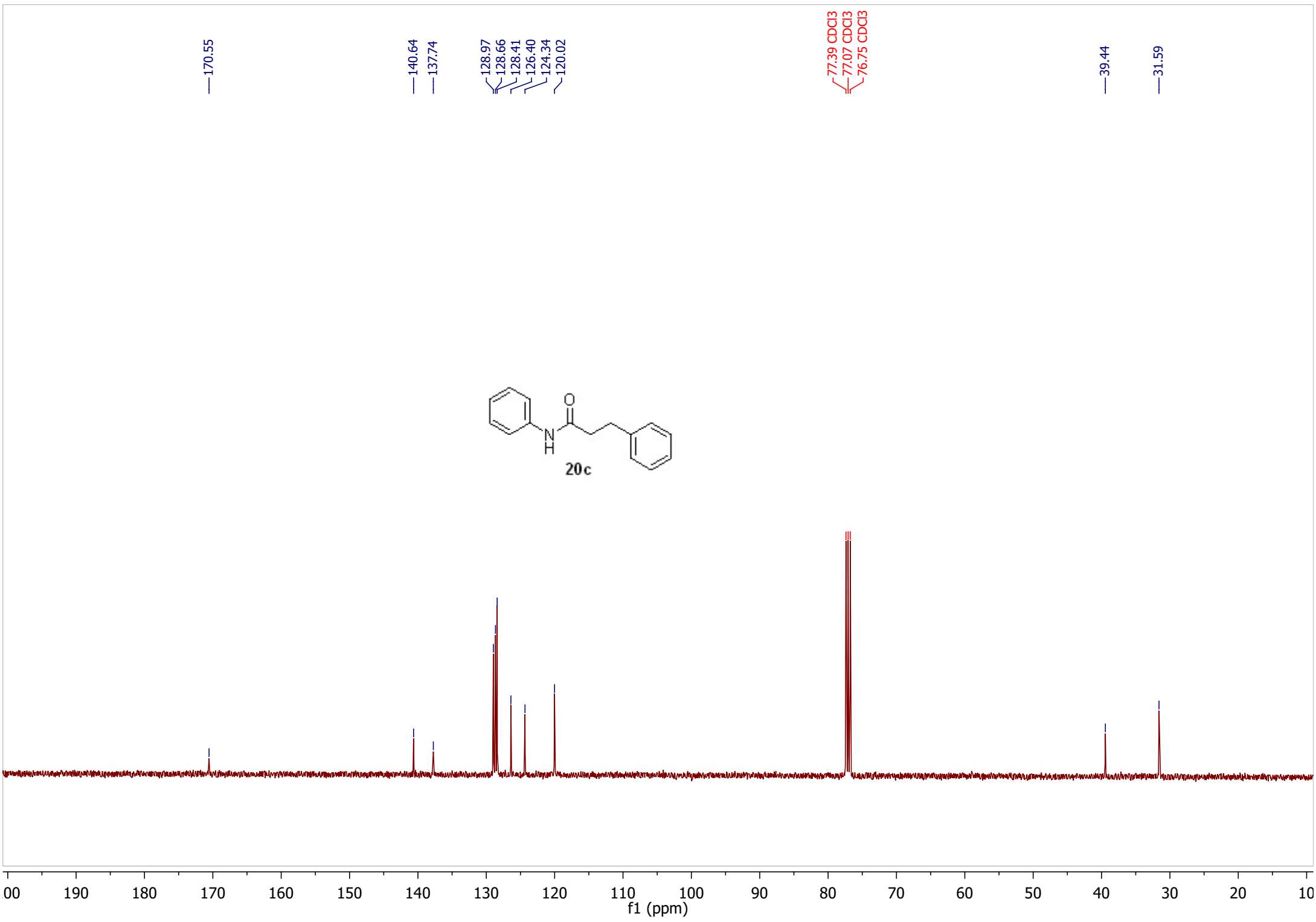


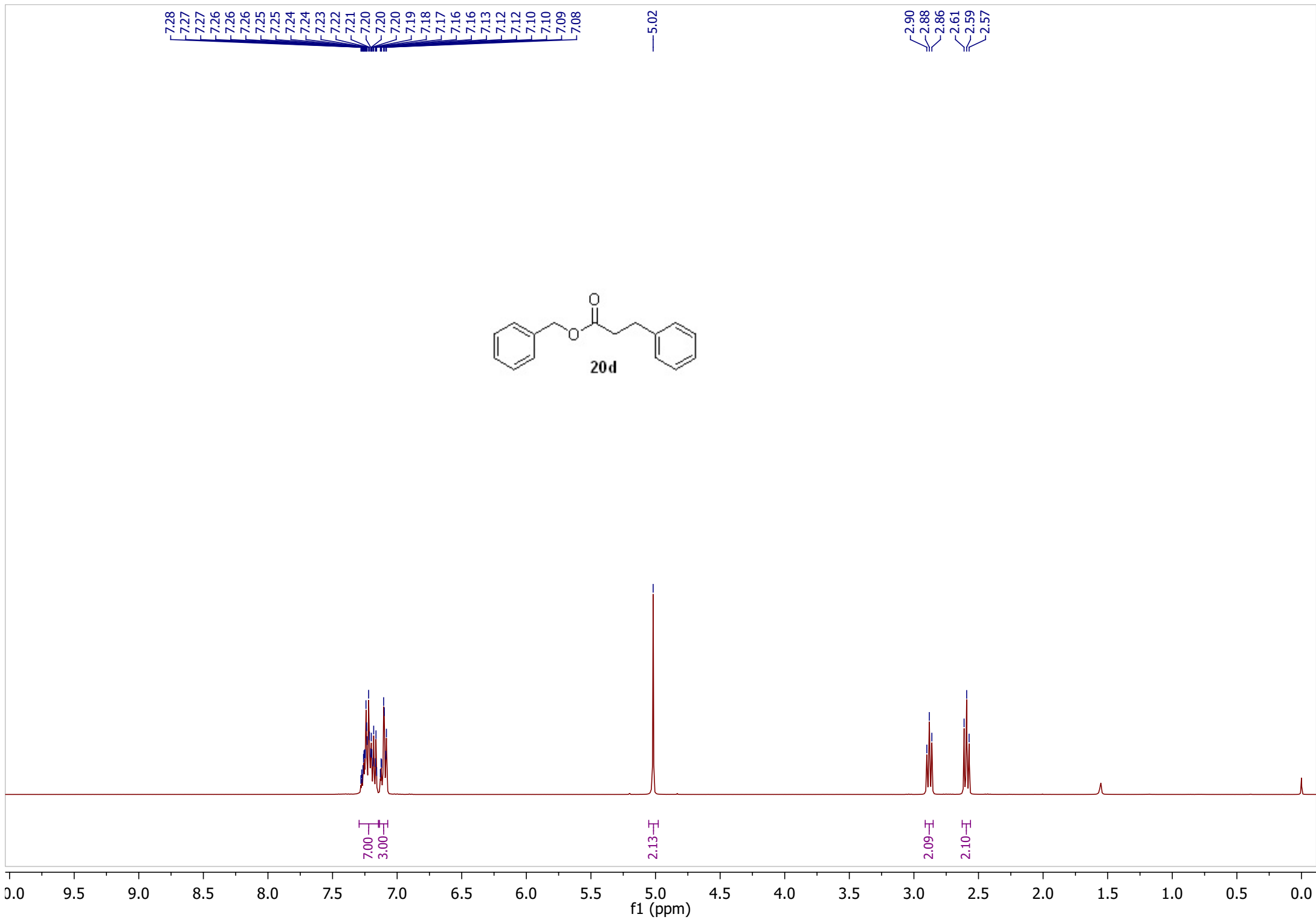


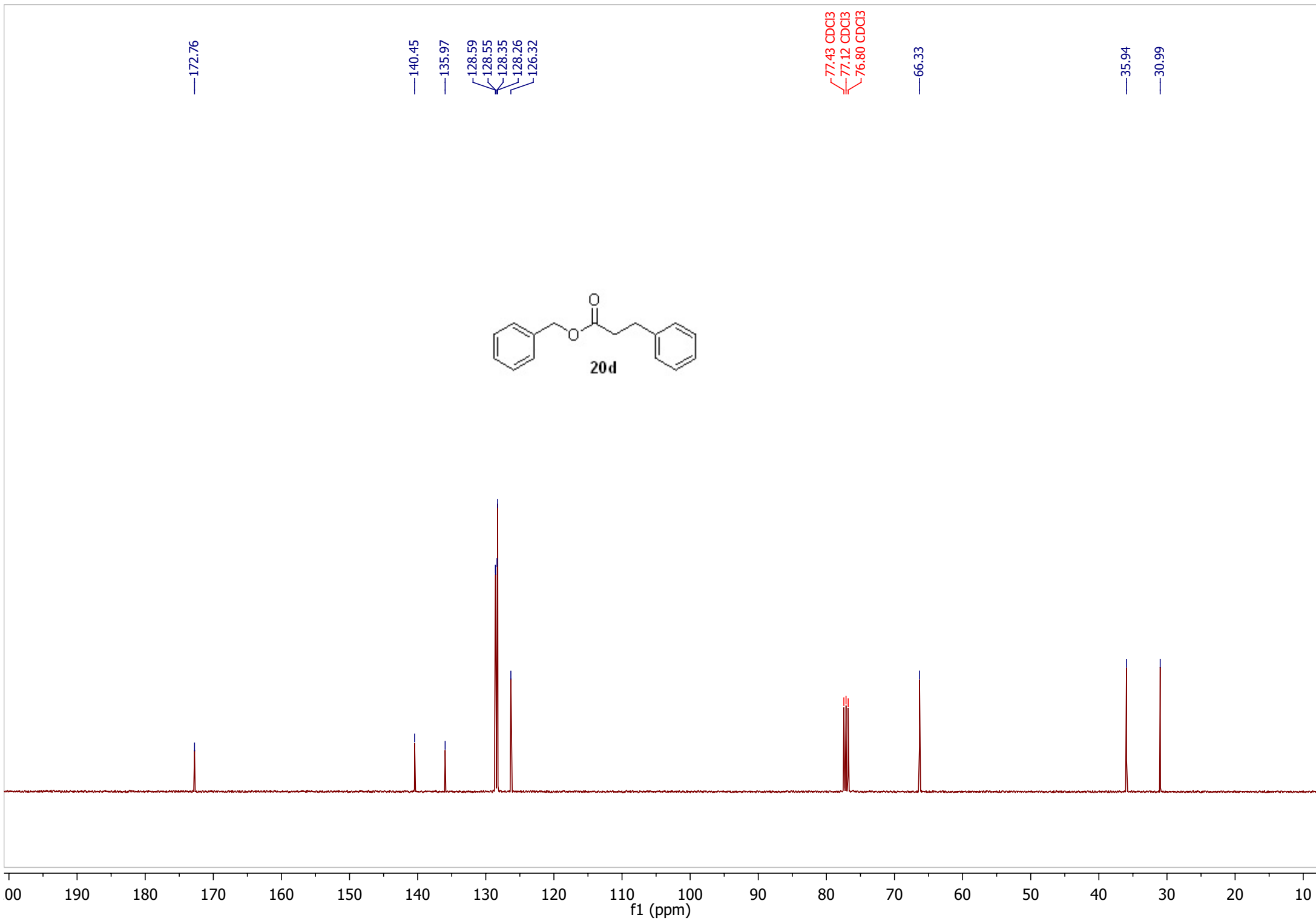
20b

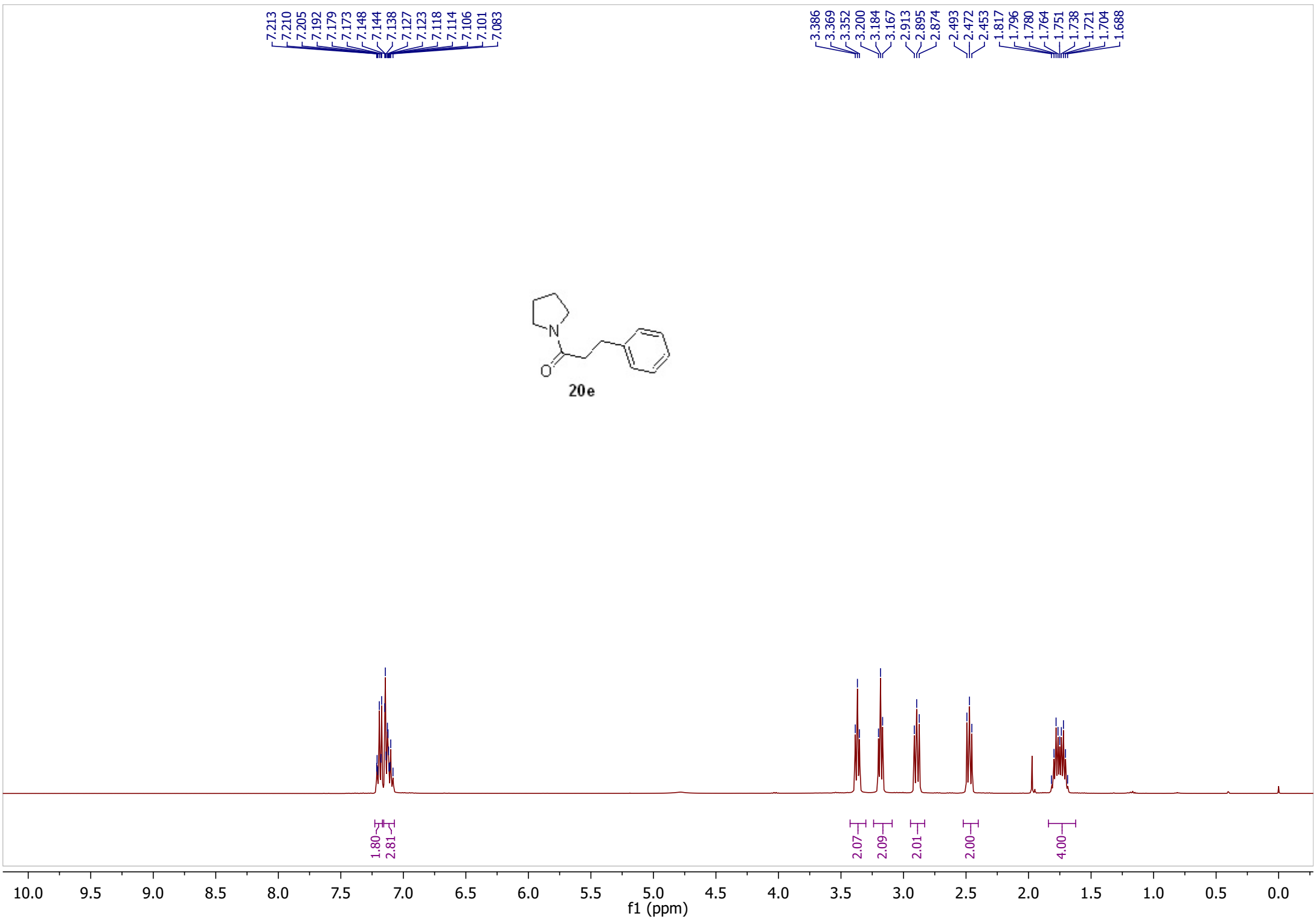


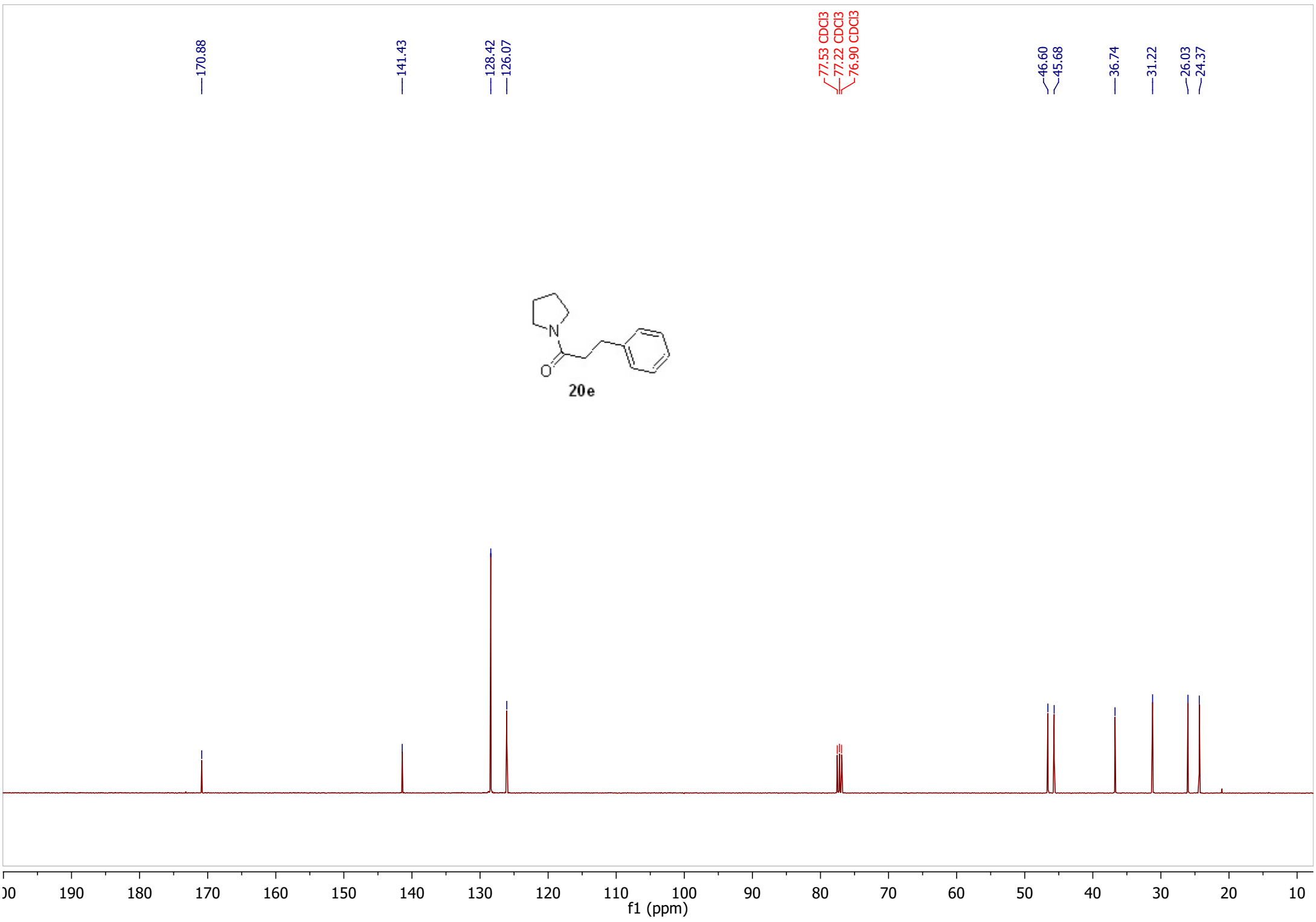


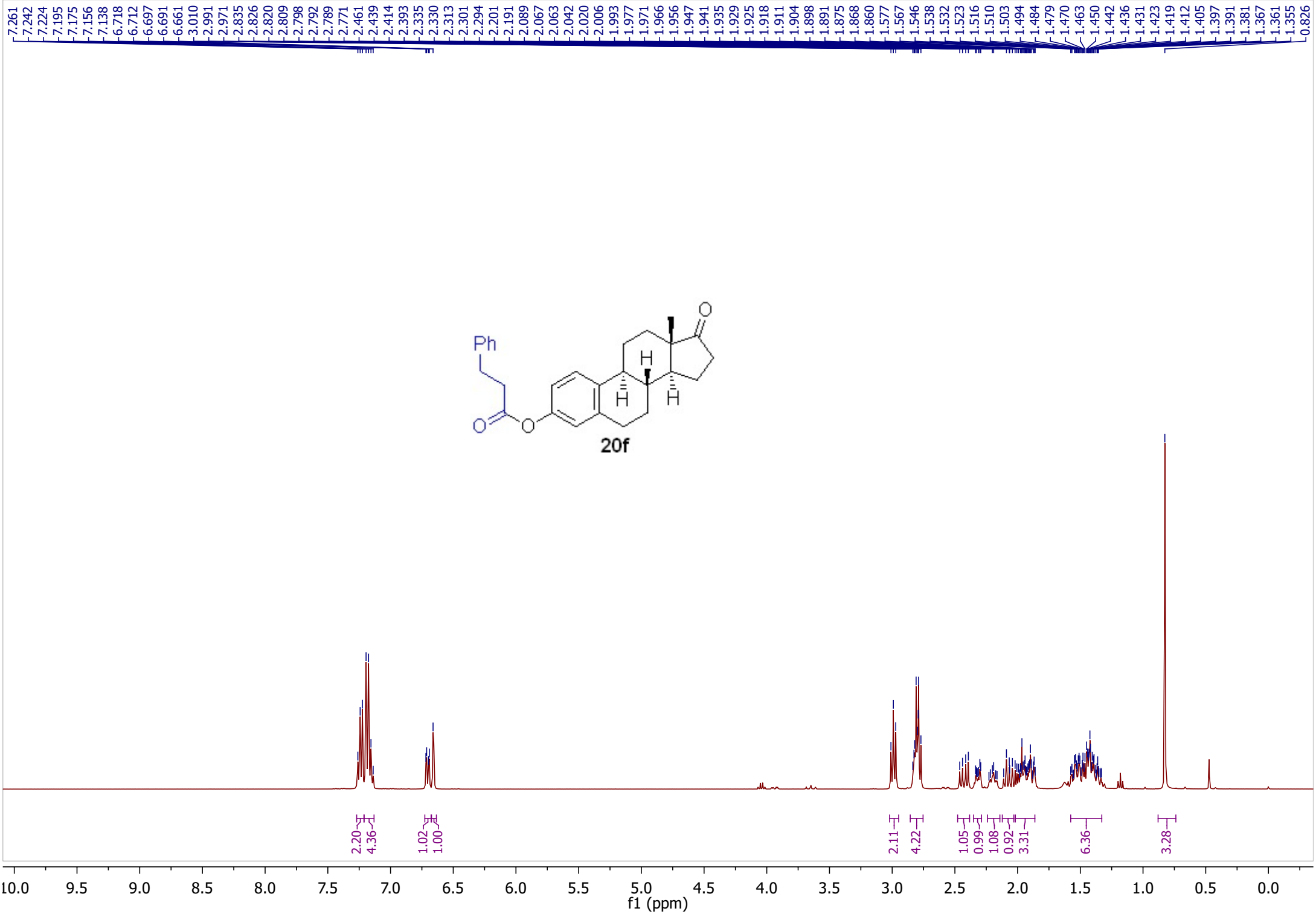


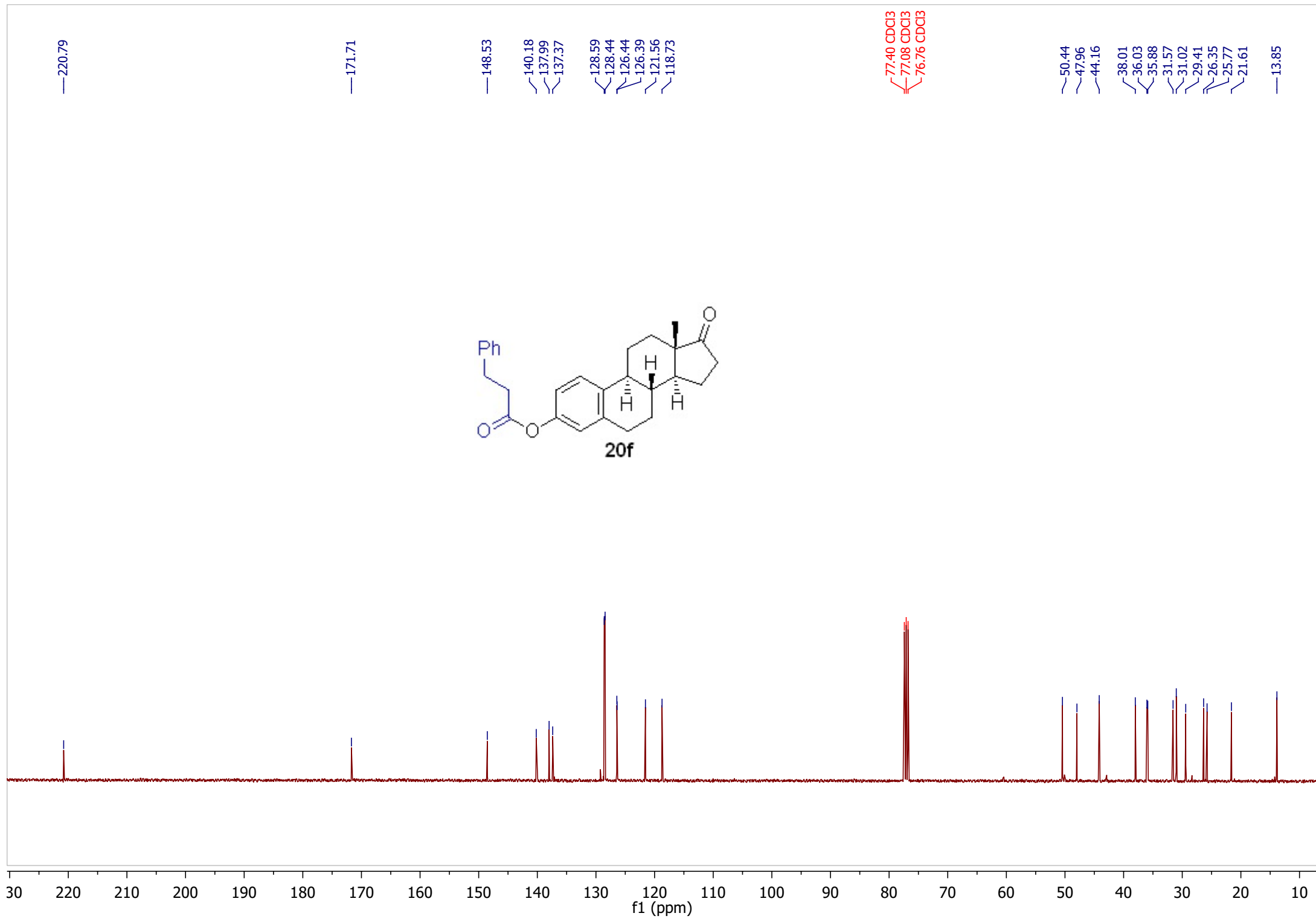


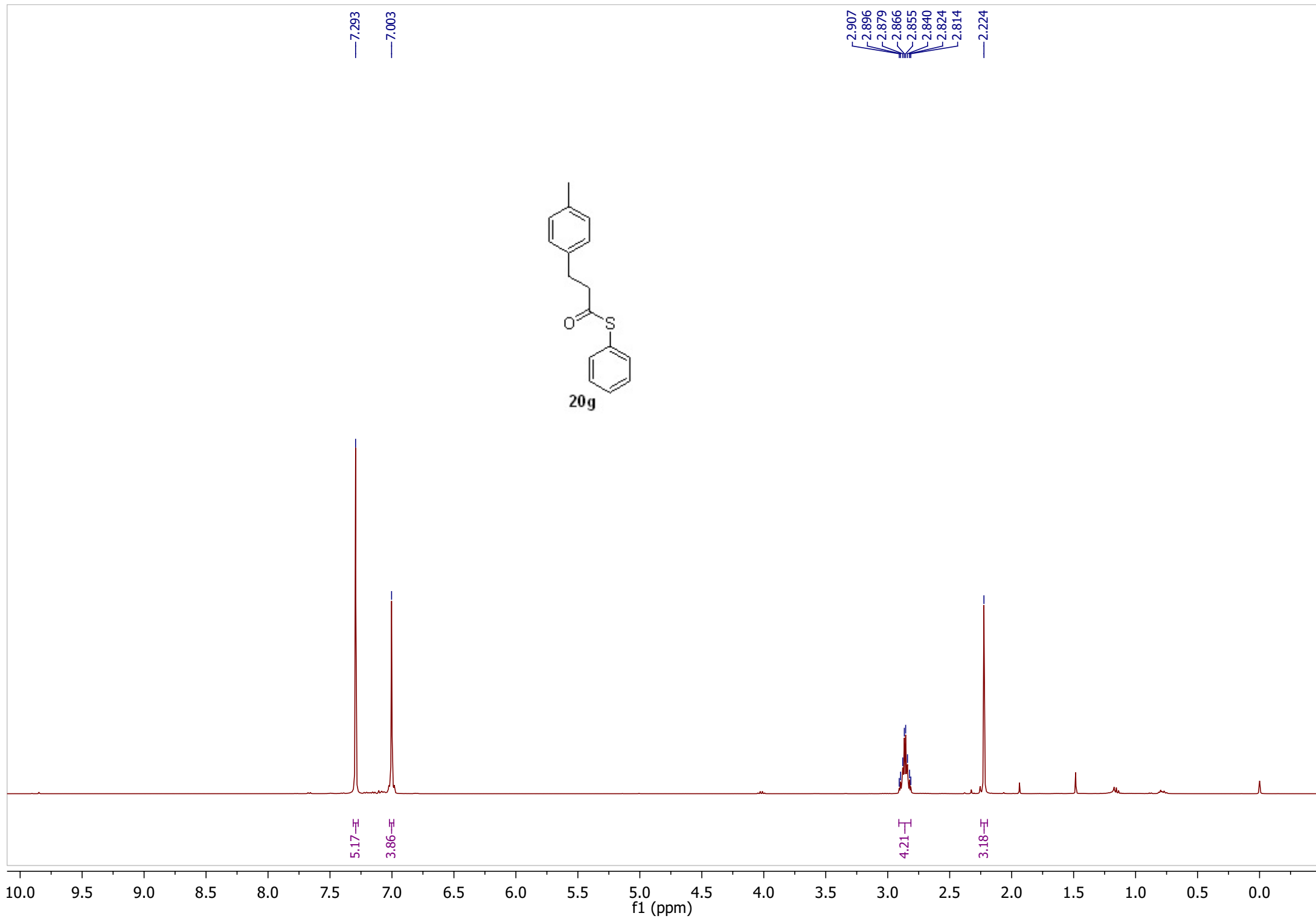
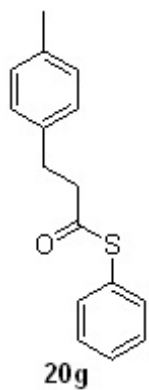












—196.76

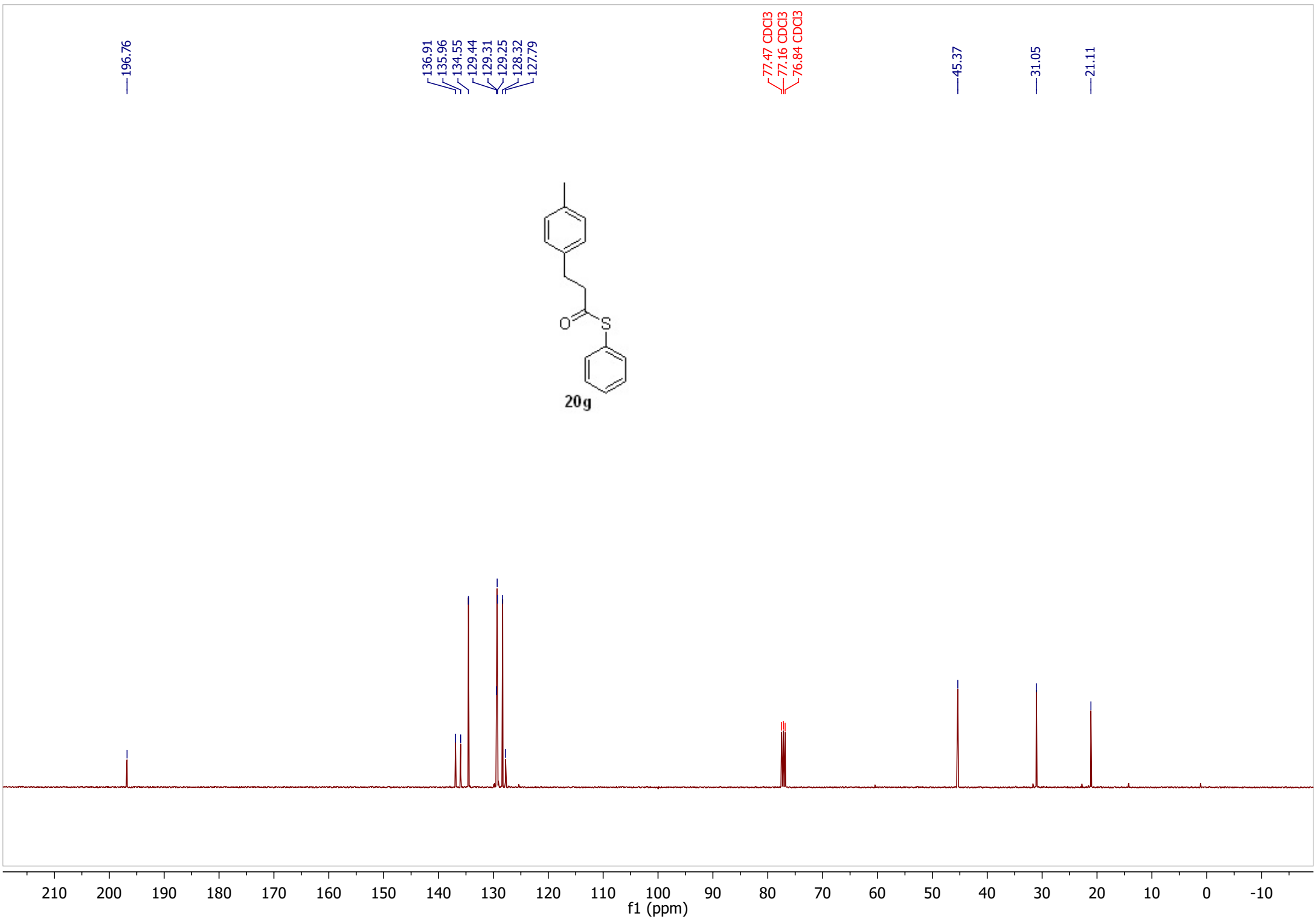
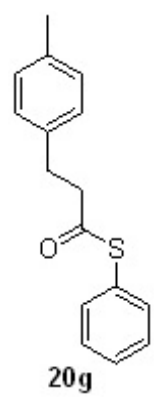
136.91
135.96
134.55
129.44
129.31
129.25
128.32
127.79

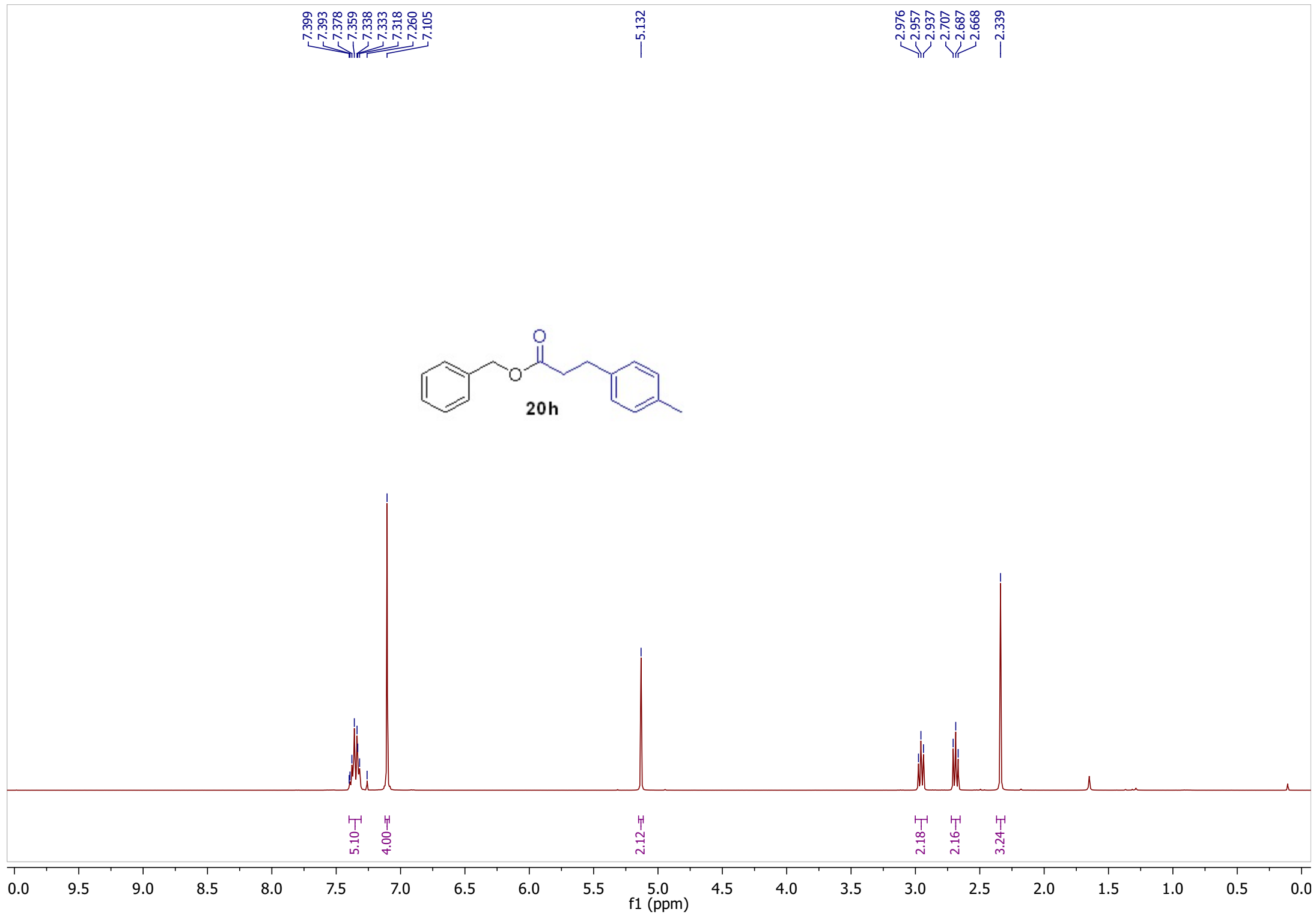
77.47 CDCl3
77.16 CDCl3
76.84 CDCl3

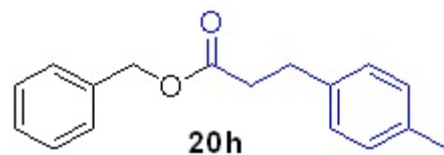
—45.37

—31.05

—21.11







—172.84

137.35
135.97
135.77
129.21
128.56
128.25
128.22
128.21

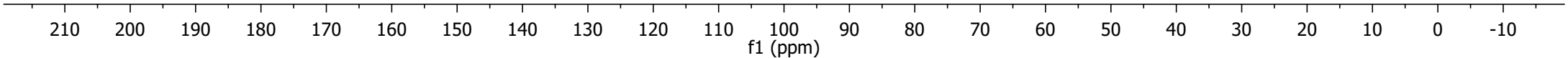
77.40 CDCl3
77.09 CDCl3
76.77 CDCl3

—66.29

—36.09

—30.57

—21.06

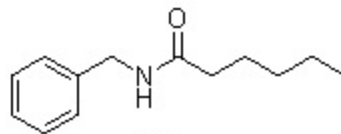


7.279
7.275
7.271
7.260
7.256
7.253
7.249
7.239
7.235
7.219
7.213
7.206
7.202
7.196
7.193
7.185
7.181
7.176

5.838

4.359
4.345

2.148
2.129
2.110
1.616
1.604
1.597
1.591
1.578
1.570
1.560
1.551
1.541
1.251
1.245
1.242
1.233
1.223
1.219
0.833
0.816
0.799



20i

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)

1.90
3.01

0.88

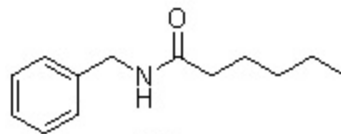
1.98

2.01

2.01

4.13

3.00



20i

—173.13

—138.40

128.69

127.81

127.48

77.38 CDCl₃

77.06 CDCl₃

76.74 CDCl₃

—43.58

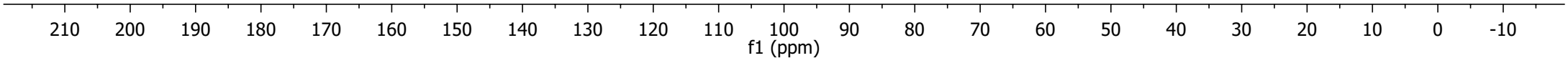
—36.75

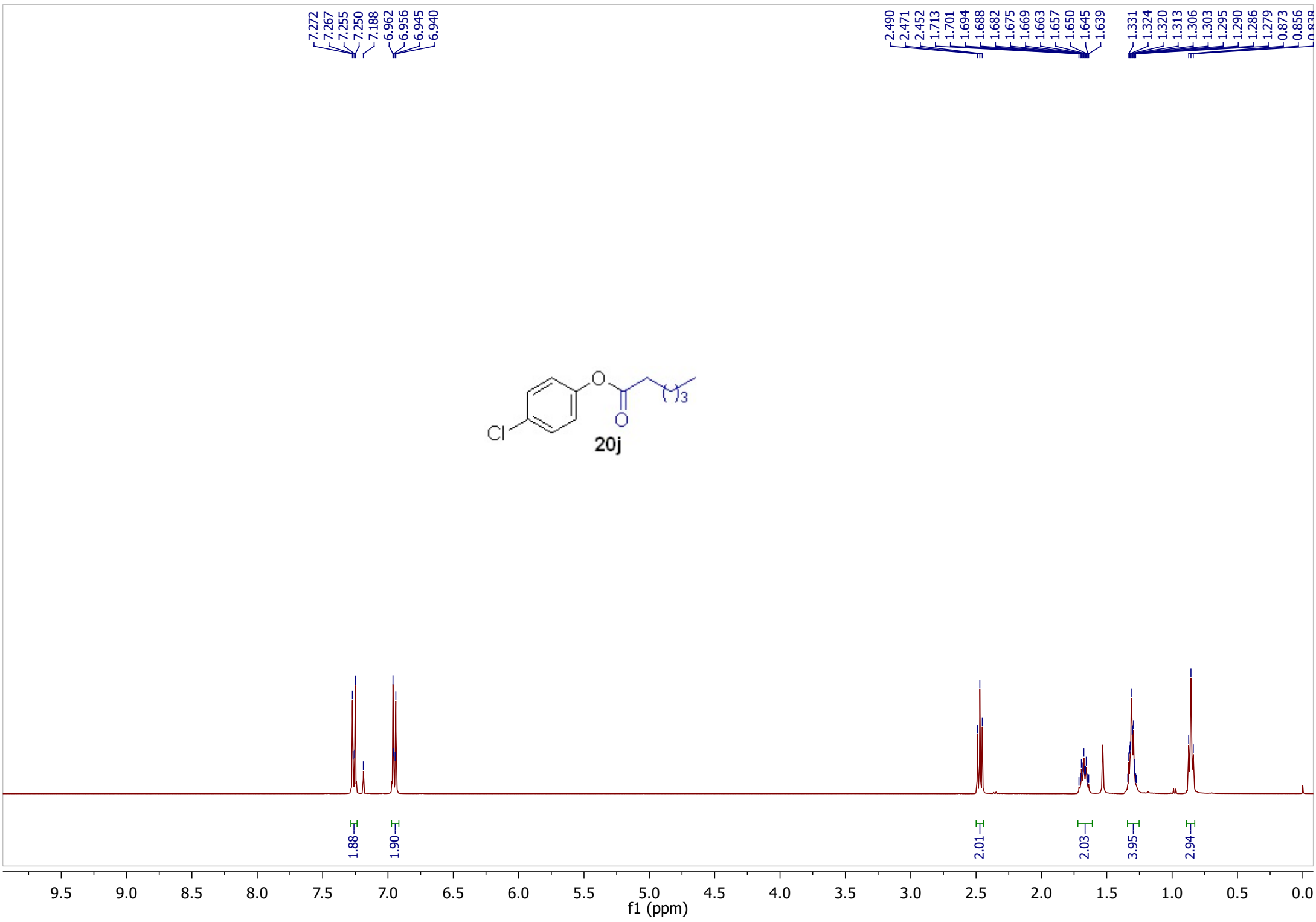
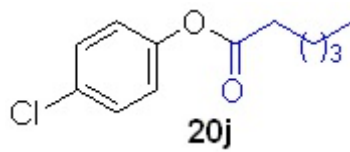
—31.49

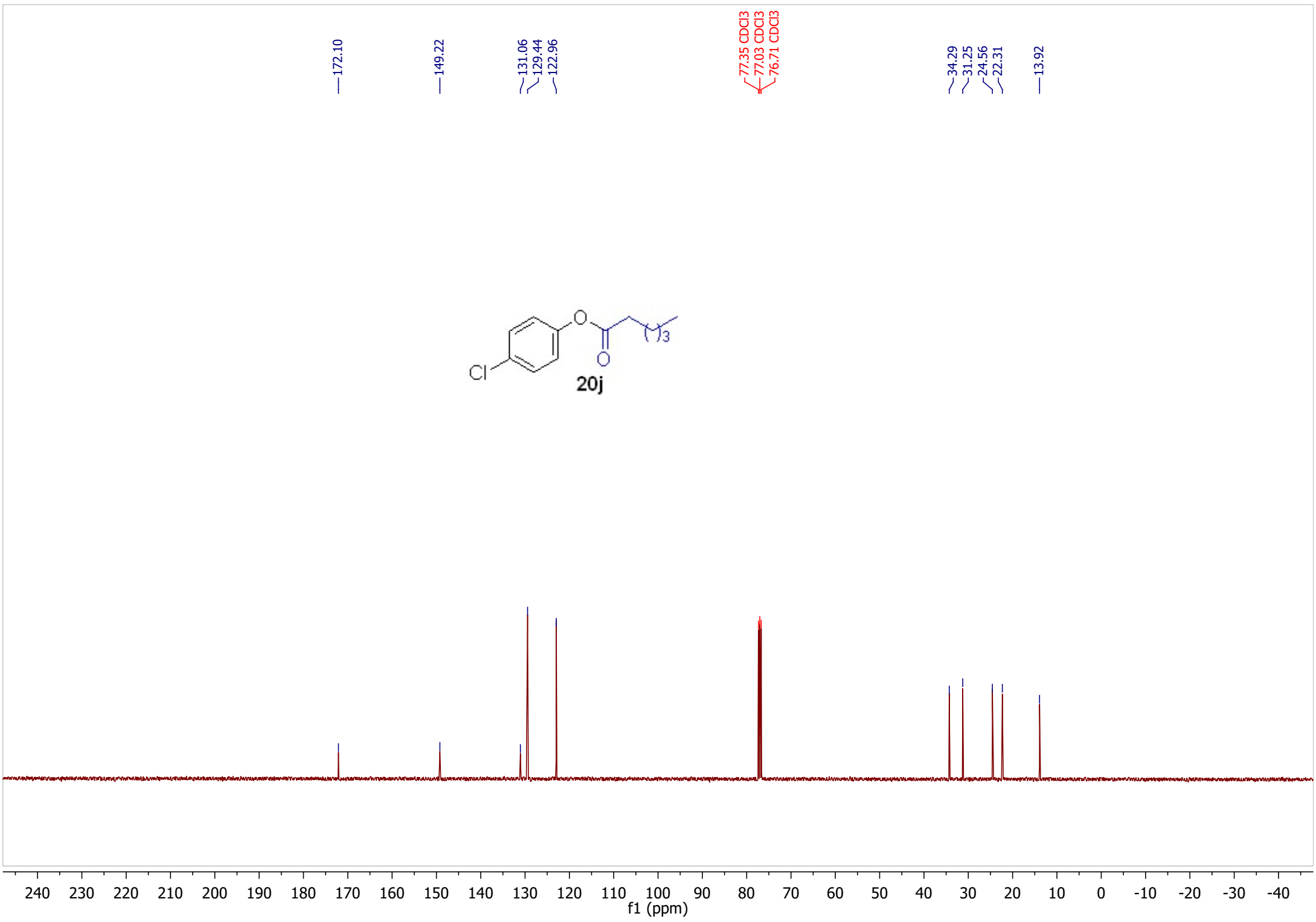
—25.48

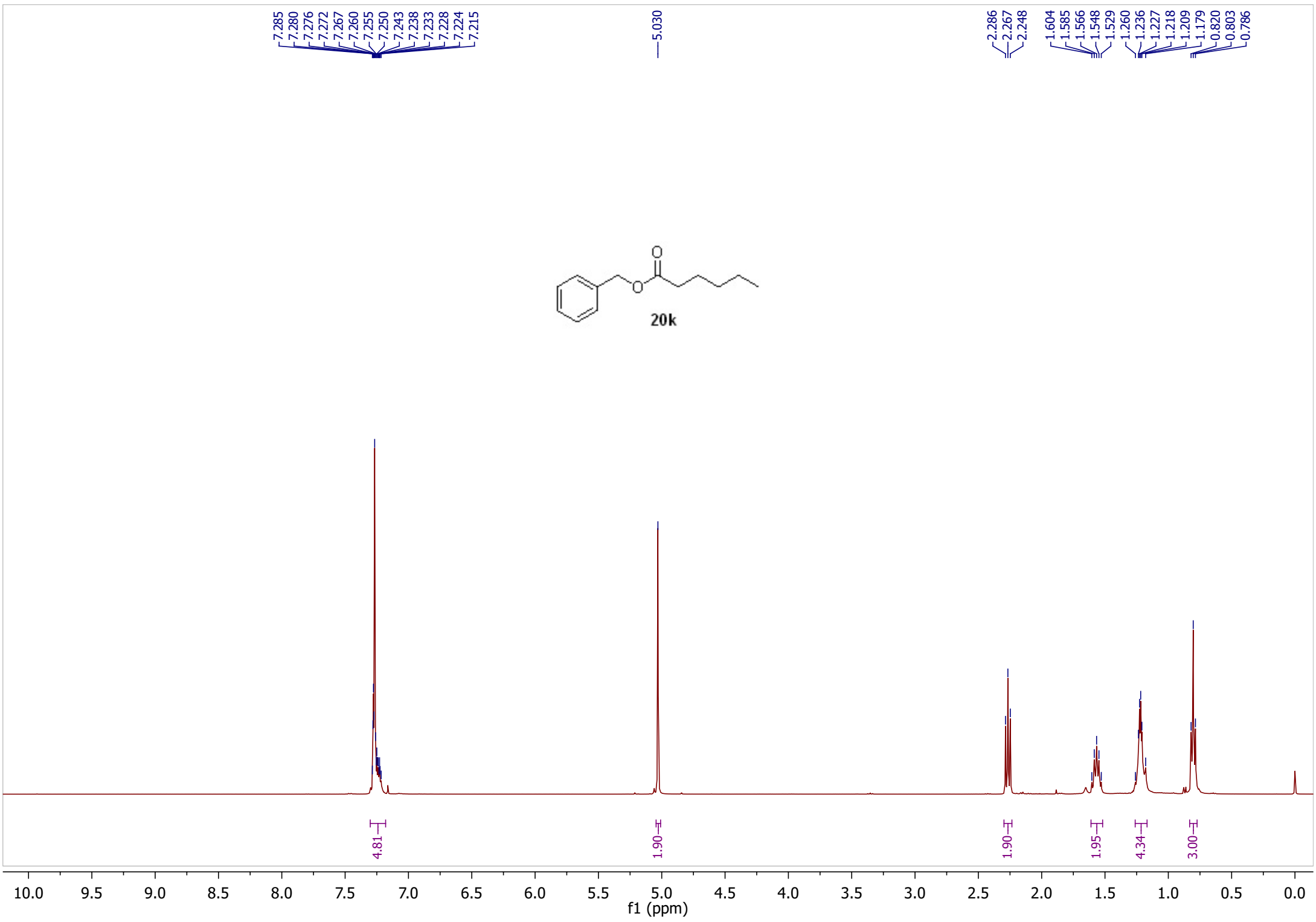
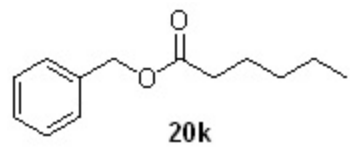
—22.41

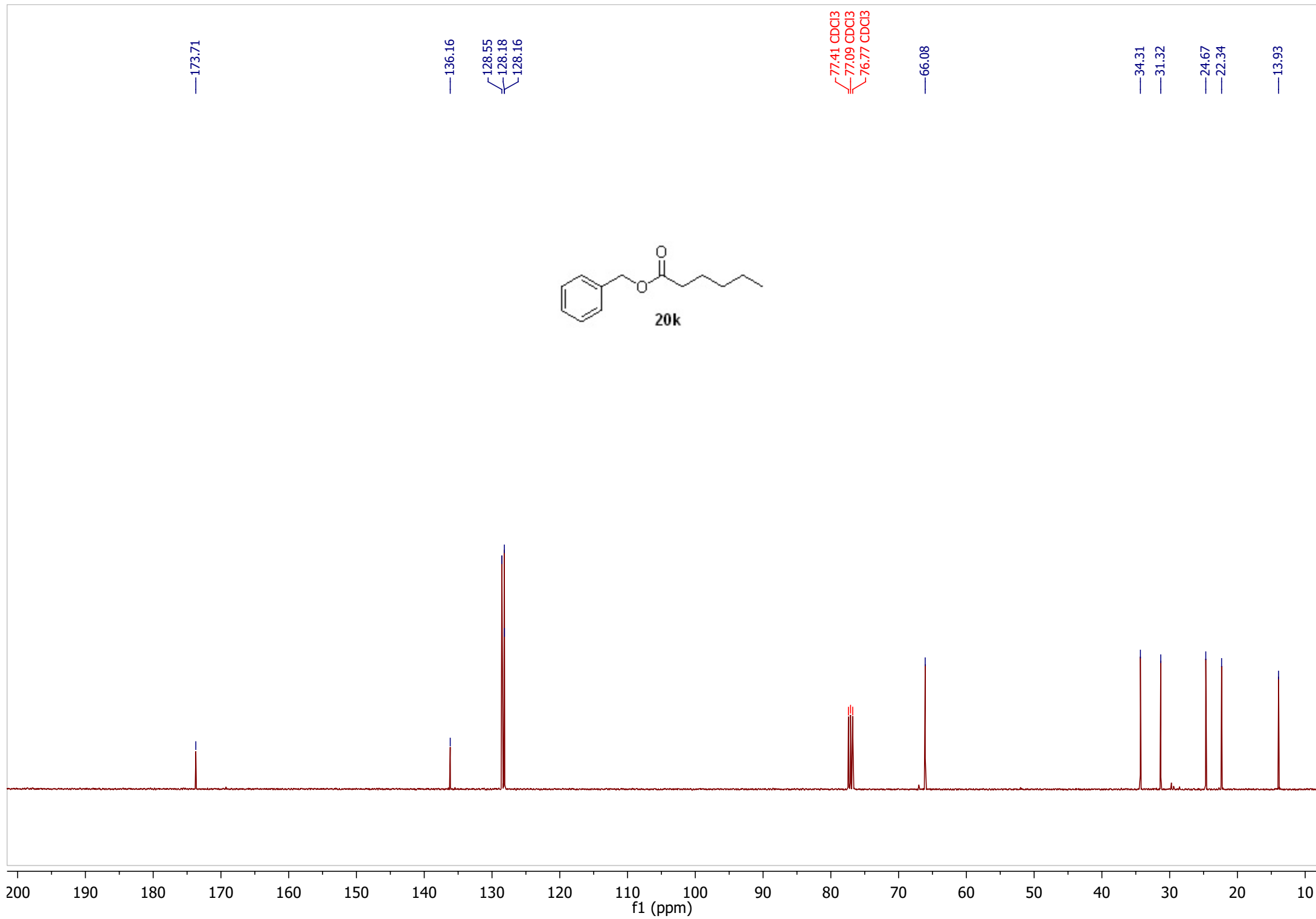
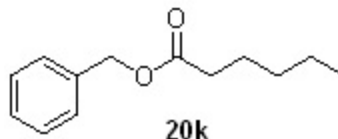
—13.96

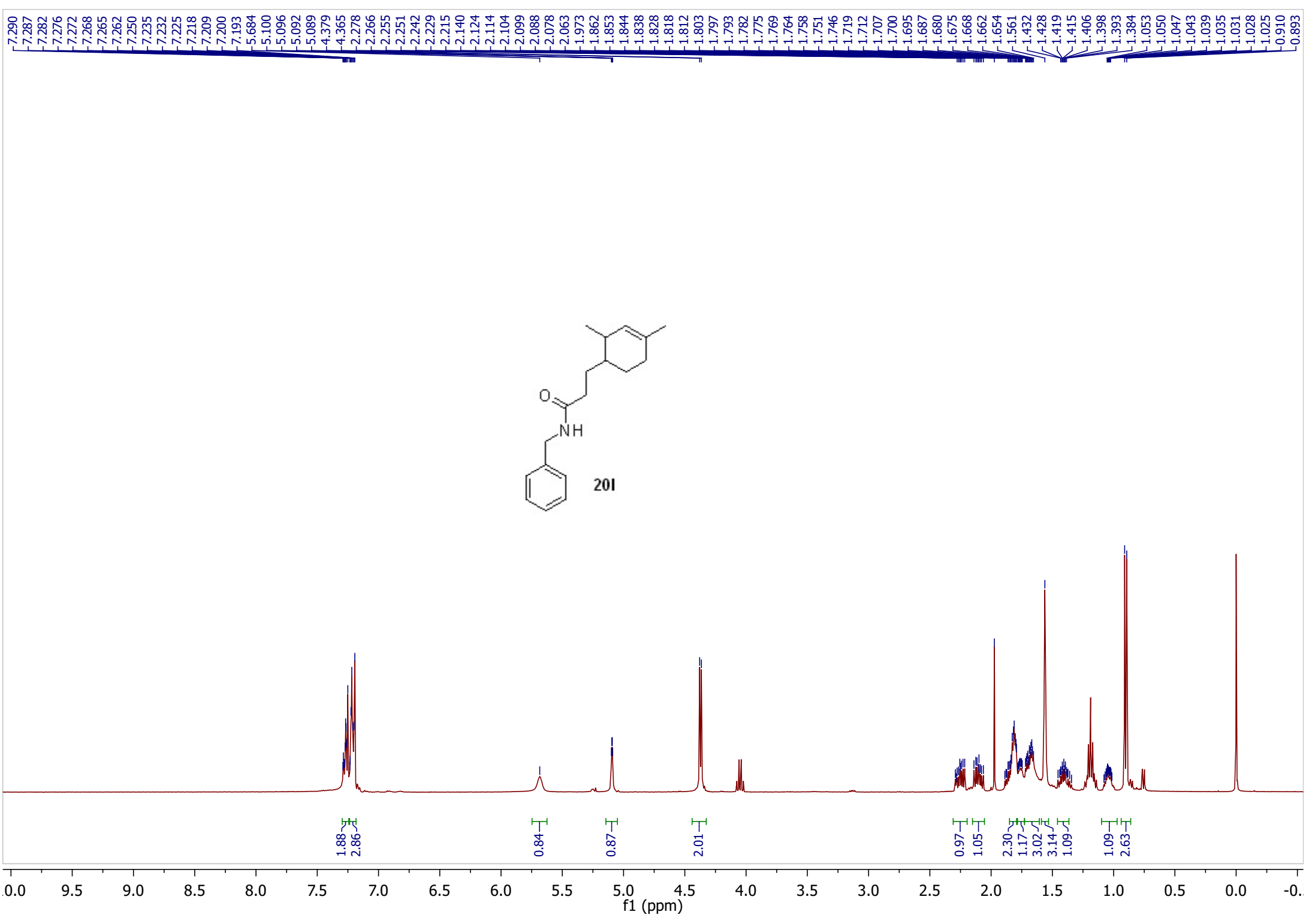


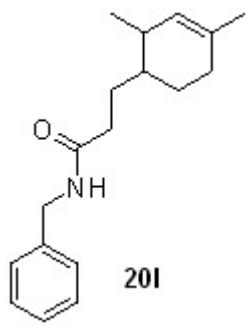










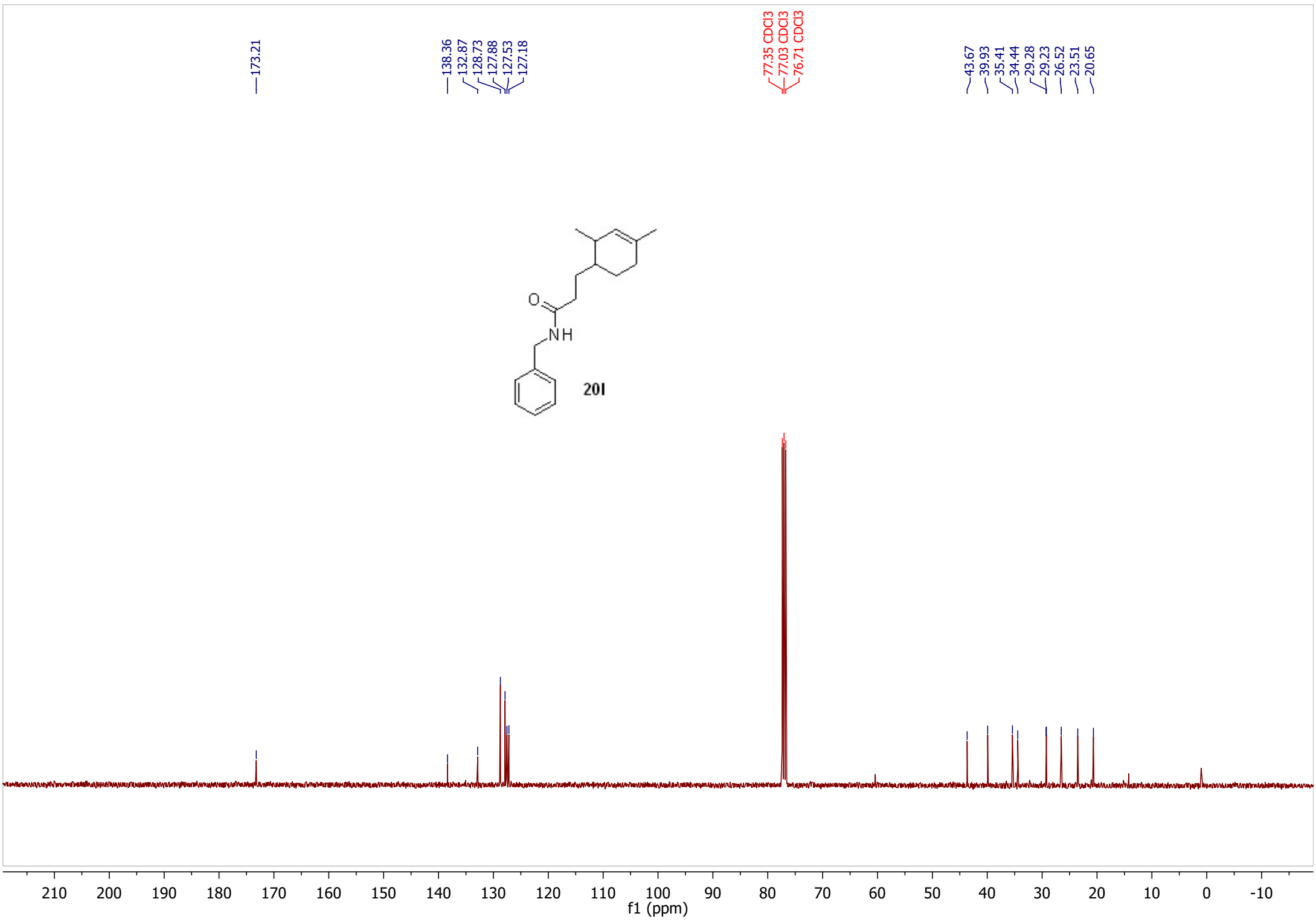


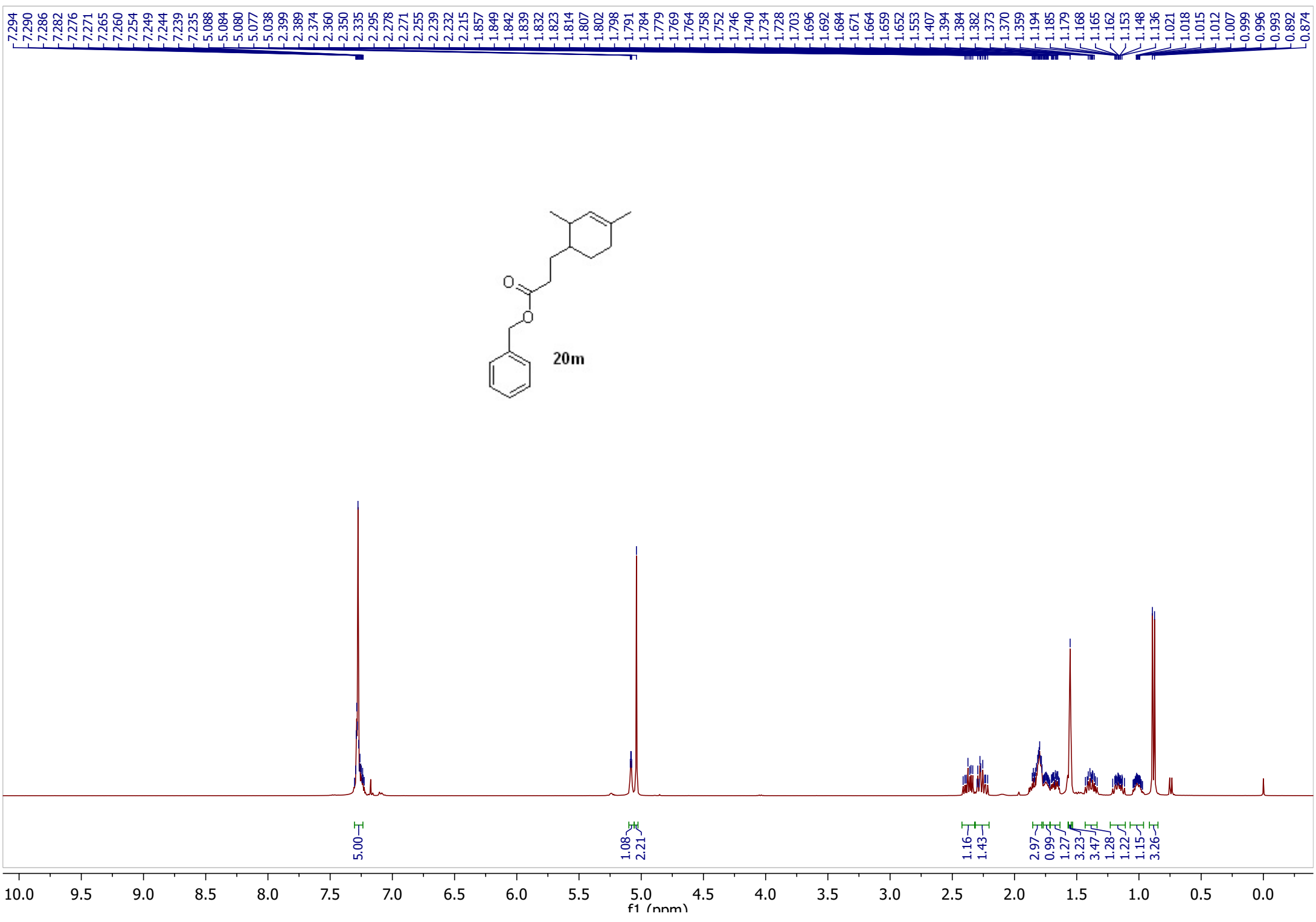
— 173.21

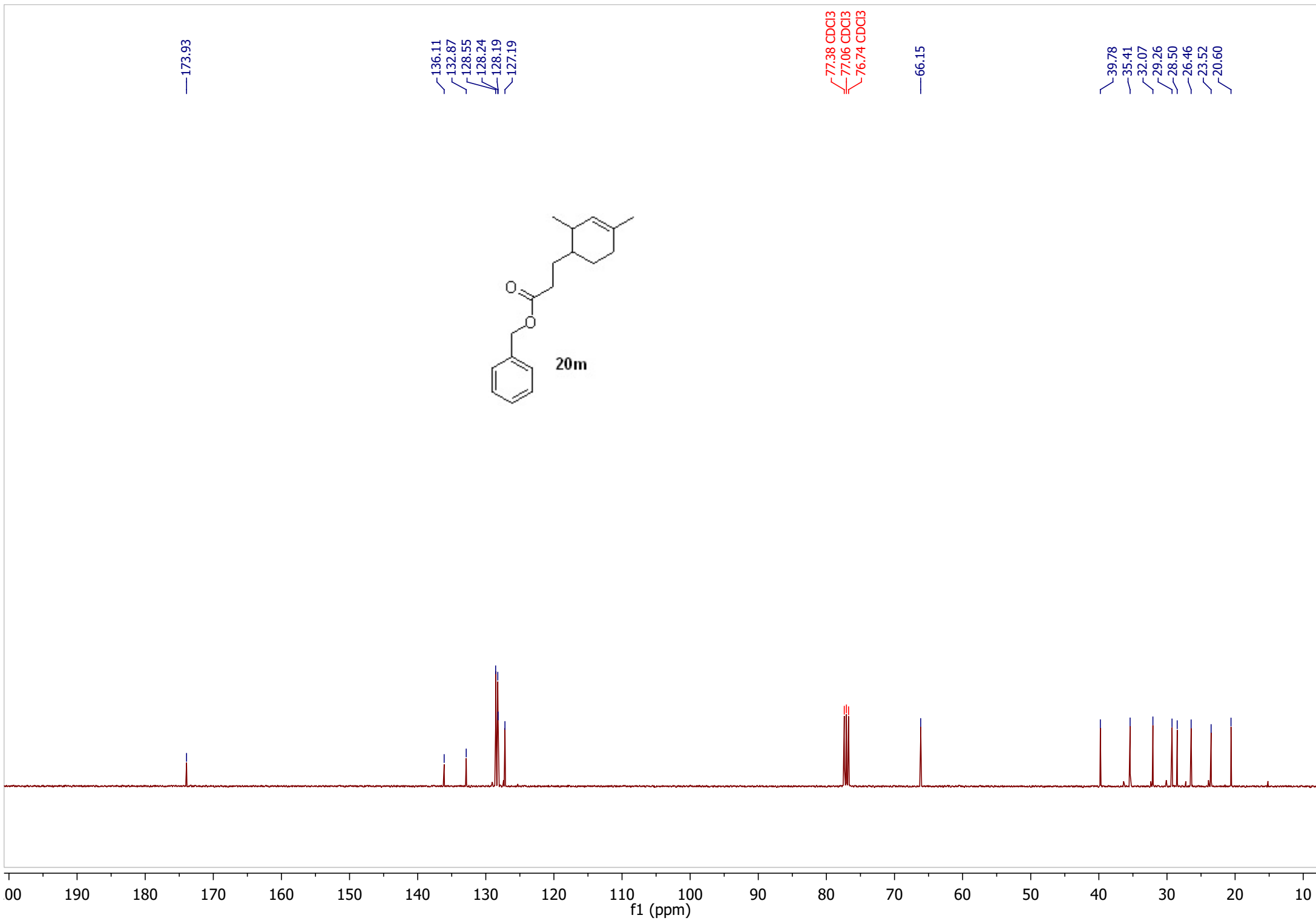
— 138.36
— 132.87
— 128.73
— 127.88
— 127.53
— 127.18

77.35 CDCl3
77.03 CDCl3
76.71 CDCl3

43.67
39.93
35.41
34.44
29.28
26.52
23.51
20.65





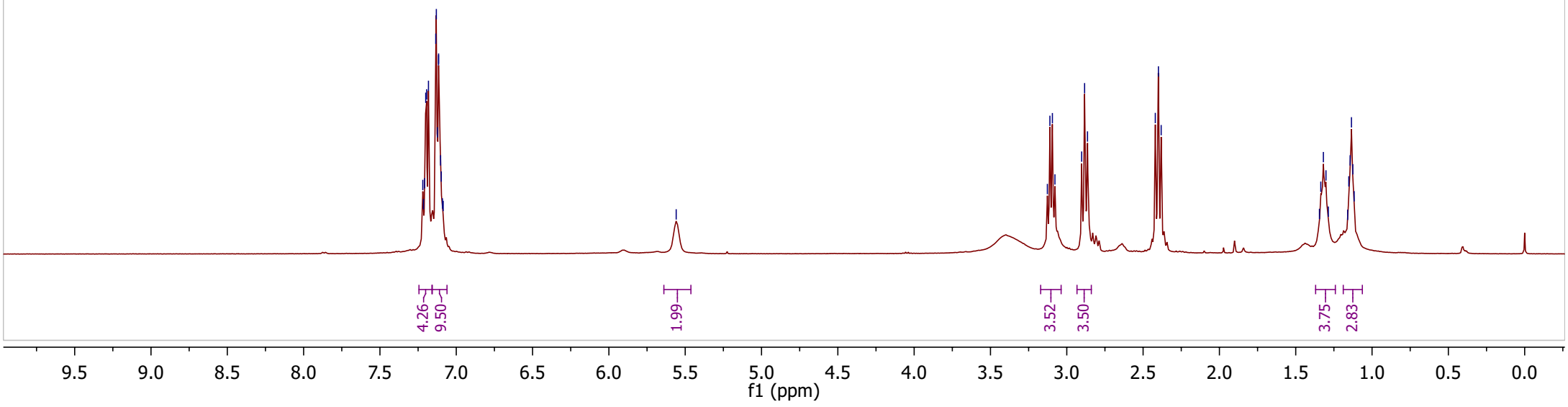
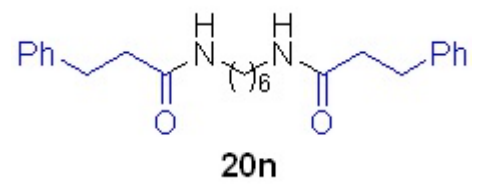


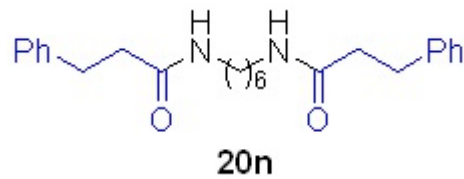
7.219
7.215
7.207
7.200
7.194
7.182
7.134
7.131
7.123
7.116
7.116
7.103
7.098
7.091
7.086

5.559

3.127
3.111
3.094
3.078
2.903
2.883
2.864
2.420
2.400
2.382

1.345
1.336
1.319
1.302
1.286
1.160
1.152
1.144
1.135
1.126
1.117





—172.23

—140.89

—128.50

—128.35

—126.21

77.37 CDCl₃

77.05 CDCl₃

76.73 CDCl₃

—38.96

—38.51

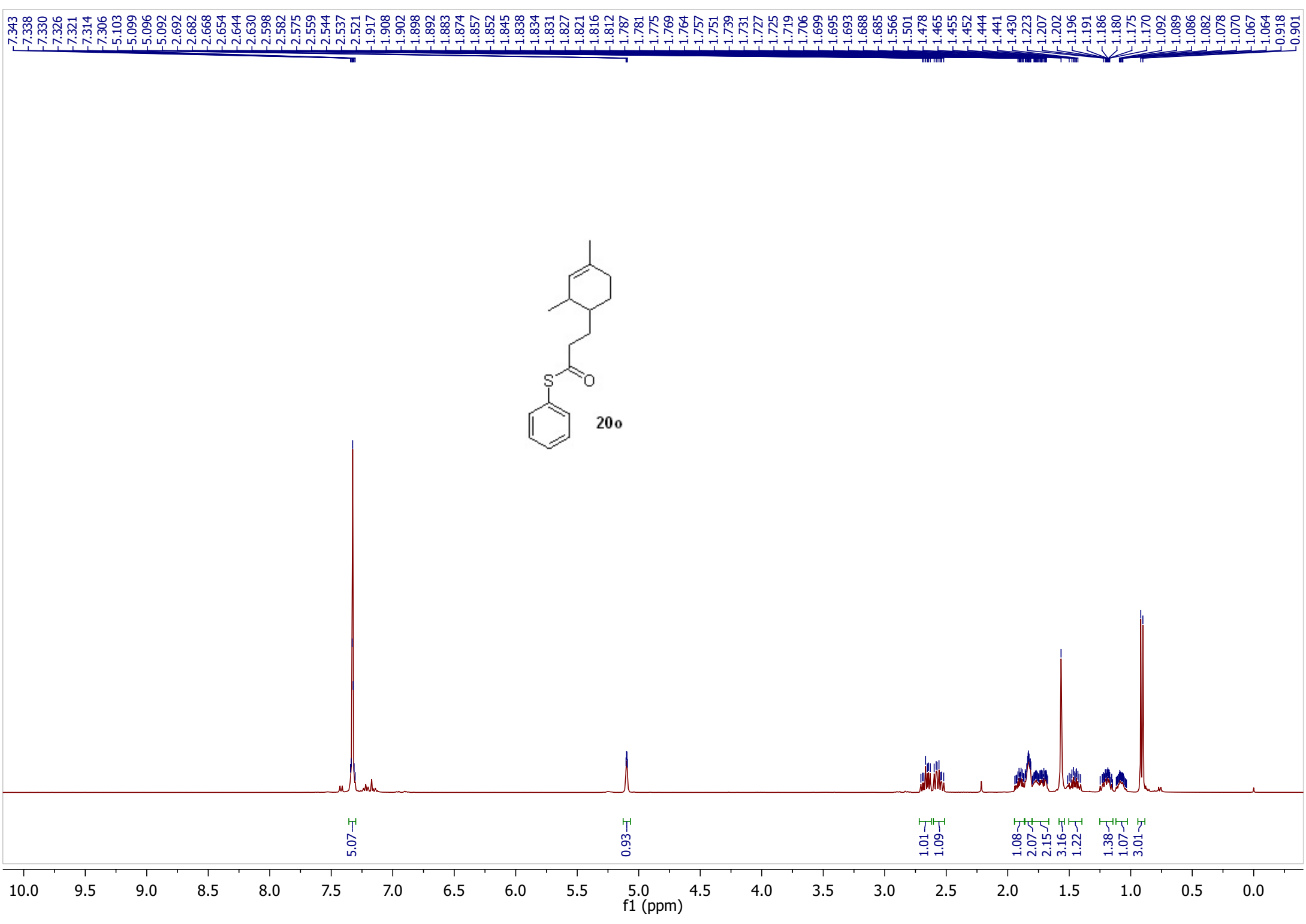
—31.80

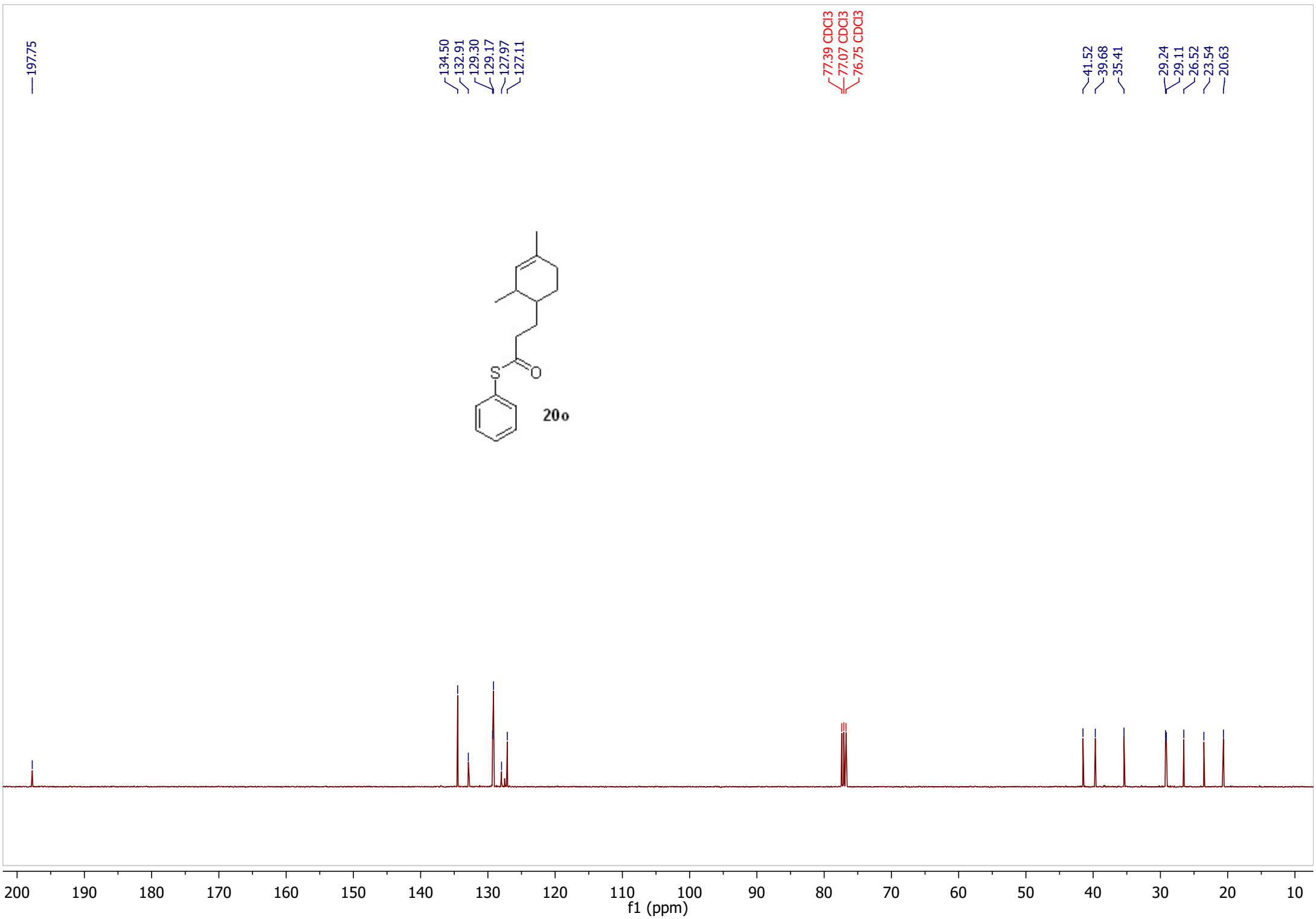
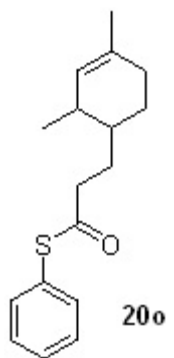
—29.30

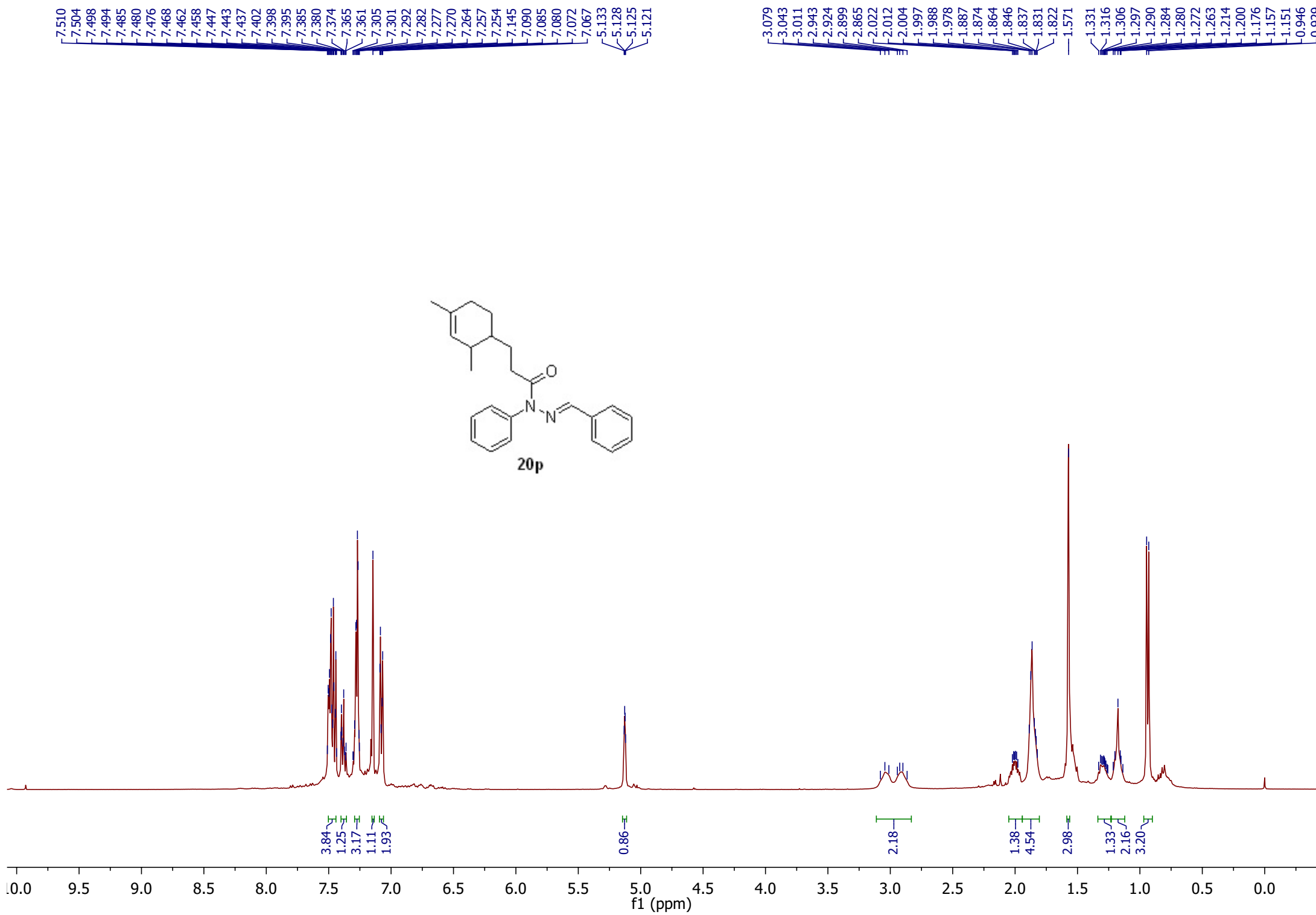
—25.84

210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10

f1 (ppm)





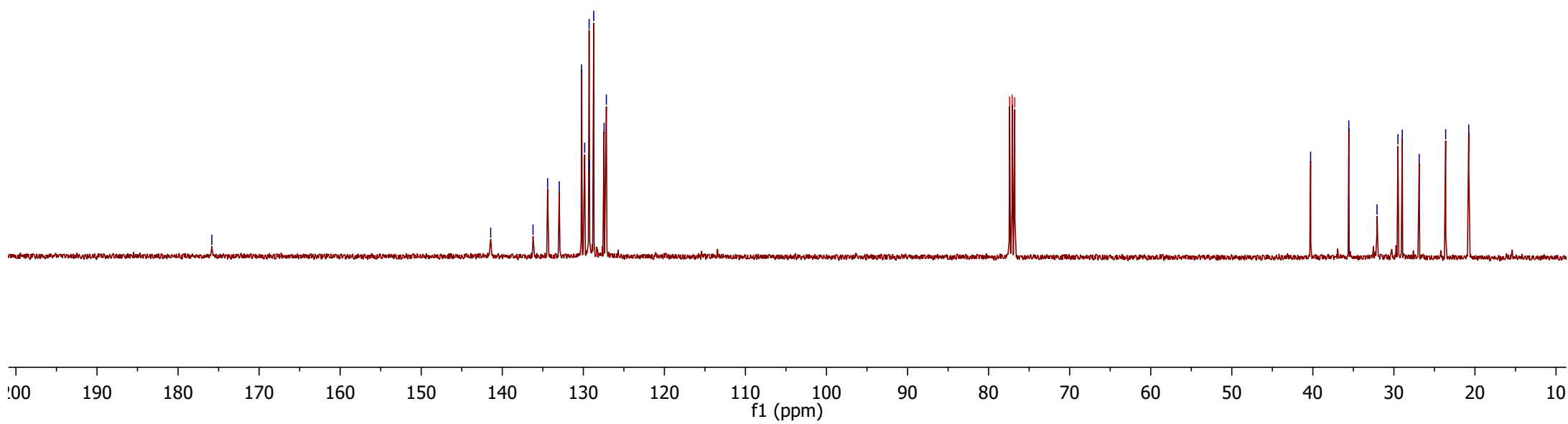
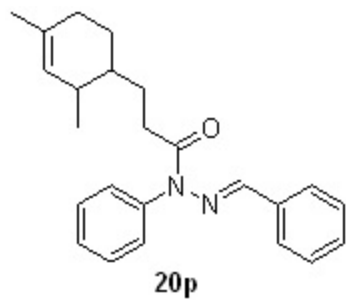


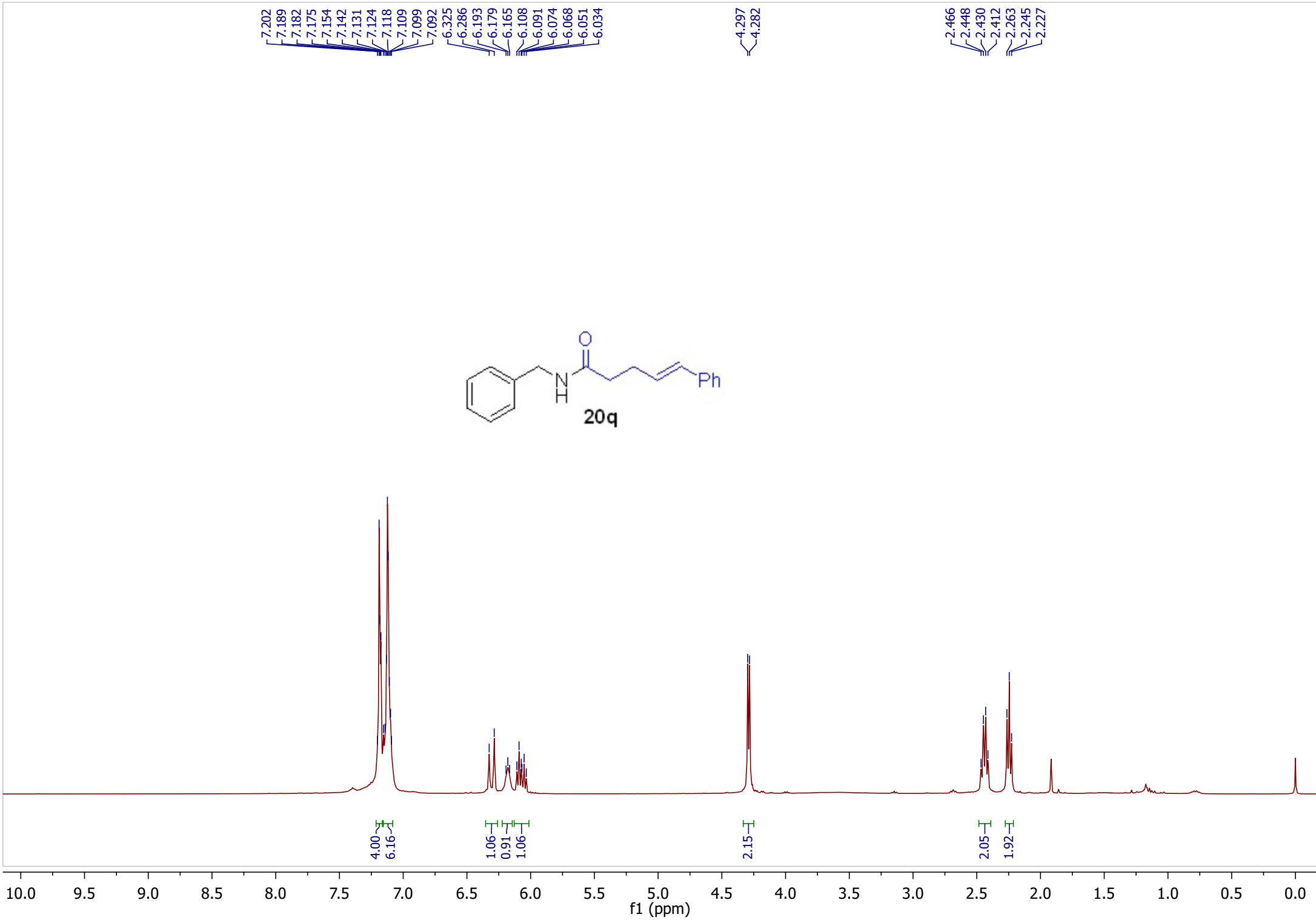
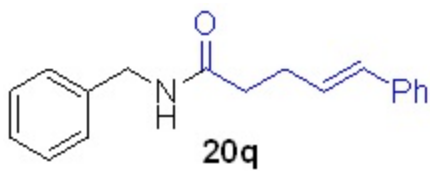
—175.829

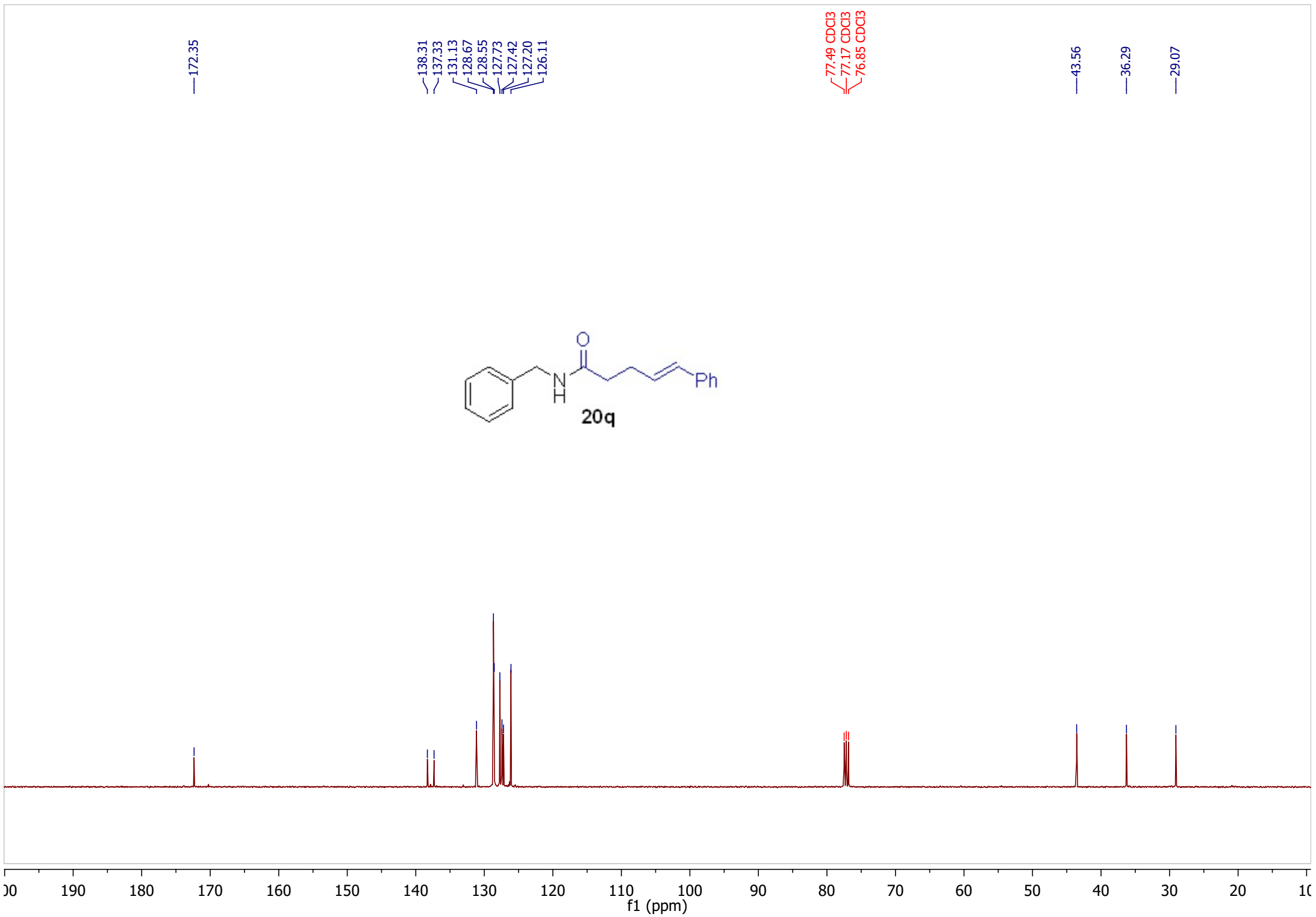
—141.427
—136.205
—134.407
—132.972
—130.221
—129.836
—129.286
—129.248
—128.717
—127.443
—127.161

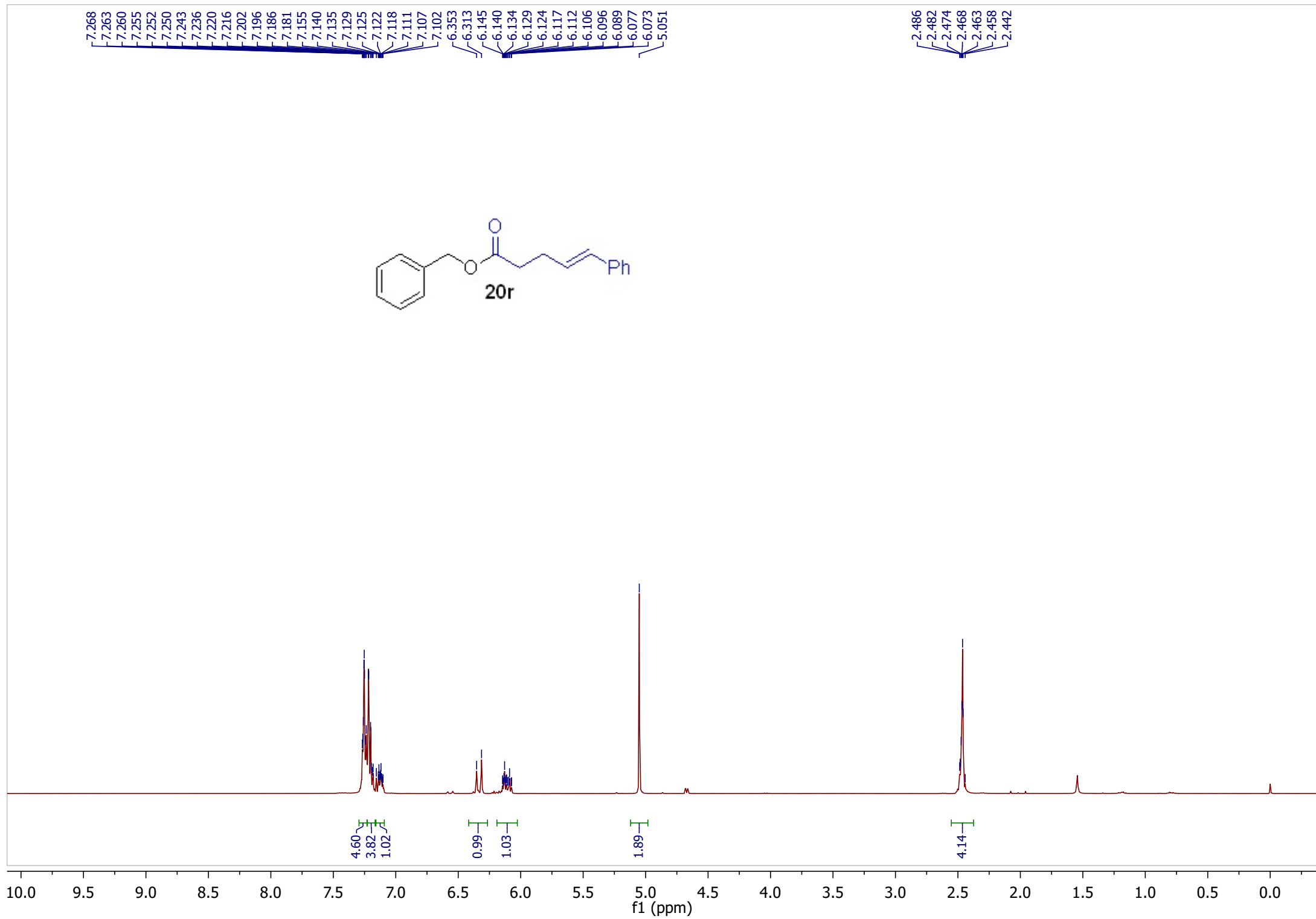
—77.413 CDCl3
—77.096 CDCl3
—76.778 CDCl3

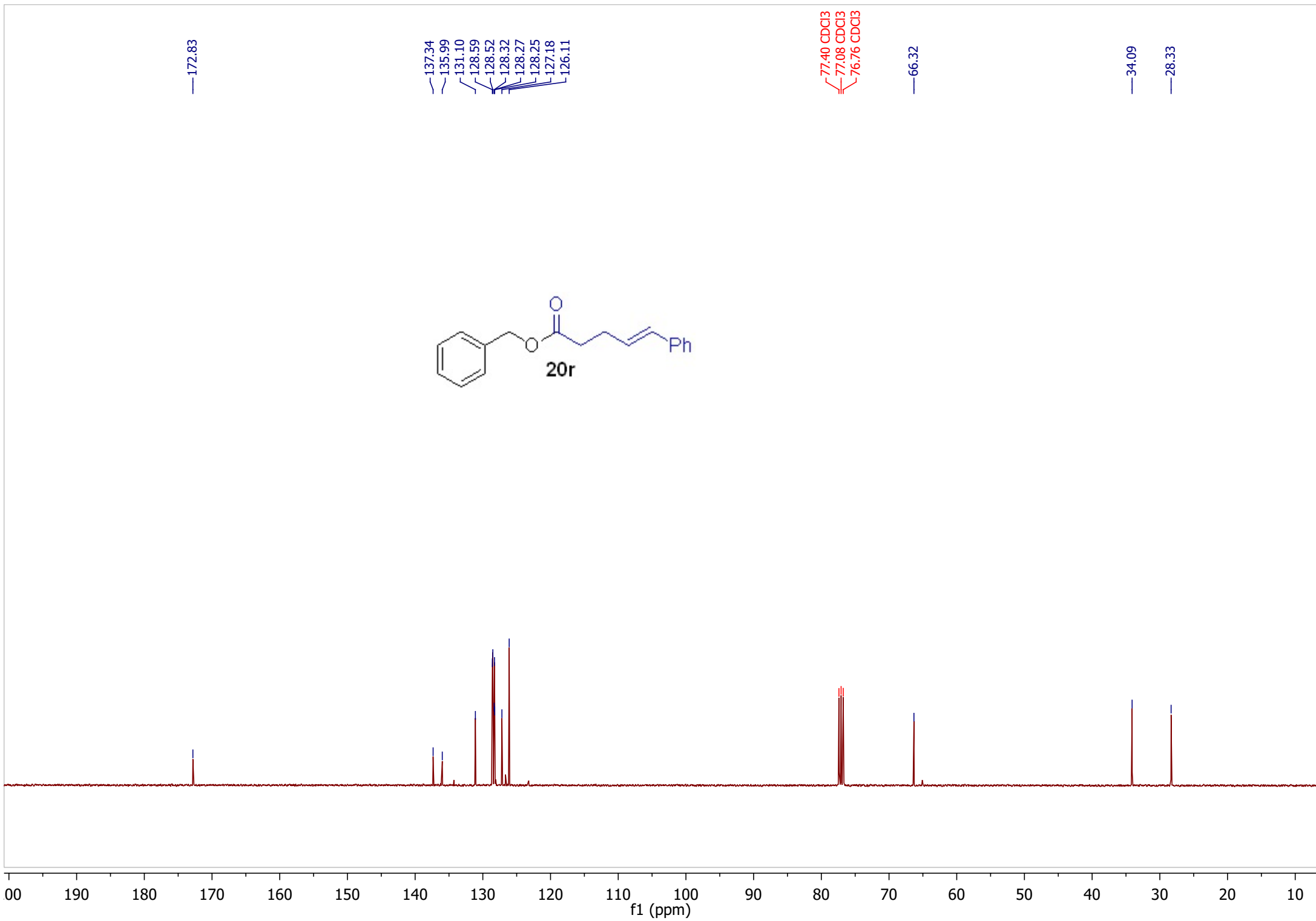
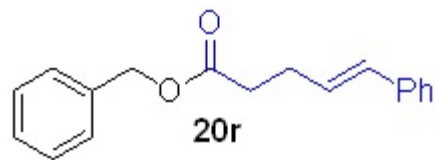
—40.277
—35.565
—32.085
—29.503
—28.972
—26.881
—23.617
—20.769

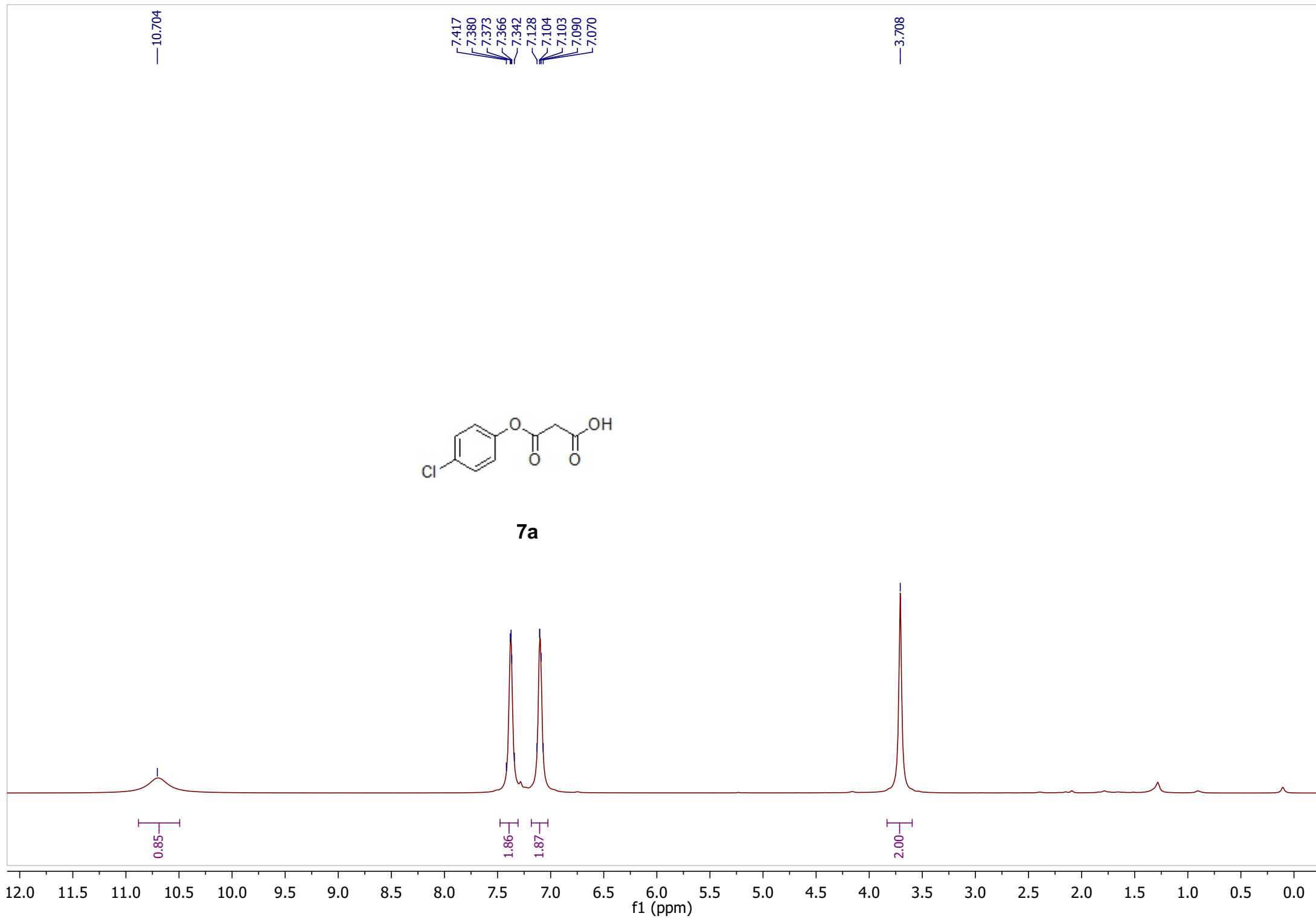


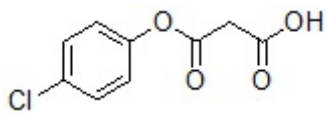




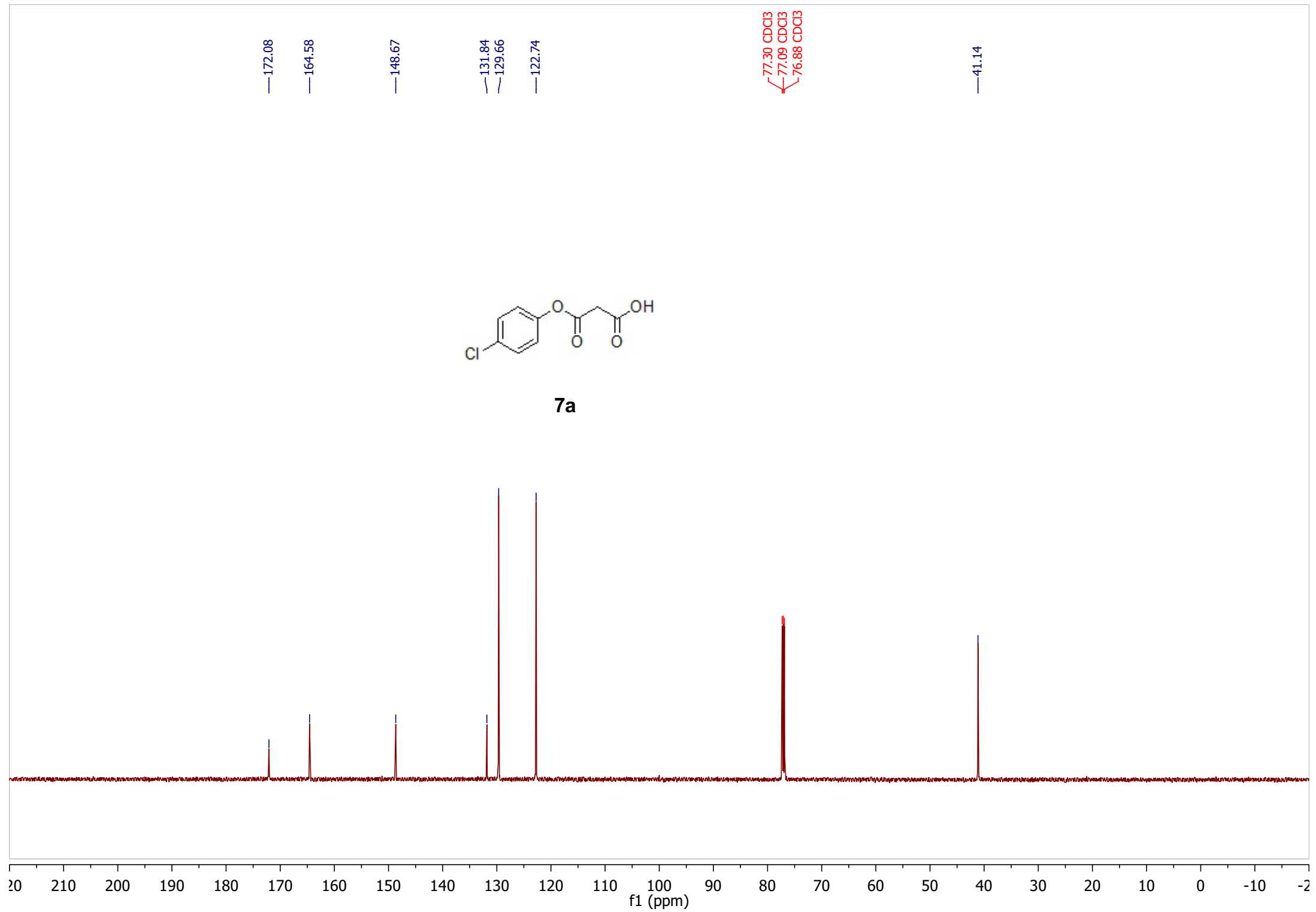


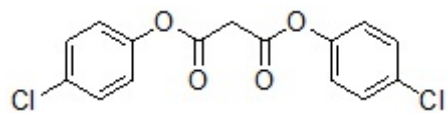




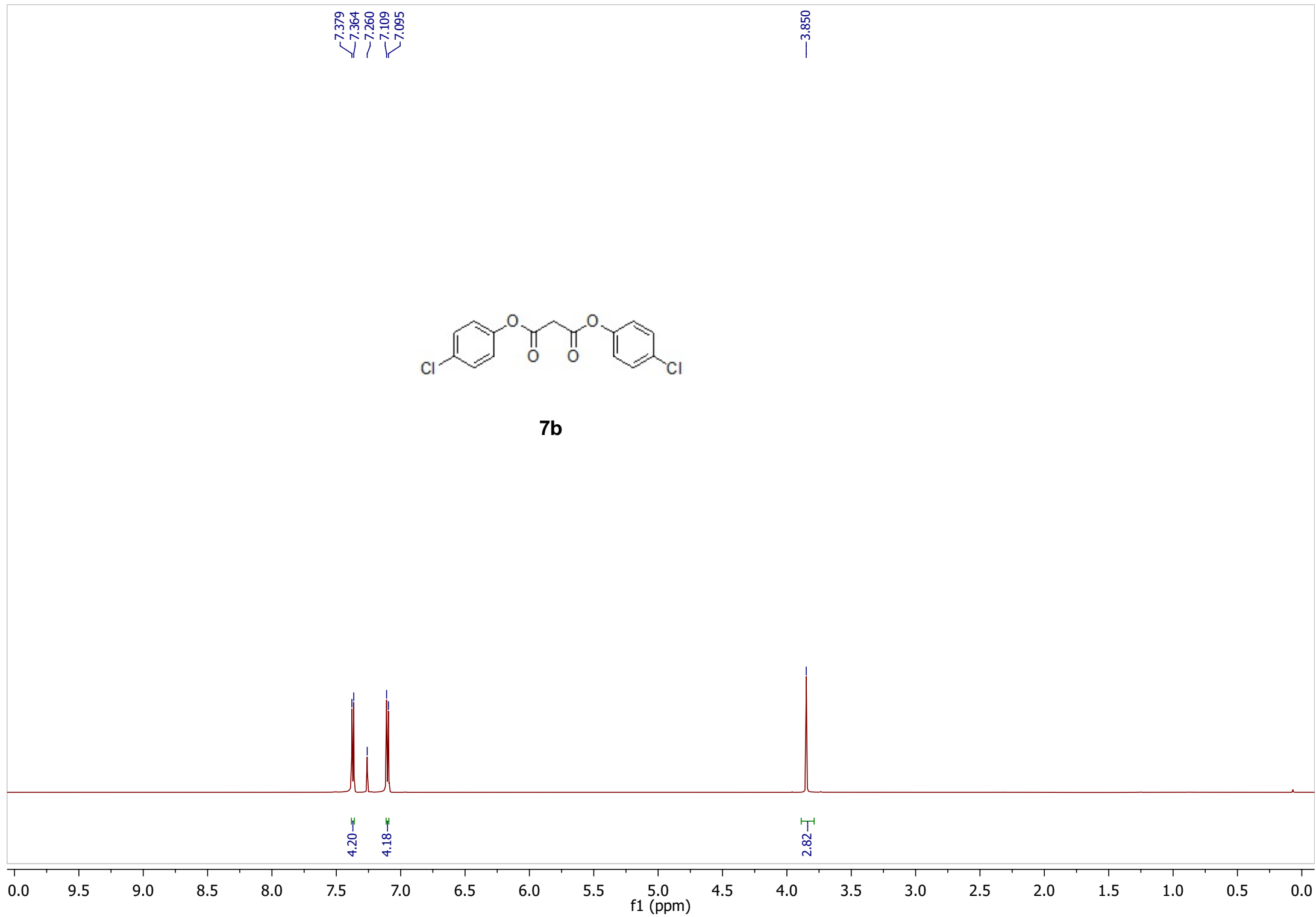


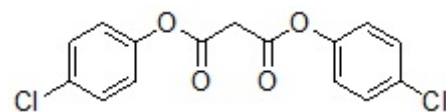
7a





7b





7b

—164.41

—148.71

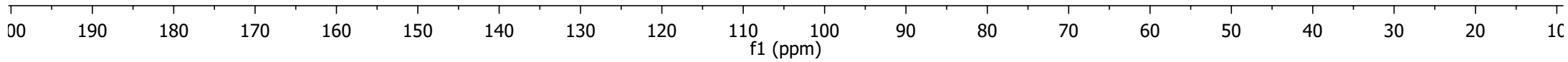
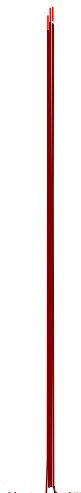
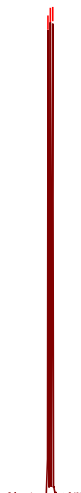
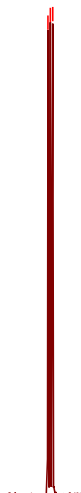
—131.90

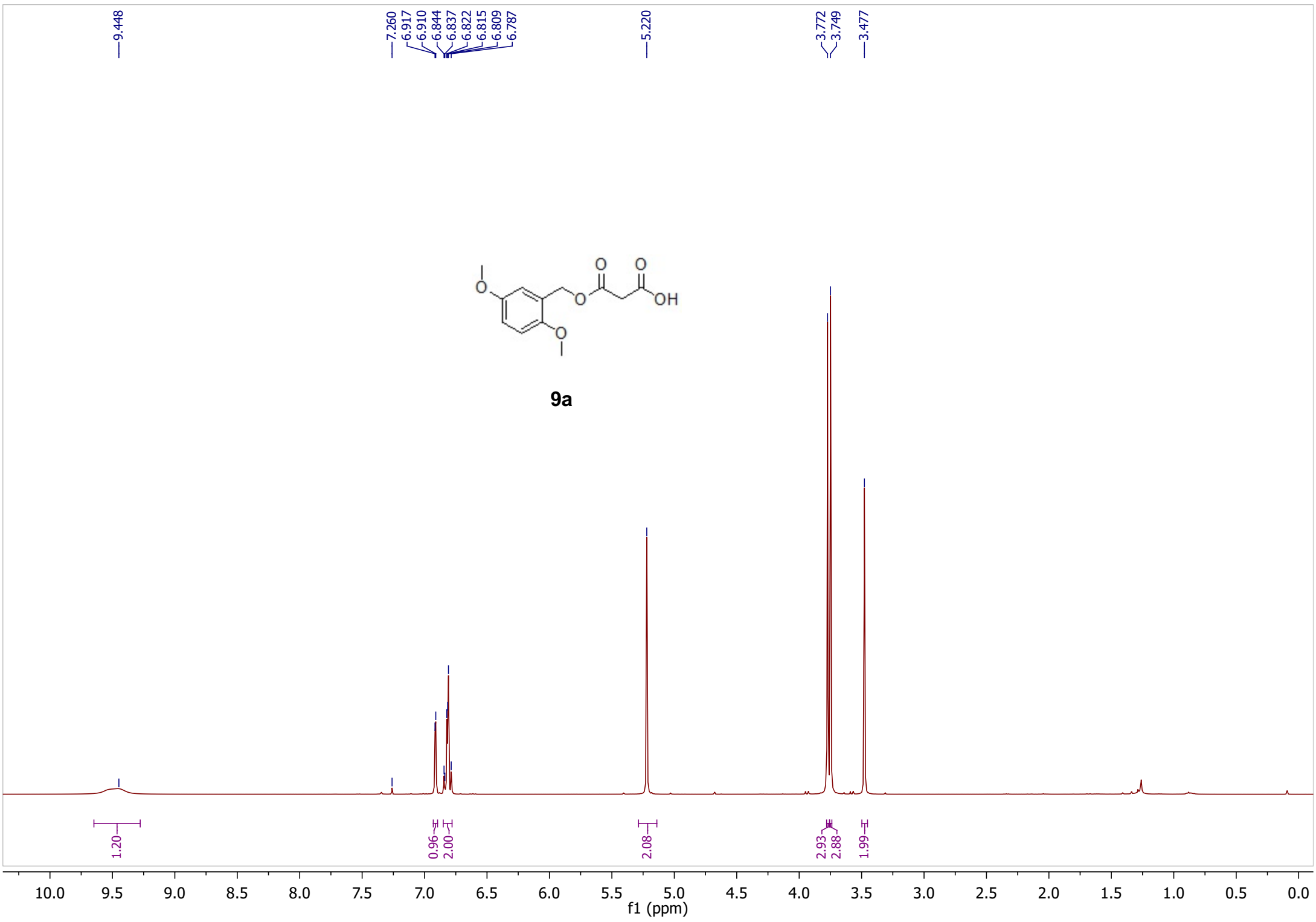
—129.71

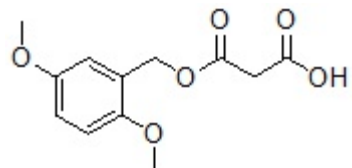
—122.70

77.25 CDCl₃
77.04 CDCl₃
76.82 CDCl₃

—41.46







9a

— 171.223
— 166.695

— 153.387
— 151.634

— 124.341

— 115.556
— 114.337
— 111.690

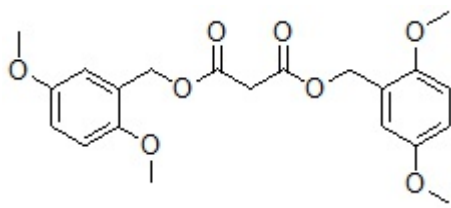
77.475 CDCl₃
77.157 CDCl₃
76.839 CDCl₃

— 62.967

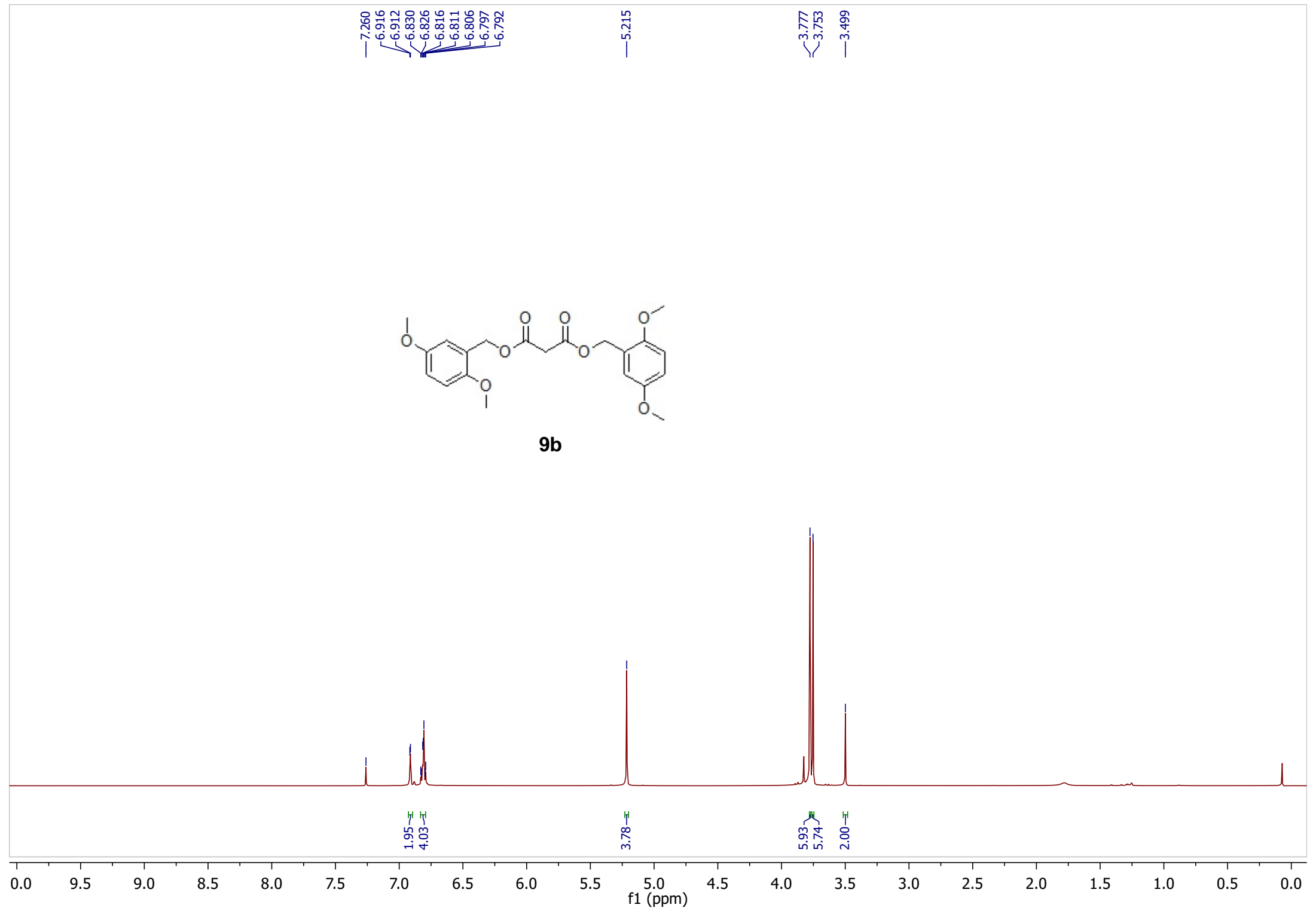
— 56.044
— 55.769

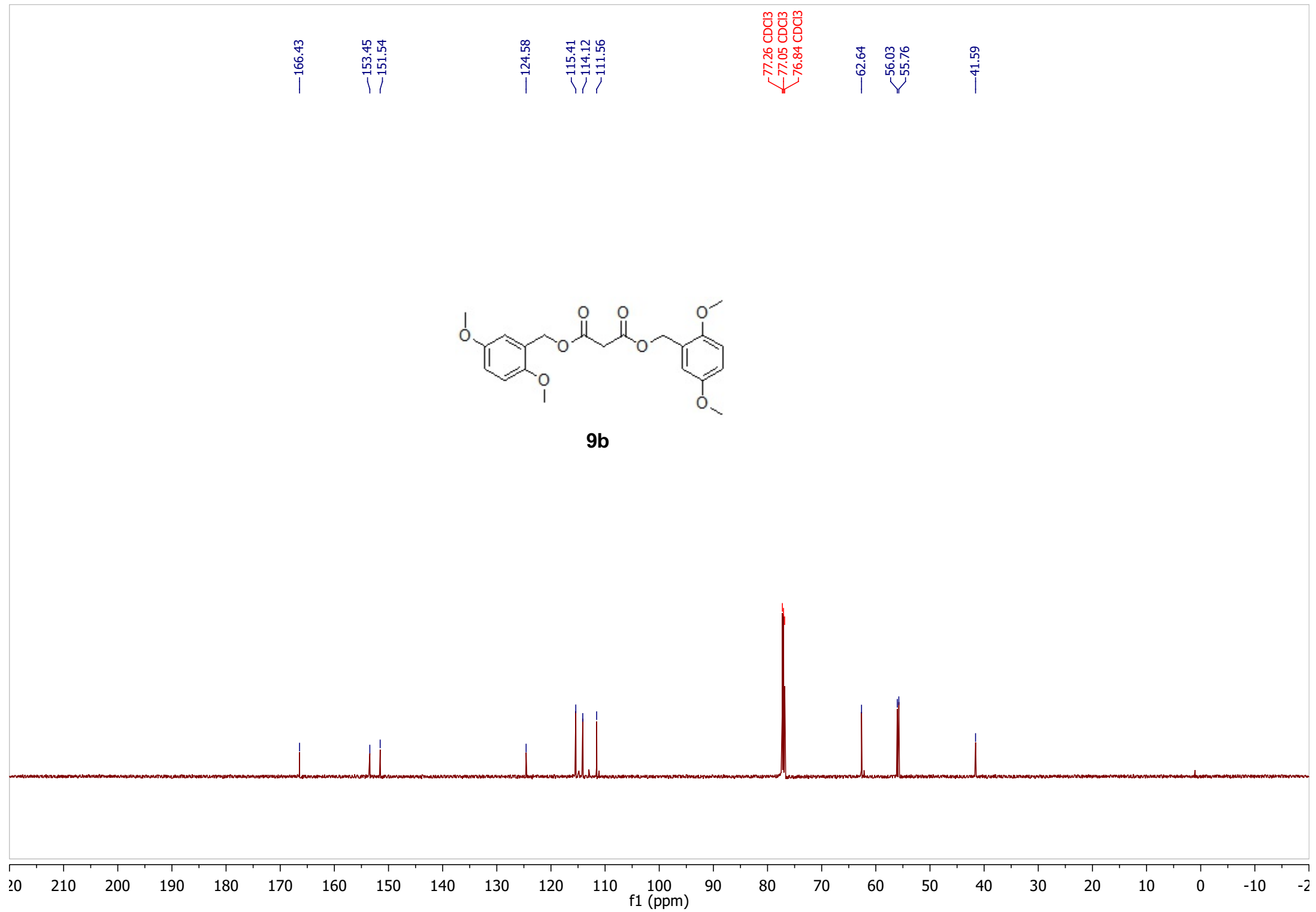
— 40.938

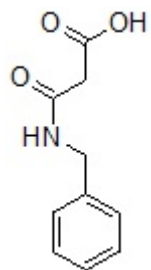
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)



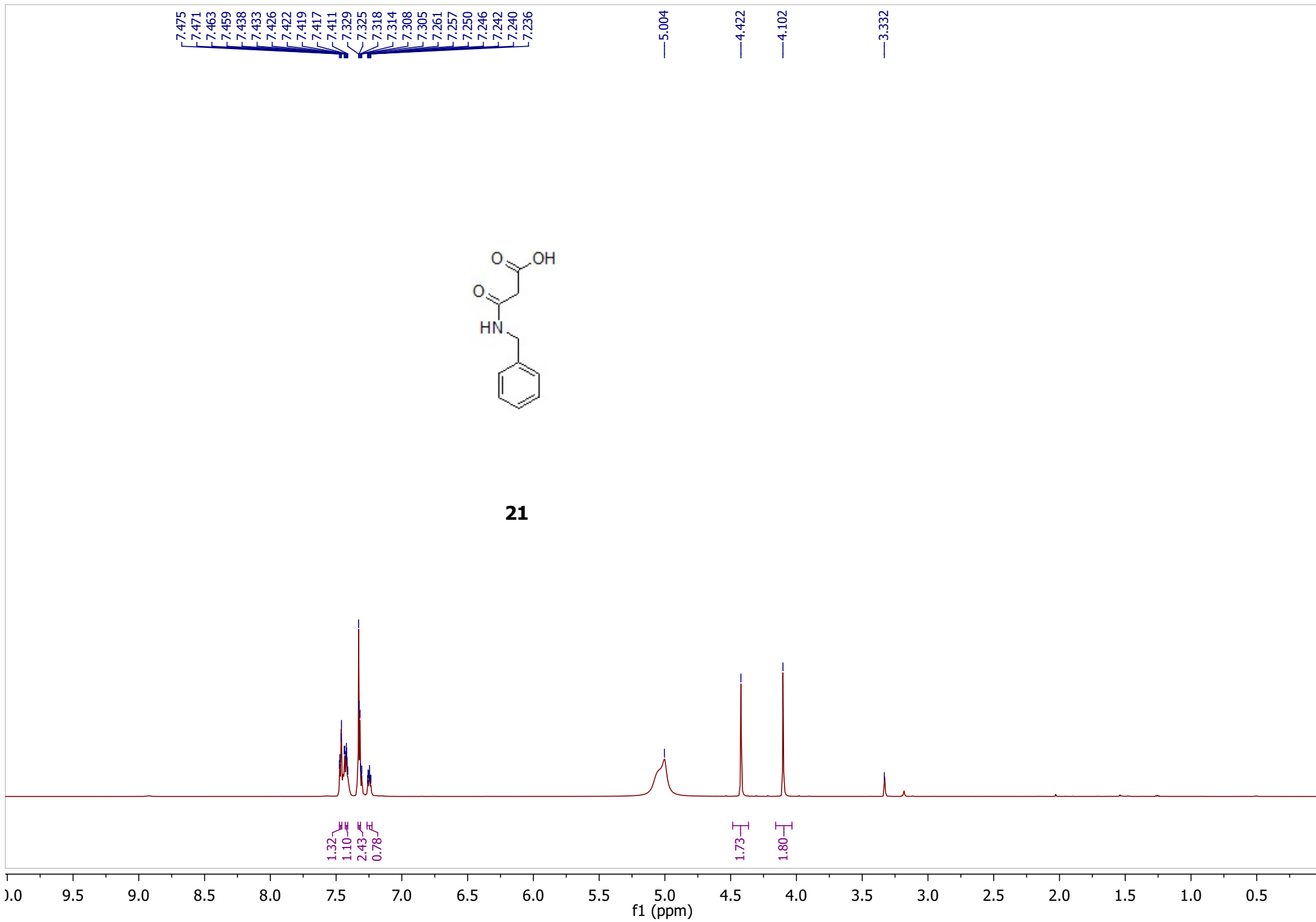
9b







21



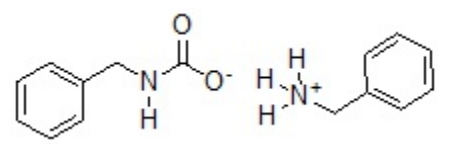
7.39
7.38
7.37
7.37
7.36
7.35
7.35
7.33
7.32
7.31
7.31
7.30
7.30
7.29
7.28

4.94

4.29

3.88

3.33



28

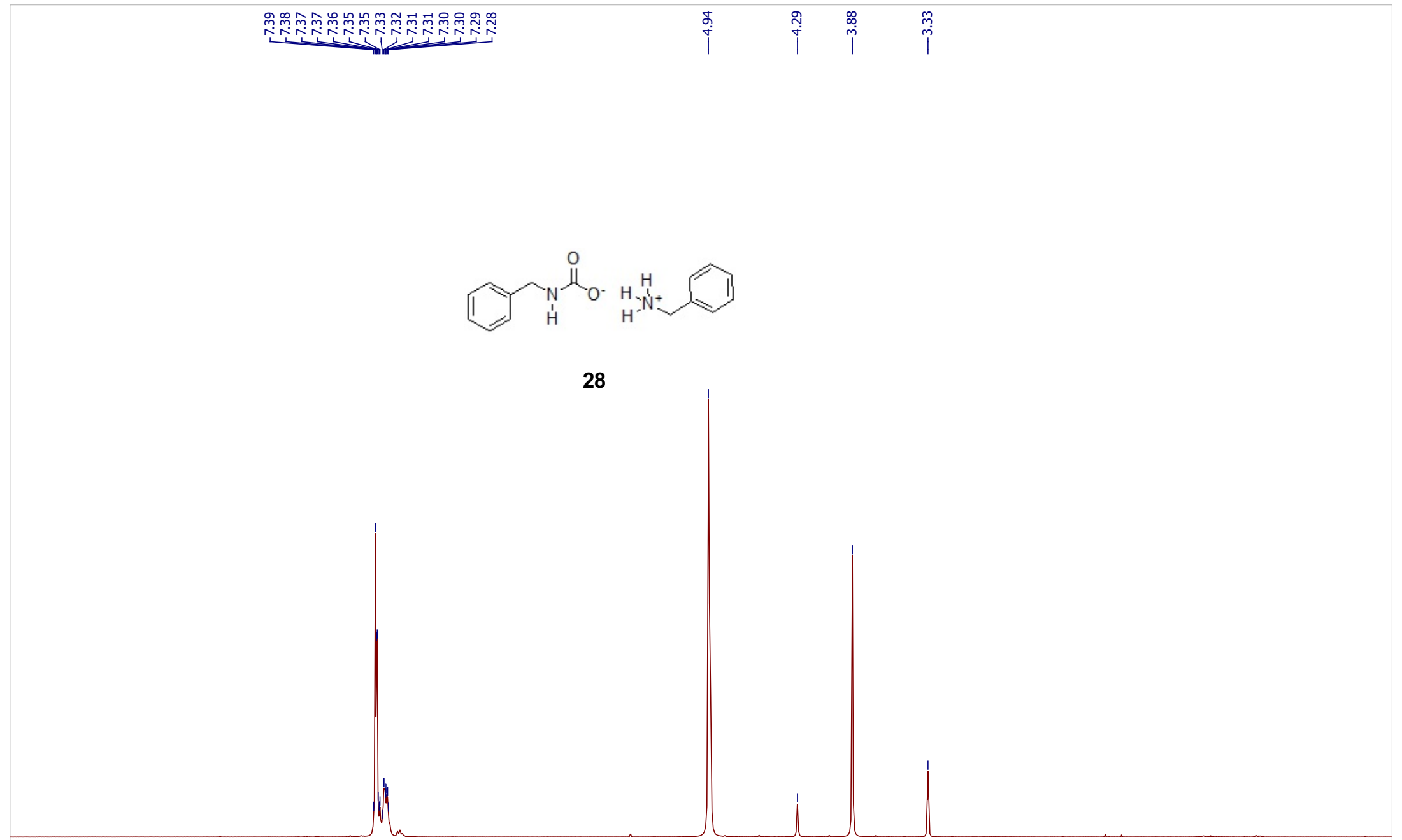
0.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0

f1 (ppm)

10.30

0.61

3.83

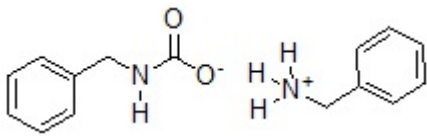


— 163.99
— 160.10

— 141.09
— 136.66

128.53
128.00
127.88
126.73
126.23

48.28 MeOD
48.06 MeOD
47.85 MeOD
47.64 MeOD
47.43 MeOD
47.21
47.00
45.08
43.75



28

