Electronic Supplementary Information (ESI):

Divergent Reaction: Metal & Oxidant Free Direct C-H Aryloxylation and Hydride Free Formal Reductive *N*-benzylation of *N*-heterocycles

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Experimental Section:

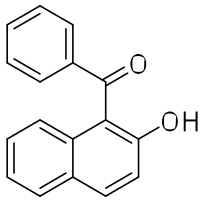
General: All reactions involving air- or moisture-sensitive reagents or intermediates were carried out in oven-dried glassware under an argon atmosphere. Dichloromethane (CH_2Cl_2) was freshly distilled from phosphorus(V)oxide (P_2O_5). Triethylamine (Et_3N) was distilled from CaH_2 and stored under argon. Commercial grade xylene, benzene and toluene were distilled before use. All other solvents and reagents were purified according to standard procedures or were used as received from Aldrich, Acros, Merck and Spectrochem. Microwave reactions were performed on CEM, Discover system. ^1H, ^{13}C NMR spectroscopy: *Varian Mercury plus 400 MHz, Bruker 600 MHz* (at 298 K). Chemical shifts, δ (in ppm), are reported relative to TMS δ (^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. IR: spectra were recorded on Perkin Elmer Instrument at normal temperature making KBr pellet grinding the sample with KBr (IR Grade). MS (ESI-HRMS): Mass spectra were recorded on a Agilent Accurate-Mass Q-TOF LC/MS 6520, and peaks are given in *m/z* (% of basis peak).

Experimental procedure:

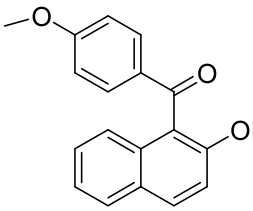
General procedure for preparation of ketones: GPI[1]

2-naphthol (1.44 g, 10 mmol) in 10 mL of $BF_3.OEt_2$ was heated at 60 ^oC to make it soluble. Then benzoic acid (1.1 eq) was added to it and resulting reaction mixture was heated at 100 ^oC for 20 h. Reaction mixture was cooled to room temperature and diluted with water (50 mL). Then the mixture was extracted with EtOAc (3 × 30 mL). Then the combined organic layers were dried (Na_2SO_4) and concentrated in vacuum. The crude product was purified by SiO_2-gel column chromatography.

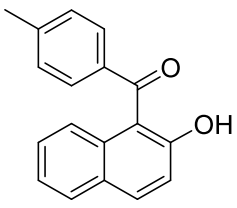
(2-Hydroxy-naphthalen-1-yl)-phenyl-methanone (1b):[3] According to GPI: 2-naphthol (1.44 g, 10.00 mmol), benzoic acid (1.34 g, 11.00 mmol) in 10 mL $BF_3.OEt_2$ for 20 h and SiO_2- column chromatography (EtOAc : Hexane, 1 : 20) gave **1b** as orange yellow crystal (1.61 g, 65%). ^1H NMR (400 MHz, $CDCl_3$) δ = 11.15 (s, 1H), 7.86 (d, *J* = 9.0 Hz, 1H), 7.68 (d, *J* = 8.0 Hz, 1H), 7.56 – 7.54 (m, 2H), 7.450 – 7.47 (m, 1H), 7.35 – 7.31 (m, 2H), 7.22 (t, *J* = 7.6 Hz, 2H), 7.17 (d, *J* = 8.9 Hz, 1H), 7.11 – 7.05 (m, 1H). LRMS (ESI) data calculated for $C_{17}H_{11}O_2^-$ $[M-H]^-$: 247.07 ; found : 247.17



(2-Hydroxy-naphthalen-1-yl)-(4-methoxy-phenyl)-methanone (3a):[2] According to GPI: 2-naphthol (0.57 g, 4.00 mmol), 4-Methoxy-benzoic acid (0.66 g, 4.40 mmol) in 4 mL $BF_3.OEt_2$ for 20 hrs and SiO_2-column chromatography (EtOAc : Hexane, 1 : 15) gave **3a** as yellow crystal, (0.49 g, 44 %). ^1H NMR (600 MHz, $CDCl_3$) δ = 10.59 (s, 1H), 7.91 (d, *J* = 9.0 Hz, 1H), 7.76 – 7.75 (m, 1H), 7.66 – 7.63 (m, 2H), 7.42 – 7.41 (d, *J* = 8.5 Hz, 1H), 7.29 – 7.27 (m, 1H), 7.23 (d, *J* = 9.0 Hz, 1H), 7.21 – 7.18 (m, 1H), 6.89 – 6.86 (m, 2H), 3.86 (s, 3H). LRMS (ESI) data calculated for $C_{18}H_{13}O_3^-$ $[M-H]^-$: 277.08 ; found : 277.20

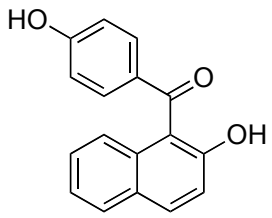


(2-Hydroxy-naphthalen-1-yl)-p-tolyl-methanone (3b): According to GPI: 2-naphthol(0.57 g, 4.00 mmol), 4-Methyl-benzoic acid (0.59 g, 4.40 mmol) in 4 mL $BF_3.OEt_2$ for 20 hrs and SiO_2-column chromatography (EtOAc : Hexane, 1 : 30) gave **3b** as yellow crystal, (0.430 gm, 41%). ^1H NMR (400 MHz, $CDCl_3$) δ = 10.99 (s, 1H), 7.91 (d, *J* = 9.0 Hz, 1H), 7.74 (d, *J* = 8.0 Hz, 1H), 7.55 – 7.53



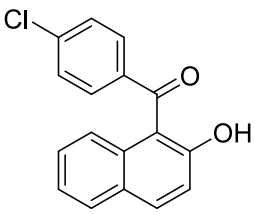
(m, 2H), 7.37 – 7.34 (m, 1H), 7.28 – 7.22 (m, 2H), 7.19 – 7.15 (m, 3H), 2.40 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ = 200.1, 160.9, 143.9, 137.7, 136.0, 132.6, 129.9, 129.4, 128.7, 128.6, 126.8, 126.5, 123.8, 119.3, 114.9, 21.9. LRMS (ESI) data calculated for $C_{18}H_{13}O_2^-$ $[M-H]^-$: 261.09 ; found : 261.19

(2-hydroxynaphthalen-1-yl)(4-hydroxyphenyl)methanone (3c):[1] According to GPI: 2-



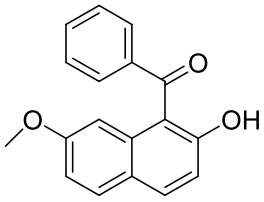
naphthol (0.58 g, 4.00 mmol), 4-hydroxy benzoic acid (0.61 g, 4.40 mmol) in 4 mL $BF_3.OEt_2$ for 16 h. and SiO_2 column chromatography (EtOAc : Hexane, 1 : 20) gave **3c** as whitish solid (0.42 g, 40%). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 10.66 (s, 1H), 7.92 (d, *J* = 9.0 Hz, 1H), 7.76 (d, *J* = 8.0 Hz, 1H), 7.63 – 7.59 (m, 2H), 7.43 – 7.41(m, 1H), 7.29 (t, *J* = 7.4 Hz, 1H), 7.25 – 7.23 (m, 1H), 7.21 – 7.19 (m, 1H), 6.84 – 6.80 (m, 2H). LRMS (ESI) data calculated for $C_{17}H_{11}O_3^-$ $[M-H]^-$: 263.07 ; found : 263.17

(4-Chloro-phenyl)-(2-hydroxy-naphthalen-1-yl)-methanone (3d):[3] According to GPI: 2-



naphthol (0.28 g, 2.0 mmol), 4-Chloro-benzoic acid (0.34 g, 2.20 mmol) in 2 mL $BF_3.OEt_2$ for 52 h and SiO_2-column chromatography (EtOAc : Hexane 1 : 30) gave **3d** as brown solid (70 mg, 12%). ^{1}H NMR (400 MHz $CDCl_3$) δ = 11.09 (s, 1H), 7.94 (d, *J* = 9.0 Hz, 1H), 7.77 – 7.75 (m, 1H), 7.59 – 7.57 (m, 2H), 7.39 – 7.37 (m, 2H), 7.31 – 7.28 (m, 2H), 7.26 – 7.18 (m, 2H). LRMS (ESI) data calculated for $C_{17}H_{10}ClO_2^-$ $[M-H]^-$: 281.03 ; found : 281.14

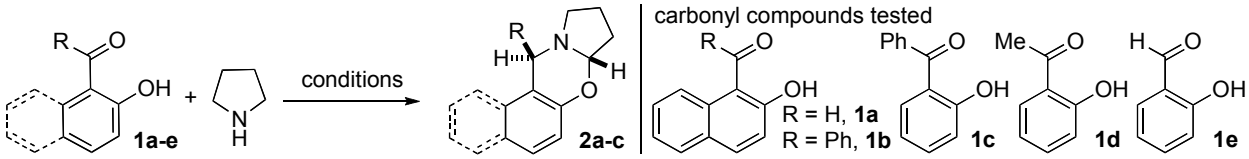
(2-Hydroxy-7-methoxy-naphthalen-1-yl)-phenyl-methanone (3e): According to GPI: 7-



Methoxy-naphthalen-2-ol (0.52 g, 3.00 mmol), benzoic acid (0.40 g, 3.30 mmol) in 3 mL $BF_3.OEt_2$ for 16 hrs and SiO_2-column chromatography (EtOAc : Hexane, 1 : 20) gave **3e** as yellow solid (0.43 g, 51%). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 11.66 (s, 1H), 7.86 – 7.84 (m, 1H), 7.63 – 7.61 (m, 3H), 7.57 – 7.53 (m, 1H), 7.45 – 7.42 (m, 2H), 7.08 (d, *J* = 8.9 Hz, 1H), 6.89 – 6.88 (m, 1H), 6.59 (s, 1H), 3.27 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ = 200.9, 163.0, 158.3, 141.0, 136.7, 134.3, 132.39, 130.2, 129.3, 128.8, 123.9, 116.7, 116.1, 113.8, 106.7, 54.6. LRMS (ESI) data calculated for $C_{18}H_{13}O_3^-$ $[M-H]^-$: 277.08 ; found : 277.19

(6-Bromo-2-hydroxy-naphthalen-1-yl)-phenyl-methanone (3f): According to GPI: 6-Bromo-naphthalen-2-ol (0.44 g, 2.00 mmol,), benzoic acid (0.26 g, 2.20 mmol) in 2 mL $BF_3.OEt_2$ for 16 hrs and SiO_2-column chromatography (EtOAc : Hexane, 1 : 15) gave **3f** as reddish brown, (0.15 g, 23%). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 11.13 (s, 1H), 7.86 (s, 1H), 7.80 (d, *J* = 8 Hz, 1H), 7.60 – 7.54 (m, 3H), 7.41 – 7.38 (m, 2H), 7.25 – 7.18 (m, 2H), 7.14 – 7.12 (m, 1H). LRMS (ESI) data calculated for $C_{17}H_{10}BrO_2^-$ $[M-H]^-$: 324.99 ; found : 325.10

Table s1. Screening of reaction condition and carbonyl compounds for direct C-H functionalization.[a]



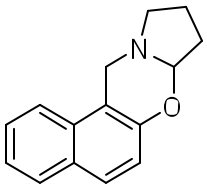
Entry	Carbonyl compound (eq.)	Solvent	Temperature	Reaction time	Additives	Isolated yield (%)
1	**1a** (0.5)	benzene	reflux	12 h	--	8
2	**1a** (0.5)	benzene	µw, 100 °C	10 min	--	38
3[b]	**1a** (0.5)	benzene	µw, 100 °C	20 min	--	42
4	**1a** (0.5)	benzene	µw, 100 °C	20 min	DBU	25
5	**1a** (0.5)	benzene	µw, 100 °C	20 min	$MgSO_4$	15
6	**1a** (0.3)	ethanol	µw, 100 °C	20 min	--	15
7	**1a** (0.5)	xylene	µw, 170 °C	20 min	4Å MS	40
8	**1a** (2.5)	benzene	µw, 100 °C	20 min	--	65
9	**1a** (2.5)	toluene	µw, 130 °C	20 min	--	72
10	**1a** (2.5)	toluene	µw, 130 °C	40 min	--	72
11	**1a** (2.5)	toluene	µw, 130 °C	20 min	KOAc	39
12	**1a** (2.5)	toluene	µw, 130 °C	20 min	PTSA	15
13	**1a** (2.5)	toluene	µw, 130 °C	20 min	Et_3N	70
14	**1b** (1.2)	toluene	µw, 130 °C	20 min	--	86
15	**1b** (1.2)	toluene	µw, 145 °C	20 min	--	96
16	**1b** (1.2)	toluene	reflux	24 h	--	49
17	**1c** (1.2)	toluene	µw, 145 °C	20 min	--	43[c]
18	**1d** (1.0)	toluene	µw, 145 °C	20 min	--	--
19	**1e** (1.0)	xylene	µw, 130 °C	20 min	--	--

[a] Reactions were carried out using 0.24 mmol of pyrrolidine in 1.5 mL of solvents. [b] 23% of reduced product was isolated along with oxazine. [c] diastereomeric ratio 3:1 was determined from 1H NMR spectroscopy.

General procedure for the preparation of Oxazine : GP II

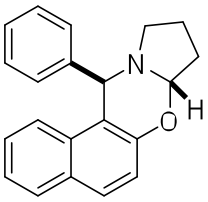
Aldehyde (2.5 eq) or ketone (0.29 mmol, 1.2 eq), tolune (1.5 mL) and amine (0.24 mmol, 1.0 eq) were added successively to an oven dried microwave reaction tube containing a stirring bar. Then the tube was sealed with cap and resulting solution was heated at 145 ^{o}C for 20 min under microwave irradiation (200 watt). Then the reaction mixture was cooled to room temperature. After that the crude mixture was transferred to round bottom flask with DCM. Then the volatiles were removed under vacuum to give gummy liquid. The liquid was subjected to SiO_2-gel column chromatography to afford analytically pure oxazine.

8,9,10,12-tetrahydro-7aH-naphtho[1,2-e]pyrrolo[2,1-b][1,3]oxazine (2a):[5] According to GP



II: 2-Hydroxy napthaldehyde (0.10 g, 0.60 mmol), pyrrolidine (20 µL, 0.24 mmol) in 1 mL of toluene 130 ^{o}C under microwave irradiation for 20 min and SiO_2 column chromatography (EtOAc : Hexane, 1 : 30) gave **2a** brown solid (39 mg, 72%). ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.77 – 7.76 (m, 1H), 7.64 – 7.63 (m, 2H), 7.49 – 7.46 (m, 1H), 7.36 – 7.34 (m, 1H), 7.01 (d, *J* = 8.9 Hz, 1H), 5.14 – 5.13 (m, 1H), 4.62 (d, *J* = 16.9 Hz, 1H), 4.28 (d, *J* = 17.0 Hz, 1H), 3.15 (td, *J* = 8.6, 3.1 Hz, 1H), 2.97 (q, *J* = 8.4 Hz, 1H), 2.24 – 2.14 (m, 2H), 2.09 – 1.96 (m, 2H). HRMS exact mass calculated for $C_{15}H_{16}NO^{+}$ ($[M+H]^{+}$) : 226.1226; Found : 226.1245.

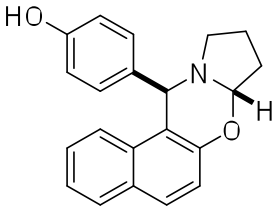
***rac*-(7aS,12R)-12-phenyl-8,9,10,12-tetrahydro-7aH-naphtho[1,2-e]pyrrolo[2,1-b][1,3]oxazine (2b):** According to GP II: Pyrrolidine (20 µL, 0.24 mmol), (2-hydroxynaphthalen-1-yl)(phenyl)methanone (72 mg, 0.29 mmol) in 1 mL of toluene 145 ^{o}C under microwave irradiation for 20 min and SiO_2 column chromatography (EtOAc : Hexane, 1 : 40) gave **2b**[4,5] as white solid (69 mg, 96%). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.70 – 7.67 (m, 1H), 7.65 (d, *J* = 9.0 Hz, 1H), 7.33 – 7.29 (m, 1H), 7.21 – 7.13 (m, 7H), 7.00 (d, *J* = 8.9 Hz, 1H), 5.38 (s, 1H), 5.02 – 4.99 (m, 1H), 3.29 – 3.22 (m, 1H), 2.84 (q, *J* = 8.3 Hz, 1H), 2.03 – 1.89 (m, 4H). HRMS exact mass calculated for $C_{21}H_{20}NO^{+}$ ($[M+H]^{+}$) : 302.1539; Found : 302.1533.

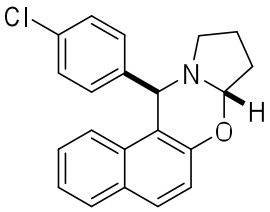


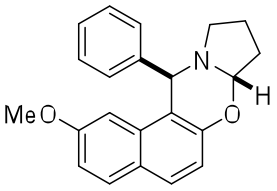
9-phenyl-2,3,3a,9-tetrahydro-1H-benzo[e]pyrrolo[2,1-b][1,3]oxazine (2c): According to GP II: 2-Hydroxy benzophenone (48 µL, 0.29 mmol), pyrrolidine (20 µL, 0.24 mmol) in 1 mL of toluene 145 ^{o}C under microwave irradiation for 20 min and SiO_2 column chromatography (EtOAc : Hexane, 1 : 40) gave diastereomeric mixture (3:1) of **2c**[4] as colorless oil (26 mg, 43%). ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.33 – 7.14 (m, 6H), 6.97 – 6.95 (m, 1H), 6.88 – 6.84 (m, 2H), 5.01 – 5.00 (m, 1H), 4.95 (s, 1H), 3.31 – 3.26 (m, 1H), 2.95 – 2.88 (m, 1H), 2.10 – 1.90 (m, 4H). HRMS exact mass calculated for $C_{17}H_{18}NO^+$ ($[M+H]^+$) : 252.1383; Found : 252.1385

***rac*-(7aS,12R)-12-(4-methoxyphenyl)-8,9,10,12-tetrahydro-7aH-naphtho[1,2-e]pyrrolo[2,1-b][1,3]oxazine (4a):** According to GP II: Pyrrolidine (40 µL, 0.49 mmol), (2-hydroxynaphthalen-1-yl)(4-methoxyphenyl)methanone (0.16 g, 0.58 mmol) in 1.5 mL of toluene 145 ^{o}C under microwave irradiation for 20 min, SiO_2 column chromatography (EtOAc : Hexane 1 : 40) gave **4a**[4, 5] as whitish solid (0.12 g, 74%). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.75 –7.73 (m, 1H), 7.70 (d, *J* = 8.9 Hz, 1H), 7.41 – 7.37 (m, 1H), 7.31 – 7.23 (m, 2H), 7.17 – 7.15 (m, 2H), 7.06 (d, *J* = 8.9 Hz, 1H), 6.81 –6.77 (m, 2H), 5.40 (s, 1H), 5.09 – 5.08 (m, 1H), 3.74 (s, 3H), 3.34 – 3.27 (m, 1H), 2.91 – 2.85 (m, 1H), 2.14 – 1.93 (m, 4H). HRMS exact mass calculated for $C_{22}H_{22}NO_2^+$ ($[M+H]^+$) : 332.1645; Found : 332.1660.

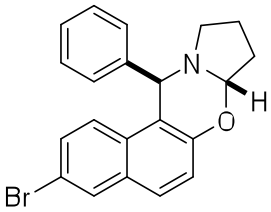
***rac*-(7aS,12R)-12-p-tolyl-8,9,10,12-tetrahydro-7aH-naphtho[1,2-e]pyrrolo[2,1-b][1,3]oxazine (4b):** According to GP II: Pyrrolidine (40 µL, 0.49 mmol) , 2-hydroxynaphthalen-1-yl)(p-tolyl)methanone (0.15 g, 0.58 mmol) in 1.5 mL of toluene 145 ^{o}C under microwave irradiation for 20 min, SiO_2 column chromatography (EtOAc : Hexane, 1 : 40) gave **4b**[4,5] as white solid (92 mg, 61%). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.62 (d, *J* = 7.7 Hz, 1H), 7.58 (d, *J* = 9.0 Hz, 1H), 7.30 – 7.27 (m, 1H), 7.17 – 7.11 (m, 2H), 7.04 – 7.02 (m, 2H), 6.98 – 6.94 (m, 3H), 5.30 (s, 1H), 5.00 – 4.98 (m, 1H), 3.19 (t, *J* = 7.6 Hz, 1H), 2.78 (q, *J* = 8.2 Hz, 1H), 2.17 (s, 3H), 2.01 – 1.82 (m, 4H). HRMS exact mass calculated for $C_{22}H_{22}NO^+$ ($[M+H]^+$) : 316.1696; Found : 316.1773.

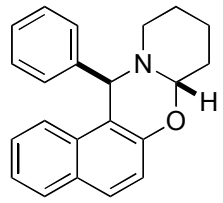
***rac*-4-((7aS,12R)-8,9,10,12-tetrahydro-7aH-naphtho[1,2-e]pyrrolo[2,1-b][1,3]oxazin-12-yl)phenol (4c):**  According to GP II: Pyrrolidine (30 µL, 0.36 mmol) , 2-hydroxynaphthalen-1-yl)(4-hydroxyphenyl)methanone (0.12 g, 0.44 mmol) in 1.5 mL of toluene and 1mL xylene 145 ^{o}C under microwave irradiation for 40 min, SiO_2 column chromatography (EtOAc : Hexane, 1 : 10) gave **4c** as white solid (55 mg, 52%). FTIR (KBr): $\tilde{\nu}$ = 3441, 2917, 2850, 1625, 1568, 1432, 1421, 1229, 991, 899, 816, 668 cm^{-1}. ^{1}H NMR (400 MHz, $CDCl_3$ with added Methanol-d_4) δ = 7.64 (d, *J* = 7.6 Hz, 1H), 7.60 (d, *J* = 9.1 Hz, 1H), 7.30 (d, *J* = 8.0 Hz, 1H), 7.19 – 7.13 (m, 2H), 6.99 – 6.94 (m, 3H), 6.63 – 6.61 (m, 2H), 5.29 (s, 1H), 5.02 – 5.01 (m, 1H), 3.24 – 3.17 (m, 1H), 2.81 – 2.75 (m, 1H), 2.01 – 1.90 (m, 4H). ^{13}C NMR (101 MHz, $CDCl_3$ with added Methanol-d_4) δ = 155.7, 150.9, 134.0, 132.0, 129.5, 128.7, 128.4, 128.0, 125.9, 122.6, 122.3, 118.1, 114.7, 110.3, 85.5, 55.6, 49.6, 31.4, 20.3. HRMS exact mass calculated for $C_{21}H_{20}NO_2^+$ ($[M+H]^+$): 318.1489; Found : 318.1488

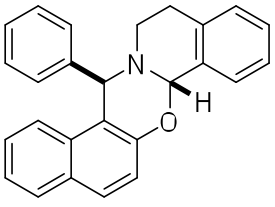
***rac*-(7aS,12R)-12-(4-chlorophenyl)-8,9,10,12-tetrahydro-7aH-naphtho[1,2-e]pyrrolo[2,1-b][1,3]oxazine (4d):**  According to GP II: Pyrrolidine (16 µL, 0.19 mmol), (4-chlorophenyl)(2-hydroxynaphthalen-1-yl)methanone (66 mg, 0.24 mmol) in 1 mL of toluene 145 ^{o}C under microwave irradiation for 40 min, SiO_2 column chromatography (EtOAc : Hexane, 1 : 40) gave **4d**[4,5] as light yellow solid (44 mg, 67%). ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.77 – 7.73 (m, 1H), 7.71 (d, *J* = 8.9 Hz, 1H), 7.32 (d, *J* = 7.8 Hz, 1H), 7.31 – 7.26 (m, 2H), 7.23 – 7.20 (m, 2H), 7.18 – 7.16 (m, 2H), 7.06 (d, *J* = 8.9 Hz, 1H), 5.40 (s, 1H), 5.01 – 4.99 (m, 1H), 3.31 (td, *J* = 8.3, 3.1 Hz, 1H), 2.90 (q, *J* = 8.3 Hz, 1H), 2.13 – 2.06 (m, 1H), 2.06 – 1.95 (m, 3H). HRMS exact mass calculated for $C_{21}H_{19}ClNO^+$ ($[M+H]^+$) : 336.1150; Found : 336.1156

***rac*-(7aS,12R)-2-methoxy-12-phenyl-8,9,10,12-tetrahydro-7aH-naphtho[1,2-e]pyrrolo[2,1-b][1,3]oxazine (4e):**  According to GP II: Pyrrolidine (30 µL, 0.36 mmol), (2-hydroxy-7-methoxynaphthalen-1-yl)(phenyl)methanone (0.12 g, 0.44 mmol) in 1.5 mL of toluene 145 ^{o}C under microwave irradiation for 40 min, crystallization and after washing (10 X 1 mL of cold EtOAc : Hexane, 1 : 30) gave **4e**[4] as white solid (94 mg, 78%). ^{1}H

NMR (400 MHz $CDCl_3$) δ = 7.65 – 7.60 (m, 2H), 7.27 – 7.21 (m, 5H), 6.94 – 6.90 (m, 2H), 6.63 (s, 1H), 5.33 (s, 1H), 5.13 – 5.09 (m, 1H), 3.61 (s, 3H), 3.38 – 3.29 (m, 1H), 2.97 – 2.91 (m, 1H), 2.16 – 1.92 (m, 4H). HRMS exact mass calculated for $C_{22}H_{22}NO_2^+$ ($[M+H]^+$) : 332.1645; Found: 332.1654

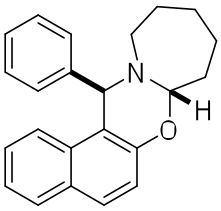
***rac*-(7aS,12R)-3-bromo-12-phenyl-8,9,10,12-tetrahydro-7aH-naphtho[1,2-e]pyrrolo[2,1-b][1,3]oxazine (4f):**  According to GP II: Pyrrolidine (20 µL, 0.24 mmol), (6-bromo-2-hydroxynaphthalen-1-yl)(phenyl)methanone (95 mg, 0.29 mmol) in 1 mL of toluene 145 ^oC under microwave irradiation for 20 min and SiO_2 column chromatography (EtOAc : Hexane, 1 : 40) gave **4f**[4] as brown solid (70 mg, 75%). ^1H NMR (600 MHz, $CDCl_3$) δ = 7.89 – 7.87 (m, 1H), 7.61 (d, *J* = 9.0 Hz, 1H), 7.33 (dd, *J* = 9.0, 2.0 Hz, 1H), 7.28 – 7.20 (m, 6H), 7.08 (d, *J* = 9.0 Hz, 1H), 5.40 (s, 1H), 5.07 – 5.06 (m, 1H), 3.35 – 3.32 (m, 1H), 2.91 – 2.87 (m, 1H), 2.11 – 1.98 (m, 4H). HRMS exact mass calculated for $C_{21}H_{19}BrNO_2^+$ ($[M+H]^+$) : 380.0645; Found : 380.0639

***rac*-(7aS,13R)-13-phenyl-7a,8,9,10,11,13-hexahydronaphtho[1,2-e]pyrido[2,1-b][1,3]oxazine (4g):**  According to GP II: Piperidine (30 µL, 0.30 mmol), (2-hydroxynaphthalen-1-yl)(phenyl)methanone (90 mg, 0.36 mmol) in 1 mL of toluene 145 ^oC under microwave irradiation for 40 min and SiO_2 column chromatography (EtOAc : Hexane, 1 : 40) gave **4g**[4,5] as white solid (43 mg, 45%). ^1H NMR (600 MHz, $CDCl_3$) δ = 7.77 – 7.76 (m, 1H), 7.73 (d, *J* = 8.8 Hz, 1H), 7.37 – 7.34 (m, 1H), 7.28 – 7.23 (m, 7H), 7.14 – 7.12 (m, 1H), 5.17 (s, 1H), 4.90 – 4.88 (m, 1H), 2.87 – 2.83 (m, 2H), 1.98 – 1.95 (m, 1H), 1.81 – 1.72 (m, 3H), 1.60 – 1.97 (m, 2H). HRMS exact mass calculated for $C_{22}H_{22}NO^+$ ($[M+H]^+$) : 316.1696; Found : 316.1696

Oxazine (4h):  According to GP II: 1,2,3,4-tetrahydroisoquinoline (42 µL, 0.33 mmol), (2-hydroxynaphthalen-1-yl)(phenyl)methanone (0.10 g, 0.40 mmol) in 1.5 mL of toluene 20 min 145 ^oC under microwave irradiation and SiO_2 column chromatography (EtOAc : Hexane, 1 : 40) gave **4h** as white solid (81 mg, 66%). FTIR (KBr): $\tilde{\nu}$ = 2921, 2855, 1621, 1597, 1463, 1402, 1257, 1235, 1137, 1091, 1067, 985, 878, 857, 748, 725, 698 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$) δ = 7.78 (d, *J* = 6.8 Hz, 1H), 7.73 (d, *J* = 9.0 Hz, 1H), 7.43 – 7.41 (m,

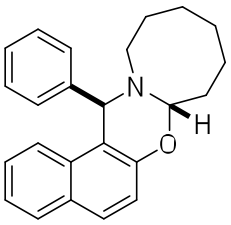
1H), 7.31 – 7.30 (m, 7H), 7.24 – 7.18 (m, 4H), 7.11 (d, *J* = 9.0 Hz, 1H), 5.65 (s, 1H), 5.43 (s, 1H), 3.40 – 3.24 (m, 2H), 3.11 – 3.08 (m, 1H), 2.89 – 2.85 (m, 1H). ^{13}C NMR (101 MHz, $CDCl_3$) δ = 152.2, 142.6, 135.2, 133.3, 132.6, 129.5, 129.3, 129.1, 129.0, 128.98, 128.96, 128.8, 128.5, 127.6, 126.8, 126.4, 123.3, 122.9, 119.1, 111.1, 82.4, 62.9, 45.6, 29.6. HRMS (ESI) exact mass calculated for $C_{26}H_{22}NO^+$ ($[M+H]^+$): 364.1696. Found: 364.1684.

***rac*-(7aS,13R)-13-Phenyl-7a,8,9,10,11,12-hexahydro-13H-7-oxa-12a-aza-cyclohepta**



[b]phenanthrene (4i): According to GP II: Hexamethyleneimine (30 µL, 0.26 mmol), (2-hydroxynaphthalen-1-yl)(phenyl)methanone (79 mg, 0.32 mmol) in 1 mL of toluene 145 °C under microwave irradiation for 40 min SiO_2 and column chromatography (EtOAc : Hexane, 1 : 40) gave **4i**[4,5] as white solid (57 mg, 66%). ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.75 – 7.74 (m, 1H), 7.71 (d, *J* = 8.9 Hz, 1H), 7.34 – 7.33 (m, 1H), 7.28 – 7.24 (m, 6H), 7.22 – 7.19 (m, 1H), 7.10 (dd, *J* = 8.9, 2.0 Hz, 1H), 5.29 (s, 1H), 4.87 – 4.85 (m, 1H), 3.26 – 3.22 (m, 1H), 2.69 – 2.67 (m, 1H), 2.22 – 2.17 (m, 1H), 1.89 – 1.82 (m, 2H), 1.78 – 1.71 (m, 2H), 1.67 – 1.64 (m, 1H), 1.50 – 1.44 (m, 1H), 1.43 – 1.35 (m, 1H). HRMS exact mass calculated for $C_{23}H_{24}NO^+$ ($[M+H]^+$) : 330.1852; Found : 330.1822

Oxazine (4j): According to GP II: Heptamethyleneimine (40 µL, 0.31 mmol), (2-hydroxynaphthalen-1-yl)(phenyl)methanone (94 mg, 0.37 mmol) in 1 mL of toluene first 40 min 100 °C then another 10 min for 145 °C under microwave irradiation and SiO_2 column chromatography (EtOAc : Hexane, 1 : 60) gave **4j**[5] as light yellow solid (41 mg, 38%). ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.76 – 7.75 (m, 1H), 7.71 (d, *J* = 8.9 Hz, 1H), 7.34 – 7.33 (m, 1H), 7.29 – 7.21 (m, 7H), 7.09 (d, *J* = 8.9 Hz, 1H), 5.28 (s, 1H), 4.70 – 4.67 (m, 1H), 3.28 – 3.23 (m, 1H), 2.59 – 2.55 (m, 1H), 1.98 – 1.87 (m, 5H), 1.55 – 1.51 (m, 2H), 1.48 – 1.43 (m, 3H). HRMS (ESI) exact mass calculated for $C_{24}H_{26}NO^+$ ($[M+H]^+$) : 344.2009; Found : 344.2098



2,4-di-tert-butyl-5a,6,7,8,9,11-hexahydrobenzo[e]pyrido[2,1-b][1,3]oxazine (4k): According to GP II: Piperidine (35 µL, 0.36 mmol), 3,5 di tert-butyl salicylaldehyde (0.10 g, 0.43 mmol) in 1.5 mL of toluene 30 min 145 ^{o}C under microwave irradiation and SiO_2 column chromatography (EtOAc : Hexane, 1:25) gave **4k** as yellowish oil (51 mg, 47%). FTIR (KBr): $\tilde{\nu}$ = 2949, 2865, 1652, 1641, 1479, 1449, 1360, 1226, 1128, 1103, 1069, 879, 870, 758 cm^{-1}. ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.15 (s, 1H), 6.81 (s, 1H), 4.85 – 4.83 (m, 1H), 4.30 (d, *J* = 16.1 Hz, 1H), 3.61 (d, *J* = 16.1 Hz, 1H), 2.94 – 2.90 (m, 1H), 2.54 – 2.51 (m, 1H), 2.01 – 1.98 (m, 1H), 1.87 – 1.83 (m, 1H), 1.78 – 1.70 (m, 3H), 1.61 – 1.57 (m, 1H), 1.40 (s, 9H), 1.29 (s, 9H). ^{13}C NMR (101 MHz, $CDCl_3$) δ = 150.3, 142.0, 136.4, 121.9, 121.7, 118.3, 86.3, 54.7, 48.2, 35.1, 34.4, 31.8, 30.7, 29.9, 25.5, 19.4. HRMS (ESI) exact mass calculated for $C_{20}H_{32}NO^{+}$ ([M+H]$^{+}$): 302.2478; Found : 302.2478.

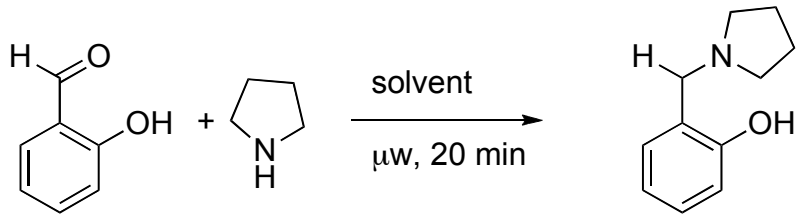
10-methyl-8,9,10,12-tetrahydro-7aH-naphtho[1,2-e]pyrrolo[2,1-b][1,3]oxazine (4l):

According to GP II: 2-Hydroxy napthaldehyde (86 mg, 0.50 mmol), 1-methyl pyrrolidine (20 µL, 0.19 mmol) in 1 mL of toluene 130 ^{o}C under microwave irradiation for 20 min and SiO_2 column chromatography (EtOAc : Hexane, 1 : 20) gave inseparable regioisomeric mixture (~5:1) of **4l** as brown solid (22 mg, 46%). FTIR (KBr): $\tilde{\nu}$ = 2960, 2920, 2853, 1635, 1516, 1467, 1434, 1396, 1261, 1228, 1095, 1074, 1016, 860, 811, 746 cm^{-1}. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.76 (d, *J* = 8.0 Hz, 1H), 7.67 (d, *J* = 8.6 Hz, 1H), 7.63 (d, *J* = 8.8 Hz, 1H), 7.50 – 7.47 (m, 1H), 7.36 – 7.32 (m, 1H), 6.99 (d, *J* = 8.9 Hz, 1H), 5.26 – 5.25 (m, 1H), 4.58 (d, *J* = 17.4 Hz, 1H), 4.27 (d, *J* = 17.5 Hz, 1H), 3.15 – 3.10 (m, 1H), 2.29 – 2.03 (m, 4H), 1.23 (d, *J* = 6.0 Hz, 3H). HRMS (ESI) exact mass calculated for $C_{16}H_{18}NO^{+}$ ([M+H]$^{+}$): 240.1383; Found: 240.1381

7a-methyl-12-phenyl-8,9,10,12-tetrahydro-7aH-naphtho[1,2-e]pyrrolo[2,1-b][1,3]oxazine (4m): According to GP II: 2-methyl pyrrolidine (34 µL, 0.33 mmol), (2-hydroxynaphthalen-1-yl)(phenyl)methanone (0.10 g, 0.40 mmol) in 1.5 mL of toluene 20 min 145 ^{o}C under microwave irradiation and SiO_2 column chromatography (EtOAc : Hexane, 1 : 30) gave inseparable regioisomeric mixture (~4:1) of **4m** as yellowish oil (85 mg, 82%). FTIR (KBr): $\tilde{\nu}$ = 2935, 2850, 1623, 1599, 1514, 1462, 1380, 1241, 1109, 1065, 972, 811, 746, 702, 623 cm^{-1}. ^{1}H NMR (400 MHz, $CDCl_3$)

δ = 7.80 – 7.78 (m, 1H), 7.75 (d, *J* = 8.9 Hz, 1H), 7.31 – 7.19 (m, 8H), 7.09 (d, *J* = 8.9 Hz, 1H), 5.50 (s, 1H), 3.52 – 3.47 (m, 1H), 2.99 (q, *J* = 8.7 Hz, 1H), 2.28 – 2.22 (m, 1H), 2.10 – 2.07 (m, 1H), 1.96 – 1.87 (m, 2H), 1.04 (s, 3H). HRMS (ESI) exact mass calculated for $C_{22}H_{22}NO^+$ ($[M+H]^+$): 316.1696. Found: 316.1685

Table S2: Optimization of formal reductive amination

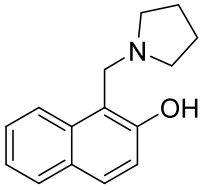


entry	pyrrolidine	solvent	temperature	Yield (%)
1	1.0 eq	Ethylene glycol	150 °C	36
2	1.0 eq	DMF	150 °C	41
3	2.0 eq	DMF	150 °C	67
4	2.0 eq	Ethylene glycol	150 °C	63
5	1.0 eq	no solvent	150 °C	30
6	2.0 eq	no solvent	150 °C	50
7	2.2 eq	m-xylene	170 °C	91

General procedure for formal reductive amination: GP III

Aldehyde or ketone (0.82 mmol, 1.0 eq.), m-xylene (1.5 mL) and amine (1.8 mmol, 2.2 eq.) were added sucssively to an oven dried microwave reaction tube conatining a stirring bar. Then the tube was sealed with cap and resulting solution was heated at 170 °C for 20 min under micorwave irradiation (200 watt). Then the reaction mixture was cooled to room temperature. Reaction mixture was quenched with aqueous 1N NH_4Cl solution (15 mL) extracted with (3×20 mL) EtOAc. Then combined organic layers ware washed with brine (30 mL) dried (Na_2SO_4), concentrated in vaccua. The crude product was purified by SiO_2-gel column chromatography.

1-((pyrrolidin-1-yl)methyl)naphthalen-2-ol (5a): According to GP III: Pyrrolidine (0.12 mL, 1.45 mmol), 2- hydroxy- naphthaldehyde (0.10 g, 0.58 mmol) in 1.5 mL m-xylene, 170 °C under microwave irradiation for 20 min and SiO_2- column chromatography (EtOAc/Hexane, 1:2) gave **5a**[4] light brown liquid (97 mg, 74 %). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 10.65 (s, 1H), 7.72 – 7.66 (m, 2H), 7.60



(d, J = 8 Hz, 1H), 7.37 – 7.34 (m, 1H), 7.22 –7.19 (m, 1H), 7.09 – 7.07 (m, 1H), 4.14 (s, 2H), 2.58 (br. s, 4H), 1.74 (br. s, 4H). HRMS (ESI) exact mass calculated for $C_{15}H_{18}NO^+$ ($[M+H]^+$) : 228.1383; Found : 228.1378

2-Pyrrolidin-1-ylmethyl-phenol (5b): According to GP III: Pyrrolidine (0.15 mL, 1.8 mmol), salicylaldehyde (86 µL, 0.82 mmol) in 1.5 mL m-xylene, 170 ^oC under microwave irradiation for 20 min and SiO_2- column chromatography (EtOAc/Hexane, 1:7) gave **5b**[5] as a light brown liquid (0.13 g, 91 %). ^1H NMR (600 MHz, $CDCl_3$) δ = 7.159 (t, J = 7.8 Hz, 1H), 6.96 (d, J = 7.20 Hz, 1H), 6.81 (d, J = 7.8 Hz, 1H), 6.75 (t, J = 7.2 Hz, 1H), 3.81 (s, 2H), 2.62 (s, 4H), 1.84 (s, 4H). HRMS (ESI) exact mass calculated for $C_{11}H_{16}NO^+$ ($[M+H]^+$) : 178.1226; Found : 178.1232

2,4-Di-tert-butyl-6-pyrrolidin-1-ylmethyl-phenol (5c): According to GP III: Pyrrolidine (78 µL, 0.95 mmol) , 3,5 ditertiary butyl salicyldehyde (0.10 g, 0.43 mmol) in 1.5 mL m-xylene, 170 ^oC under microwave irradiation for 20 min and SiO_2-column chromatography (EtOAc/Hexane, 1:30) gave **5c**[6] yellow solid product (92 mg, 74 %). ^1H NMR (600 MHz, $CDCl_3$) δ = 7.21 (d, J = 2.4 Hz, 1H), 6.84 (d, J = 2.4 Hz, 1H), 3.80 (s, 2H), 2.63 (s, 4H), 1.85 – 1.84 (m, 4H), 1.43 (s, 9H), 1.29 (s, 9H). HRMS (ESI) exact mass calculated for $C_{19}H_{32}NO^+$ ($[M+H]^+$) : 290.2478; Found : 290.2486

2-[(Ethyl-methyl-amino)-methyl]-6-methoxy-phenol (5d): According to GP III: Pyrrolidine (0.12 mL, 1.45 mmol), *ortho*-vanillin (0.10 g, 0.66 mmol) in 1.5 mL m-xylene, 170 ^oC under microwave irradiation for 20 min and SiO_2-column chromatography (EtOAc/Hexane, 1:1) gave **5d** yellow liquid product (83 mg, 61 %). FTIR (KBr): ῡ = 3451, 2933, 2833, 1639, 1478, 1414, 1240, 1075, 733, 709 cm^{-1}. ^1H NMR (600 MHz, $CDCl_3$) δ = 6.80 (d, J = 7.8 Hz, 1H), 6.71 (t, J = 7.8 Hz, 1H), 6.60 (d, J = 7.2 Hz, 1H), 3.87 (s, 3H), 3.84 (s, 2H), 2.64 (br. s, 4H), 1.84-1.83 (m, 4H). ^{13}C NMR (100 MHz, $CDCl_3$), δ = 148.07, 147.59, 122.78, 120.24, 118.60, 111.03, 58.63, 56.08, 53.63, 23.83. HRMS (ESI) exact mass calculated for $C_{12}H_{18}NO_2^+$($[M+H]^+$): 208.1332, found: 208.1332.

1-Piperidin-1-ylmethyl-naphthalen-2-ol (5e): According to GP III: Piperidine (0.13 mL, 1.28 mmol), 2- hydroxy- naphthaldehyde (0.10 g, 0.58 mmol) in 1.5 mL m-xylene, 170 ^{o}C under microwave irradiation for 20 min and SiO_2-column chromatography (EtOAc/Hexane, 1:5) gave **5e**[7] light brown liquid product (83 mg, 60 %). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.79 (d, *J* = 8.8 Hz, 1H), 7.73 (d, *J* = 8.0 Hz, 1H), 7.66 (d, *J* = 8.4 Hz, 1H), 7.43-7.39 (m, 1H), 7.28-7.24 (m, 1Hz), 7.07 (d, *J* = 8.8 Hz, 1H), 4.10 (s, 2H), 3.45-1.51 (m, 10H). HRMS (ESI) exact mass calculated for $C_{16}H_{20}NO^+$ ($[M+H]^+$) : 242.1539; Found : 242.1544

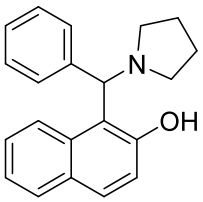
2-Piperidin-1-ylmethyl-phenol (5f): According to GP III: Piperidine (0.18 mL, 1.82 mmol), salicylaldehyde (87 µL, 0.82 mmol) in 1.5 mL *m*-xylene, 170 ^{o}C under microwave irradiation for 20 min and SiO_2-column chromatography (EtOAc/Hexane, 1:15) gave **5f**[8] light brown liquid product (0.13 g, 80 %). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.17 – 7.13 (m, 1H), 6.96 – 6.94 (m, 1H), 6.81 (dd, J_1 = 8.4 Hz, J_2 = 0.8 Hz, 1H), 6.77 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 3.66 (s, 2H), 2.70-2.20 (m, 3H), 1.65-1.61(m, 5H), 1.48-1.41 (m, 2H). HRMS (ESI) exact mass calculated for $C_{12}H_{18}NO^+$ ($[M+H]^+$) : 192.1383; Found : 192.1397

2,4-Di-tert-butyl-6-piperidin-1-ylmethyl-phenol (5g): According to GP III: Pipyridine (93 µL, 0.95 mmol), 3,5 ditertiary butyl salicyldehyde (0.10 g, 0.43 mmol) in 1.5 mL m-xylene, 170 ^{o}C under microwave irradiation for 20 min and SiO_2-column chromatography (EtOAc/Hexane, 1:30) gave **5g**[9] yellow solid product (89 mg, 67 %). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.20 (d, *J* = 2.0 Hz, 1H), 6.81 (d, *J* = 2.4 Hz, 1H), 3.63 (s, 2H), 2.98-1.43 (m, 10H), 1.42 (s, 9H), 1.27(s, 9H). HRMS (ESI) exact mass calculated for $C_{20}H_{34}NO^+$ ($[M+H]^+$) : 304.2635; Found : 304.2690

2-Azepan-1-ylmethyl-phenol (5h): According to GP III: Hexamethyleneimine (0.20 mL, 1.8 mmol), salicylaldehyde (86 µL, 0.82 mmol) in 1.5 mL m-xylene, 170 ^{o}C under microwave irradiation for 20 min and SiO_2-column chromatography (EtOAc/Hexane, 1:20) gave **5h** light brown liquid product (0.12 g, 72 %). FTIR (KBr): ῡ = 3472, 2926, 2848, 1635, 1479, 1258, 1145, 1045, 1033, 753 cm^{-1}. ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.17-7.14 (m, 1H), 6.94 (d, *J* = 7.8 Hz, 1H), 6.81 (d, *J* = 7.8 Hz, 1H), 6.77-6.74 (m, 1H), 3.78 (s, 2H), 2.70 (s, 4H), 1.69 (d, *J* = 4.8 Hz, 4H), 1.64-1.63 (m, 4H).

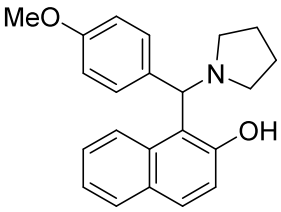
^{13}C NMR (100 MHz, $CDCl_3$), δ = 158.6, 128.8, 128.5, 122.4, 118.97, 116.2, 62.2, 55.4, 27.8, 26.8. HRMS (ESI) exact mass calculated for $C_{13}H_{20}NO^+$ ($[M+H]^+$): 206.1539, found: 206.1538.

1-(Phenyl-pyrrolidin-1-yl-methyl)-naphthalen-2-ol (5i): According to GP III: Pyrrolidine (72



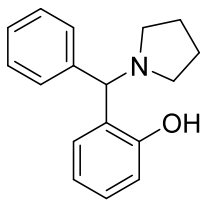
μL, 0.88 mmol), (2-Hydroxynaphthalen-1-yl)-phenyl-methanone (0.10 g, 0.40 mmol) in 1.5 mL m-xylene, 170 °C under microwave irradiation for 20 min and SiO_2-column chromatography (EtOAc/Hexane 1:20) gave **5i**[5] colourless solid product (75 mg, 62 %). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.87 (d, *J* = 8.4 Hz, 1H), 7.69 (d, *J* = 8.0 Hz, 1H), 7.65 (d, *J* = 9.2 Hz, 1H), 7.61 – 7.59 (m, 2H), 7.38-7.34 (m, 1H), 7.27-7.14 (m, 5H), 5.12 (s, 1H), 3.25-2.04 (m, 4H), 1.85 (s, 4H). HRMS (ESI) exact mass calculated for $C_{21}H_{22}NO^+$ ($[M+H]^+$) : 304.1696; Found : 304.1696

1-[(4-Methoxy-phenyl)-pyrrolidin-1-yl-methyl]-naphthalen-2-ol (5j): According to GP III:

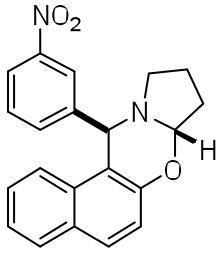


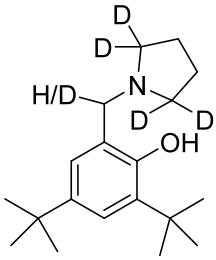
Pyrrolidine (64 μL, 0.79 mmol), (2-Hydroxy-naphthalen-1-yl)-(4-methoxy-phenyl)-methanone (0.10 g, 0.36 mmol) in 1.5 mL m-xylene, 170 °C under microwave irradiation for 20 min and SiO_2-column chromatography (EtOAc/Hexane, 1:7) gave **5j**[5] light brown liquid (73 mg, 61 %). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.84 (d, *J* = 8.4 Hz, 1H), 7.70-7.64 (m, 2H), 7.51- 7.49 (m, 2H), 7.38-7.34 (m, 1H), 7.24-7.13 (m, 2H), 6.78 (d, *J* = 7.2 Hz, 2H), 5.08 (s, 1H), 3.71 (s, 3H), 3.25-2.04 (m, 4H), 1.84 (br. s, 4H). HRMS (ESI) exact mass calculated for $C_{22}H_{24}NO_2^+$ ($[M+H]^+$) : 334.1802; Found : 334.1808

2-(2-Methylene-1-pyrrolidin-1-yl-pent-3-enyl)-phenol (5k): According to GPIII: Pyrrolidine



(92 μL, 1.12 mmol), 2- HydroxyBenzophenone (0.10 g, 0.51 mmol) in 1.5 mL m-xylene, 170 °C under microwave irradiation for 20 min and SiO2-column chromatography (EtOAc/Hexane, 1:30) gave **5k**[10] light brown liquid (0.10 g, 80 %). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.47 – 7.45 (m, 2H), 7.29 - 7.27 (m, 2H), 7.23-7.20 (m, 1H), 7.11-7.07 (m, 1H), 6.95 (d, *J* = 7.2 Hz, 1H), 6.84 (d, *J* = 8.0 Hz, 1H), 6.70 (t, *J* = 7.6 Hz, 1H), 4.37 (s, 1H), 2.63-2.49 (m, 4H), 1.87 – 1.83 (m, 4H). HRMS (ESI) exact mass calculated for $C_{17}H_{20}NO^+$ ($[M+H]^+$) : 254.1539; Found : 254.1540

***rac*-(7aS,12R)-12-(3-nitrophenyl)-8,9,10,12-tetrahydro-7aH-naphtho[1,2-e]pyrrolo[2,1-b][1,3]oxazine (15):**  According to GPII: 1-((3-nitrophenyl)(pyrrolidin-1-yl)methyl)naphthalen-2-ol (0.21 g, 0.60 mmol) (2-hydroxynaphthalen-1-yl)(phenyl)methanone (75 mg, 0.30 mmol) in 2 mL toluene 145 ^{o}C under microwave irradiation for 20 min and SiO_2 column chromatography (EtOAc : Hexane, 1 : 40) gave **2b** (19 mg, 21%) Compound **15**[5] was obtained after preparative TLC. (10 mg, 9 %). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 8.25 (s, 1H), 8.10 – 8.08 (m, 1H), 7.80 – 7.75 (m, 2H), 7.46 – 7.37 (m, 2H), 7.30 – 7.28 (m, 3H), 7.12 – 7.06 (m, 1H), 5.51 (s, 1H), 4.96 – 4.94 (m, 1H), 3.40 – 3.36 (m 1H), 2.96 – 2.92 (m, 1H), 2.15 – 2.03 (m, 4H). HRMS (ESI) exact mass calculated for $C_{21}H_{19}N_2O_3^+$ ($[M+H]^+$) : 347.1390; Found : 347.1395

2-[(2,2,5,5-$^{2}H_4$)pyrrolidin-1-ylmethyl]phenol (18) and **2-[(2,2,5,5-$^{2}H_4$)pyrrolidin-1-yl($^{2}H_1$)methyl]phenol (19):**  According to GP III: Deuterated Pyrrolidine (0.02 mL, 0.22 mmol), 3,5 ditertiary butyl salicyldehyde (23 mg, 0.10 mmol) in 0.34 mL m-xylene 170 ^{o}C under microwave irradiation for 20 min and SiO_2-column chromatography (EtOAc/Hexane 1:50 to 1:30) gave inseparable 1:1 mixture of **18** & **19** as yellowish solid (15 mg, 50 %). FTIR (KBr): ῡ = 3450, 2955, 2924, 2862, 2204, 2078, 1639, 1567, 1558, 1479, 1433, 1360, 1248, 1235, 1203, 1123, 1002, 878, 819, 792, 647, 647, 524 cm^{-1}. ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.2 (d, *J* = 2.4 Hz, 1H), 6.82 (d, *J* = 1.8 Hz, 1H), 3.79 (s, 1H), 3.76 (s, 0.5 H), 1.82 (s, 4H), 1.42 (s, 9H), 1.28 (s, 9H). ^{13}C NMR (150 MHz, $CDCl_3$), δ = 154.72, 140.36, 135.48, 122.87, 122.81, 122.02, 59.84, 35.06, 34.35, 31.94, 29.84, 23.76. HRMS (ESI) exact mass calculated for (**18**) $C_{19}H_{28}D_4NO$ ($[M+H]^+$): 294.2729; found: 294.2733. HRMS (ESI) exact mass calculated for (**19**) $C_{19}H_{27}D_5NO$ ($[M+H]^+$): 295.2792; found: 295.2796.

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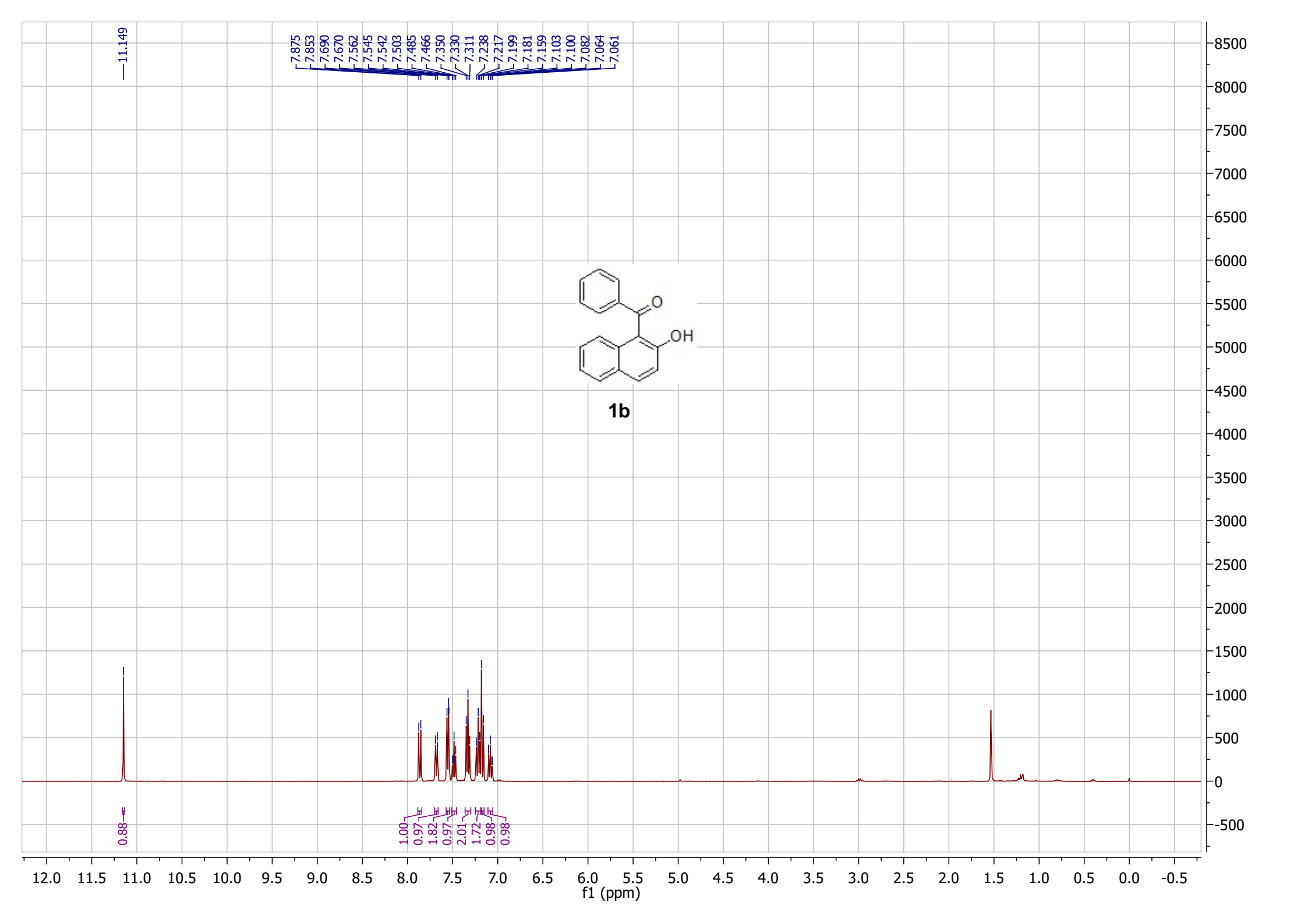
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7.082
7.064
7.061
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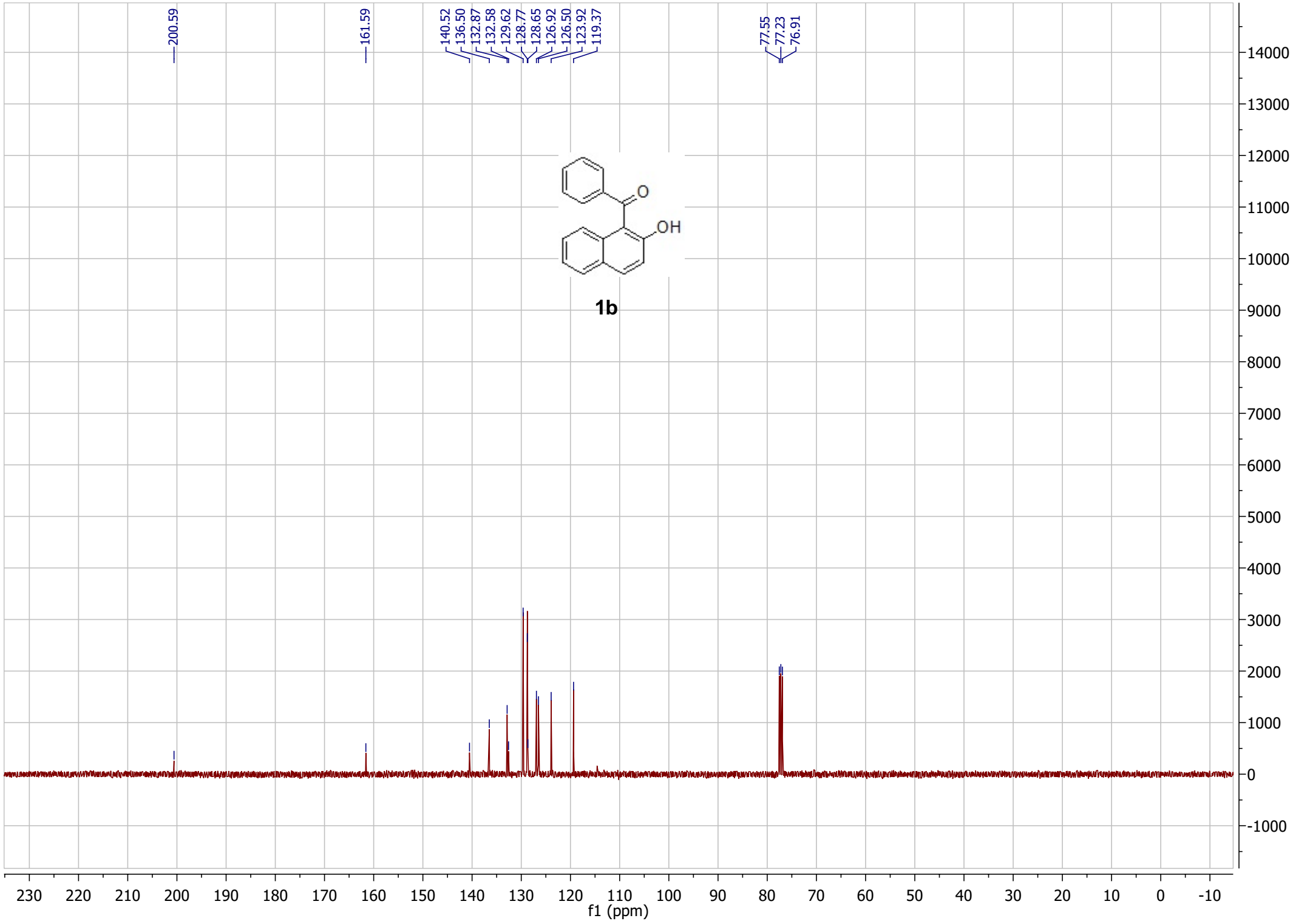
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O
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1b
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13000
12000
11000
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9000
8000
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6000
5000
4000
3000
2000
1000
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220
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200
190
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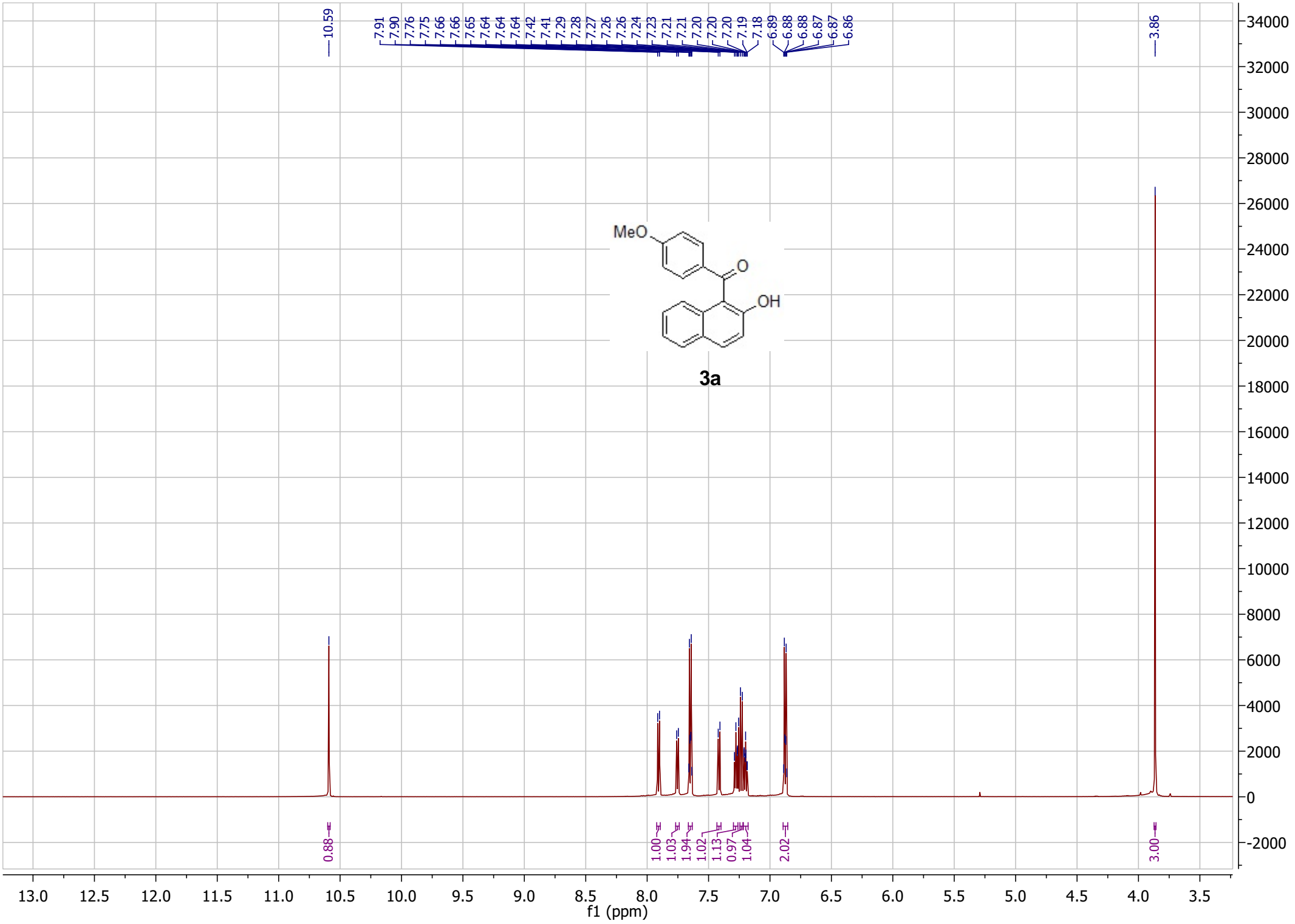
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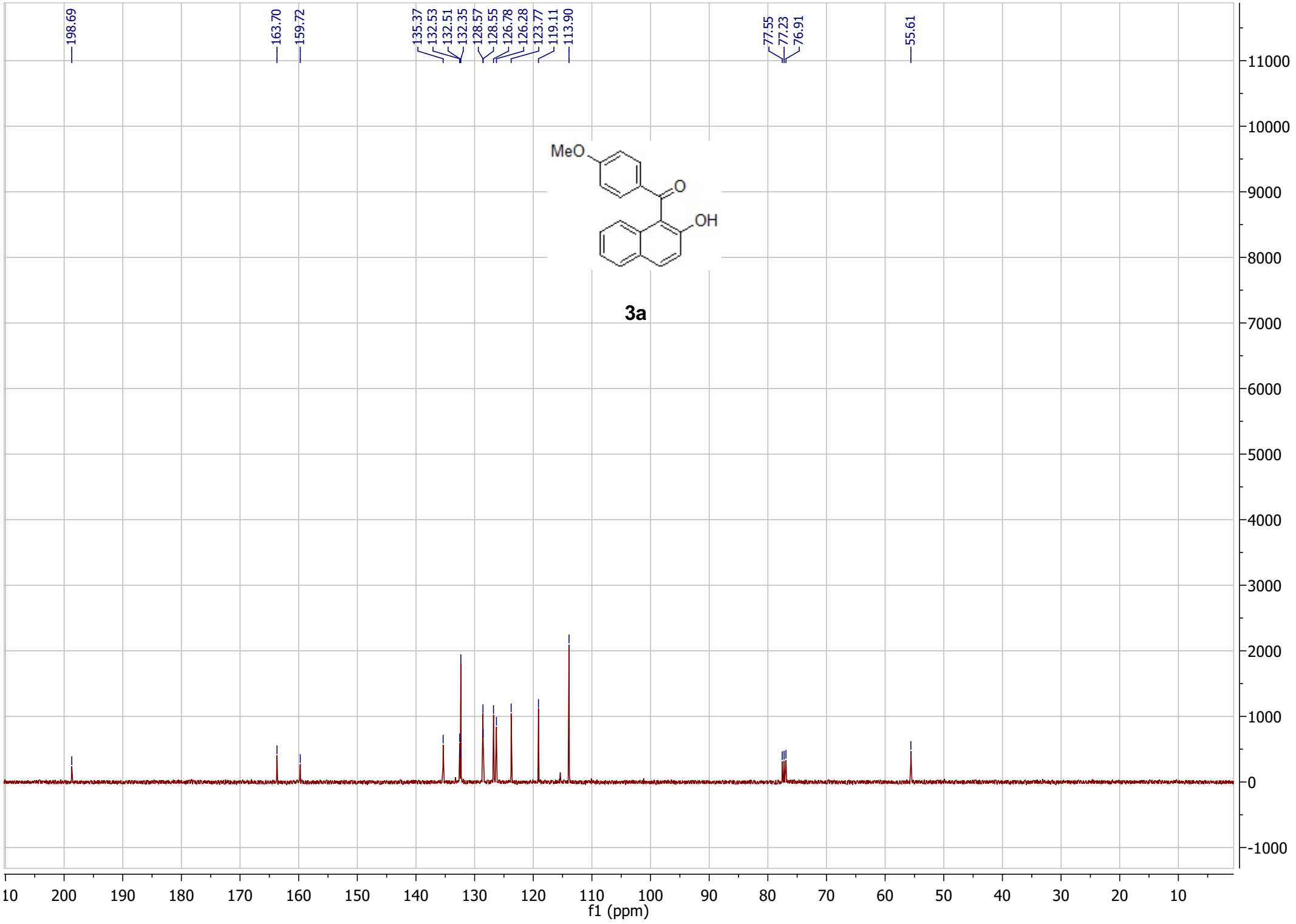
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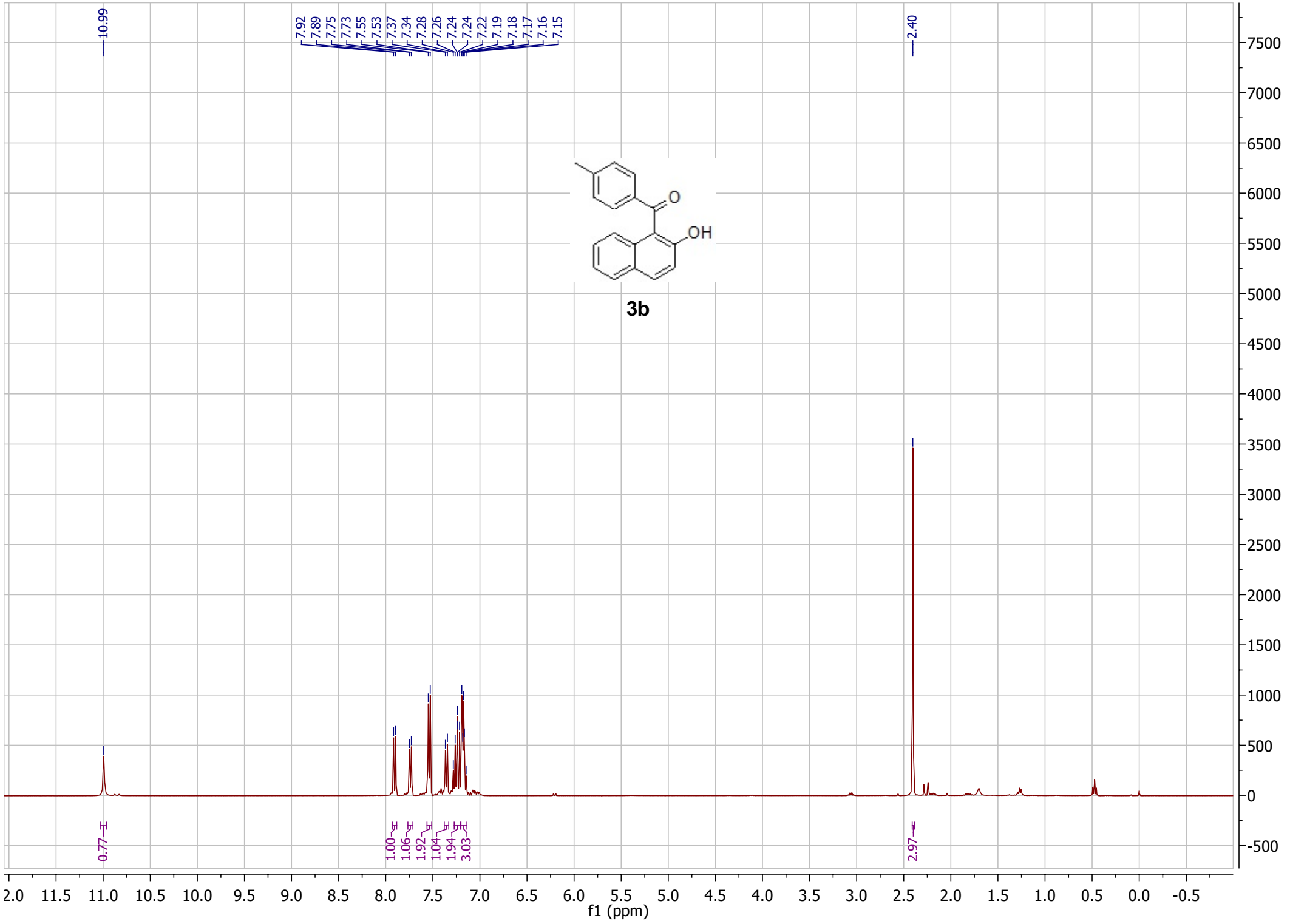
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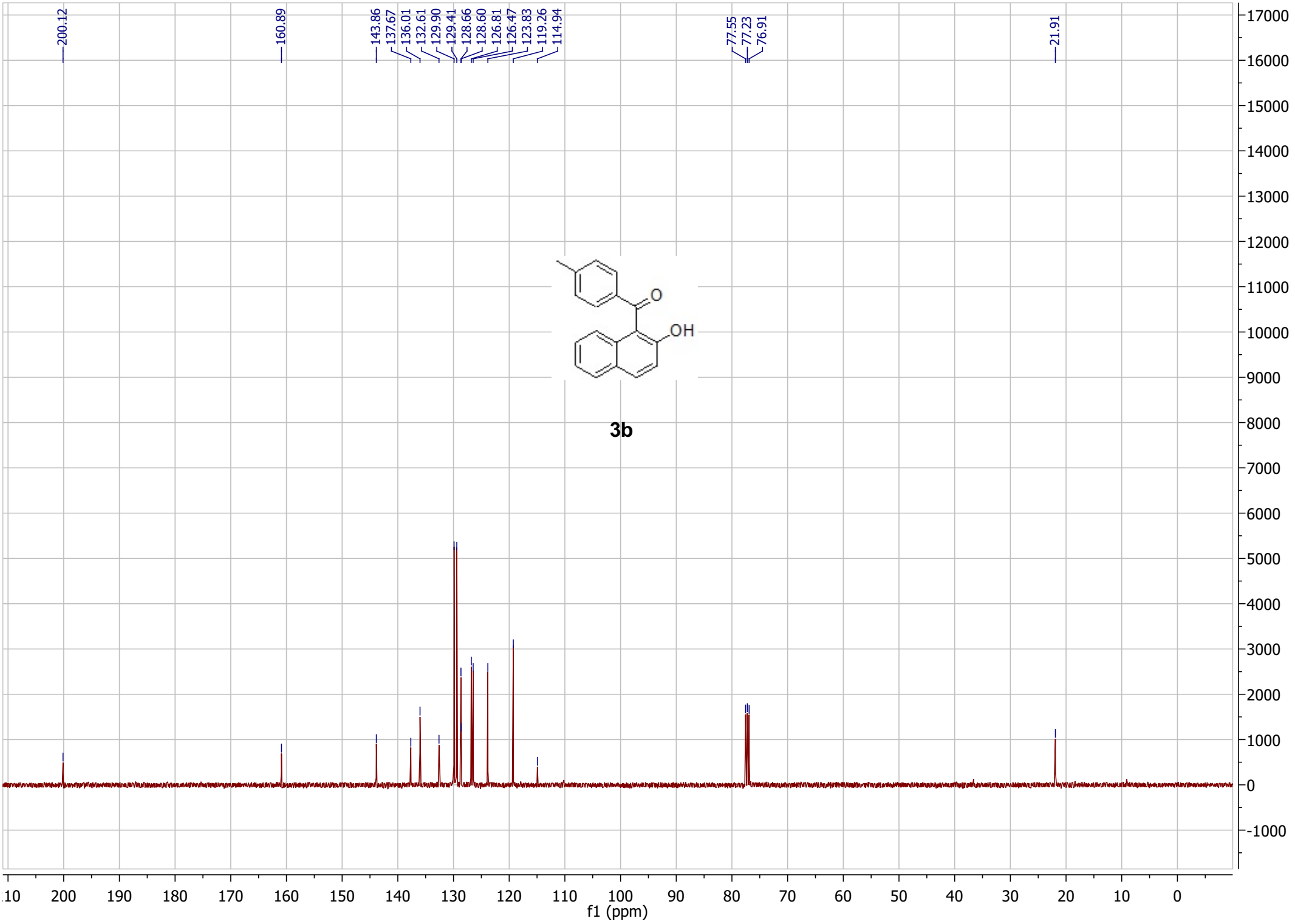
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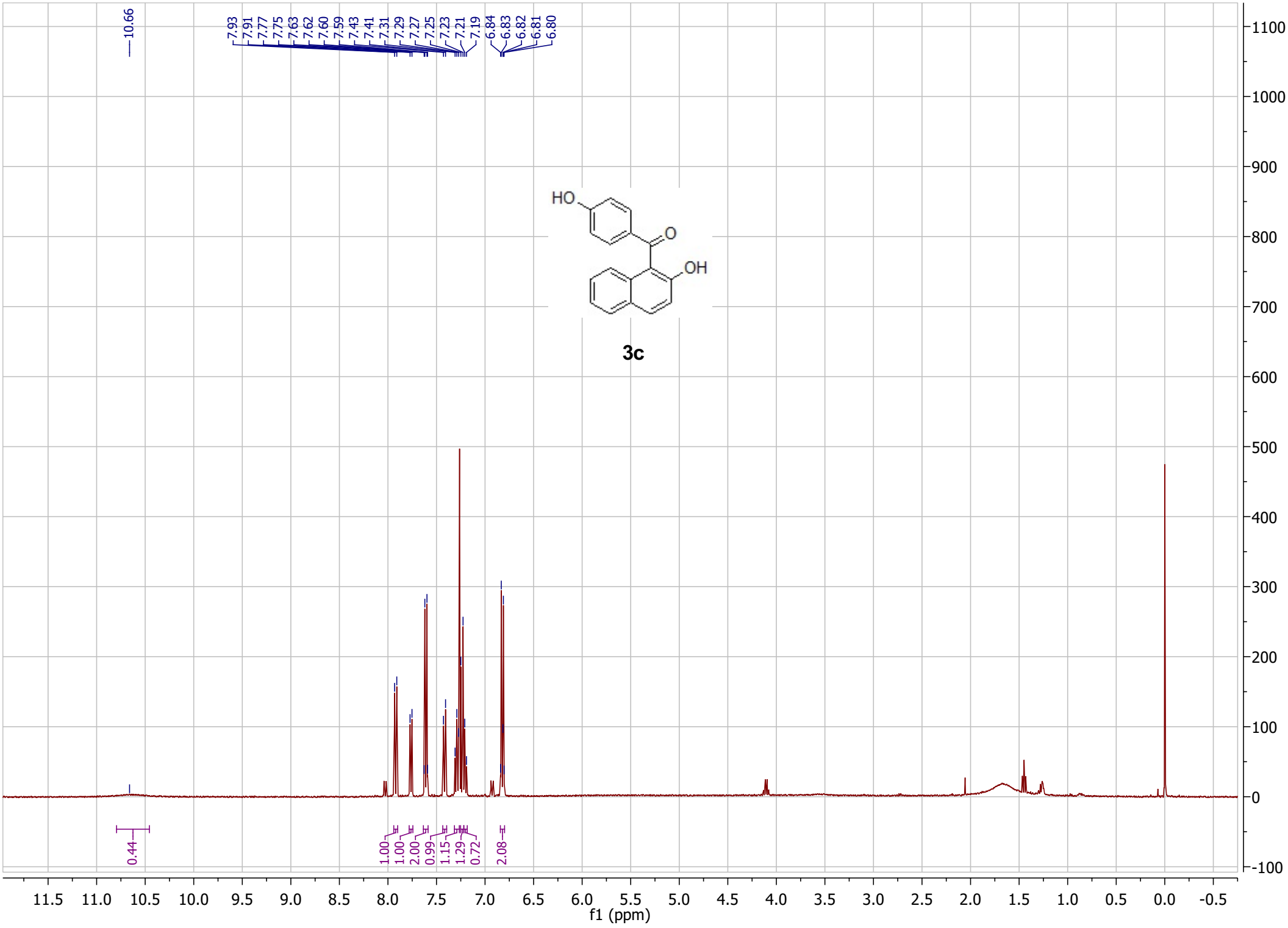
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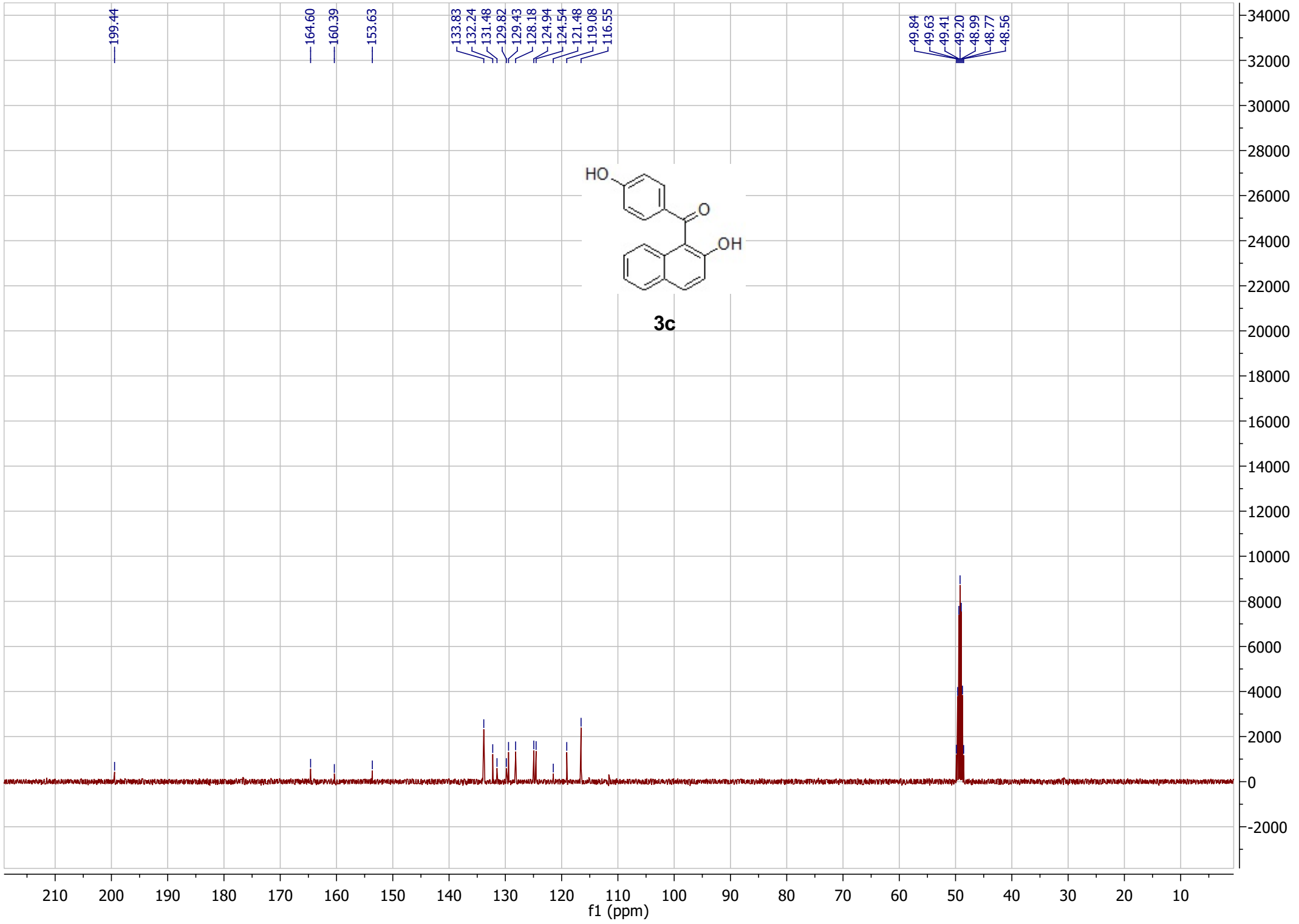
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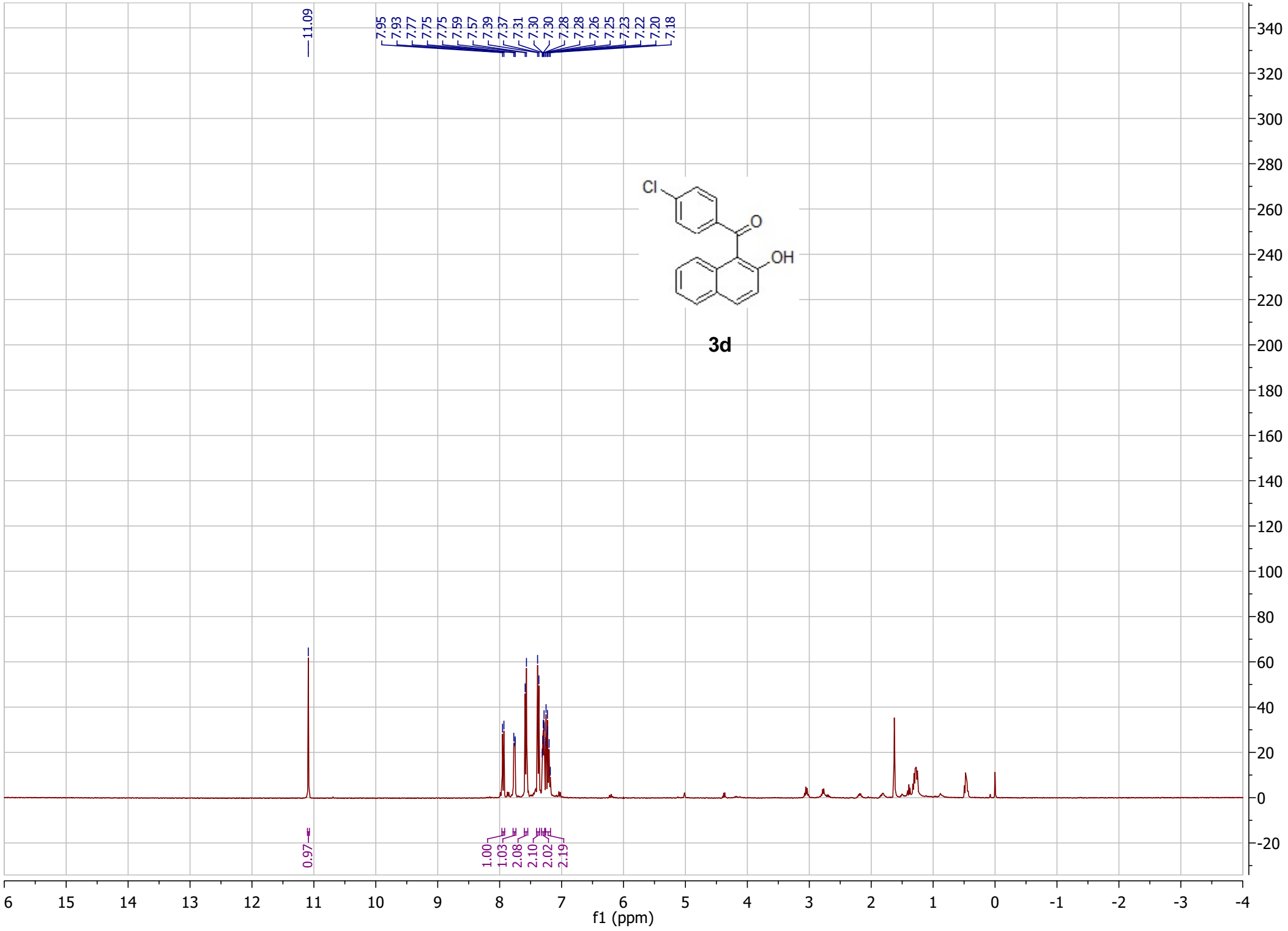
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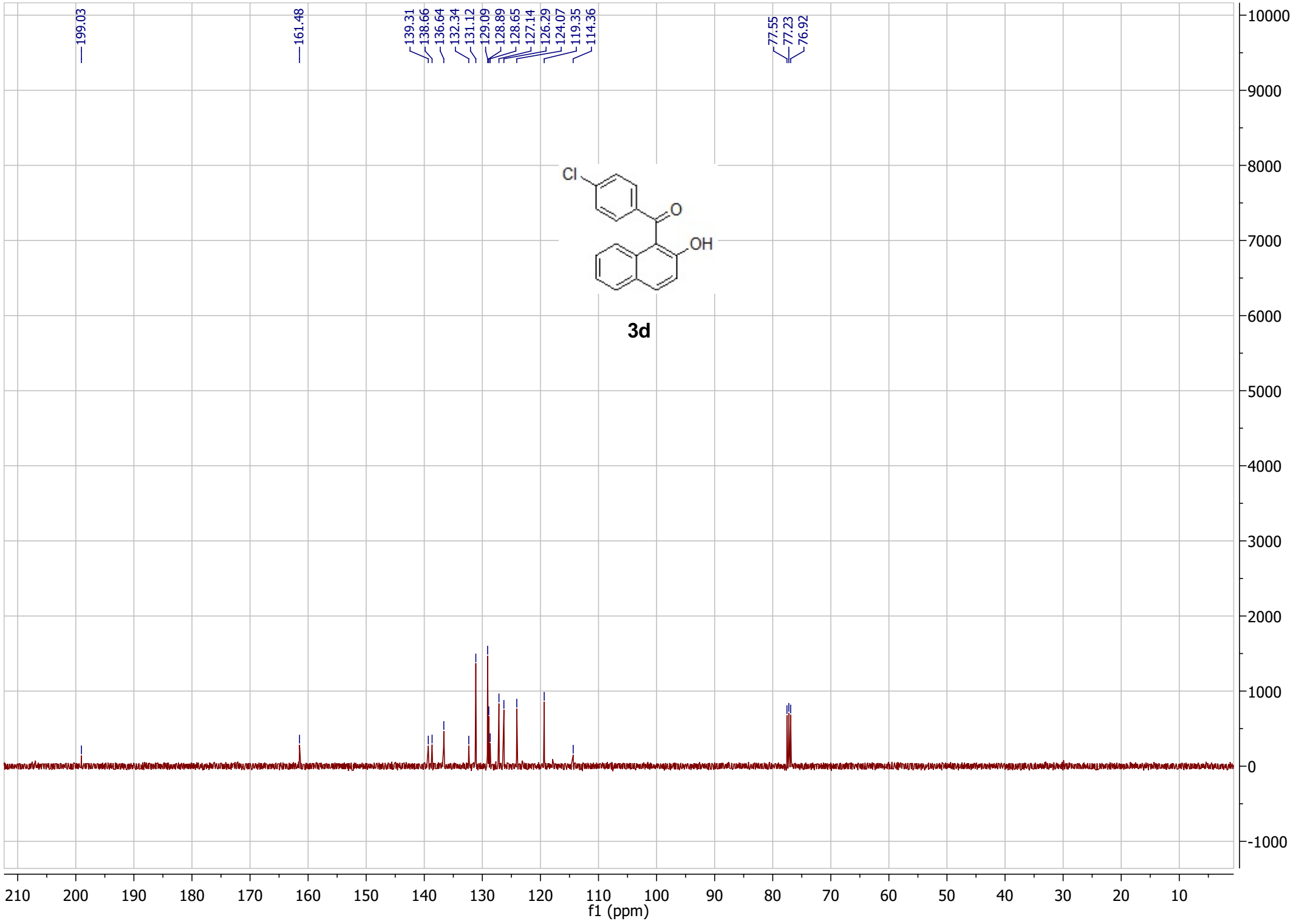
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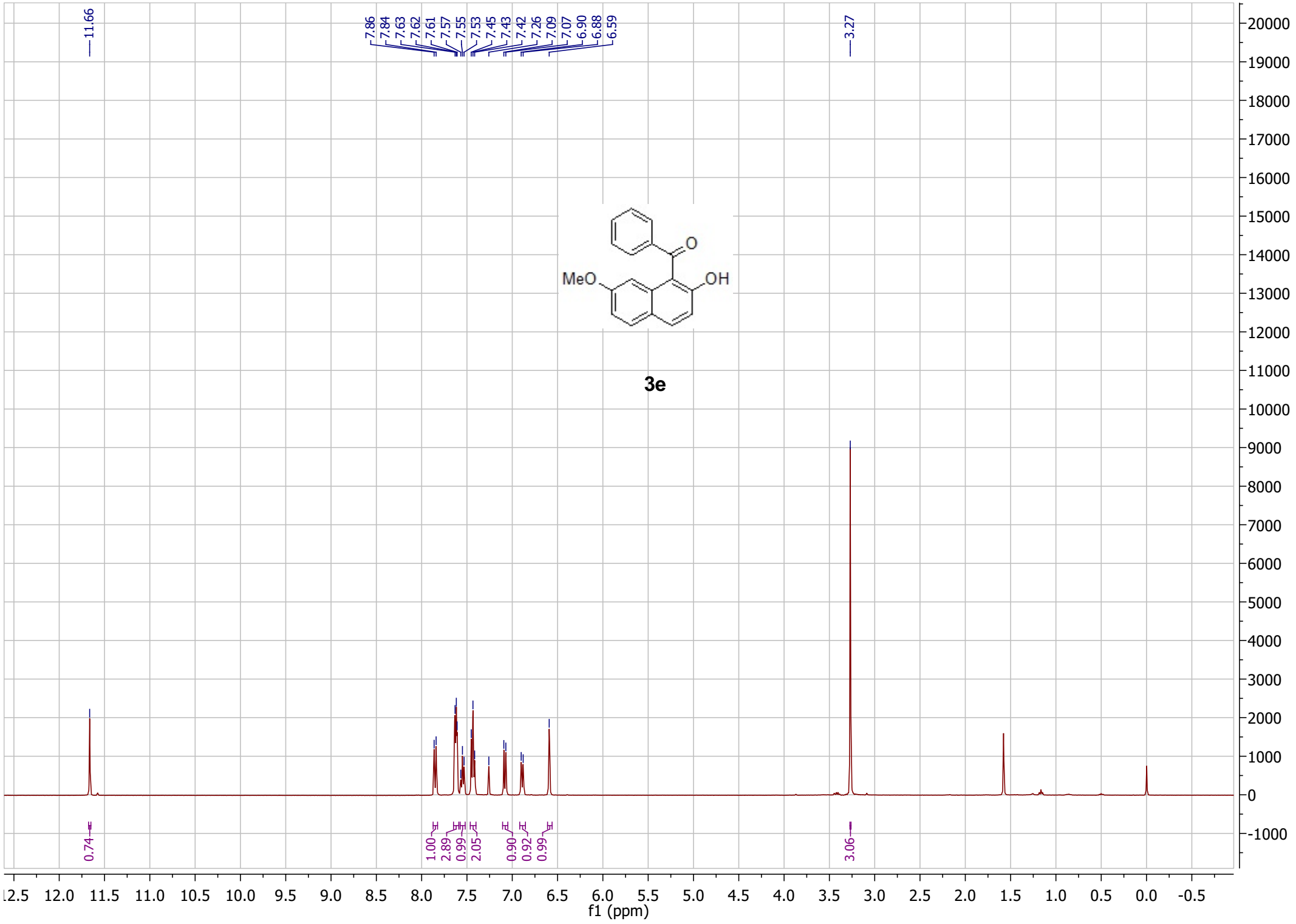
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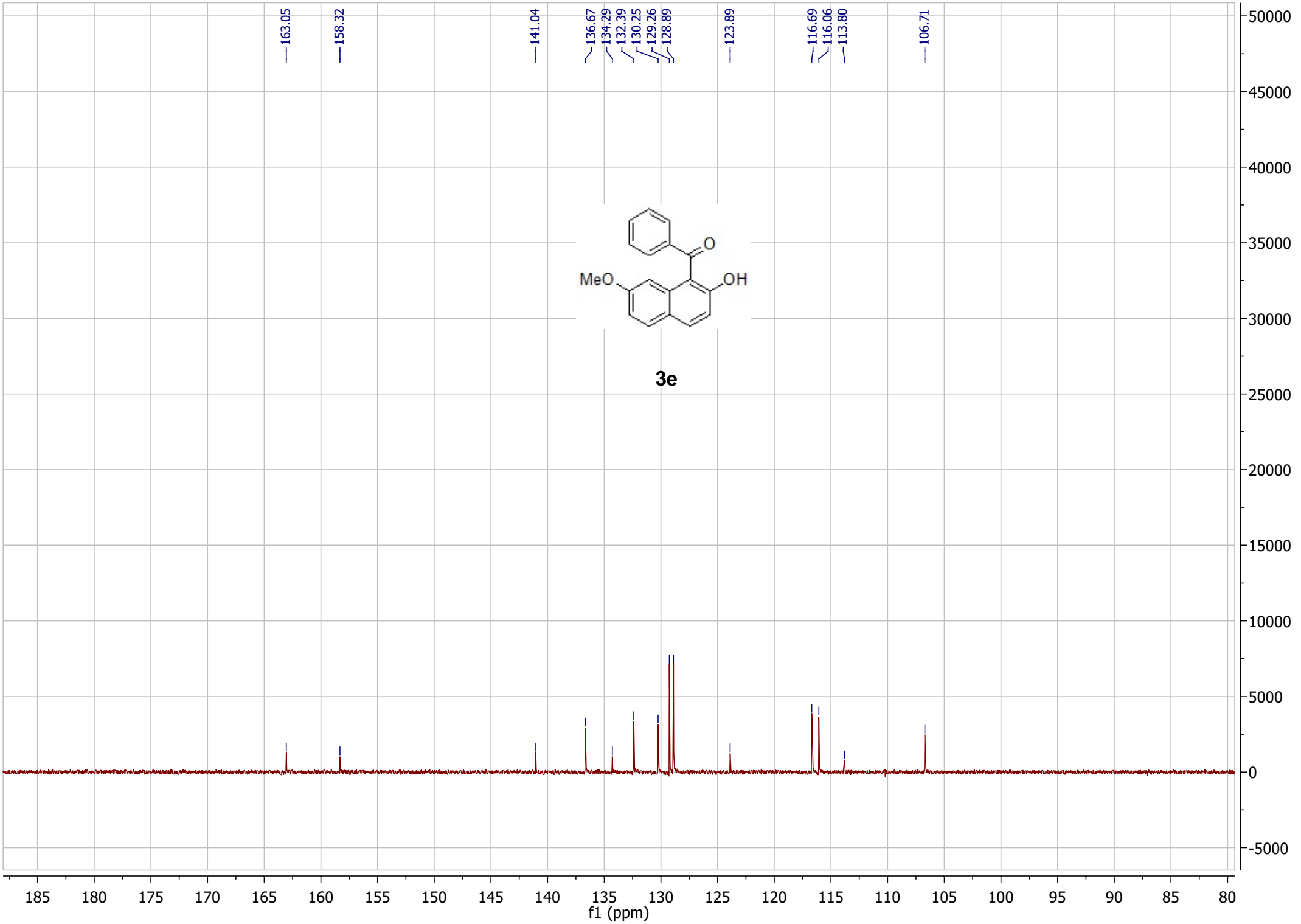
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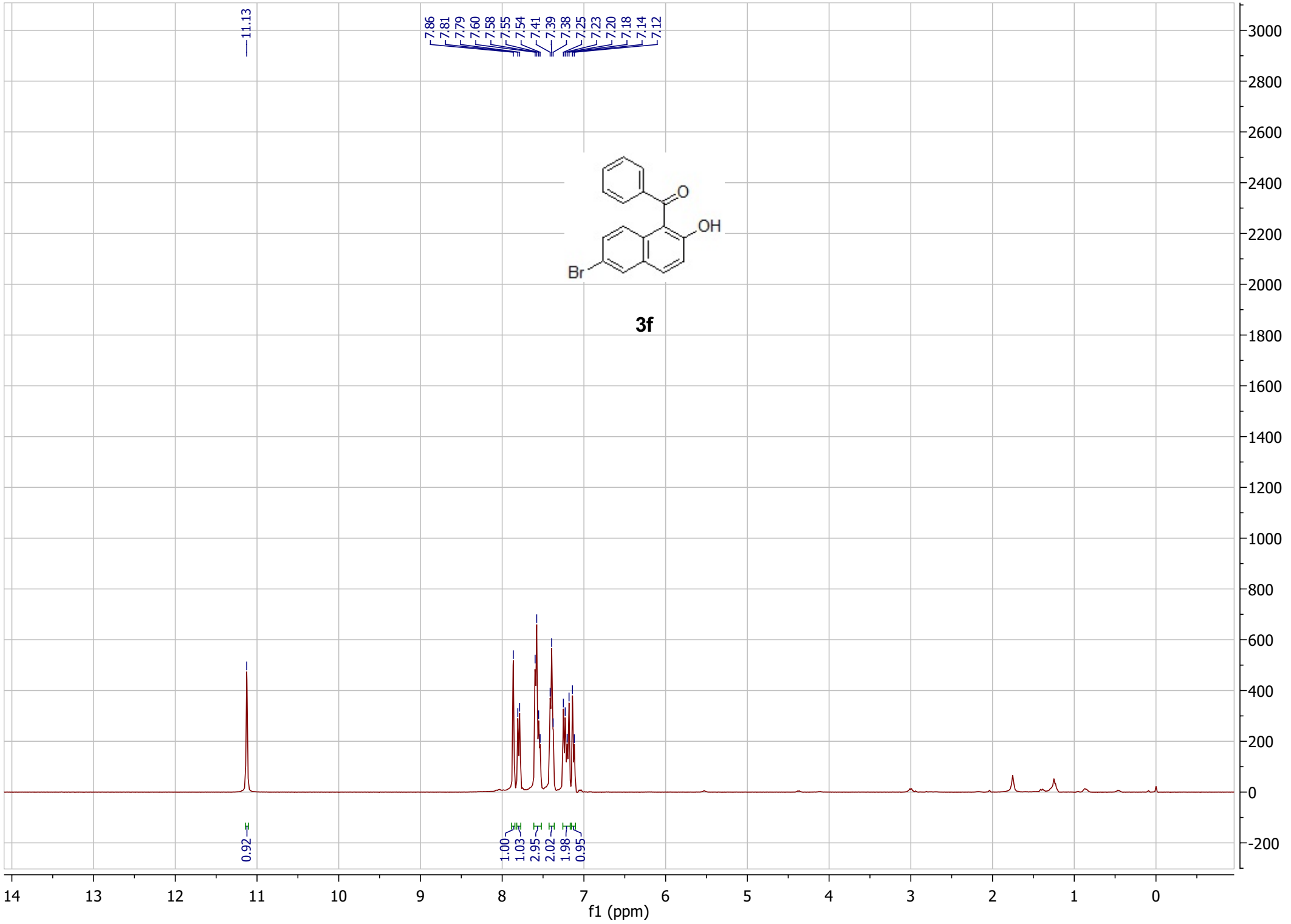
110
105
100
95
90
85
80
f1 (ppm)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7.23
7.20
7.18
7.14
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O
OH
Br
3f
0.92
1.00
1.03
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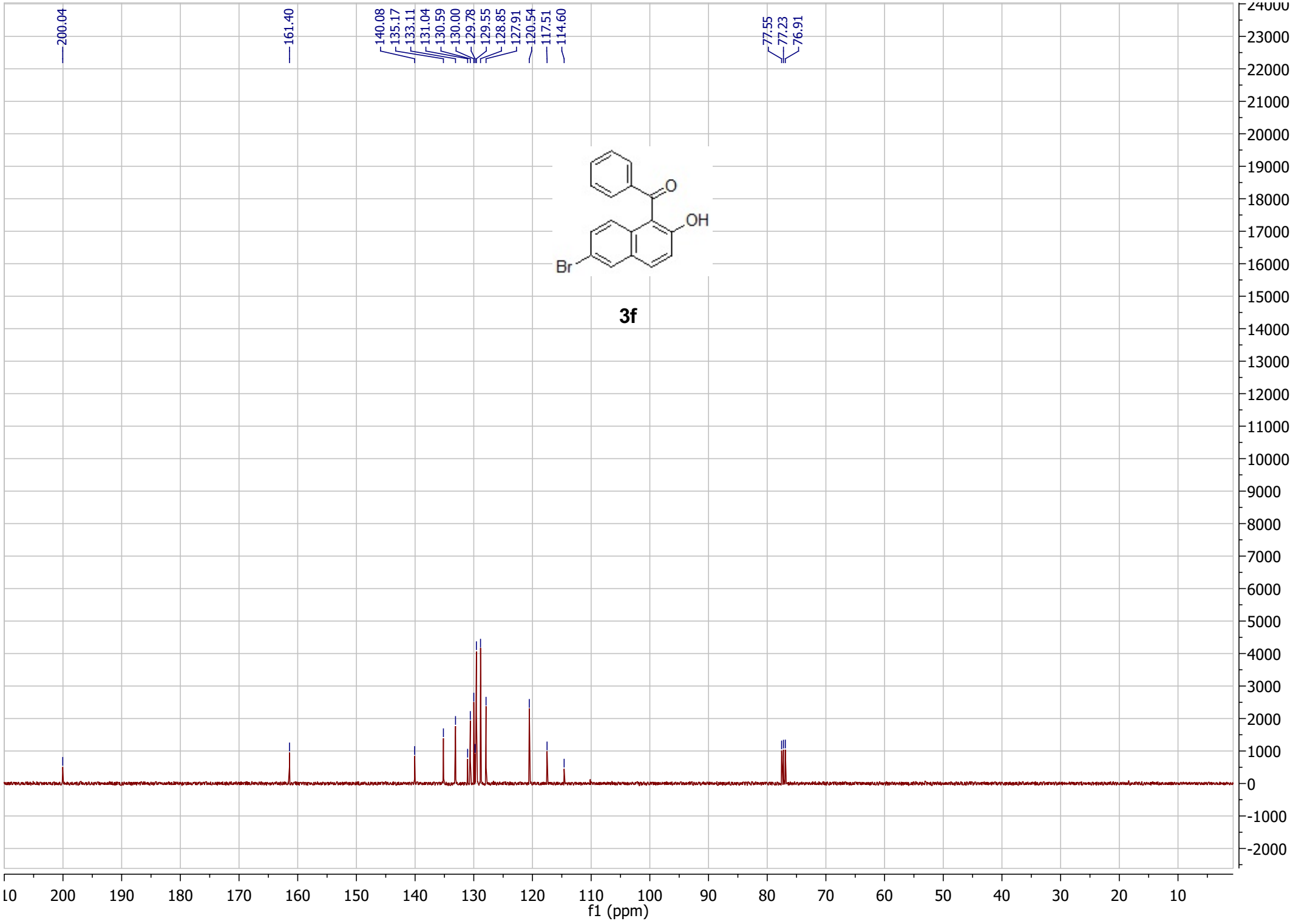
2.02
1.98
0.95
f1 (ppm)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130.00
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120.54
117.51
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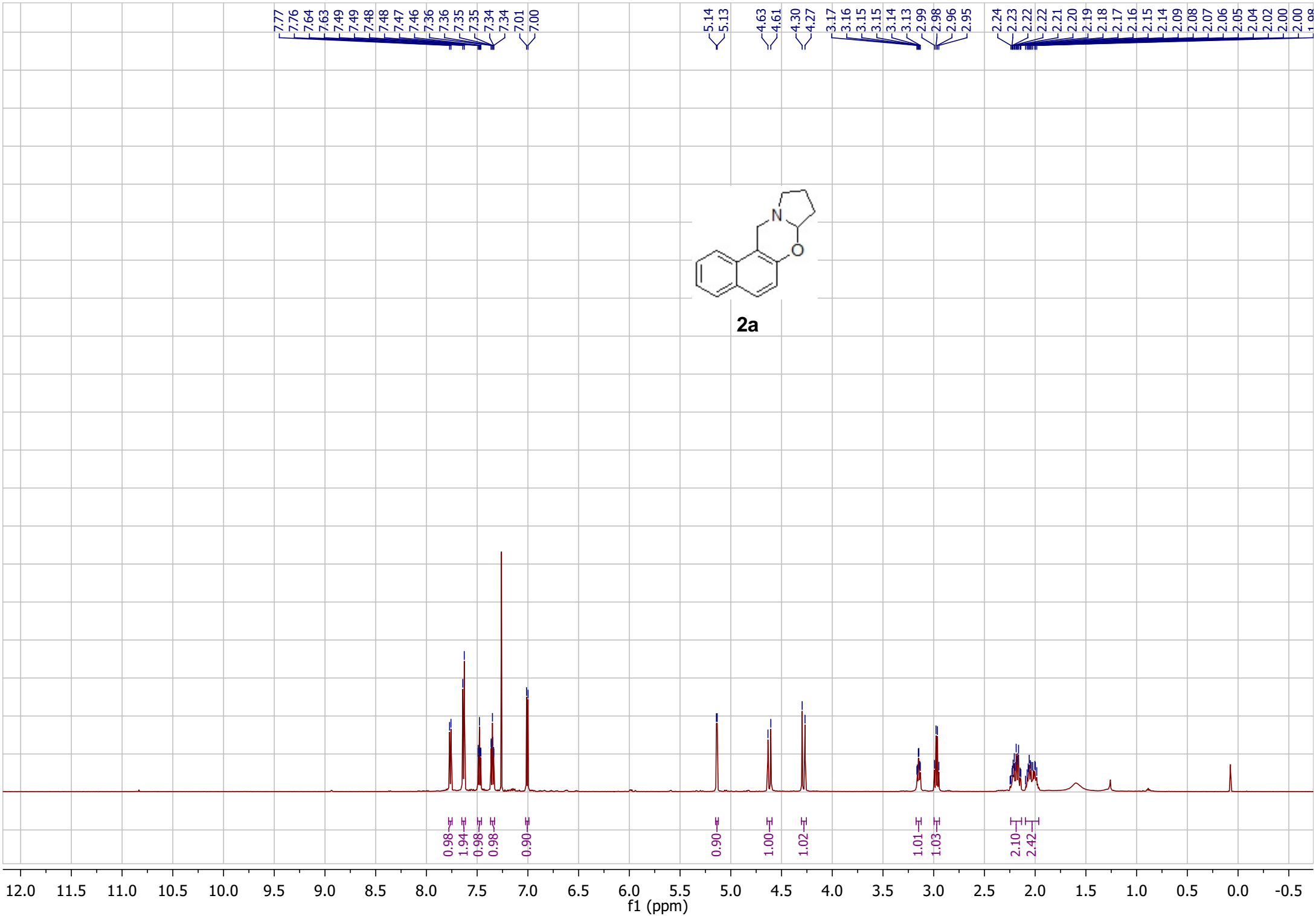
77.55
77.23
76.91
O
OH
Br
3f
f1 (ppm)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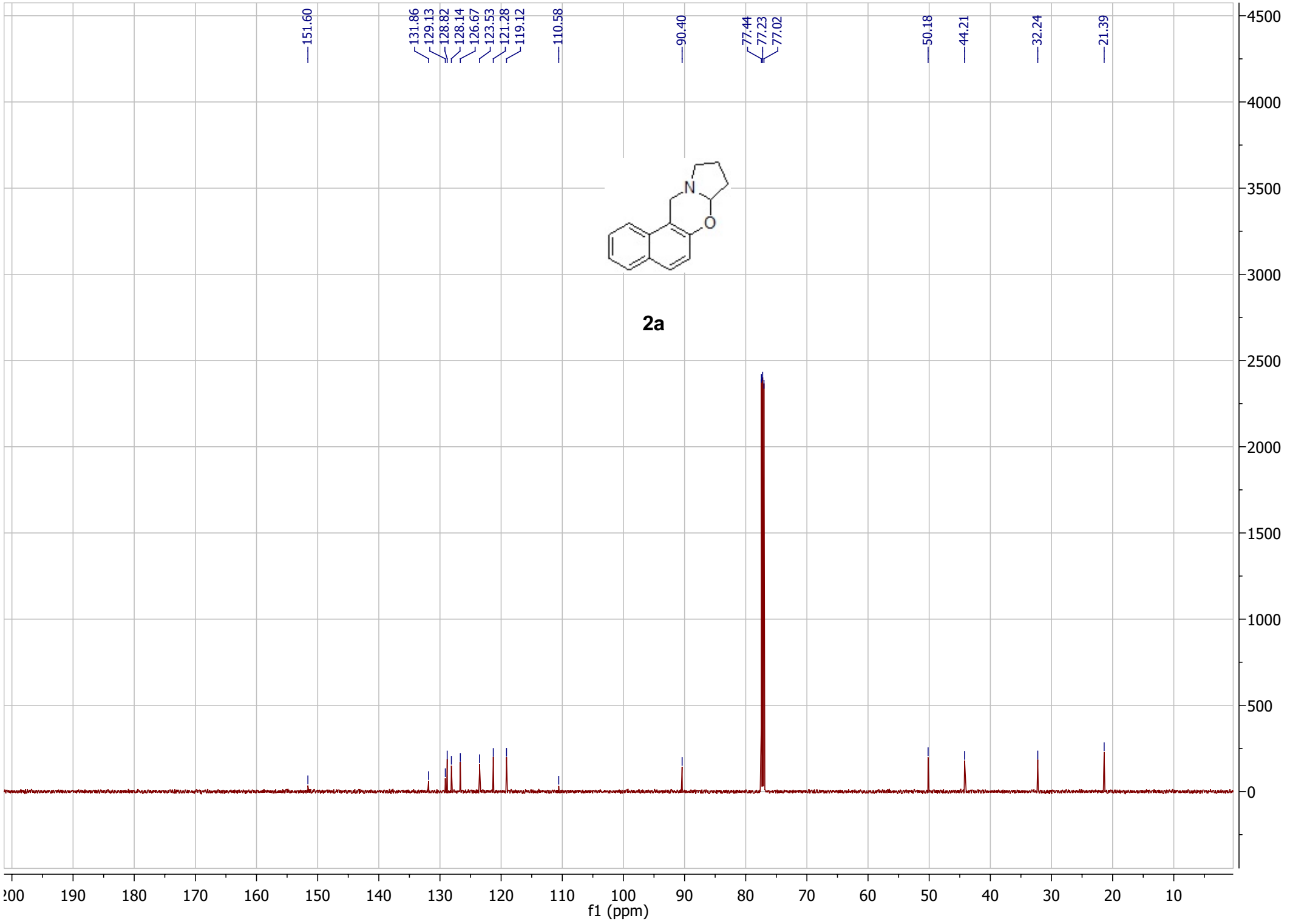
2.24 2.23 2.22 2.22 2.21 2.20 2.19 2.18 2.17 2.16 2.15 2.14 2.09 2.08 2.07 2.06 2.05 2.04 2.02 2.00 2.00
N
O
2a
0.98 1.94 0.98 0.98 0.90
0.90 1.00 1.02 1.01 1.03 2.10 2.42
12.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5
f1 (ppm)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77.23
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50.18
44.21
32.24
21.39
N
O
2a
4500
4000
3500
3000
2500
2000
1500
1000
500
0
200
190
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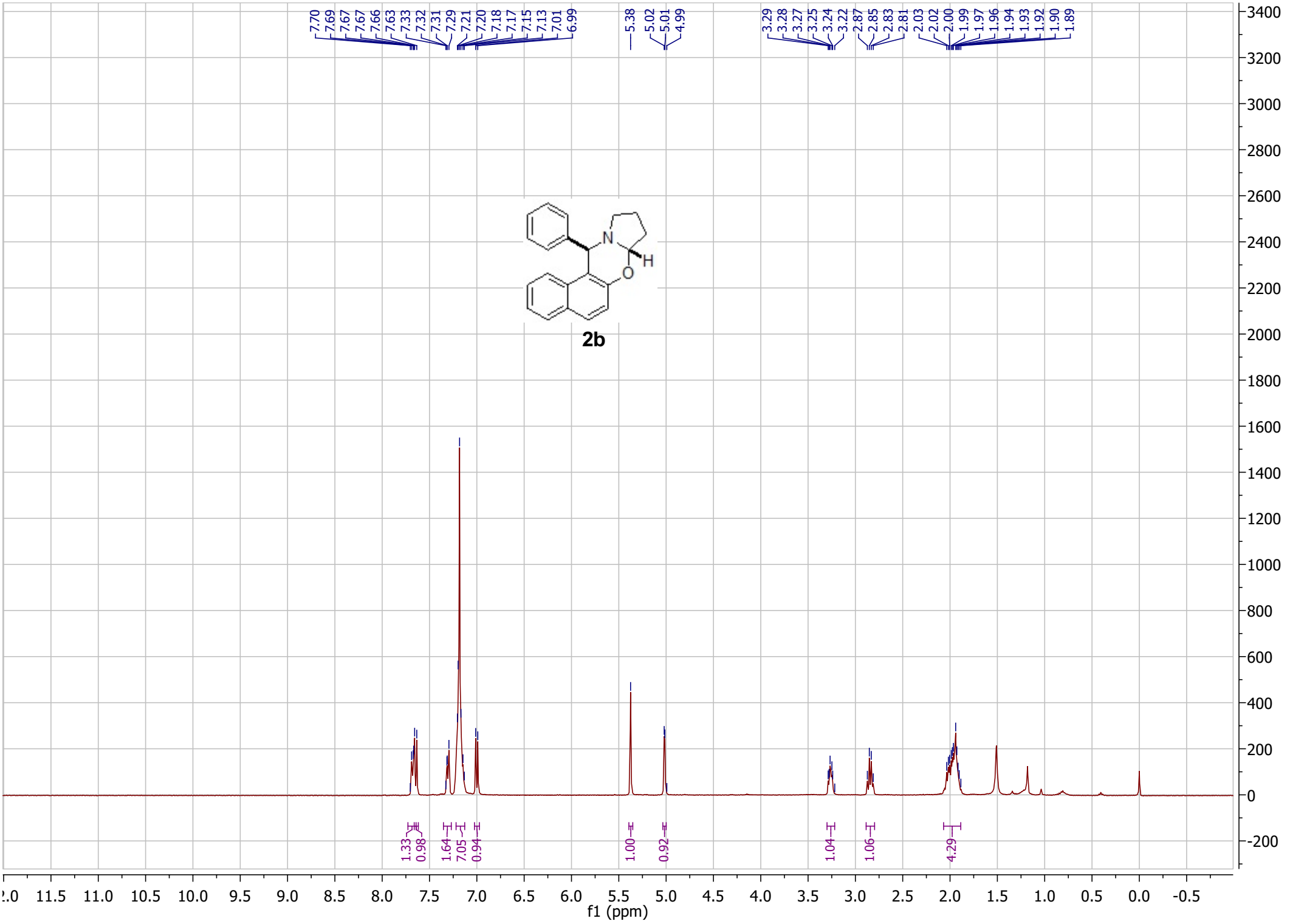
f1 (ppm)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7.15
7.13
7.01
6.99
5.38
5.02
5.01
4.99
3.29
3.28
3.27
3.25
3.24
3.22
2.87
2.85
2.83
2.81
2.03
2.02
2.00
1.99
1.97
1.96
1.94
1.93
1.92
1.90
1.89
N
H
O
2b
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7.05
0.94
1.00
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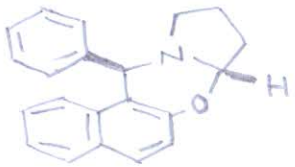
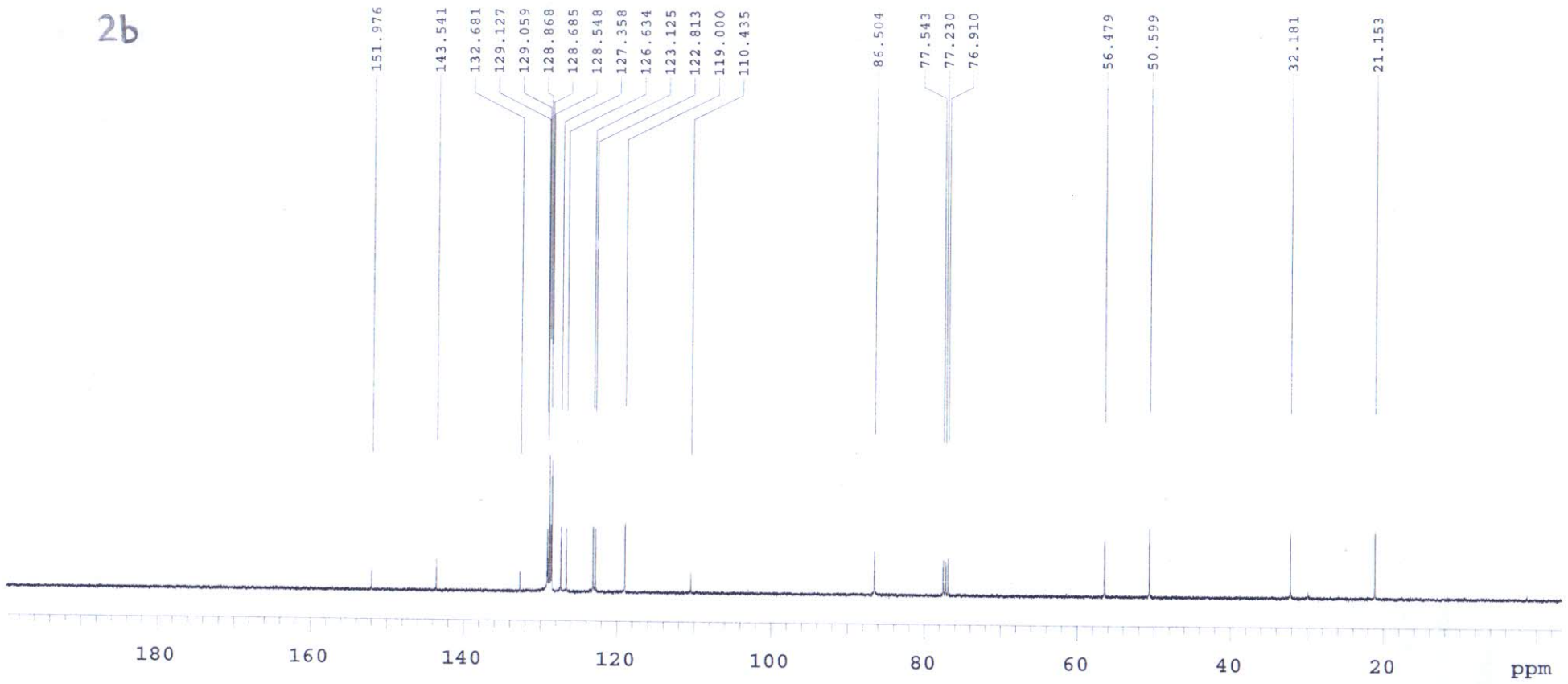
1.04
1.06
4.29
f1 (ppm)



2b



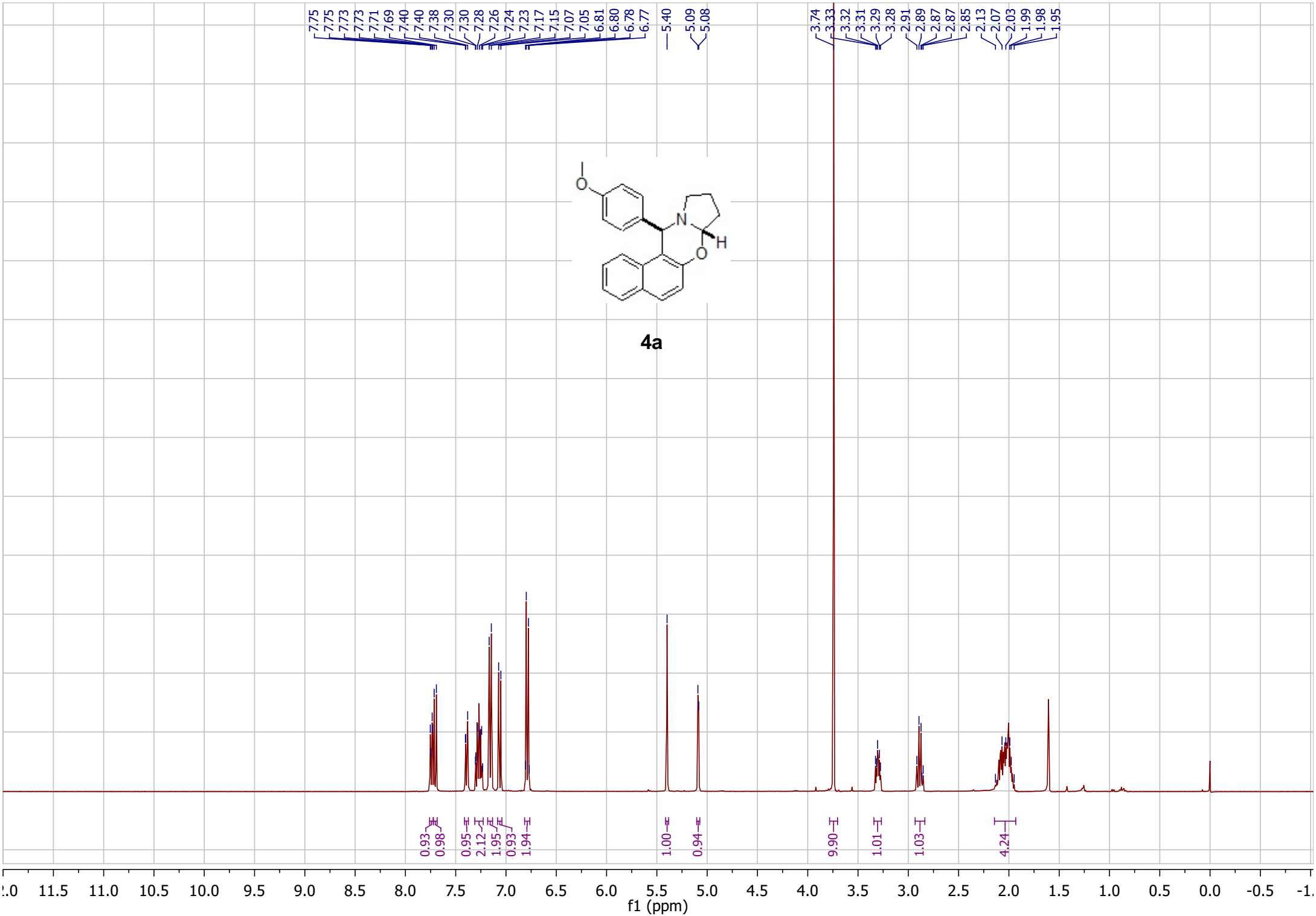
PULSE SEQUENCE
Relax. delay 1.000 sec
Pulse 45.0 degrees
Acq. time 1.304 sec
Width 25125.6 Hz
240 repetitions

OBSERVE C13, 100.5425932
DECOUPLE H1, 399.8529994
Power 42 dB
continuously on
WALTZ-16 modulated

DATA PROCESSING
Line broadening 0.5 Hz
FT size 65536
Total time 9 minutes

CKJ-2-65A-13C

Solvent: cdcl3
Temp. 25.0 C / 298.1 K
Operator: chem
Mercury-400 "IITG-NMR"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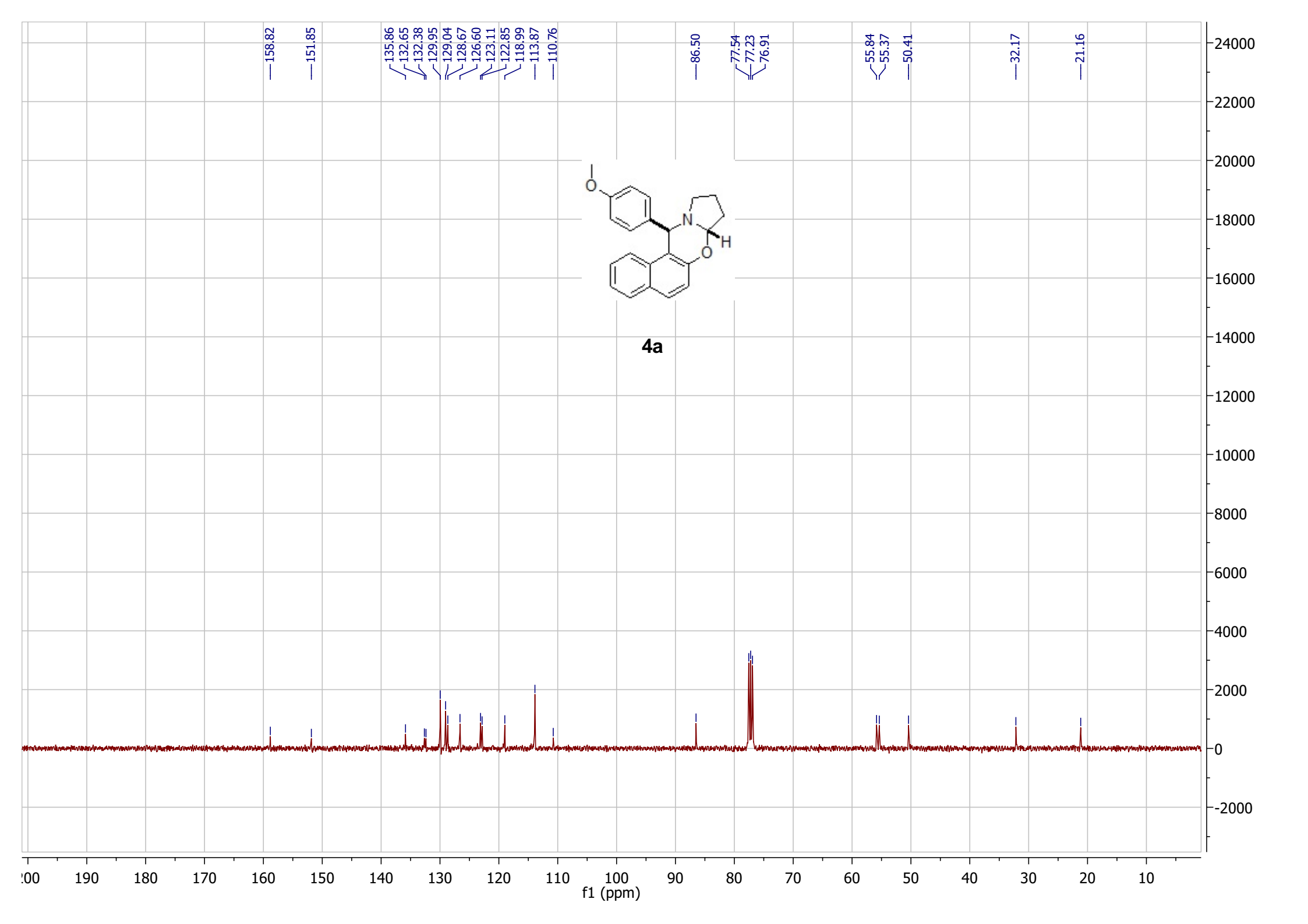
7.24
7.23
7.17
7.15
7.07
7.05
6.81
6.80
6.78
6.77
5.40
5.09
5.08
3.74
3.33
3.32
3.31
3.29
3.28
2.91
2.89
2.87
2.87
2.85
2.13
2.07
2.03
1.99
1.98
1.95
O
N
H
O
0.93
0.98
0.95
2.12
1.95
0.93
1.94
1.00
0.94
9.90
1.01
1.03
4.24
f1 (ppm)

4a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126.60
123.11
122.85
118.99
113.87
110.76
86.50
77.54
77.23
76.91
55.84
55.37
50.41
32.17
21.16
O
N
H
O
4a
24000
22000
20000
18000
16000
14000
12000
10000
8000
6000
4000
2000
0
-2000
200
190
180
170
160
150
140
130
120
110
100
90
80
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60
50
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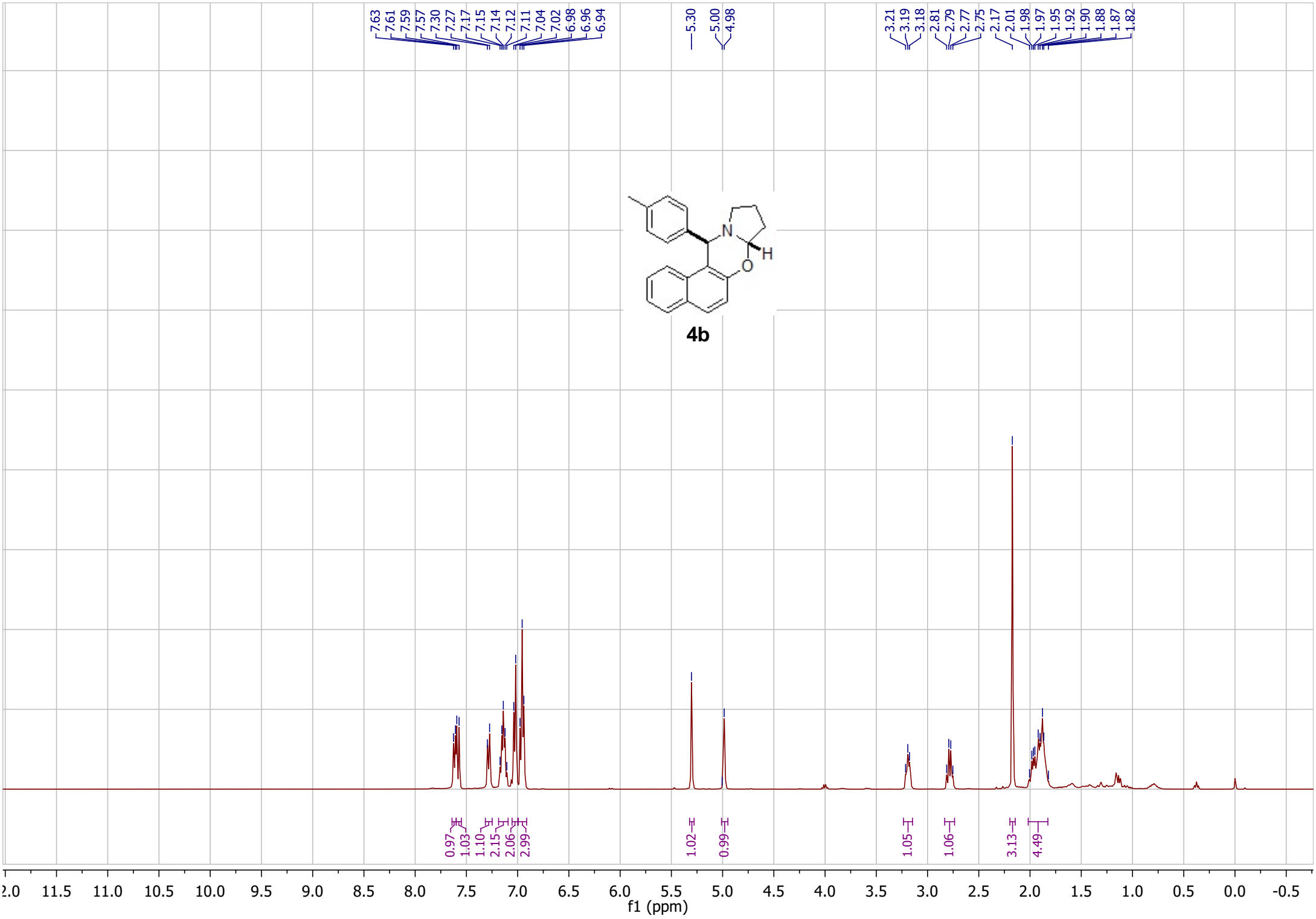
30
20
10
f1 (ppm)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7.15
7.14
7.12
7.11
7.04
7.02
6.98
6.96
6.94
5.30
5.00
4.98
3.21
3.19
3.18
2.81
2.79
2.77
2.75
2.17
2.01
1.98
1.97
1.95
1.92
1.90
1.88
1.87
1.82
N
H
O
4b
0.97
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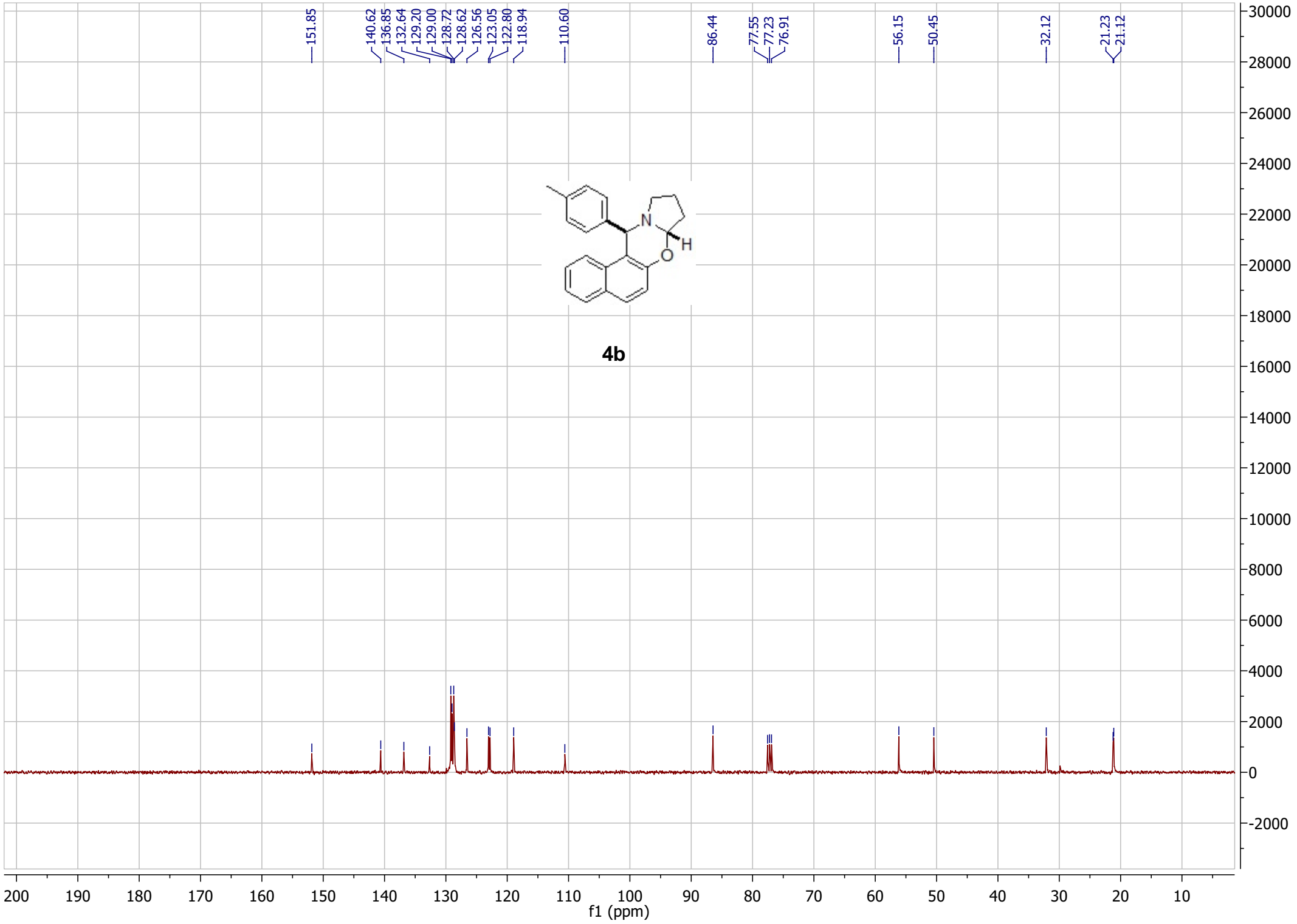
1.02
0.99
1.05
1.06
3.13
4.49
f1 (ppm)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77.55
77.23
76.91
56.15
50.45
32.12
21.23
21.12
N
O
H
4b
30000
28000
26000
24000
22000
20000
18000
16000
14000
12000
10000
8000
6000
4000
2000
0
-2000
200
190
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130
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100
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80
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60
50
40
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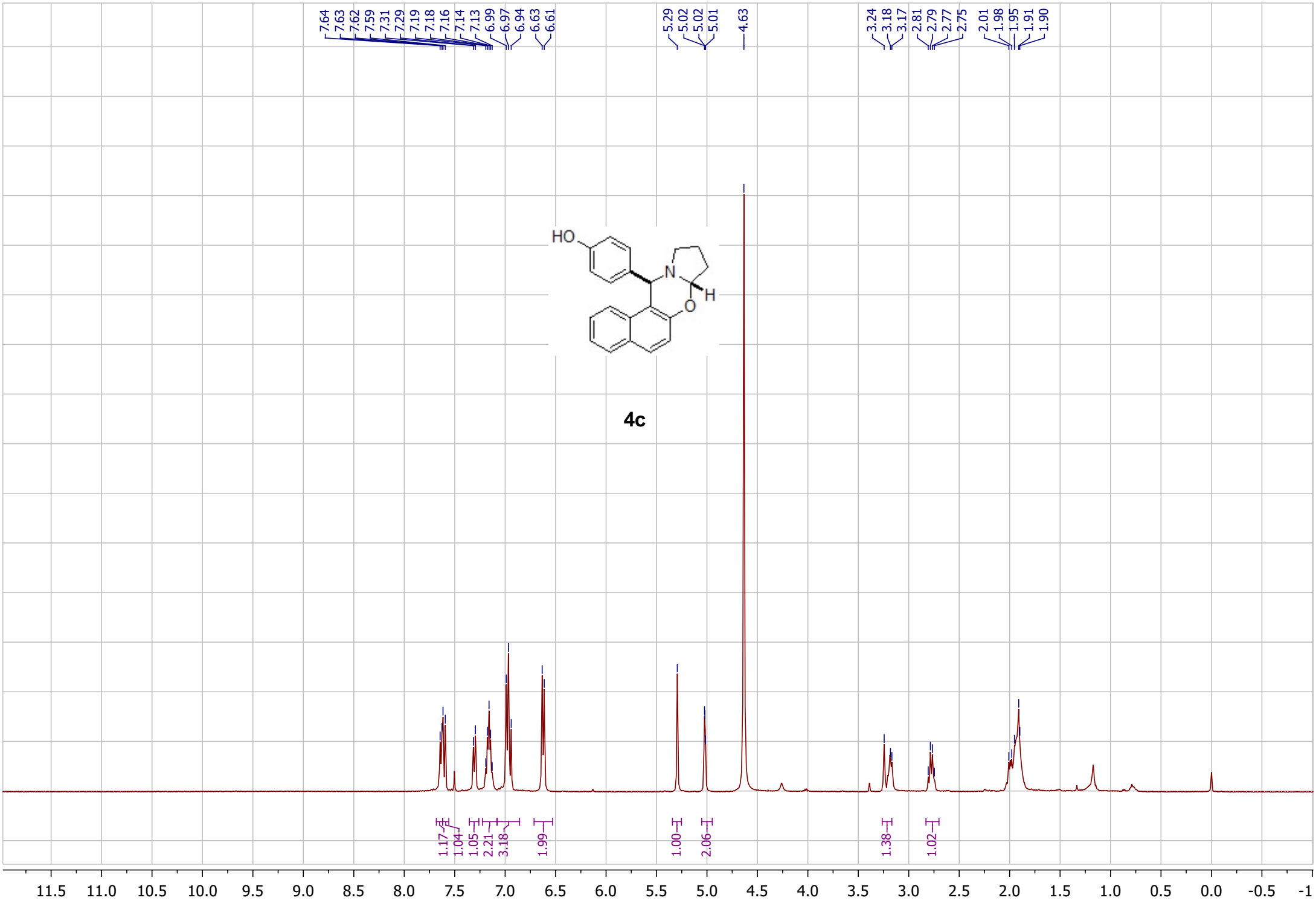
20
10
f1 (ppm)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6.97
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6.63
6.61
5.29
5.02
5.02
5.01
4.63
3.24
3.18
3.17
2.81
2.79
2.77
2.75
2.01
1.98
1.95
1.91
1.90
HO
N
H
O
4c
1.17
1.04
1.05
2.21
3.18
1.99
1.00
2.06
1.38
1.02
11.5
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
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3.0
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2.0
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1.0
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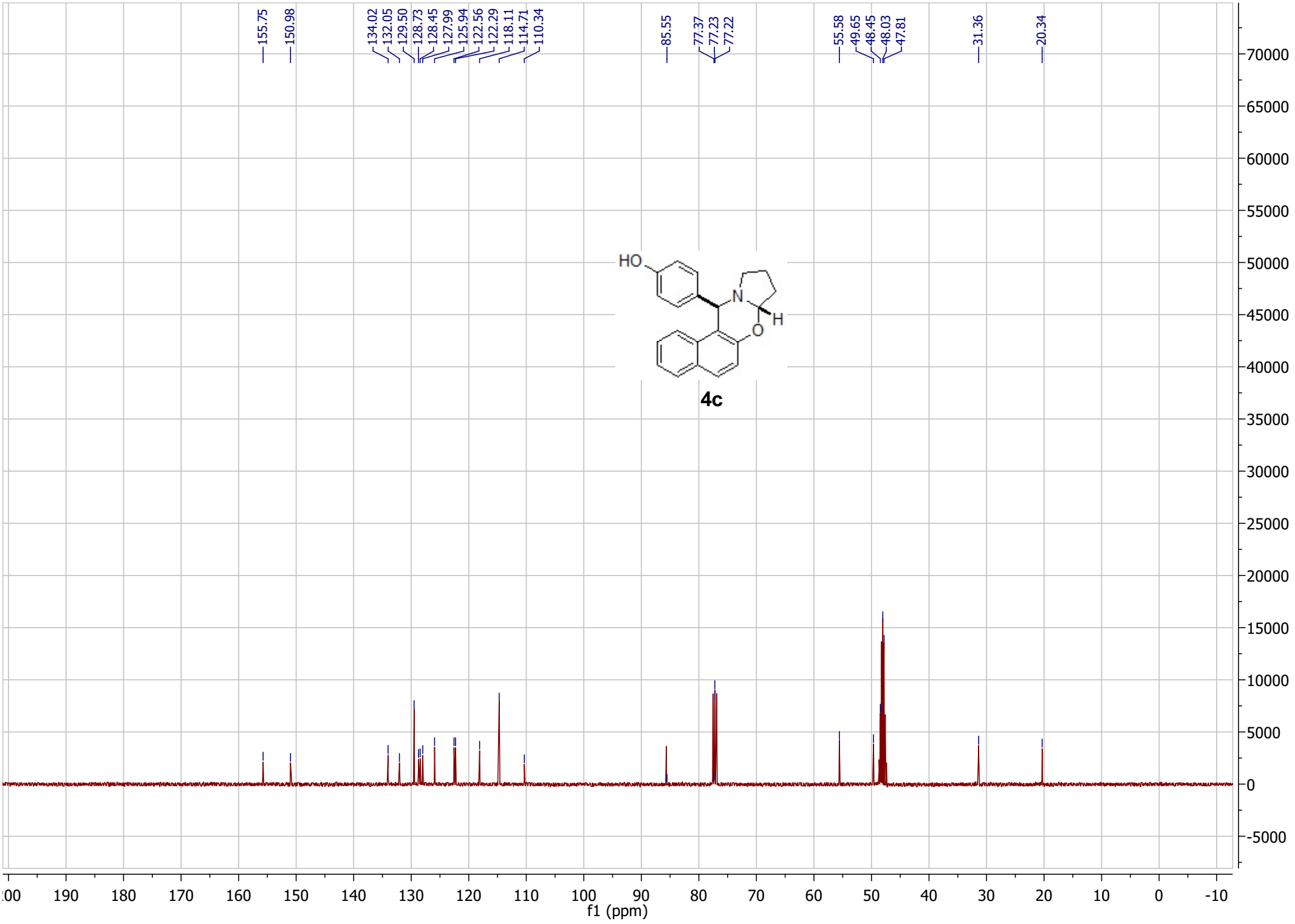
0.0
-0.5
-1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122.29
118.11
114.71
110.34
85.55
77.37
77.23
77.22
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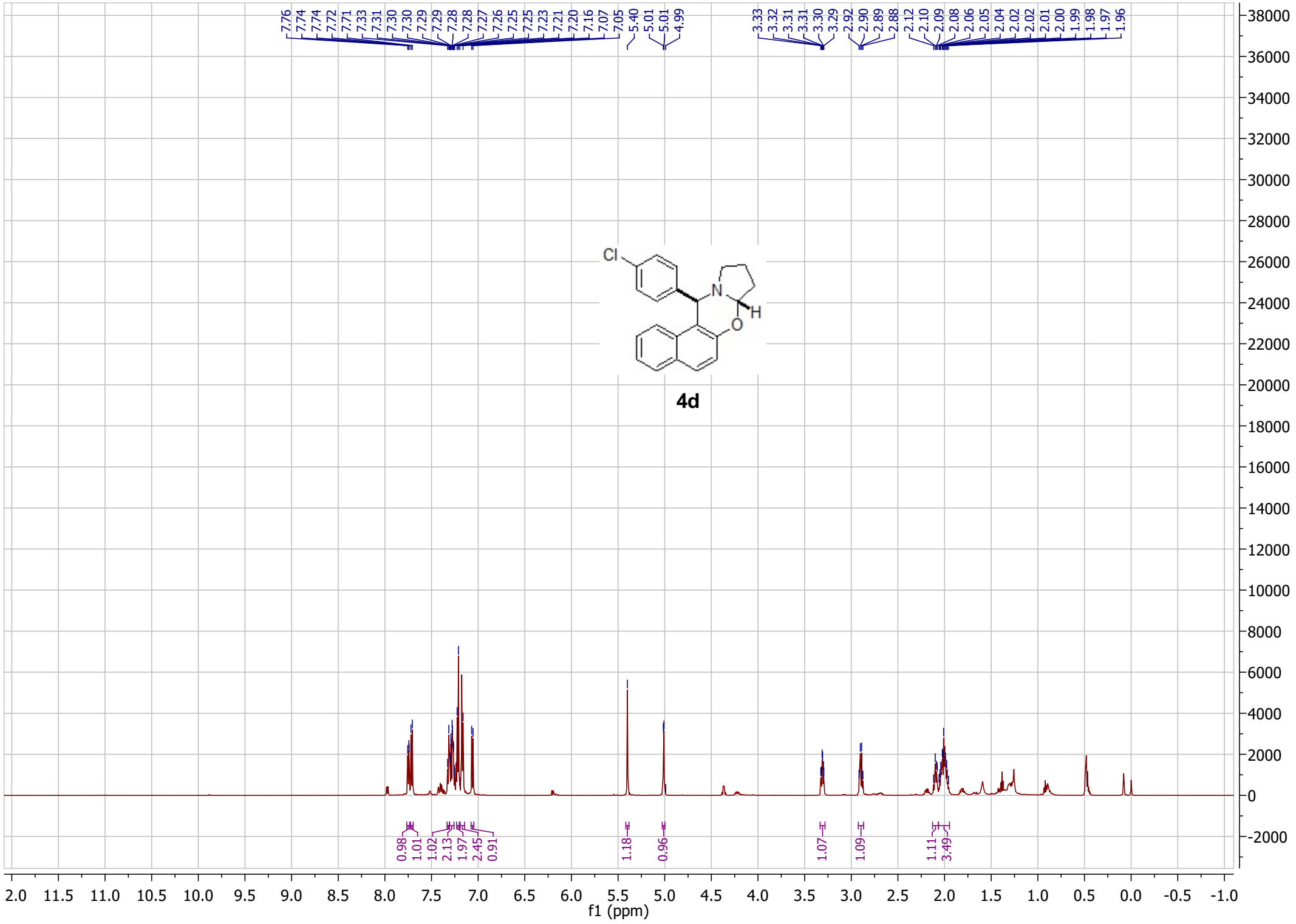
48.45
48.03
47.81
31.36
20.34
HO
N
O
H
4c
f1 (ppm)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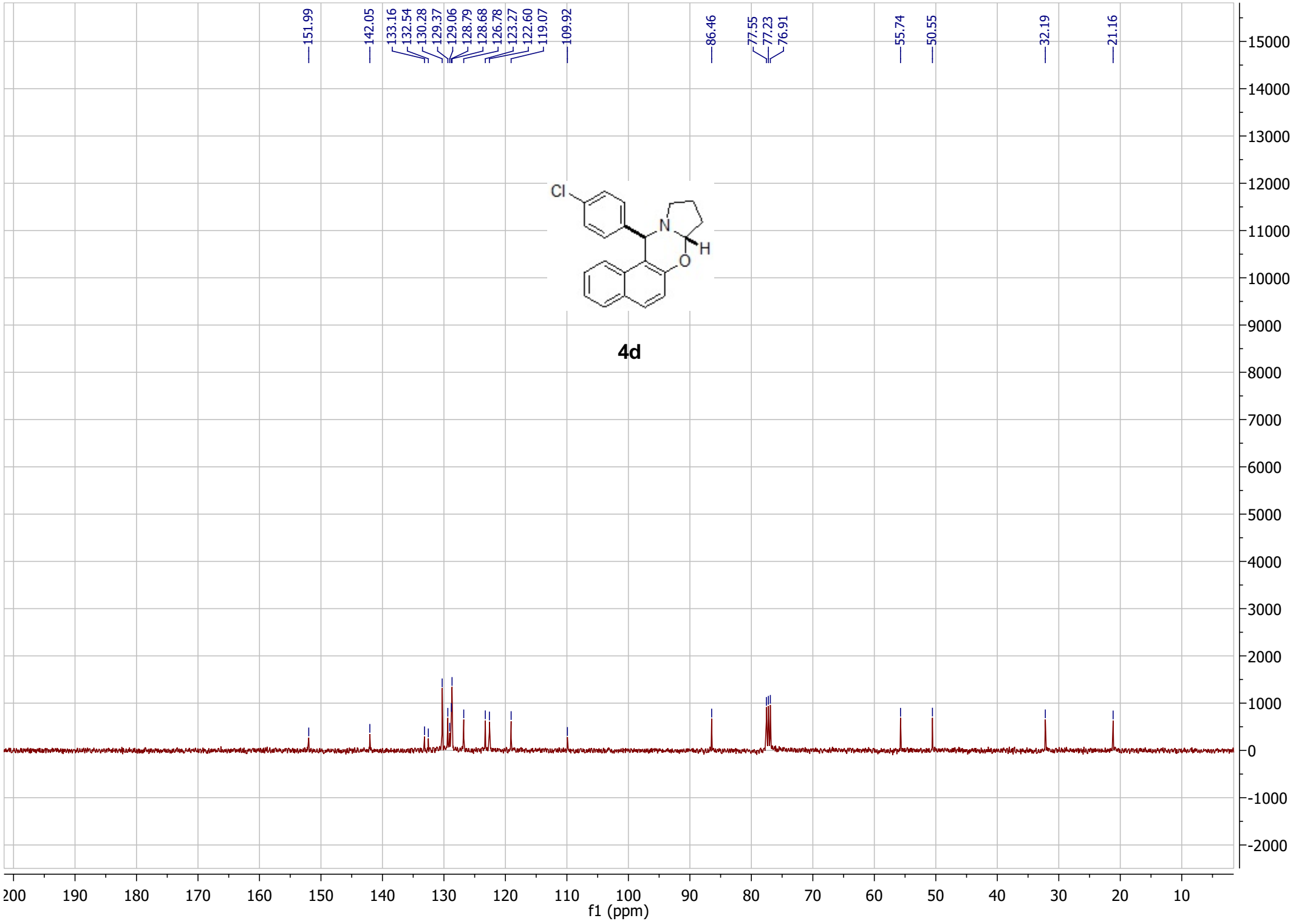
3.33 3.32 3.31 3.31 3.30 3.29 2.92 2.90 2.89 2.88 2.12 2.10 2.09 2.08 2.06 2.05 2.04 2.02 2.02 2.01 2.00 1.99 1.98 1.97 1.96
Cl
N
H
O
4d
0.98 1.01 1.02 2.13 1.97 2.45 0.91 1.18 0.96 1.07 1.09 1.11 3.49
38000 36000 34000 32000 30000 28000 26000 24000 22000 20000 18000 16000 14000 12000 10000 8000 6000 4000 2000 0 -2000
2.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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77.55
77.23
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55.74
50.55
32.19
21.16
Cl
N
H
O
4d
15000
14000
13000
12000
11000
10000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
-1000
-2000
200
190
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100
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80
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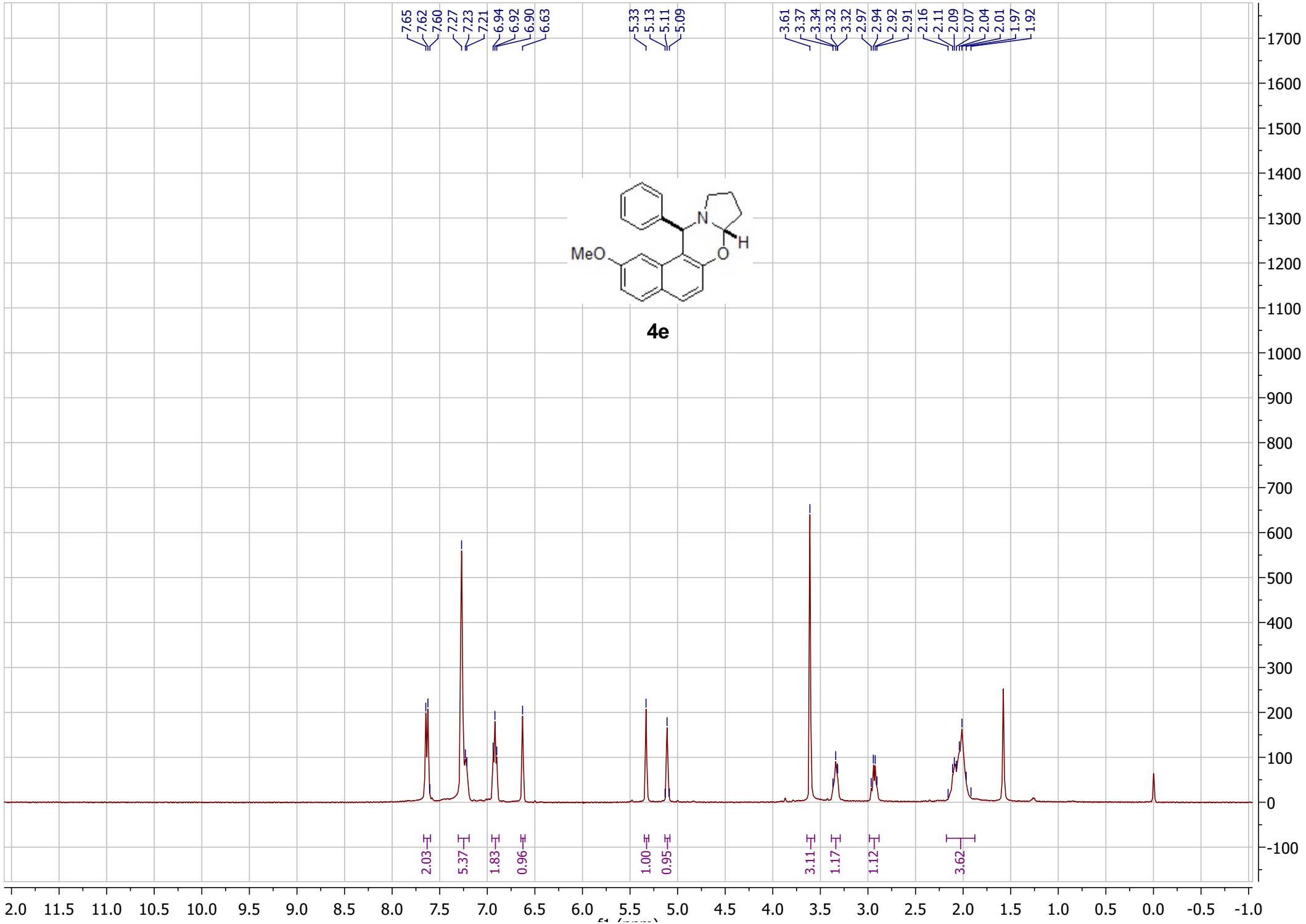
60
50
40
30
20
10
f1 (ppm)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3.32
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2.92
2.91
2.16
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2.09
2.07
2.04
2.01
1.97
1.92
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N
O
H
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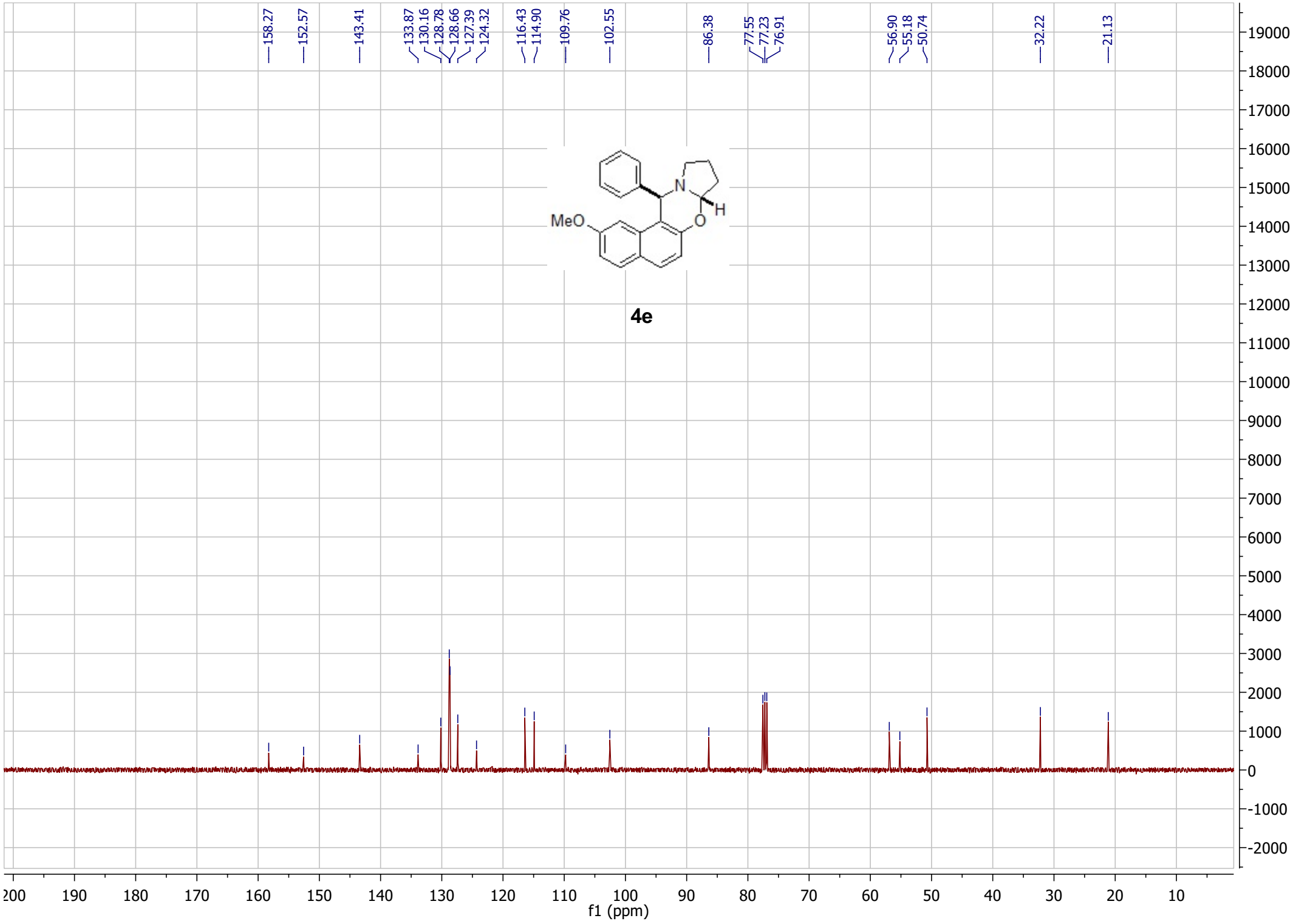
1.12
3.62
f1 (ppm)

4e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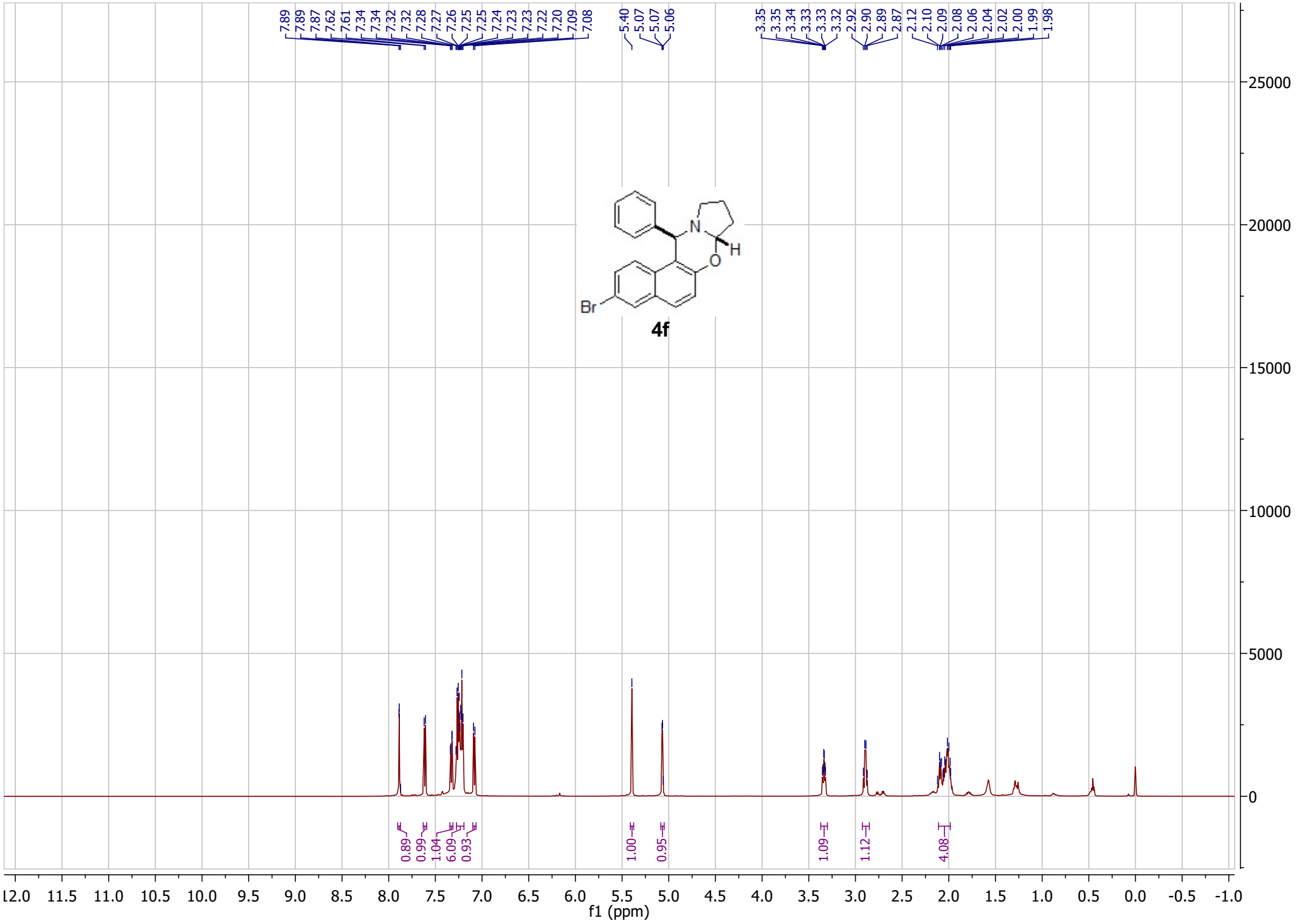
21.13
MeO
N
O
H
4e
f1 (ppm)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7.08
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2.04
2.02
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H
O
Br
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6.09
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4.0
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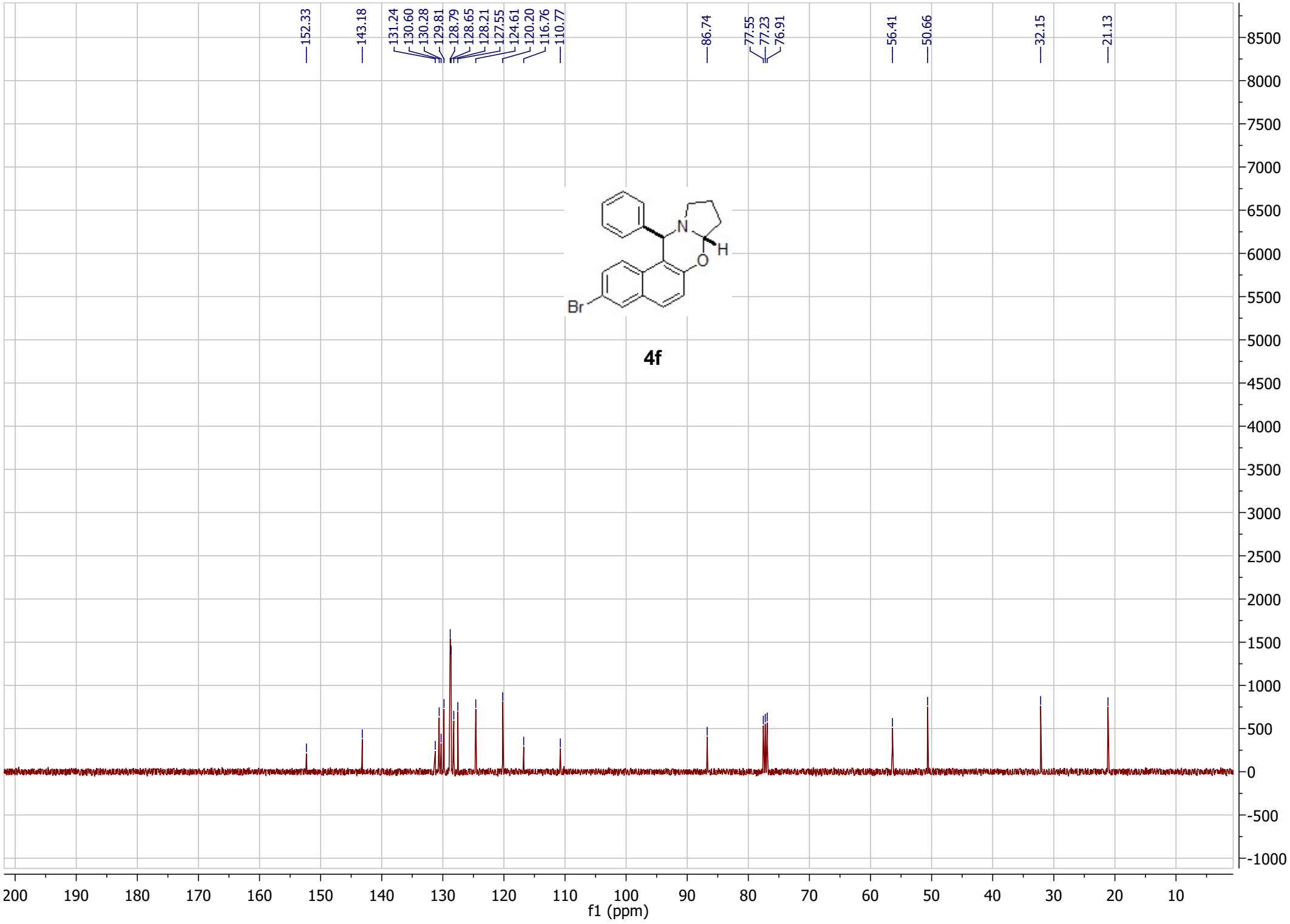
f1 (ppm)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56.41
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N
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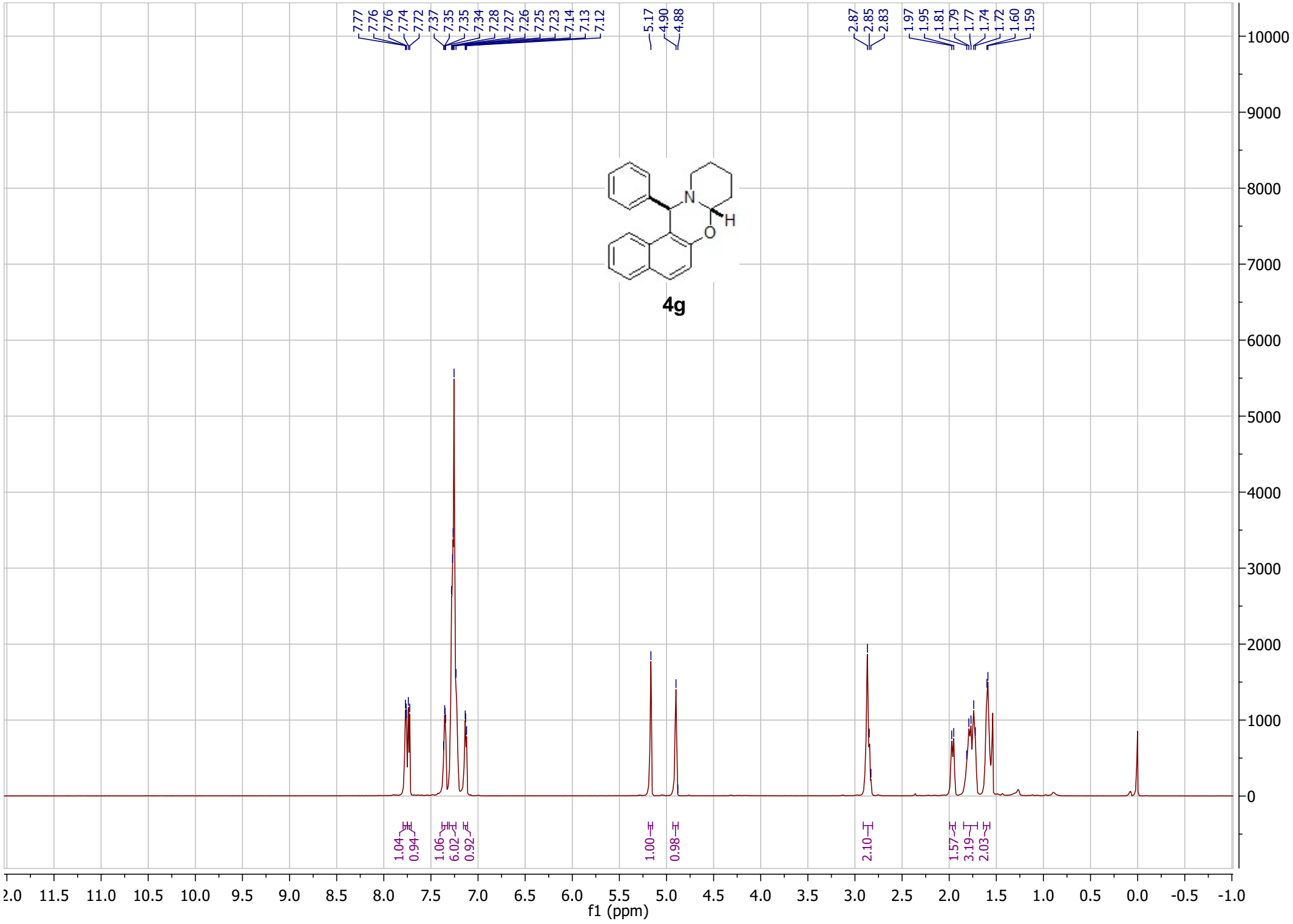
H
Br
4f
f1 (ppm)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7.23
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7.13
7.12
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2.87
2.85
2.83
1.97
1.95
1.81
1.79
1.77
1.74
1.72
1.60
1.59
N
H
O
4g
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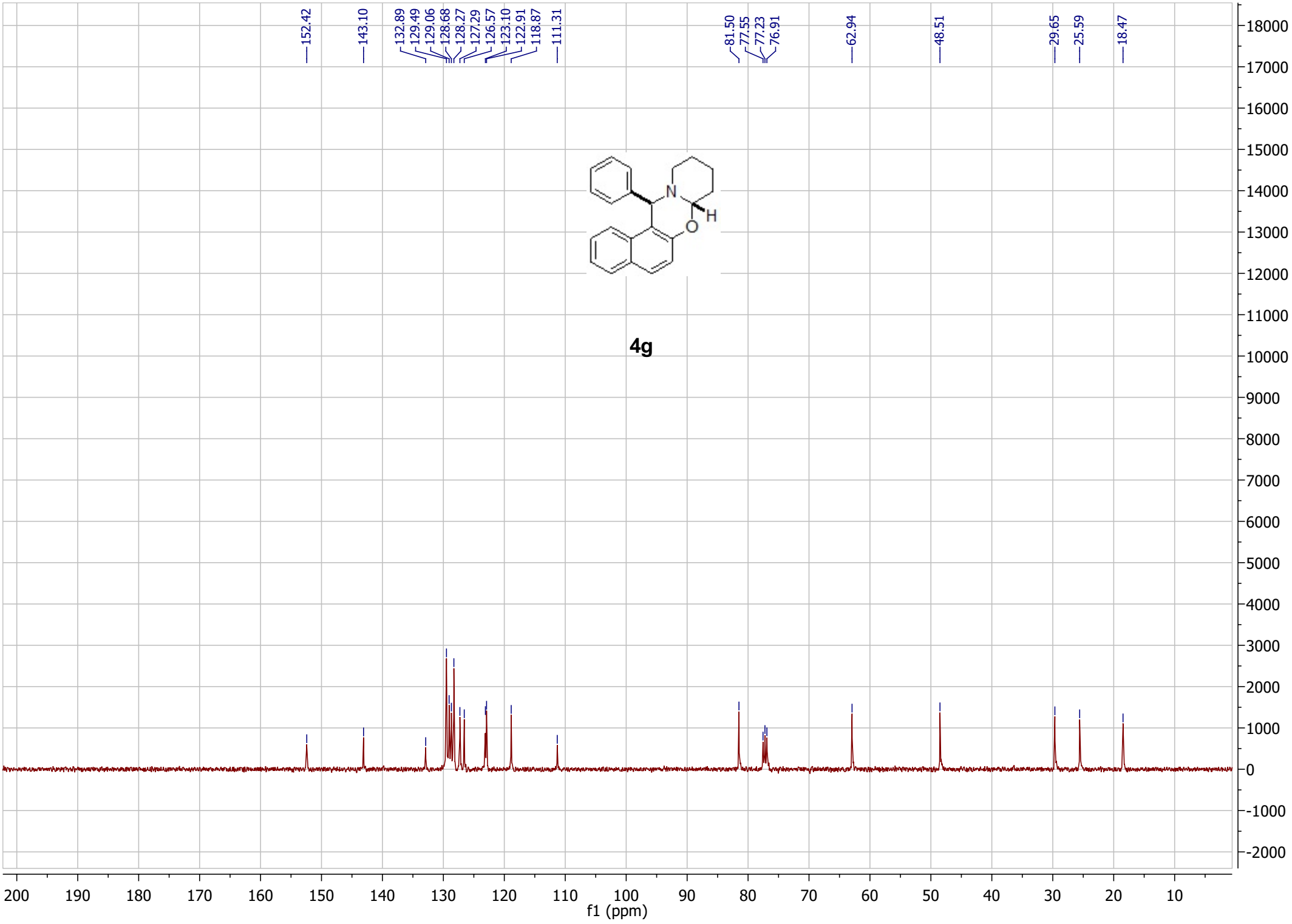
2.03
f1 (ppm)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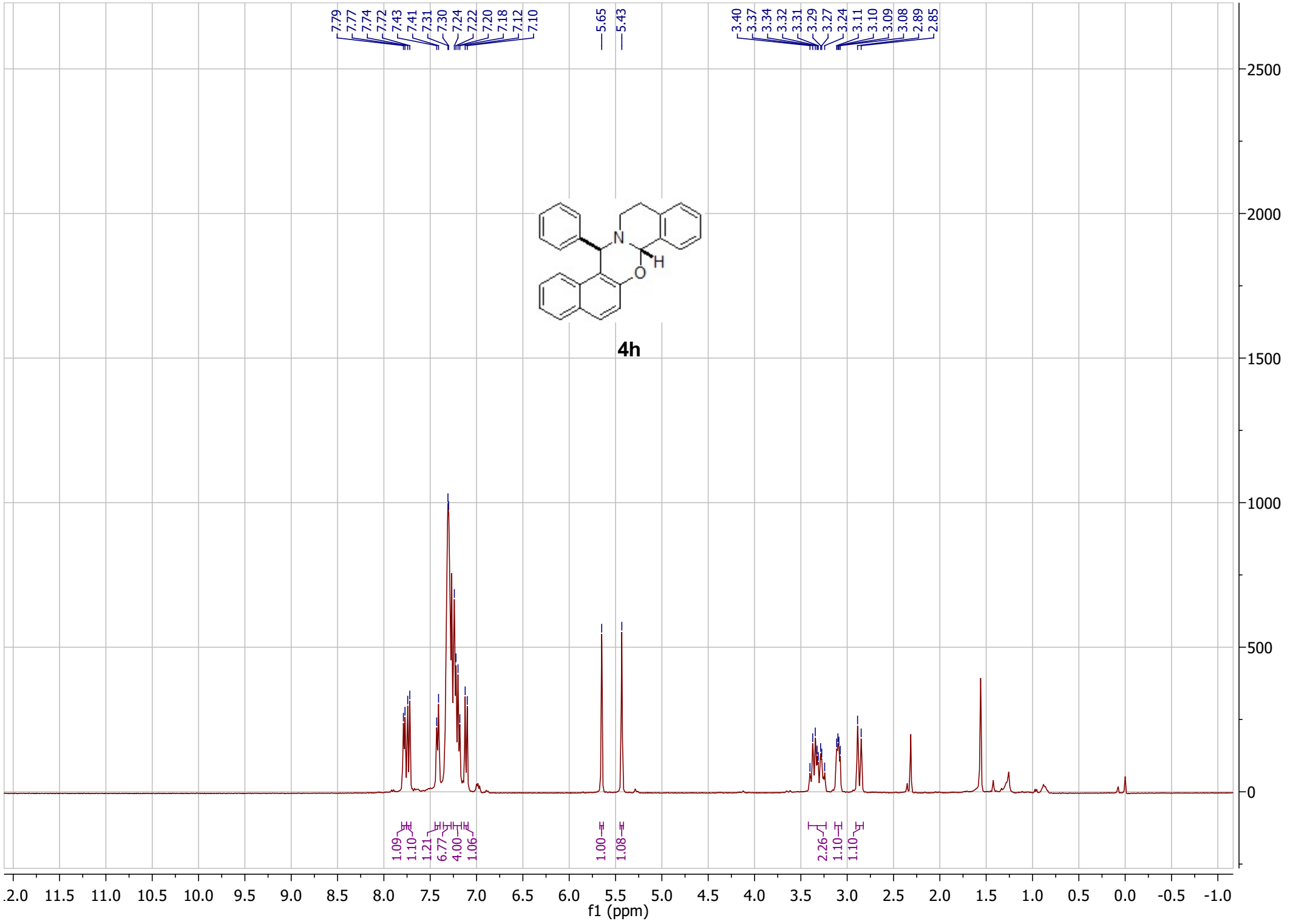
N
O
H
4g
f1 (ppm)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7.12
7.10
5.65
5.43
3.40
3.37
3.34
3.32
3.31
3.29
3.27
3.24
3.11
3.10
3.09
3.08
2.89
2.85
N
O
H
4h
1.09
1.10
1.21
6.77
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1.08
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2000
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500
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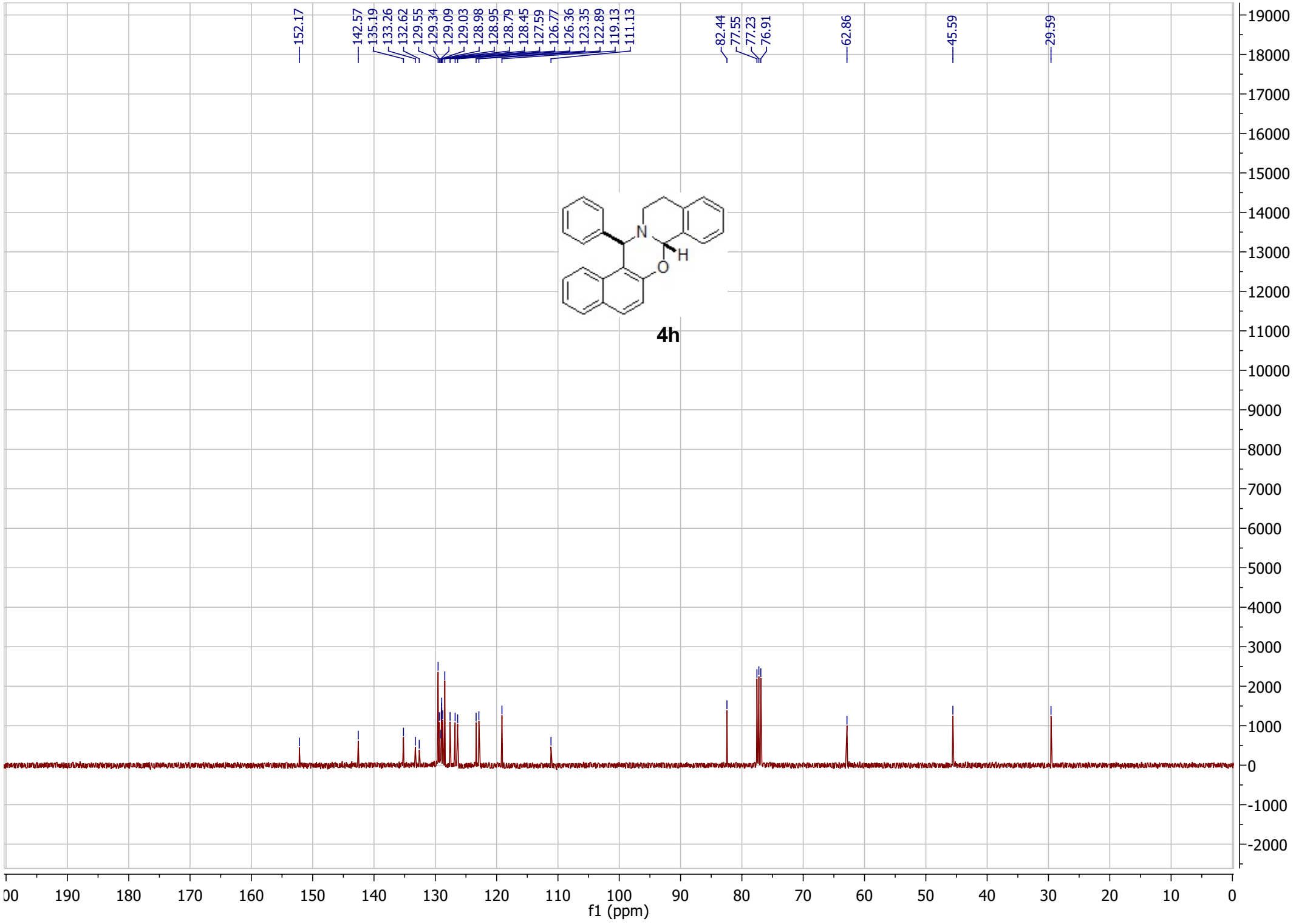
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123.35
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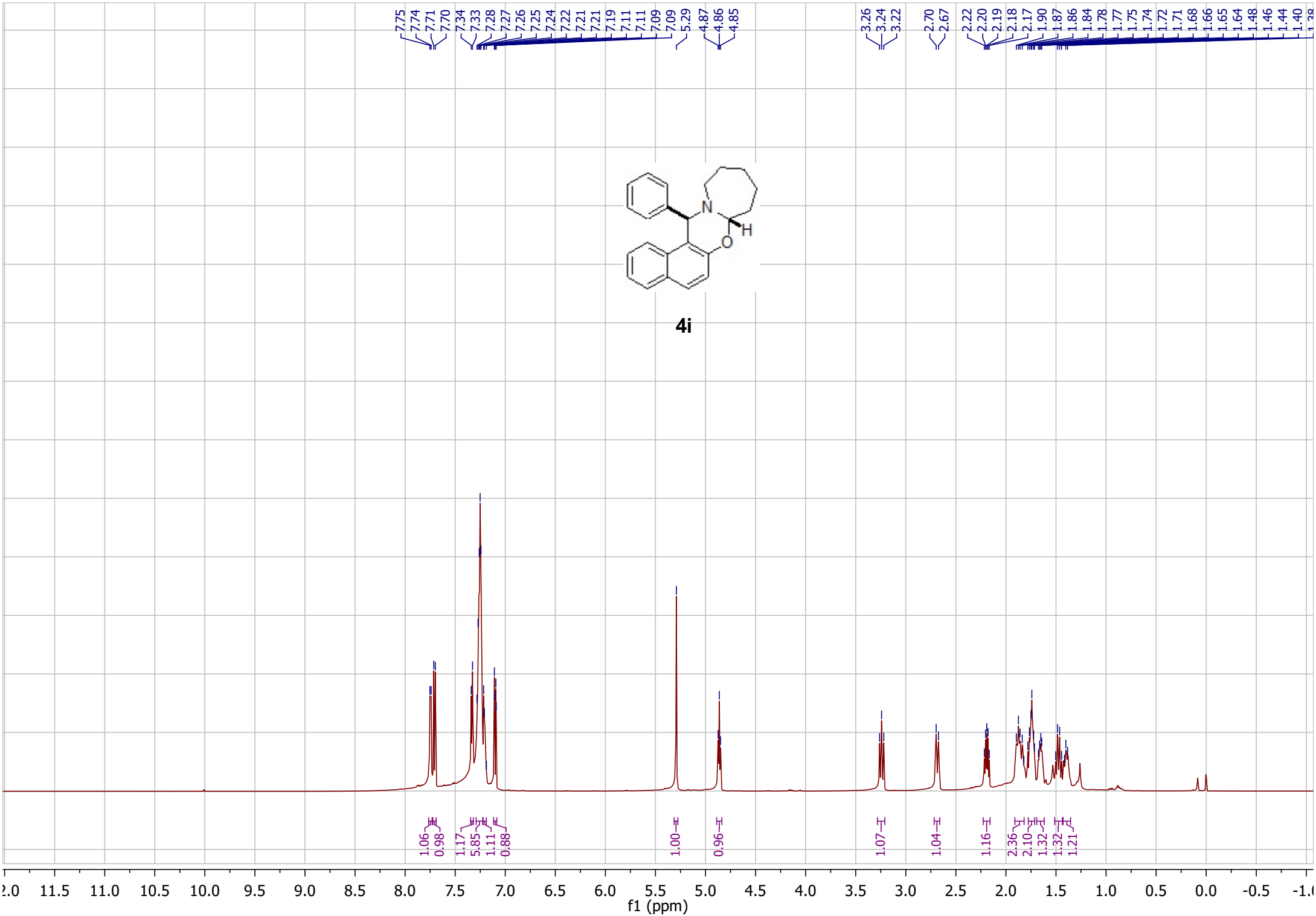
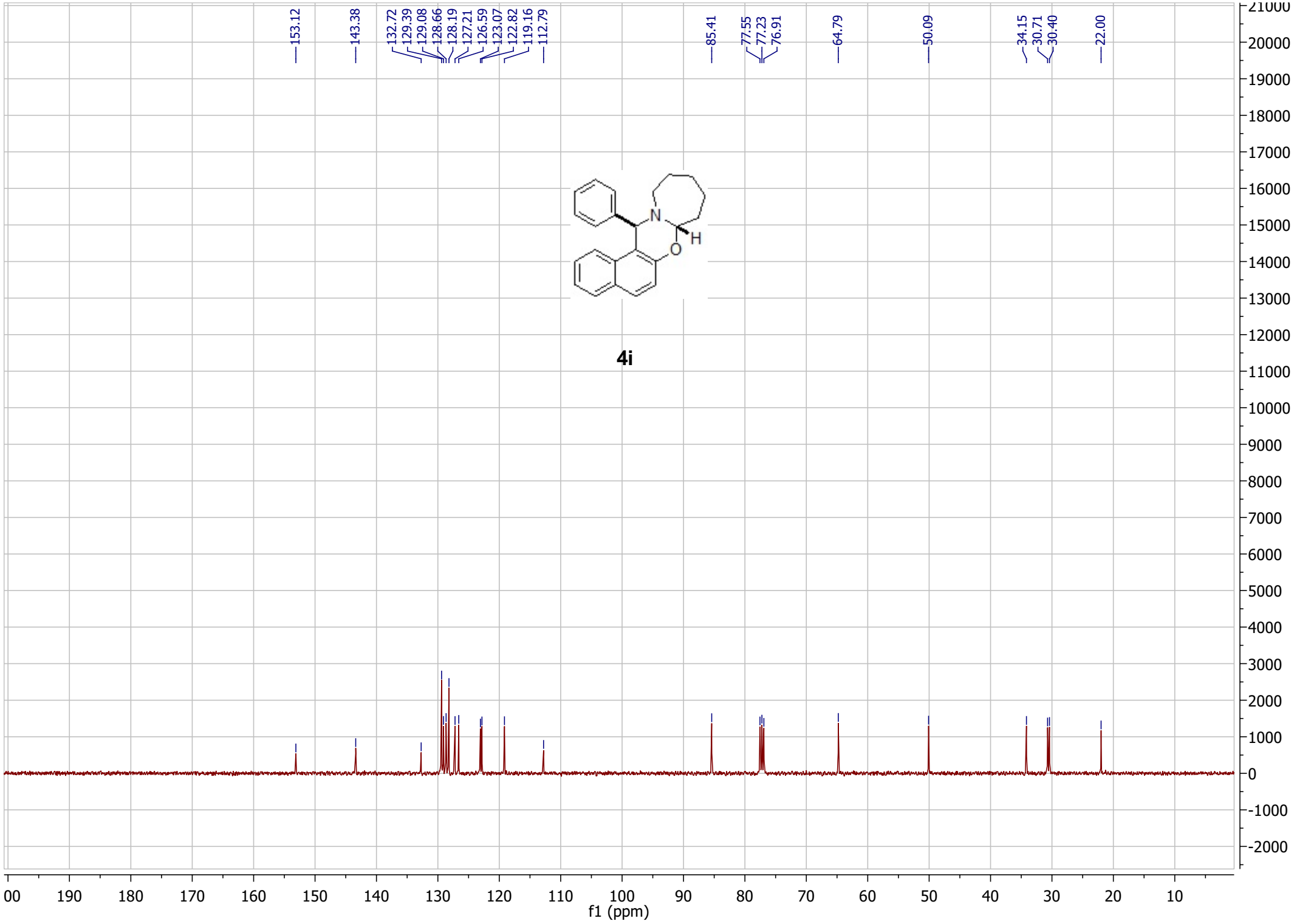
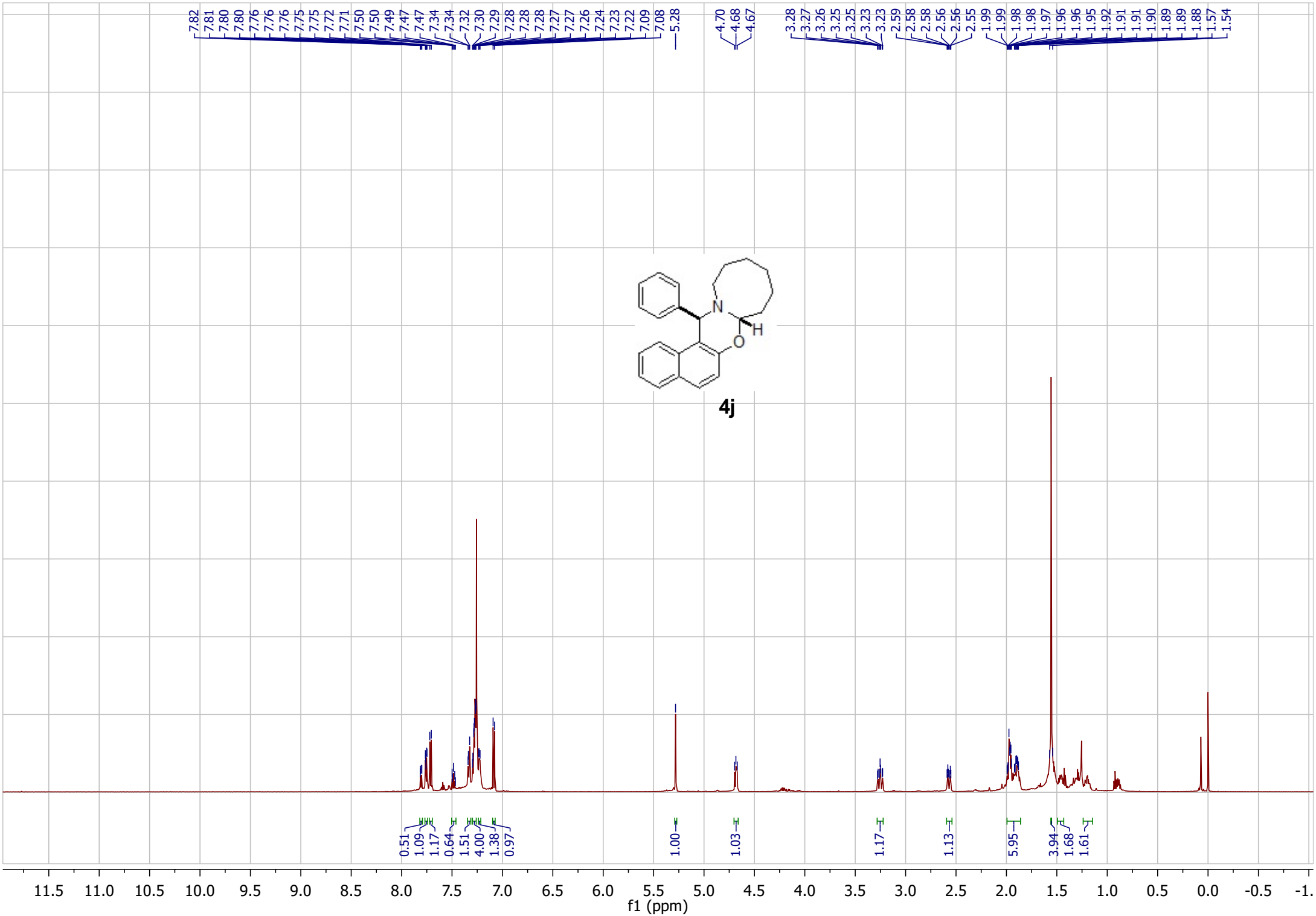
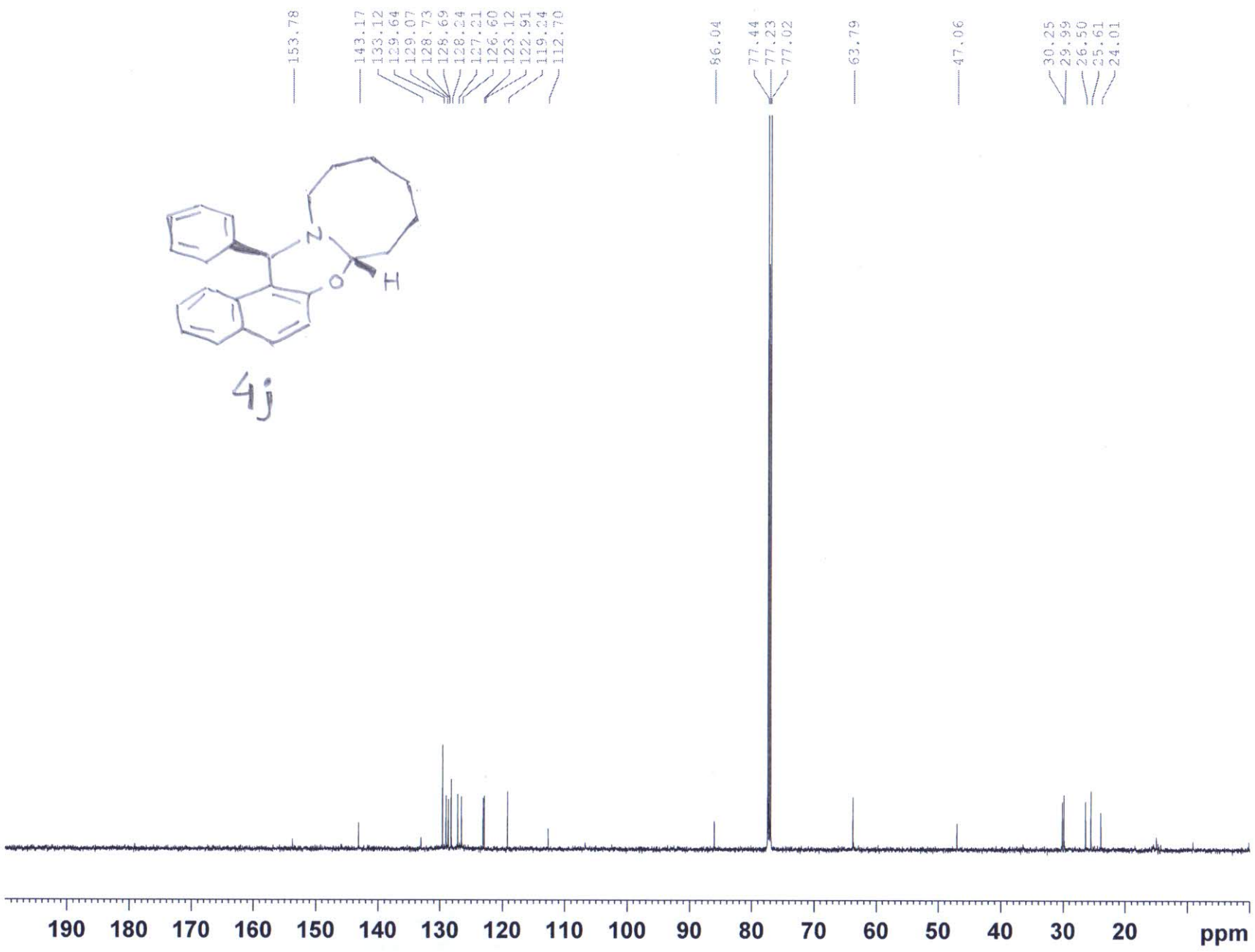
77.23
76.91
62.86
45.59
29.59
N
H
O
4h
f1 (ppm)


7.75 7.74 7.71 7.70 7.34 7.33 7.28 7.27 7.26 7.25 7.24 7.22 7.21 7.21 7.19 7.11 7.11 7.09 7.09 5.29 4.87 4.86 4.85
3.26 3.24 3.22 2.70 2.67 2.22 2.20 2.19 2.18 2.17 1.90 1.87 1.86 1.84 1.78 1.77 1.75 1.74 1.72 1.71 1.68 1.66 1.65 1.64 1.48 1.46 1.44 1.40
N
H
O
4i
1.06 0.98 1.17 5.85 1.11 0.88 1.00 0.96 1.07 1.04 1.16 2.36 2.10 1.32 1.32 1.21
2.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


153.12
143.38
132.72
129.39
129.08
128.66
128.19
127.21
126.59
123.07
122.82
119.16
112.79
85.41
77.55
77.23
76.91
64.79
50.09
34.15
30.71
30.40
22.00
N
O
H
4i
f1 (ppm)


7.82 7.81 7.80 7.80 7.76 7.76 7.76 7.75 7.75 7.72 7.71 7.50 7.50 7.49 7.47 7.47 7.34 7.34 7.32 7.30 7.29 7.28 7.28 7.28 7.27 7.27 7.26 7.24 7.23 7.22 7.09 7.08 5.28
4.70 4.68 4.67
3.28 3.27 3.26 3.25 3.25 3.23 3.23 2.59 2.58 2.58 2.56 2.56 2.55 1.99 1.99 1.98 1.98 1.97 1.96 1.96 1.95 1.92 1.91 1.91 1.90 1.89 1.89 1.88 1.57 1.54
N
H
O
4j
0.51 1.09 1.17 0.64 1.51 4.00 1.38 0.97
1.00
1.03
1.17
1.13
5.95
3.94 1.68 1.61
11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.
f1 (ppm)

CKJ-2-92A-13C



Current Data Parameters

NAME	CKJ-2-92A-13C
EXPNO	1
PROCNO	1

F2 - Acquisition Parameters

Date_	20140813
Time	16.59
INSTRUM	spect
PROBHD	5 mm PABBO BB/
PULPROG	zgpg30
TD	32768
SOLVENT	CDCl3
NS	621
DS	2
SWH	36057.691 Hz
FIDRES	1.100393 Hz
AQ	0.4543829 sec
RG	65.24
DW	13.867 usec
DE	6.50 usec
TE	299.8 K
D1	2.00000000 sec
D11	0.03000000 sec
TD0	1

======== CHANNEL f1 ========

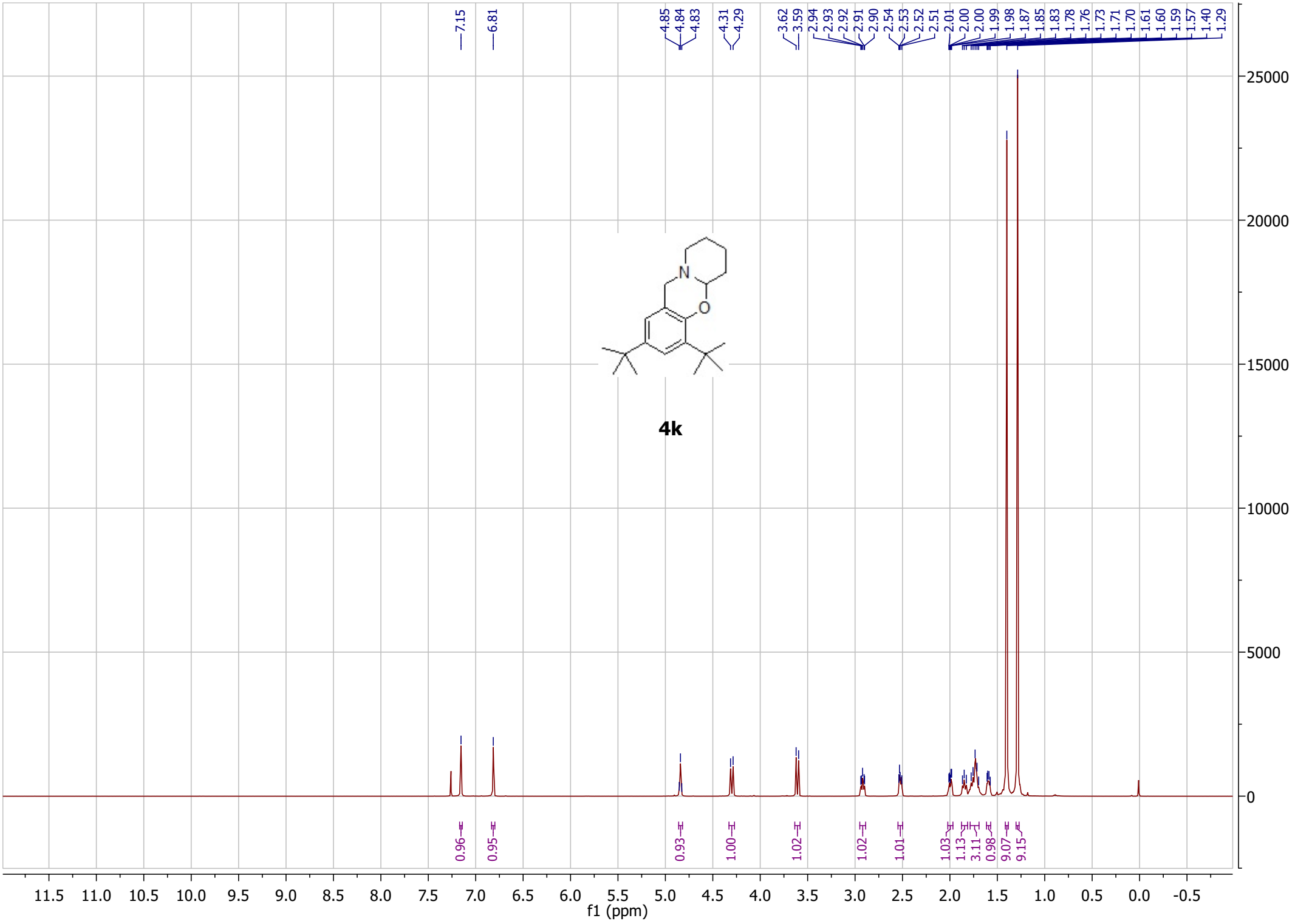
SFO1	150.9279571 MHz
NUC1	13C
P1	10.50 usec
PLW1	95.00000000 W

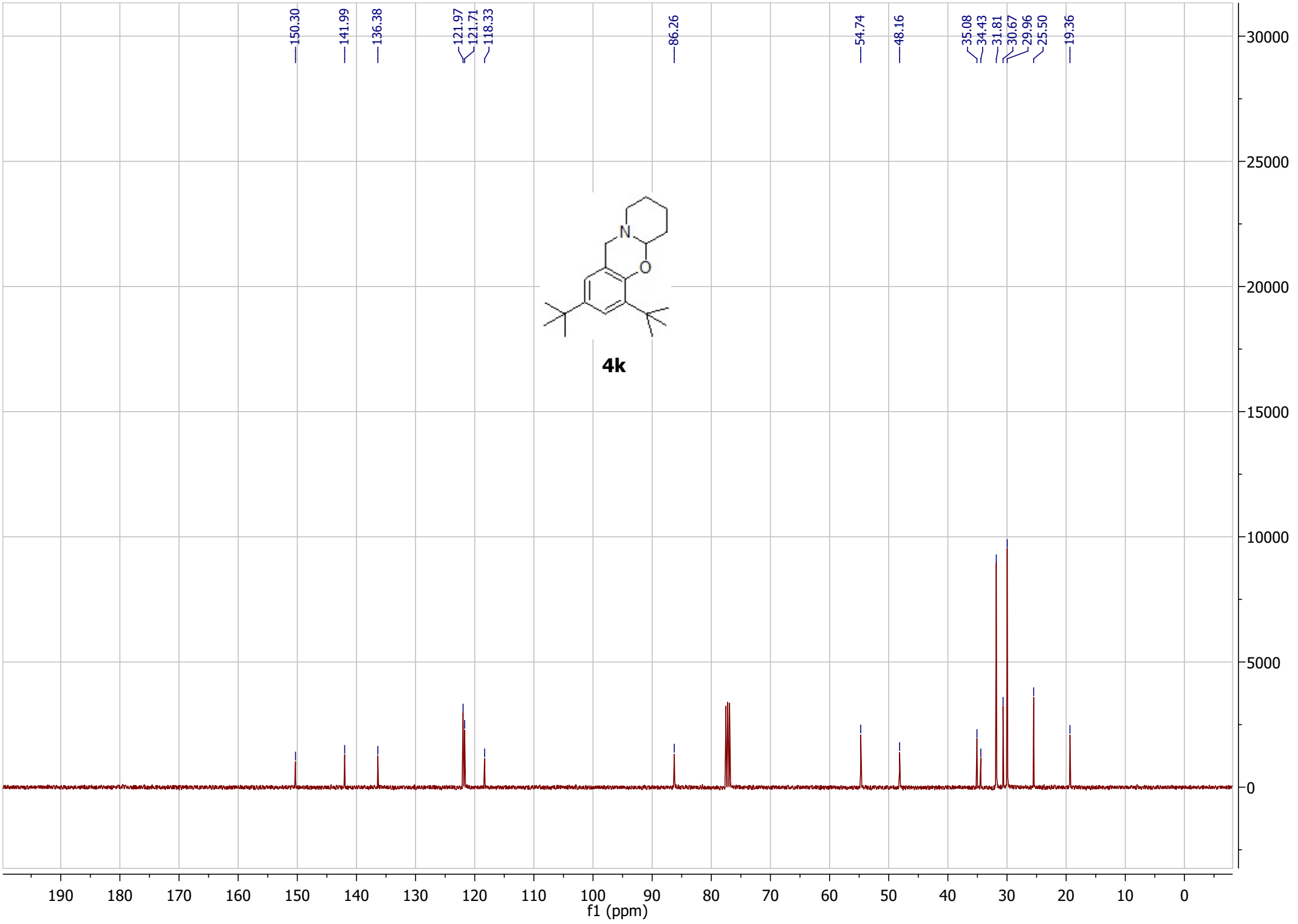
======== CHANNEL f2 ========

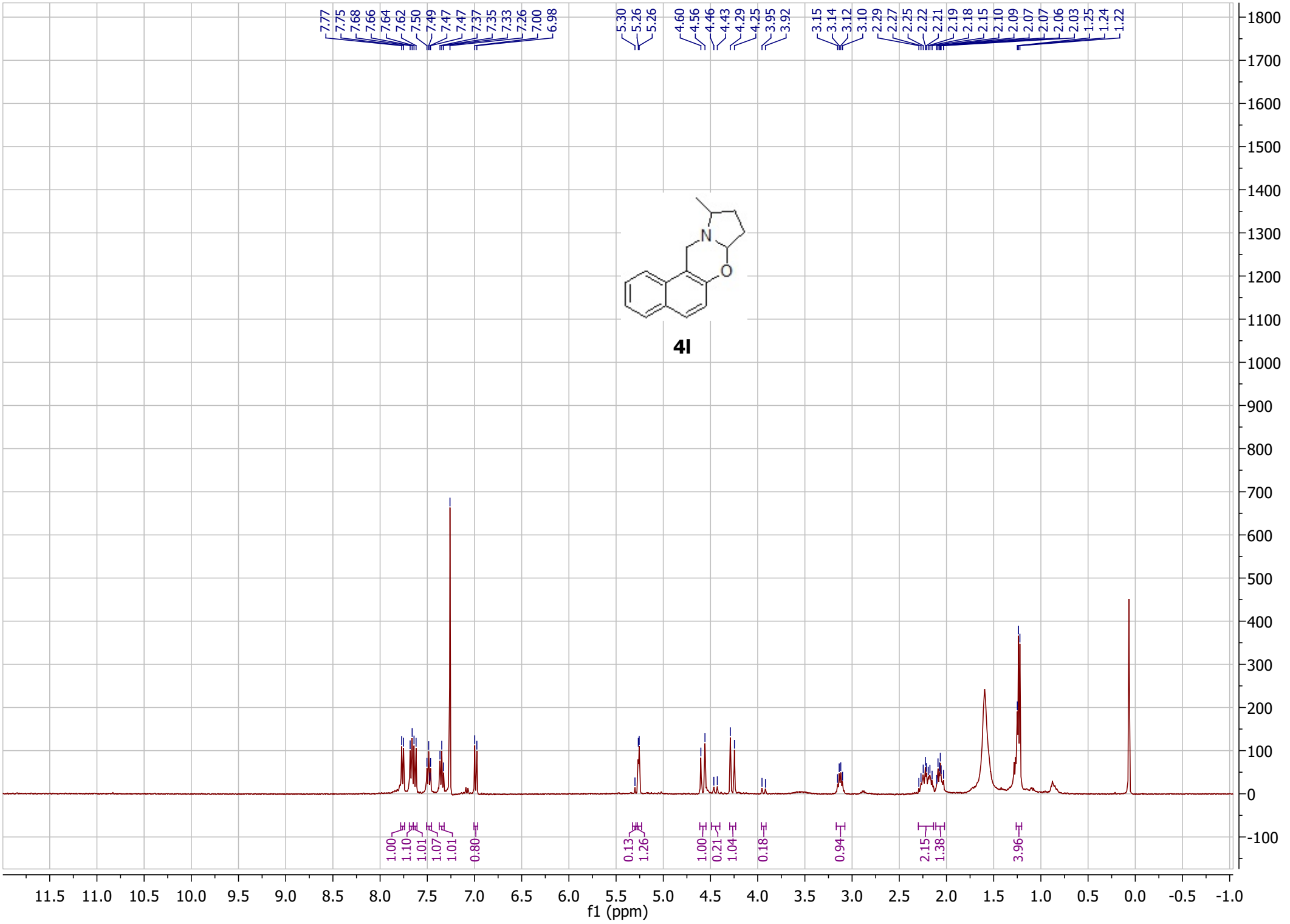
SFO2	600.1724007 MHz
NUC2	1H
CPDPRG[2	waltz16
PCPD2	70.00 usec
PLW2	21.00000000 W
PLW12	0.61714000 W
PLW13	0.30239999 W

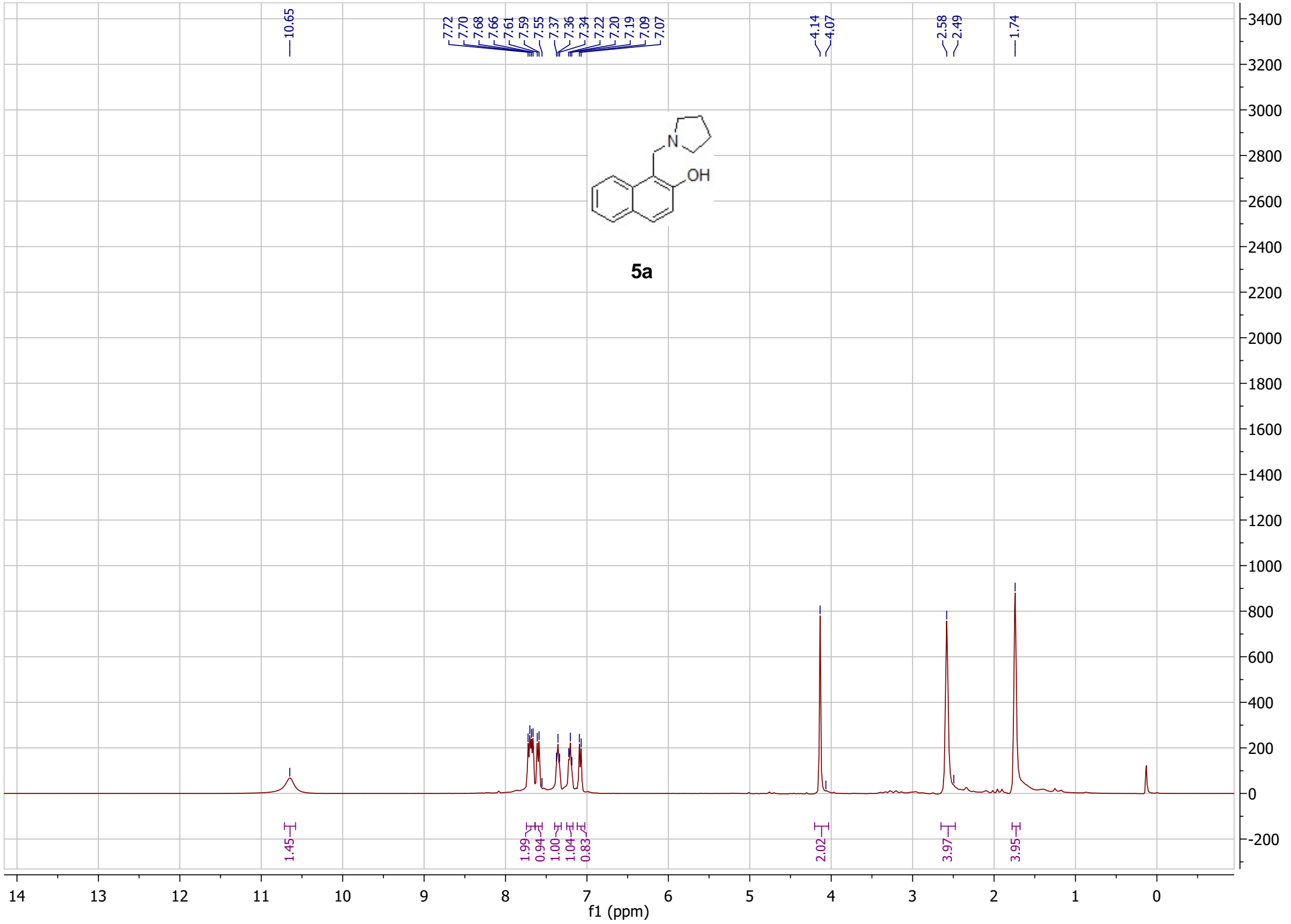
F2 - Processing parameters

SI	16384
SF	150.9128336 MHz
WDW	EM
SSB	0
LB	1.00 Hz
GB	0
PC	1.40

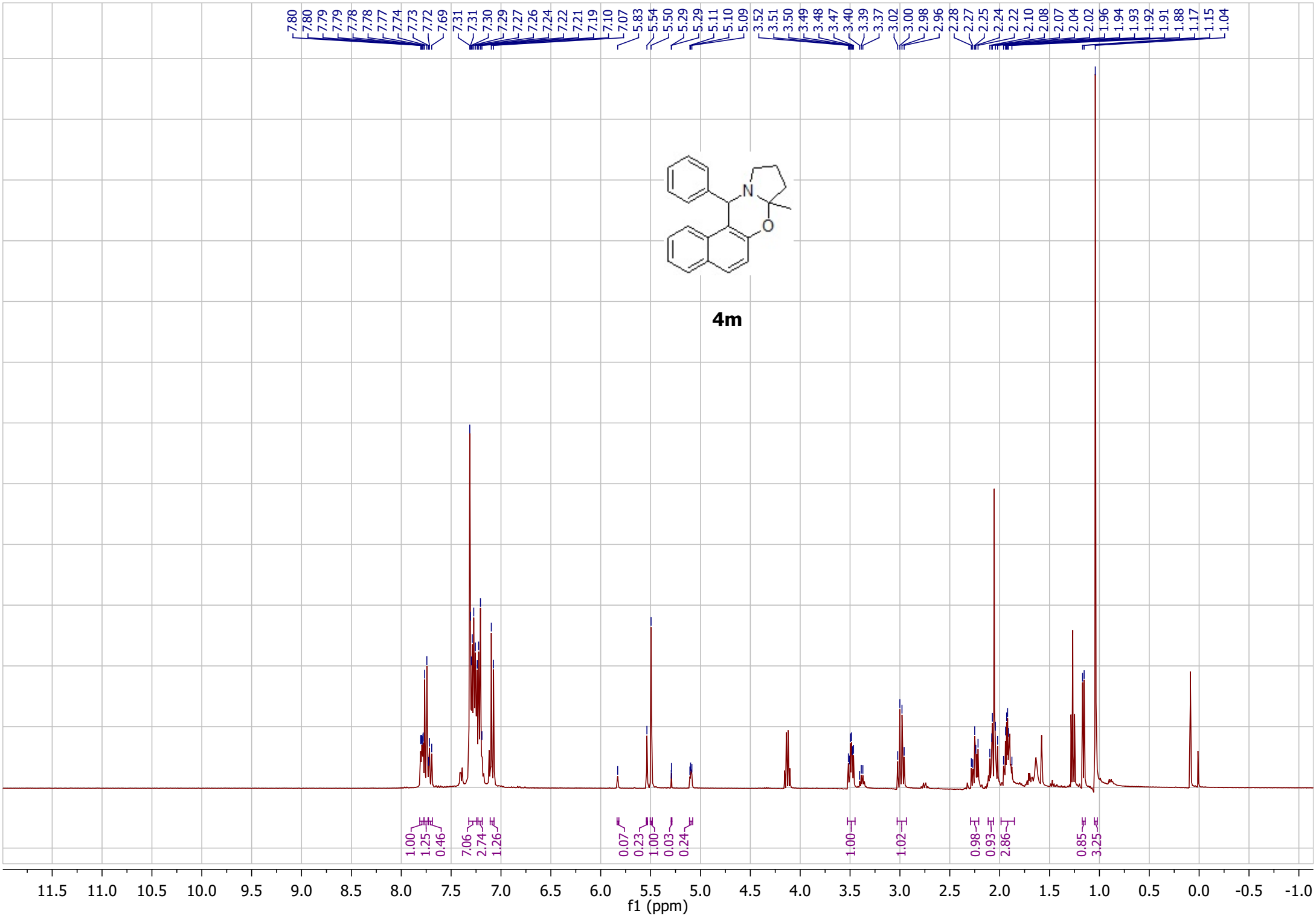

7.15
6.81
4.85
4.84
4.83
4.31
4.29
3.62
3.59
2.94
2.93
2.92
2.91
2.90
2.54
2.53
2.52
2.51
2.01
2.00
2.00
1.99
1.98
1.87
1.85
1.83
1.78
1.76
1.73
1.71
1.70
1.61
1.60
1.59
1.57
1.40
1.29
N
O
4k
0.96
0.95
0.93
1.00
1.02
1.02
1.01
1.03
1.13
3.11
0.98
9.07
9.15
25000
20000
15000
10000
5000
0
11.5
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
-0.5
f1 (ppm)

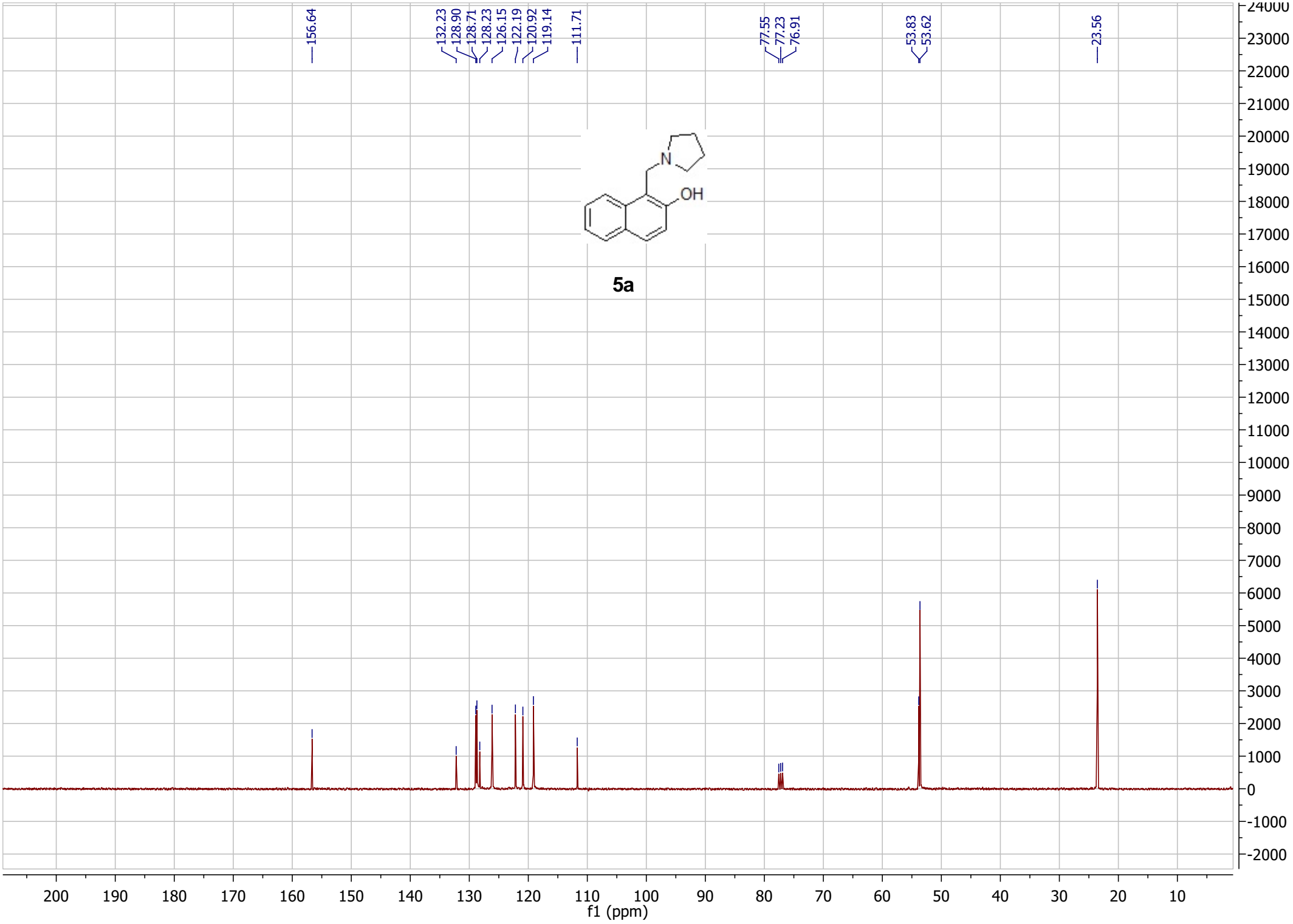

150.30
141.99
136.38
121.97
121.71
118.33
86.26
54.74
48.16
35.08
34.43
31.81
30.67
29.96
25.50
19.36
N
O
4k
30000
25000
20000
15000
10000
5000
0
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


7.77
7.75
7.68
7.66
7.64
7.62
7.50
7.49
7.47
7.47
7.37
7.35
7.33
7.26
7.00
6.98
5.30
5.26
5.26
4.60
4.56
4.46
4.43
4.29
4.25
3.95
3.92
3.15
3.14
3.12
3.10
2.29
2.27
2.25
2.22
2.21
2.19
2.18
2.15
2.10
2.09
2.07
2.07
2.06
2.03
1.25
1.24
1.22
N
O
4l
1.00
1.10
1.01
1.07
1.01
0.80
0.13
1.26
1.00
0.21
1.04
0.18
0.94
2.15
1.38
3.96
f1 (ppm)

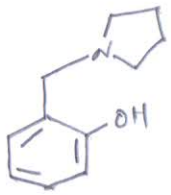

10.65
7.72
7.70
7.68
7.66
7.61
7.59
7.55
7.37
7.36
7.34
7.22
7.20
7.19
7.09
7.07
4.14
4.07
2.58
2.49
1.74
N
OH
1.45
1.99
0.94
1.00
1.04
0.83
2.02
3.97
3.95
f1 (ppm)

5a


7.80 7.80 7.79 7.79 7.78 7.78 7.77 7.74 7.73 7.72 7.69
7.31 7.31 7.30 7.29 7.27 7.26 7.24 7.22 7.21 7.19 7.10 7.07 5.83 5.54 5.50 5.29 5.29 5.11 5.10 5.09
3.52 3.51 3.50 3.49 3.48 3.47 3.40 3.39 3.37 3.02 3.00 2.98 2.96
2.28 2.27 2.25 2.24 2.22 2.10 2.08 2.07 2.04 2.02 1.96 1.94 1.93 1.92 1.91 1.88 1.17 1.15 1.04
N
O
4m
1.00 1.25 0.46 7.06 2.74 1.26 0.07 0.23 1.00 0.03 0.24 1.00 1.02 0.98 0.93 2.86 0.85 3.25
11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

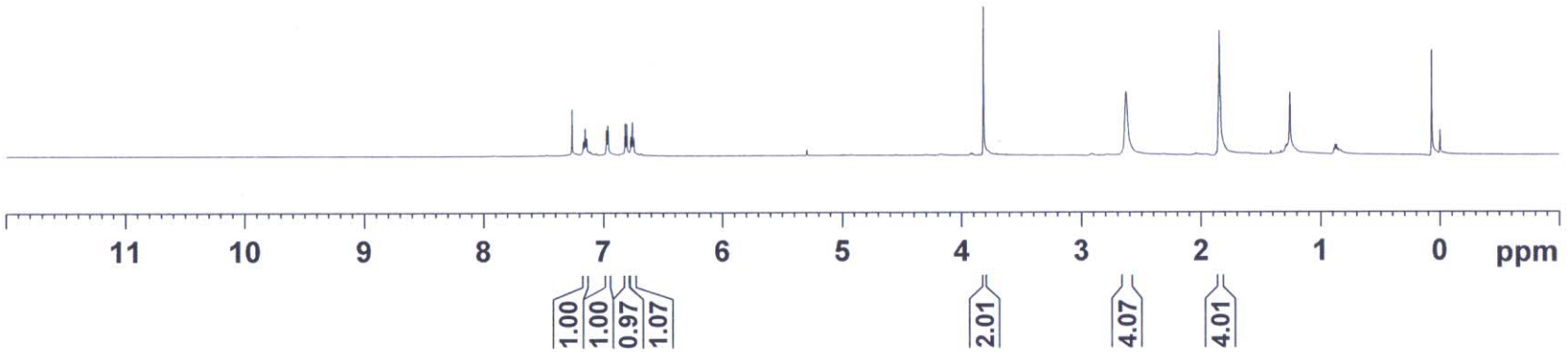

156.64
132.23
128.90
128.71
128.23
126.15
122.19
120.92
119.14
111.71
77.55
77.23
76.91
53.83
53.62
23.56
N
OH
5a
f1 (ppm)

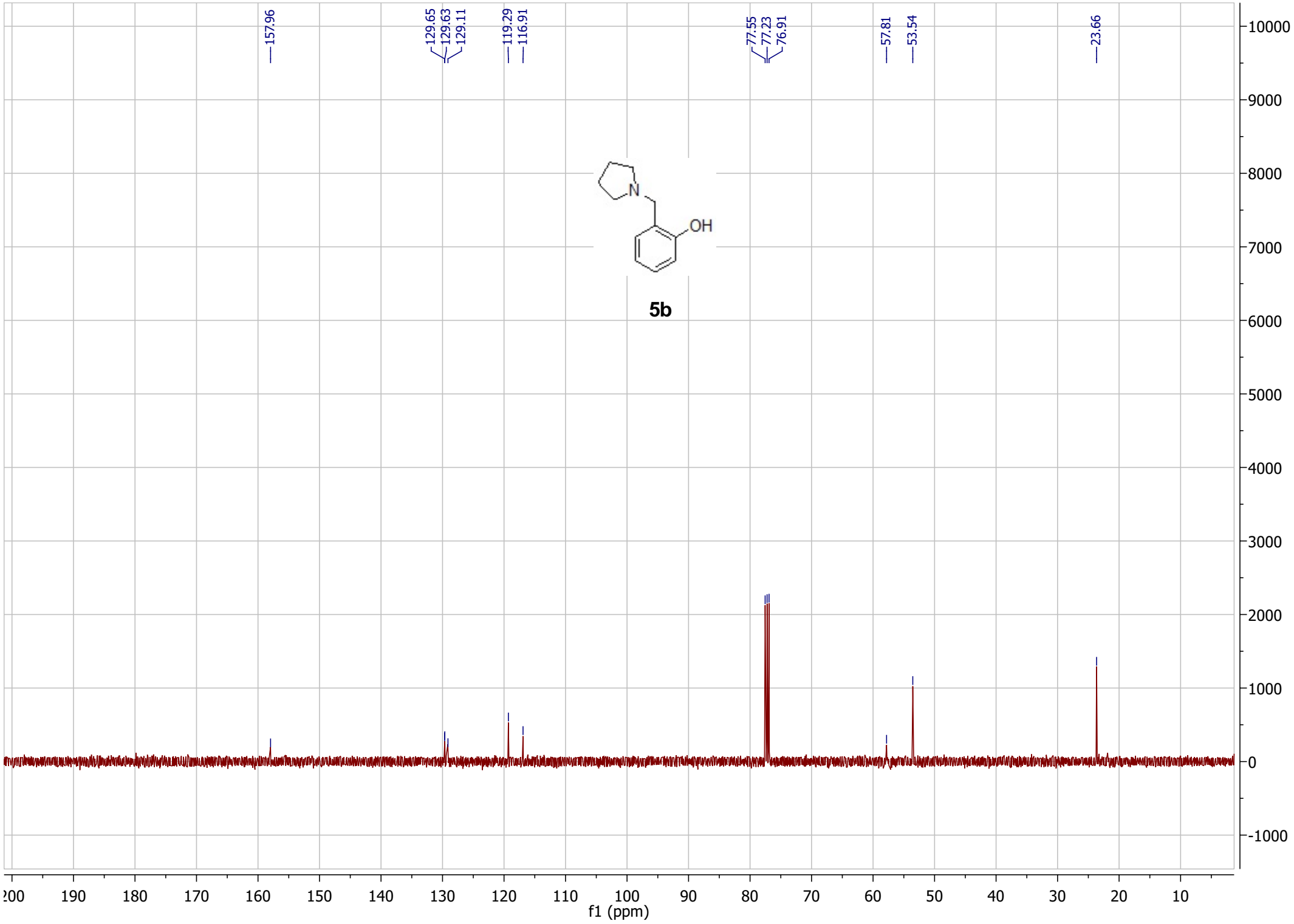
CKJ-AH-02-04



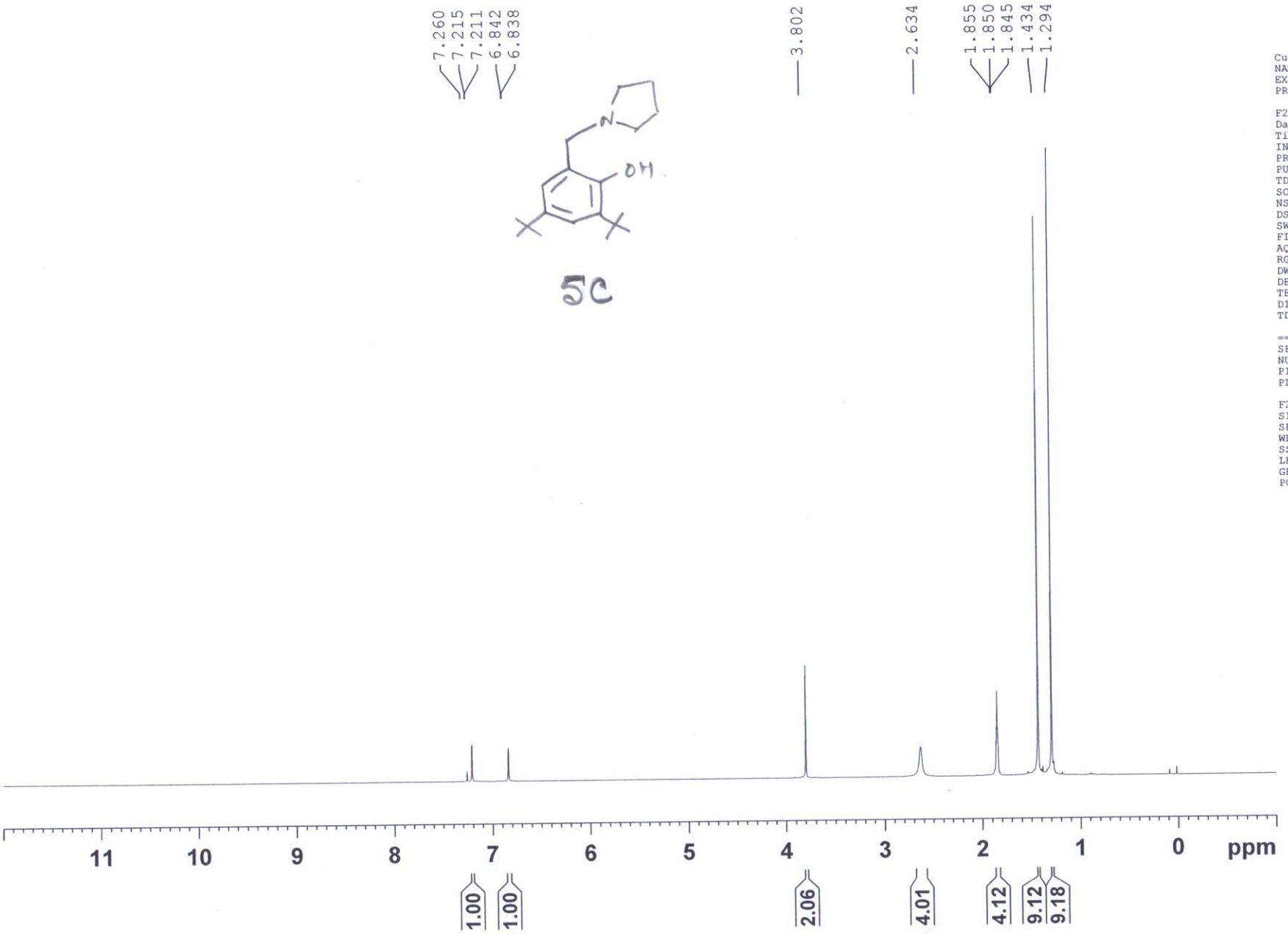
5b

```
Current Data Parameters
NAME        CKJ-AH-02-04
EXPNO                  1
PROCNO                 1

F2 - Acquisition Parameters
Date_           20140409
Time               15.14
INSTRUM            spect
PROBHD   5 mm PABBO BB/
PULPROG             zg30
TD                 32768
SOLVENT            CDCl3
NS                    16
DS                     2
SWH            12019.230 Hz
FIDRES         0.366798 Hz
AQ             1.3631488 sec
RG                 99.36
DW                41.600 usec
DE                 6.50 usec
TE                302.1 K
D1           1.00000000 sec
TD0                    1

======== CHANNEL f1 ========
SFO1         600.1737063 MHz
NUC1                   1H
P1                 12.00 usec
PLW1         21.00000000 W

F2 - Processing parameters
SI                 16384
SF           600.1700160 MHz
WDW                   EM
SSB       0
LB                 0.30 Hz
GB        0
PC                 1.00
```

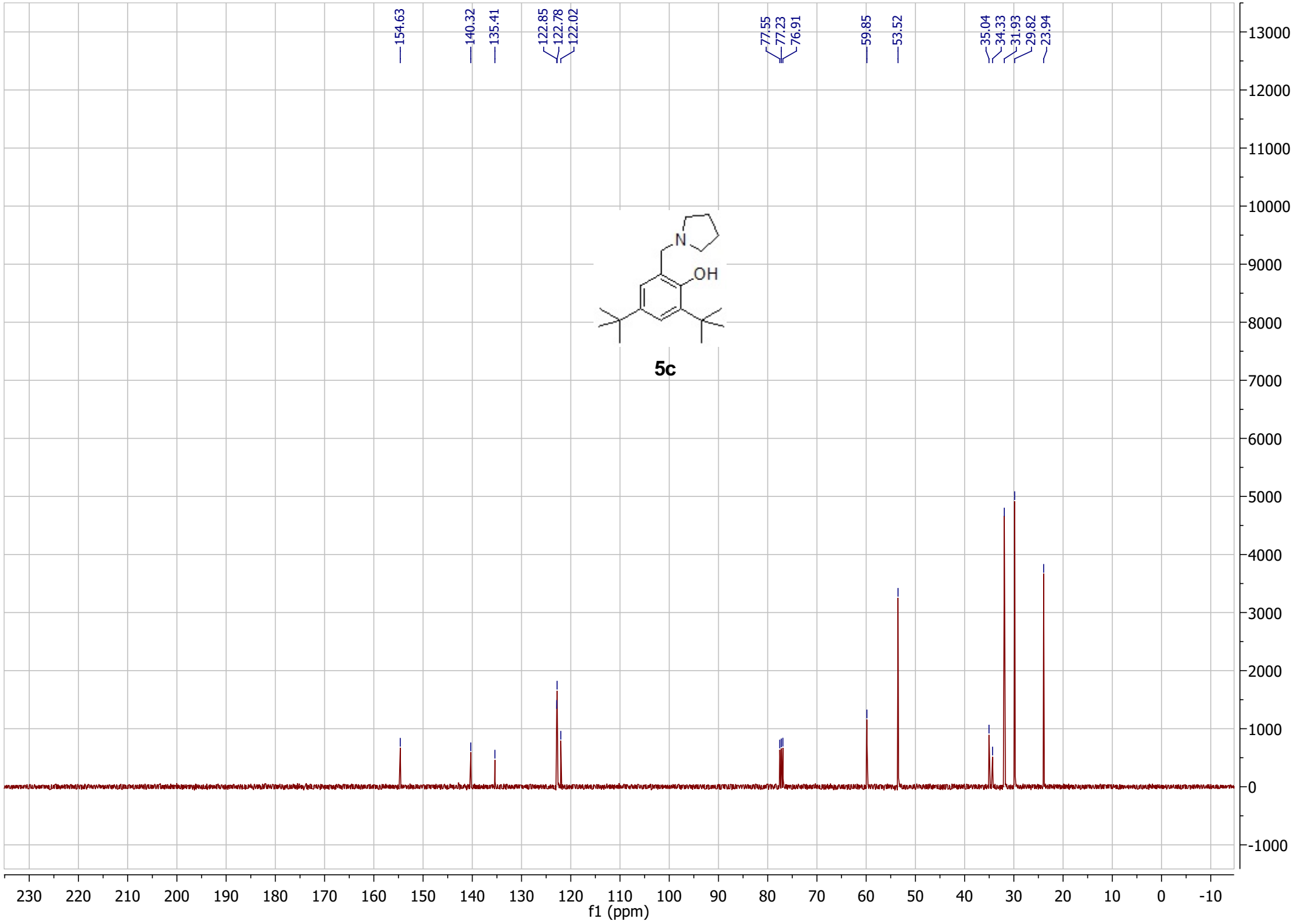



157.96
129.65
129.63
129.11
119.29
116.91
77.55
77.23
76.91
57.81
53.54
23.66
N
OH
5b
10000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
-1000
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
f1 (ppm)

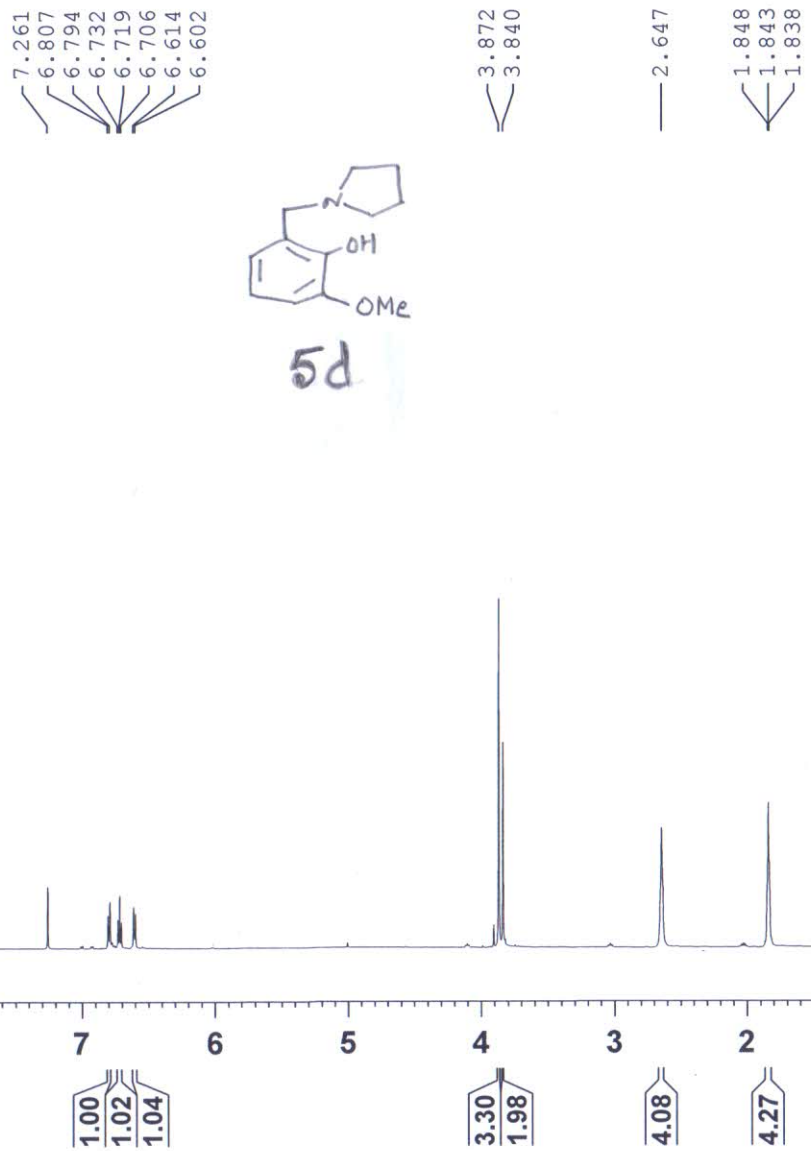
CKJ-AH-2-08B-1H



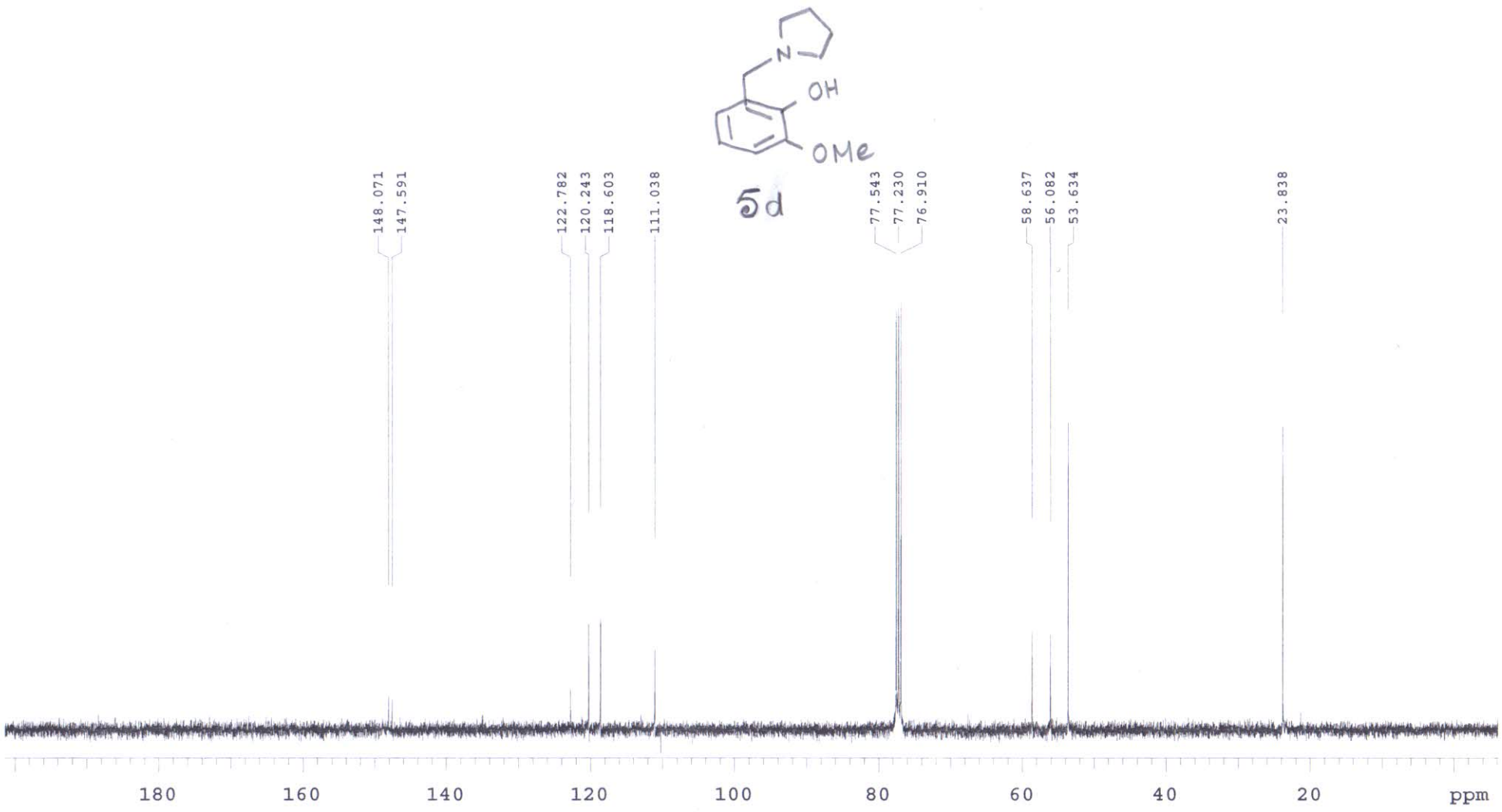

```
Current Data Parameters
NAME      CKJ-AH-2-08B-1H
EXPNO                   1
PROCNO                  1

F2 - Acquisition Parameters
Date_            20140416
Time                14.03
INSTRUM             spect
PROBHD   5 mm PABBO BB/
PULPROG             zg30
TD                 32768
SOLVENT            CDCl3
NS                    16
DS                     2
SWH           12019.230 Hz
FIDRES         0.366798 Hz
AQ             1.3631488 sec
RG                 27.82
DW                41.600 usec
DE                 6.50 usec
TE                301.4 K
D1            1.00000000 sec
TD0                    1

======== CHANNEL f1 ========
SFO1         600.1737063 MHz
NUC1                   1H
P1                 12.00 usec
PLW1         21.00000000 W

F2 - Processing parameters
SI                 16384
SF           600.1700141 MHz
WDW                   EM
SSB       0
LB                 0.30 Hz
GB        0
PC                 1.00
```


154.63
140.32
135.41
122.85
122.78
122.02
77.55
77.23
76.91
59.85
53.52
35.04
34.33
31.93
29.82
23.94
N
OH
5c
f1 (ppm)

CKJ-AH-2-09D1- 1H




```
Current Data Parameters
NAME      CKJ-AH-2-09D1- 1H
EXPNO                    1
PROCNO                   1

F2 - Acquisition Parameters
Date_             20140509
Time                 15.35
INSTRUM              spect
PROBHD   5 mm PABBO BB/
PULPROG               zg30
TD                   32768
SOLVENT              CDCl3
NS                      16
DS                       2
SWH              12019.230 Hz
FIDRES           0.366798 Hz
AQ               1.3631488 sec
RG                     113
DW                41.600 usec
DE                 6.50 usec
TE                299.0 K
D1             1.00000000 sec
TD0                      1

======== CHANNEL f1 ========
SFO1          600.1737063 MHz
NUC1                    1H
P1                  12.00 usec
PLW1          21.00000000 W

F2 - Processing parameters
SI                   16384
SF            600.1700145 MHz
WDW                     EM
SSB       0
LB                   0.30 Hz
GB        0
PC                   1.00
```



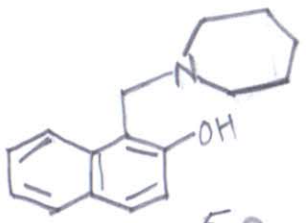
PULSE SEQUENCE
Relax. delay 1.000 sec
Pulse 45.0 degrees
Acq. time 1.304 sec
Width 25125.6 Hz
2044 repetitions

OBSERVE C13, 100.5425817
DECOUPLE H1, 399.8529994
Power 42 dB
continuously on
WALTZ-16 modulated

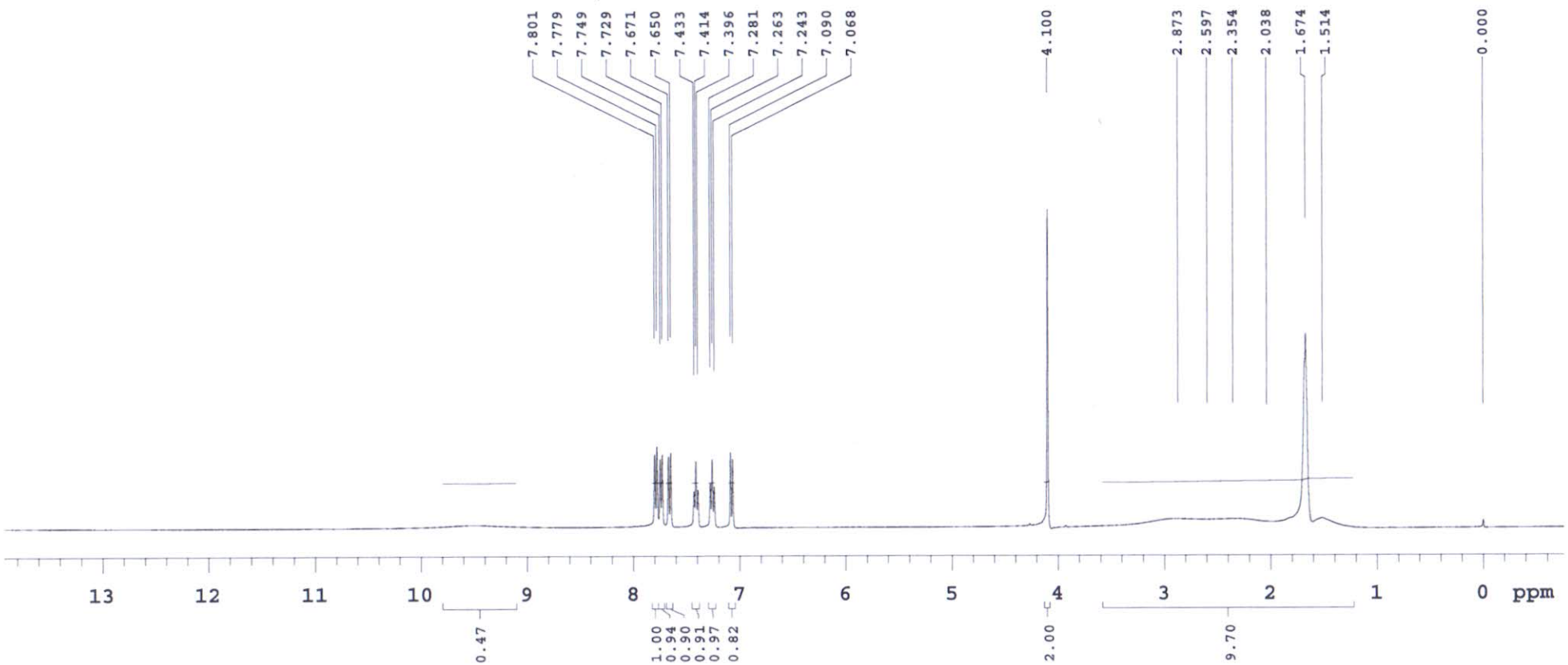
DATA PROCESSING
Line broadening 0.5 Hz
FT size 65536
Total time 78 minutes

CKJ-AH-2-09D2-13C

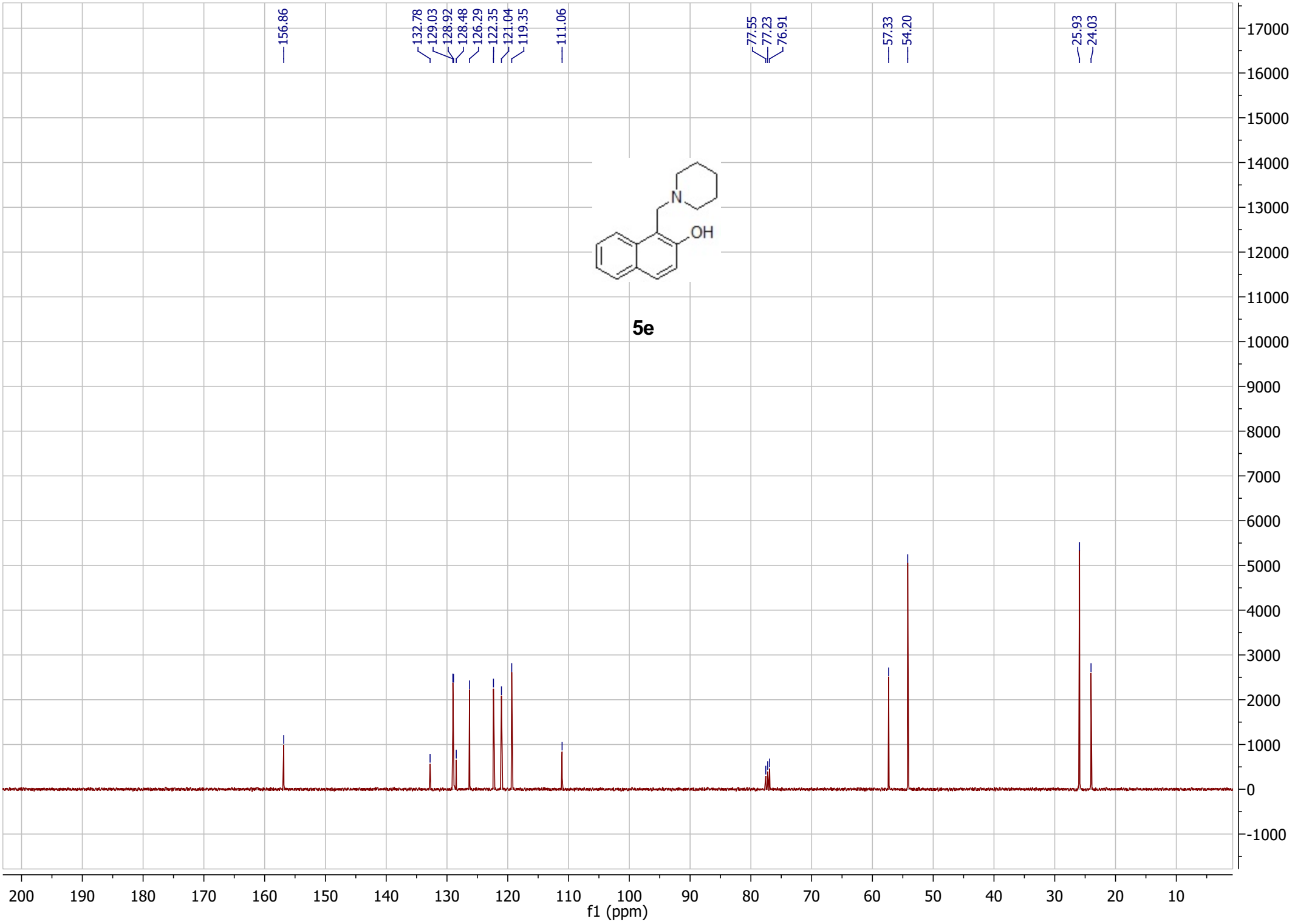
Solvent: cdcl3
Temp. 25.0 C / 298.1 K
Operator: chem
Mercury-400 "IITG-NMR"

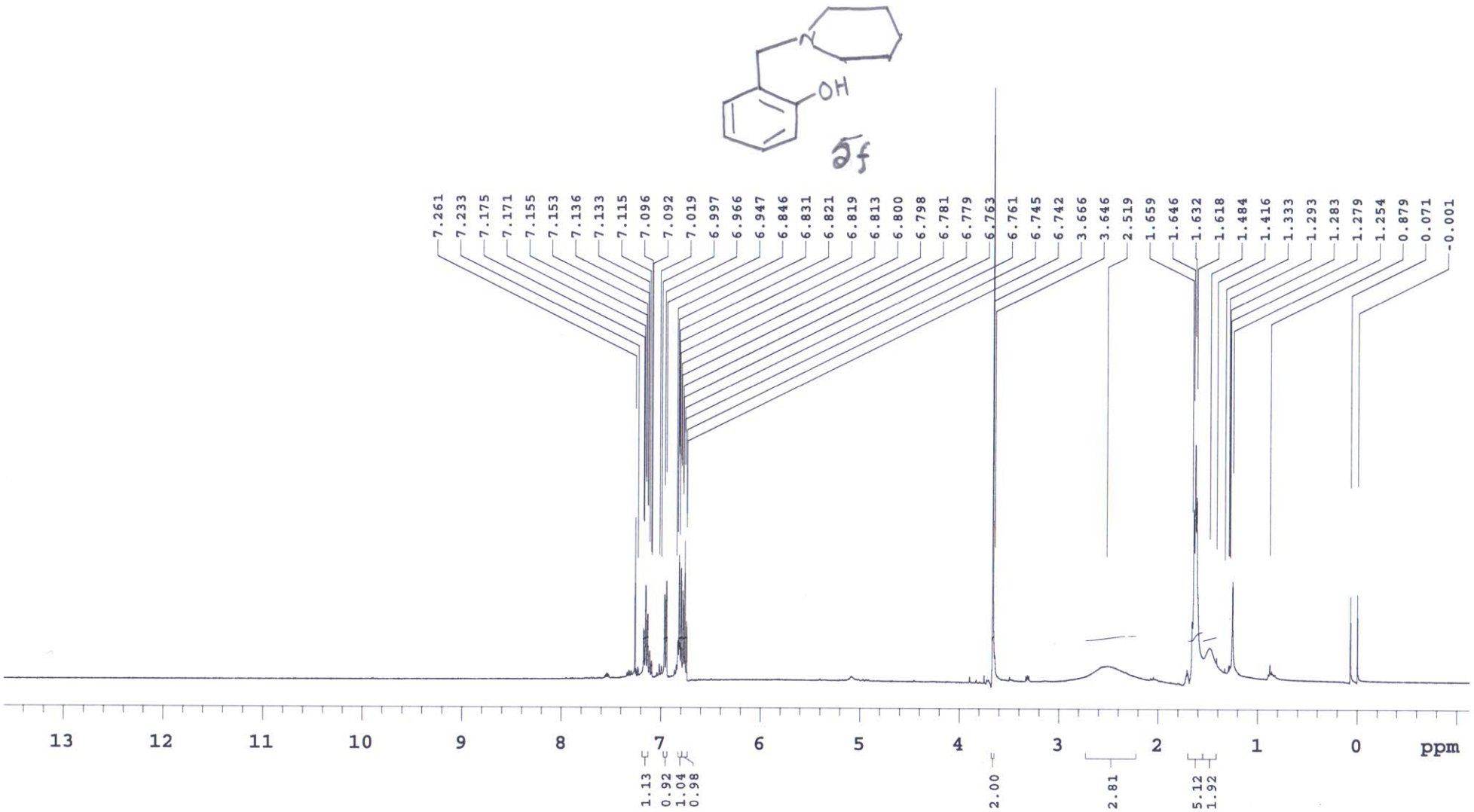


5e

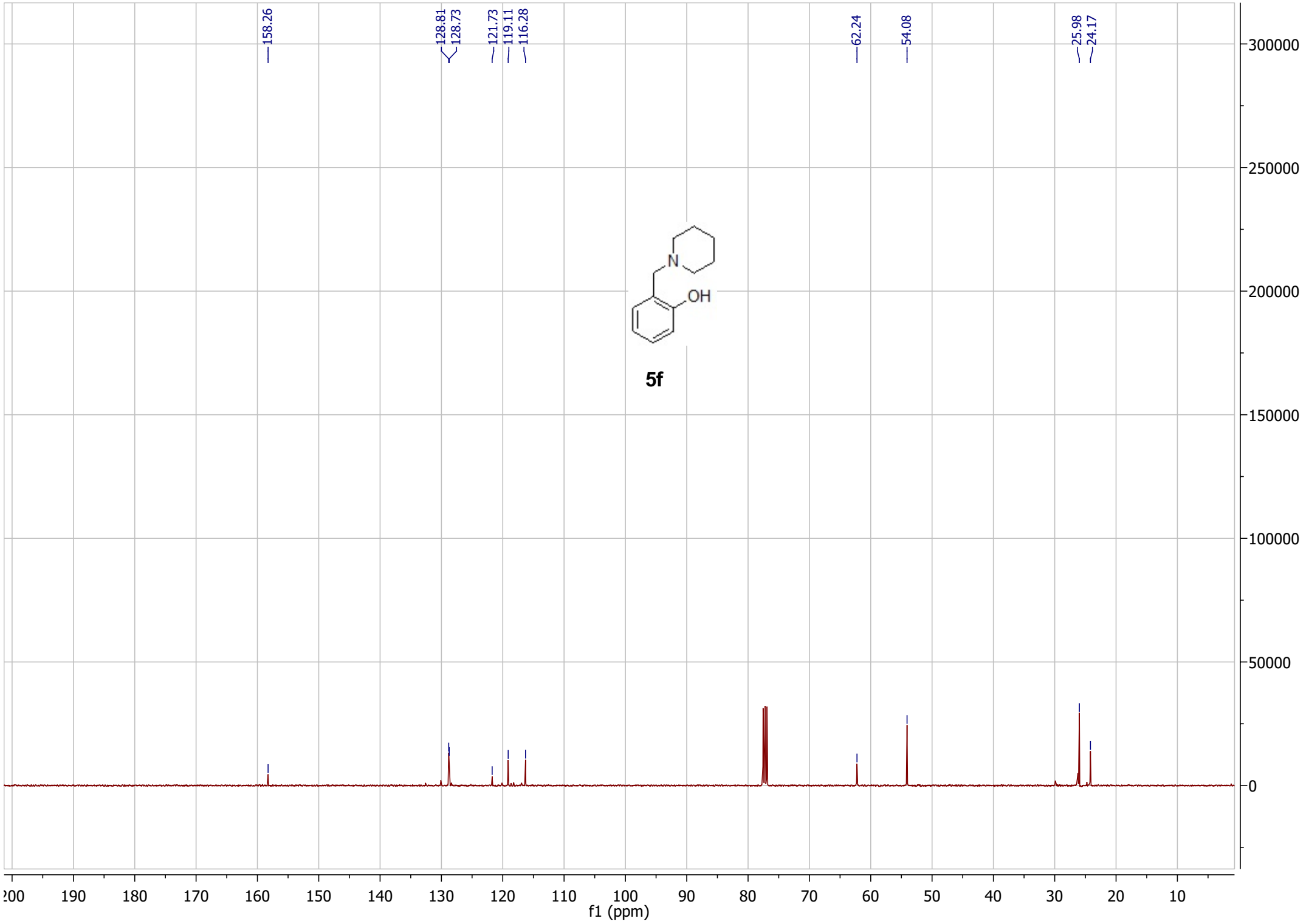


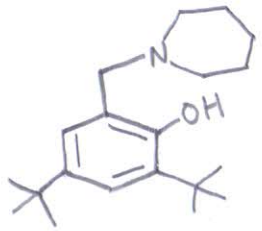
PULSE SEQUENCE Relax. delay 1.000 sec Pulse 45.0 degrees Acq. time 2.561 sec Width 8000.0 Hz 32 repetitions	OBSERVE H1, 399.8509714	DATA PROCESSING FT size 65536 Total time 1 minutes		CKJ-AH-2-11 Solvent: cdcl3 Temp. 25.0 C / 298.1 K Operator: chem File: CKJ-AH-2-11 Mercury-400 "IITG-NMR"


156.86
132.78
129.03
128.92
128.48
126.29
122.35
121.04
119.35
111.06
77.55
77.23
76.91
57.33
54.20
25.93
24.03
N
OH
5e
17000
16000
15000
14000
13000
12000
11000
10000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
-1000
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
f1 (ppm)



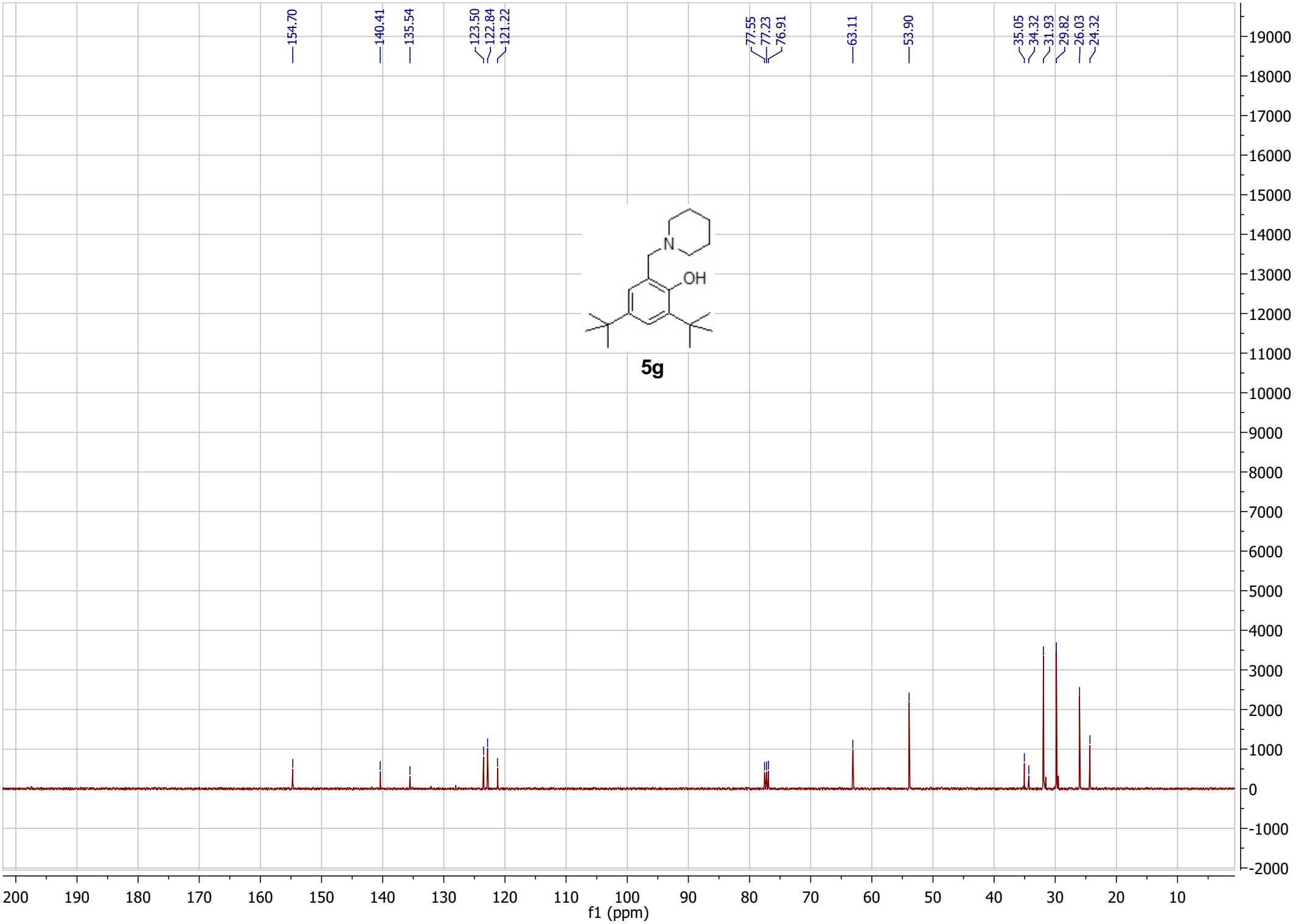
PULSE SEQUENCE Relax. delay 1.000 sec Pulse 45.0 degrees Acq. time 2.561 sec Width 10000.0 Hz 32 repetitions	OBSERVE H1, 399.8509644	DATA PROCESSING FT size 65536 Total time 1 minutes		CKJ-AH-2-12C Solvent: cdcl3 Temp. 25.0 C / 298.1 K Operator: chem File: CKJ-AH-2-12C Mercury-400 "IITG-NMR"


158.26
128.81
128.73
121.73
119.11
116.28
62.24
54.08
25.98
24.17
N
OH
5f
300000
250000
200000
150000
100000
50000
0
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
f1 (ppm)

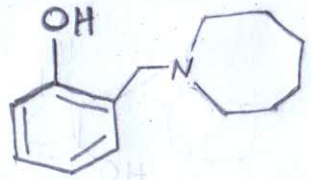


5g

PULSE SEQUENCE Relax. delay 1.000 sec Pulse 45.0 degrees Acq. time 2.561 sec Width 6398.0 Hz 32 repetitions	OBSERVE H1, 399.8509637	DATA PROCESSING FT size 32768 Total time 1 minutes		CKJ-AH-2-13A Solvent: cdcl3 Temp. 25.0 C / 298.1 K Operator: chem File: CKJ-AH-2-13A Mercury-400 "IITG-NMR"

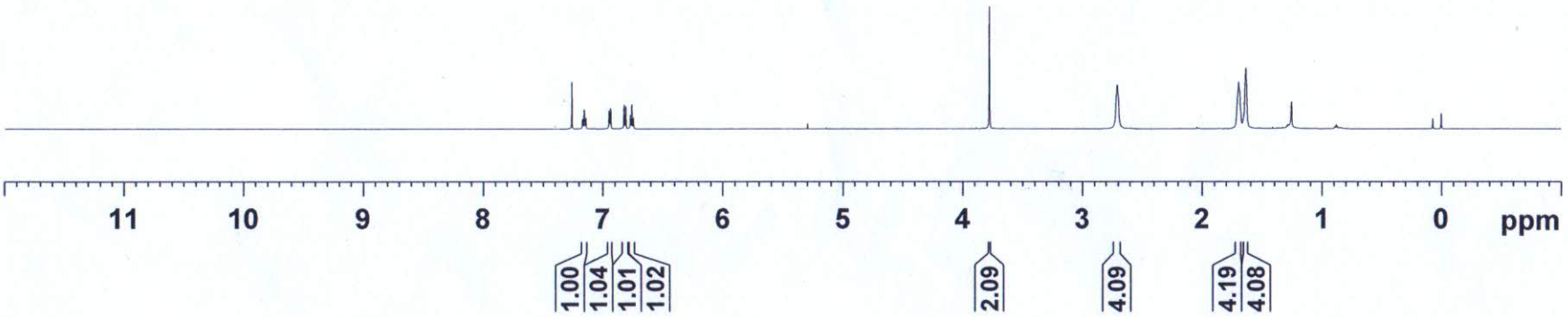

154.70
140.41
135.54
123.50
122.84
121.22
77.55
77.23
76.91
63.11
53.90
35.05
34.32
31.93
29.82
26.03
24.32
N
OH
5g
f1 (ppm)

CKJ-AH-2-16C-1H

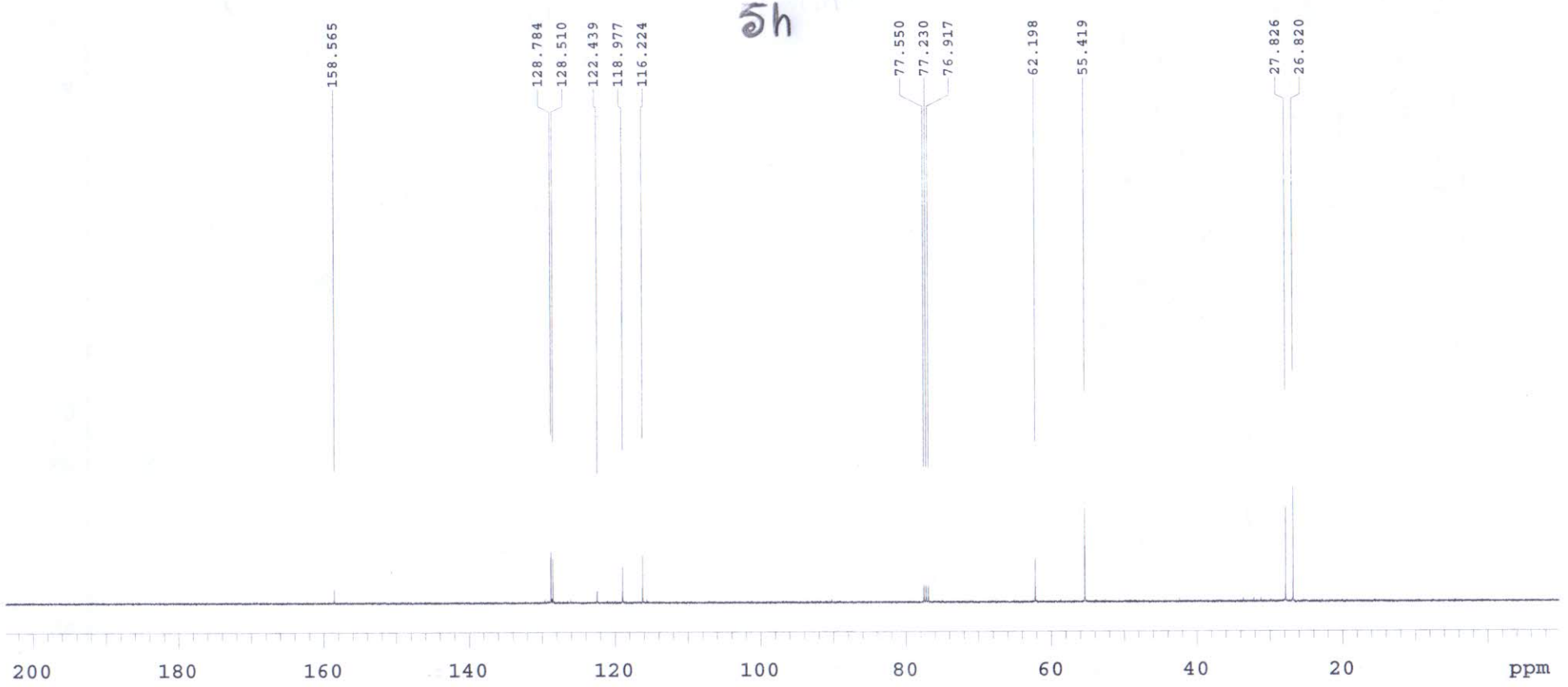


5h

```
Current Data Parameters
NAME      CKJ-AH-2-16C-1H
EXPNO                   1
PROCNO                  1

F2 - Acquisition Parameters
Date_            20140417
Time                11.09
INSTRUM             spect
PROBHD   5 mm PABBO BB/
PULPROG              zg30
TD                  32768
SOLVENT             CDCl3
NS                     16
DS                      2
SWH           12019.230 Hz
FIDRES         0.366798 Hz
AQ             1.3631488 sec
RG                 99.36
DW                41.600 usec
DE                 6.50 usec
TE                301.8 K
D1           1.00000000 sec
TD0                     1

======== CHANNEL f1 ========
SFO1         600.1737063 MHz
NUC1                   1H
P1                 12.00 usec
PLW1        21.00000000 W

F2 - Processing parameters
SI                  16384
SF           600.1700148 MHz
WDW                    EM
SSB      0
LB                 0.30 Hz
GB       0
PC                 1.00
```



5h

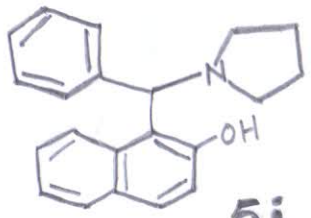


PULSE SEQUENCE
Relax. delay 1.000 sec
Pulse 45.0 degrees
Acq. time 1.304 sec
Width 25125.6 Hz
384 repetitions

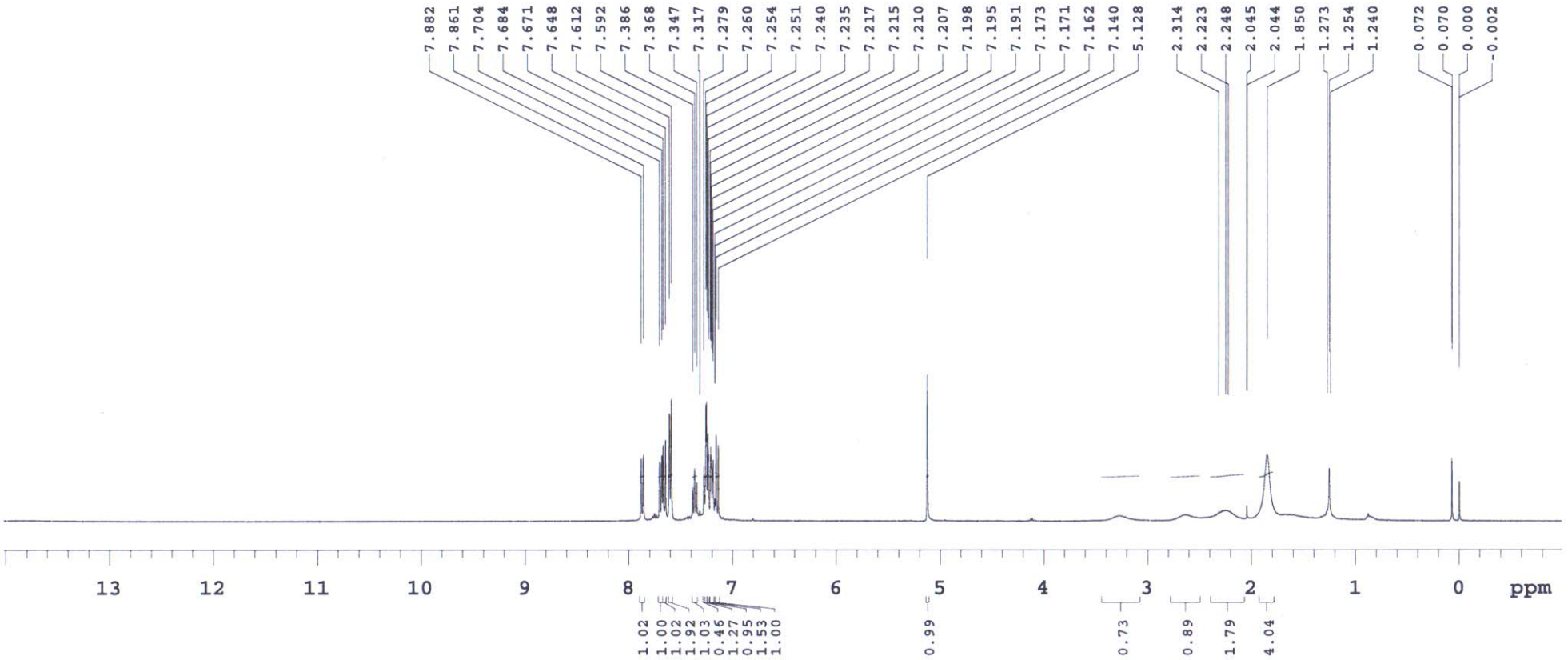
OBSERVE C13, 100.5425878
DECOUPLE H1, 399.8529994
Power 42 dB
continuously on
WALTZ-16 modulated

DATA PROCESSING
Line broadening 0.5 Hz
FT size 65536
Total time 14 minutes

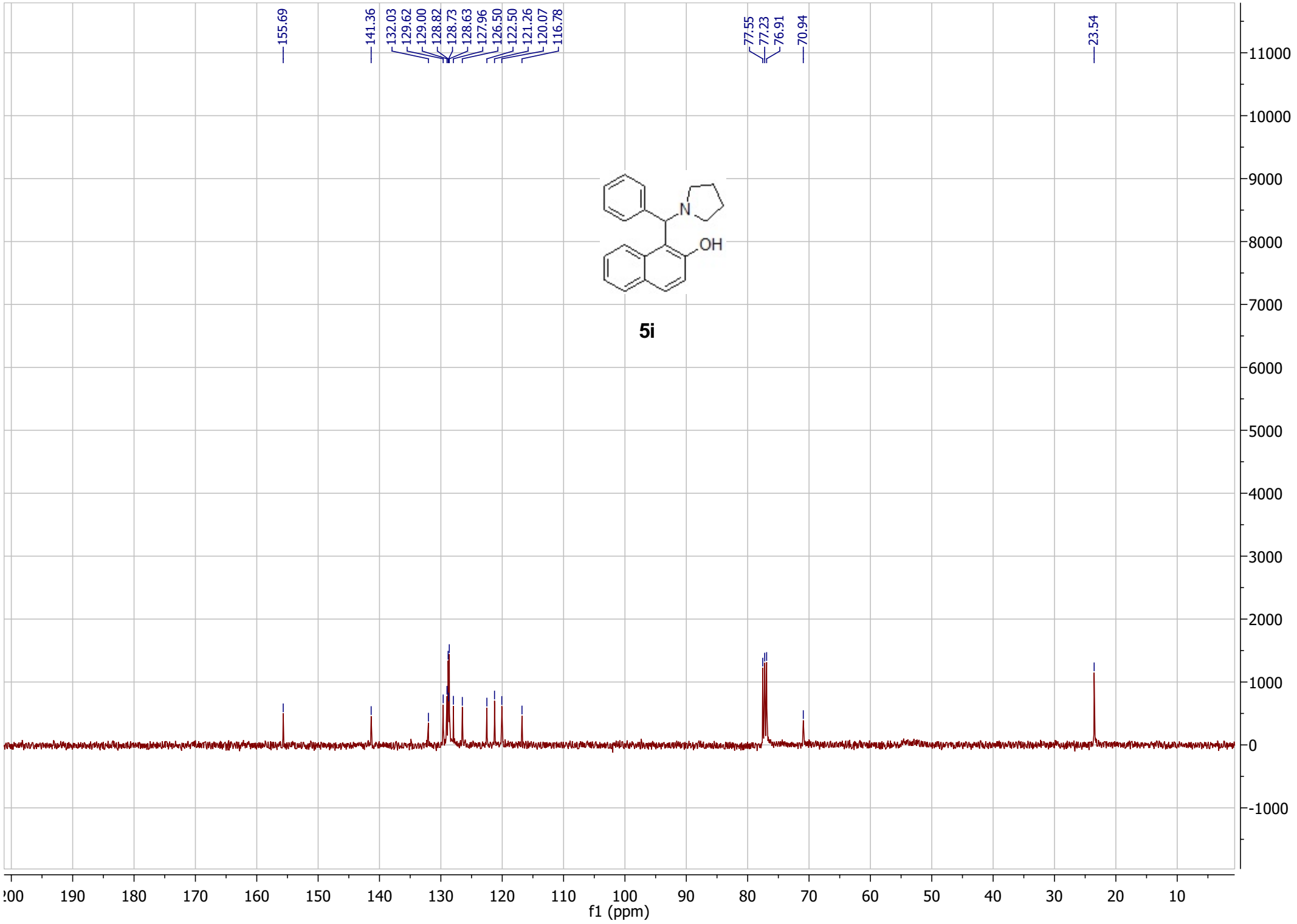
CKJ-AH-2-16C-13C
Solvent: cdcl3
Temp. 25.0 C / 298.1 K
Operator: chem
Mercury-400 "IITG-NMR"

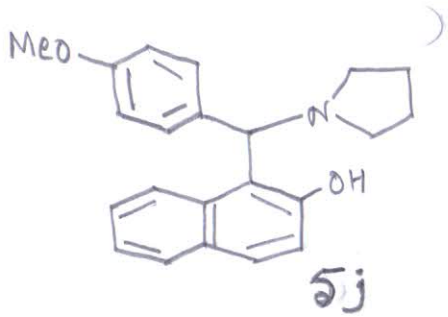


5i

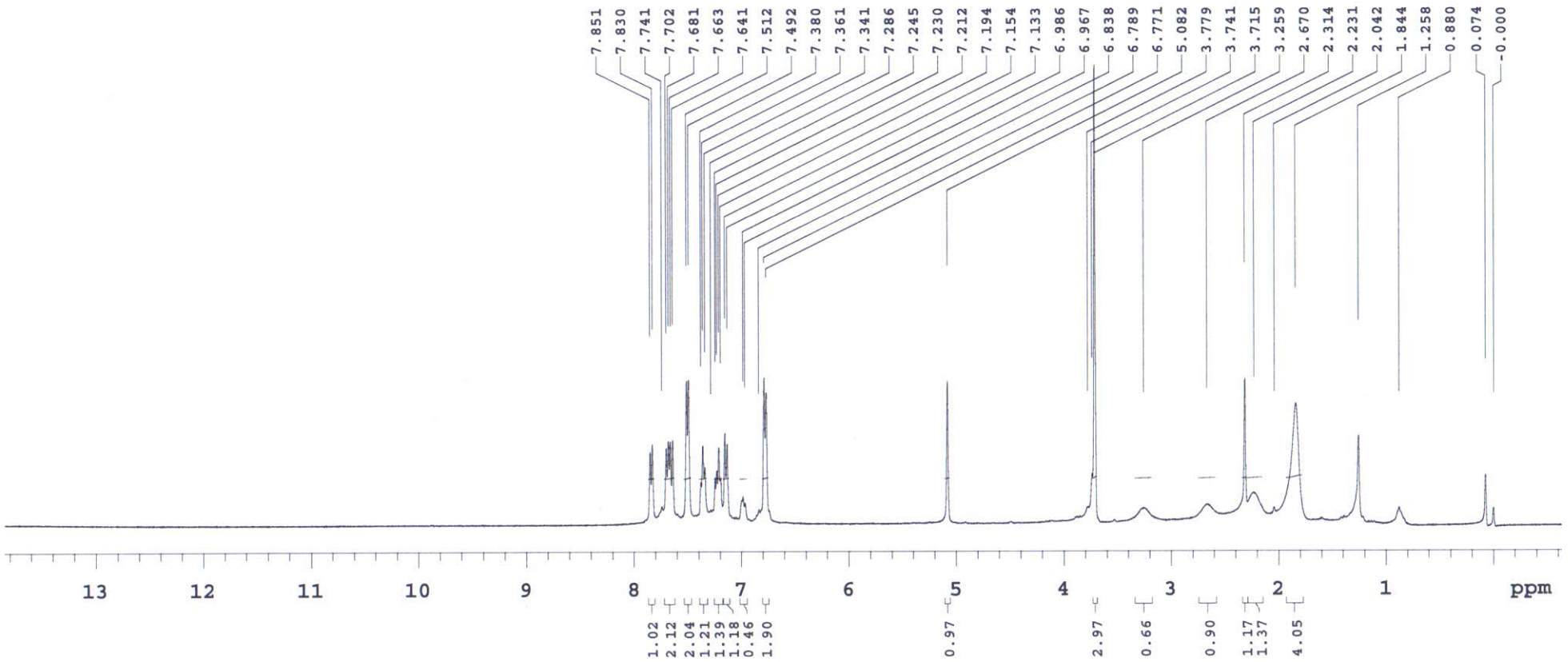


PULSE SEQUENCE Relax. delay 1.000 sec Pulse 45.0 degrees Acq. time 2.561 sec Width 8000.0 Hz 32 repetitions	OBSERVE H1, 399.8509667	DATA PROCESSING FT size 65536 Total time 1 minutes		CKJ-AH-2-10C Solvent: cdcl3 Temp. 25.0 C / 298.1 K Operator: chem File: CKJ-AH-2-10C Mercury-400 "IITG-NMR"

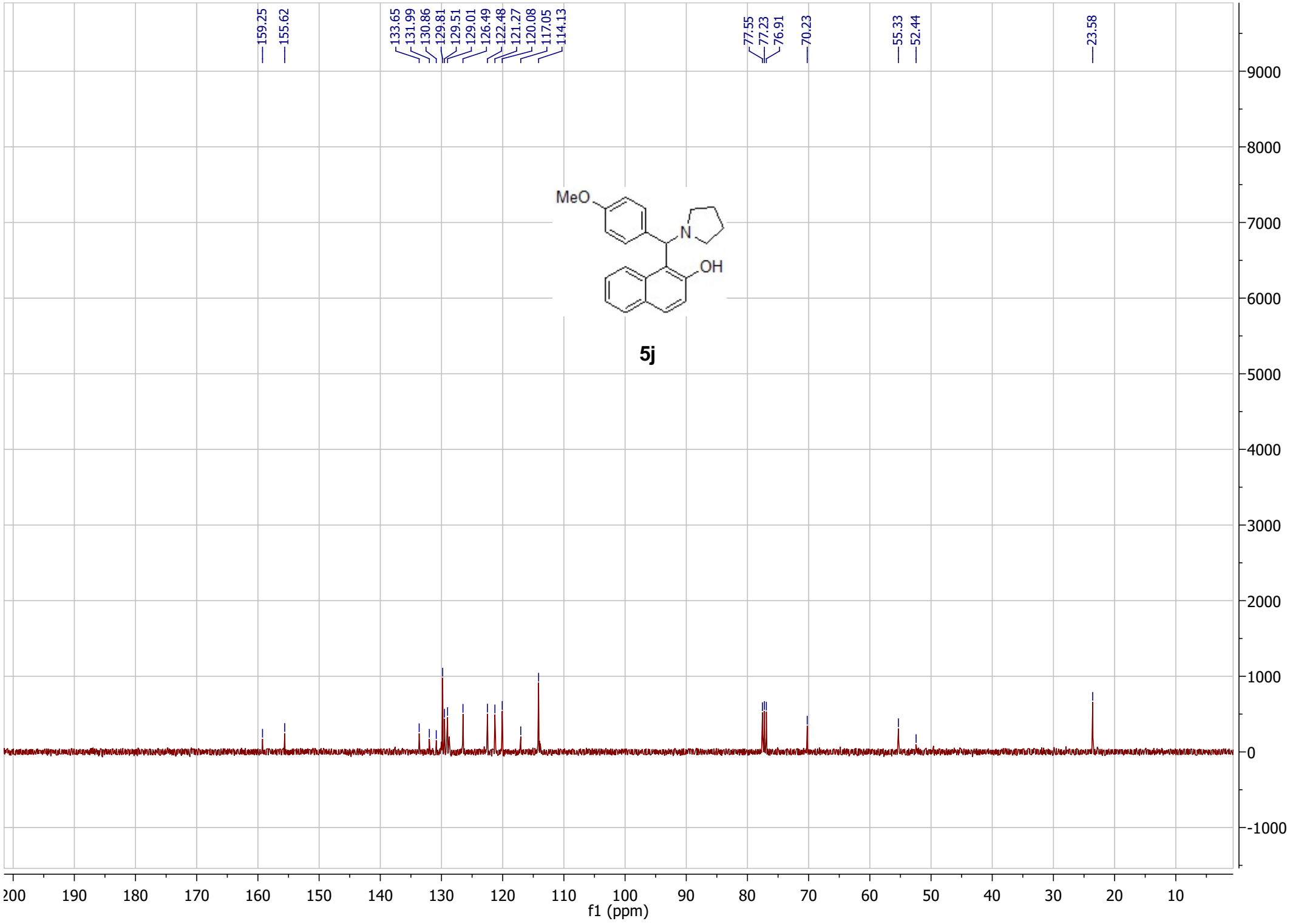

155.69
141.36
132.03
129.62
129.00
128.82
128.73
128.63
127.96
126.50
122.50
121.26
120.07
116.78
77.55
77.23
76.91
70.94
23.54
N
OH
5i
11000
10000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
-1000
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
f1 (ppm)

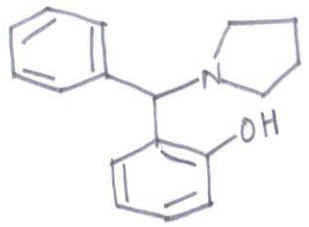


5j

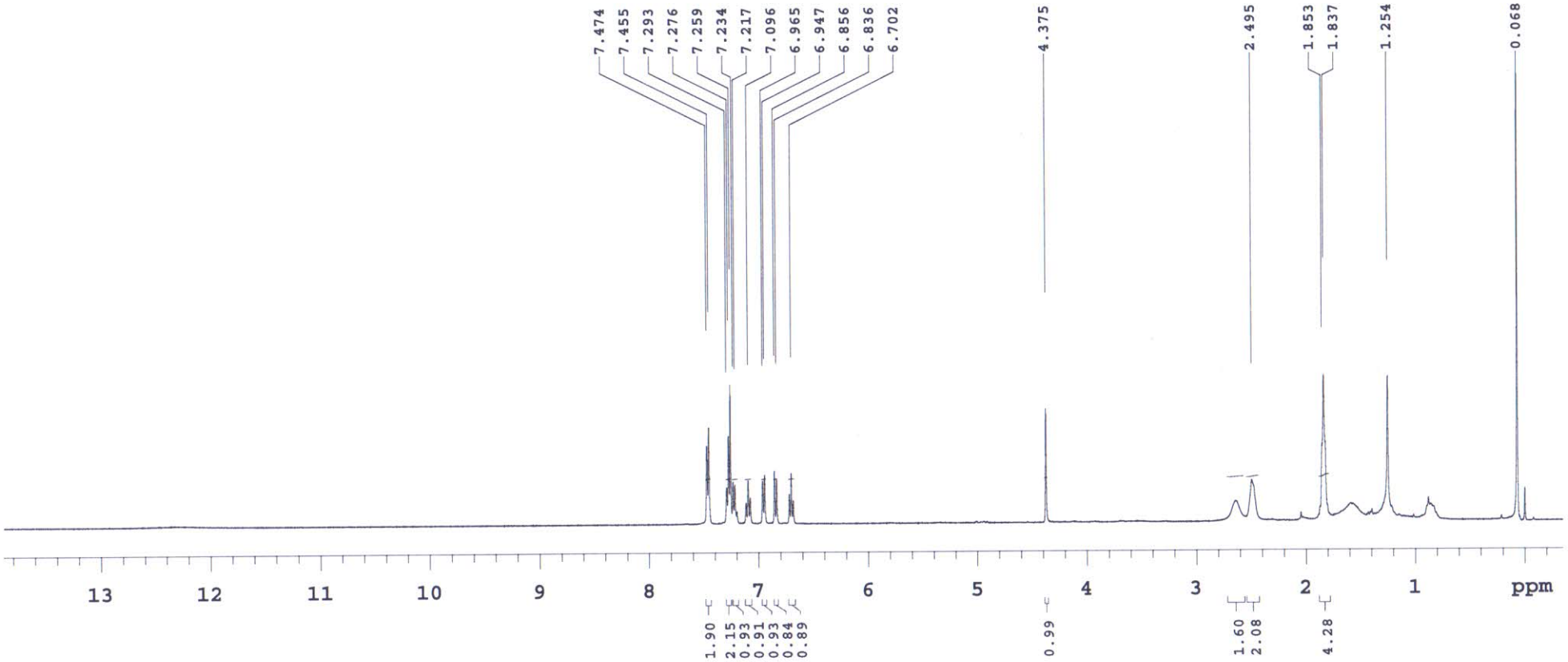


PULSE SEQUENCE Relax. delay 1.000 sec Pulse 45.0 degrees Acq. time 2.561 sec Width 10000.0 Hz 32 repetitions	OBSERVE H1, 399.8509693	DATA PROCESSING FT size 65536 Total time 1 minutes		CKJ-AH-2-14C Solvent: cdcl3 Temp. 25.0 C / 298.1 K Operator: chem File: CKJ-AH-2-14C Mercury-400 "IITG-NMR"

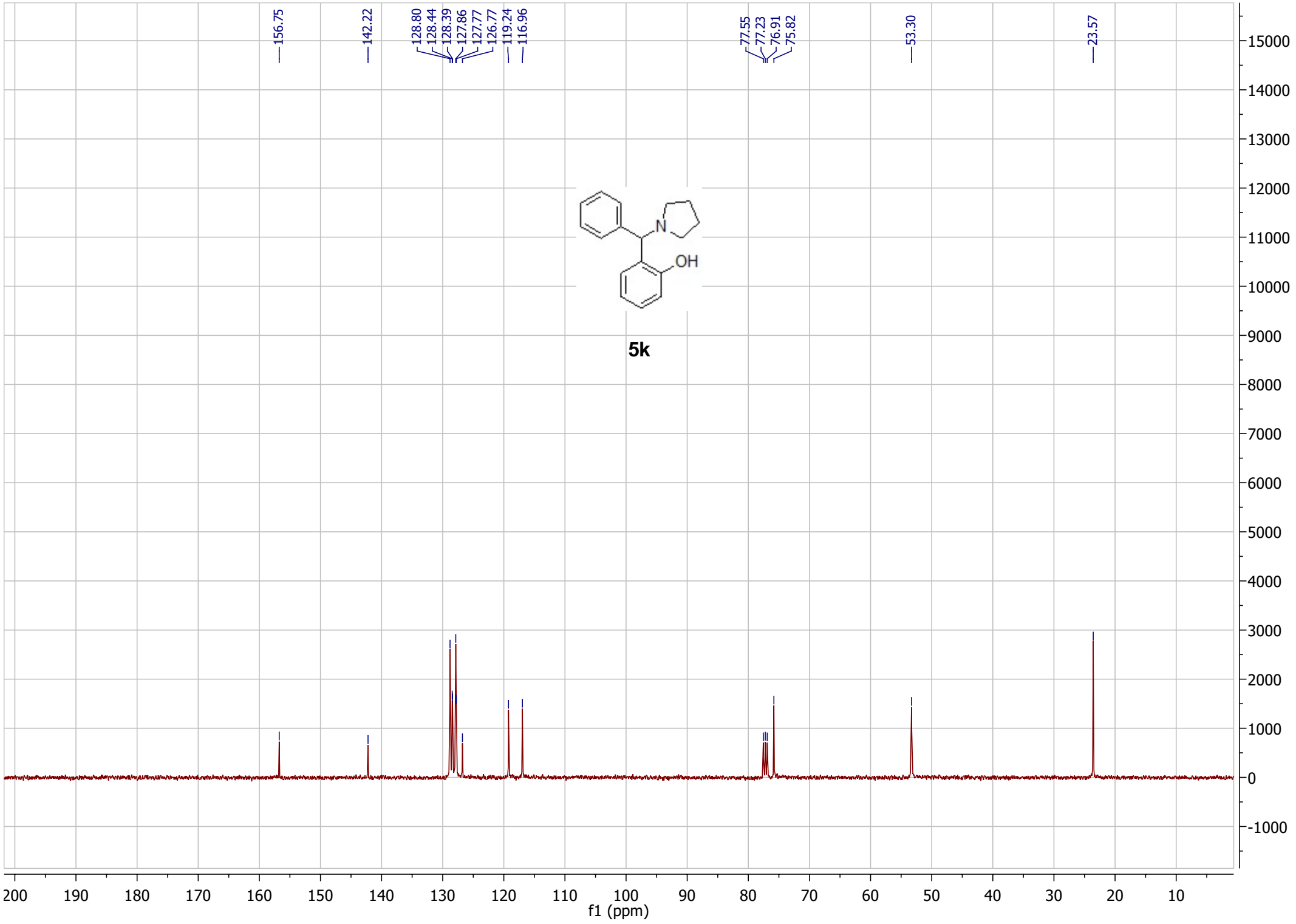

159.25
155.62
133.65
131.99
130.86
129.81
129.51
129.01
126.49
122.48
121.27
120.08
117.05
114.13
77.55
77.23
76.91
70.23
55.33
52.44
23.58
MeO
N
OH
5j
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
-1000
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
f1 (ppm)

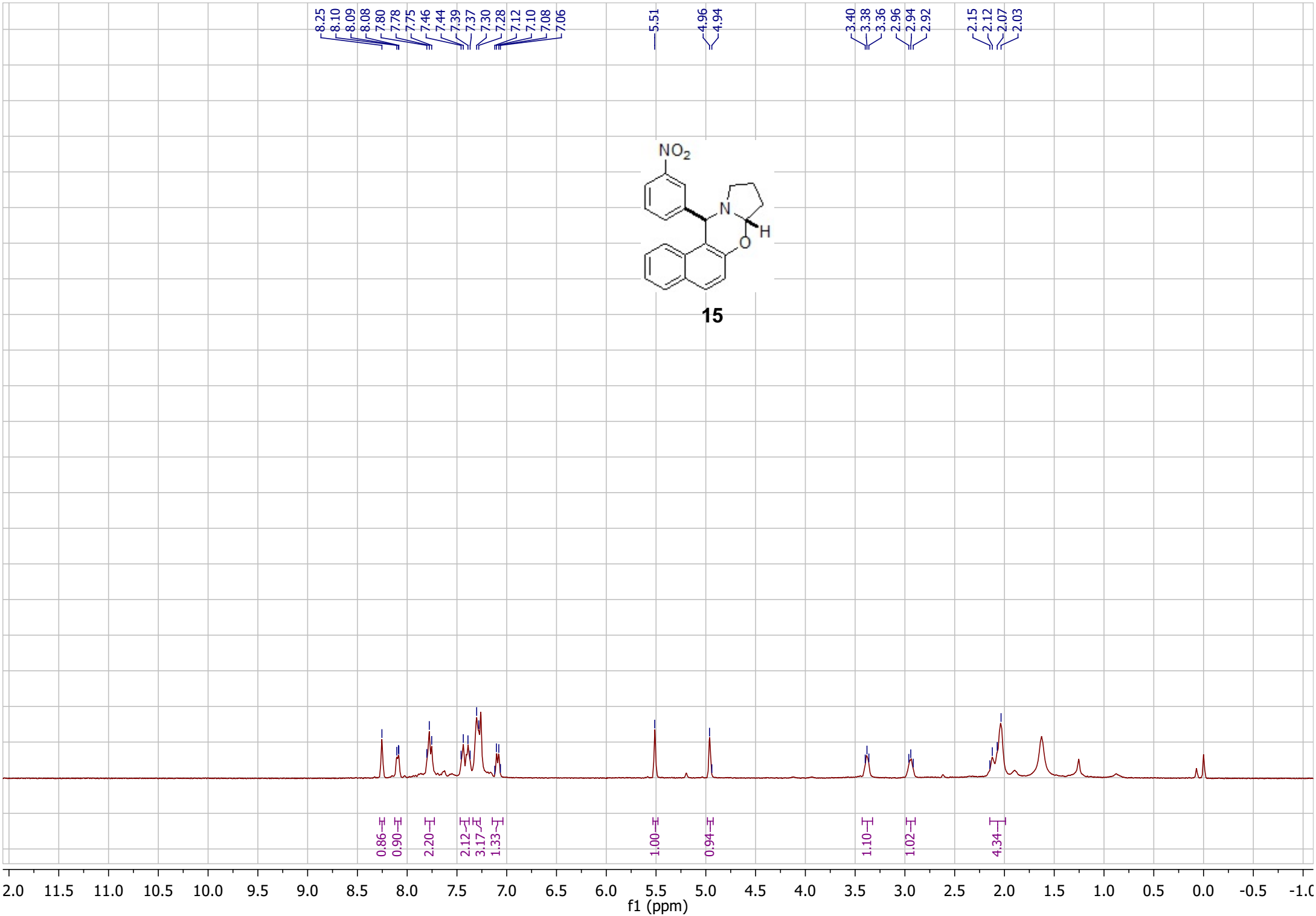


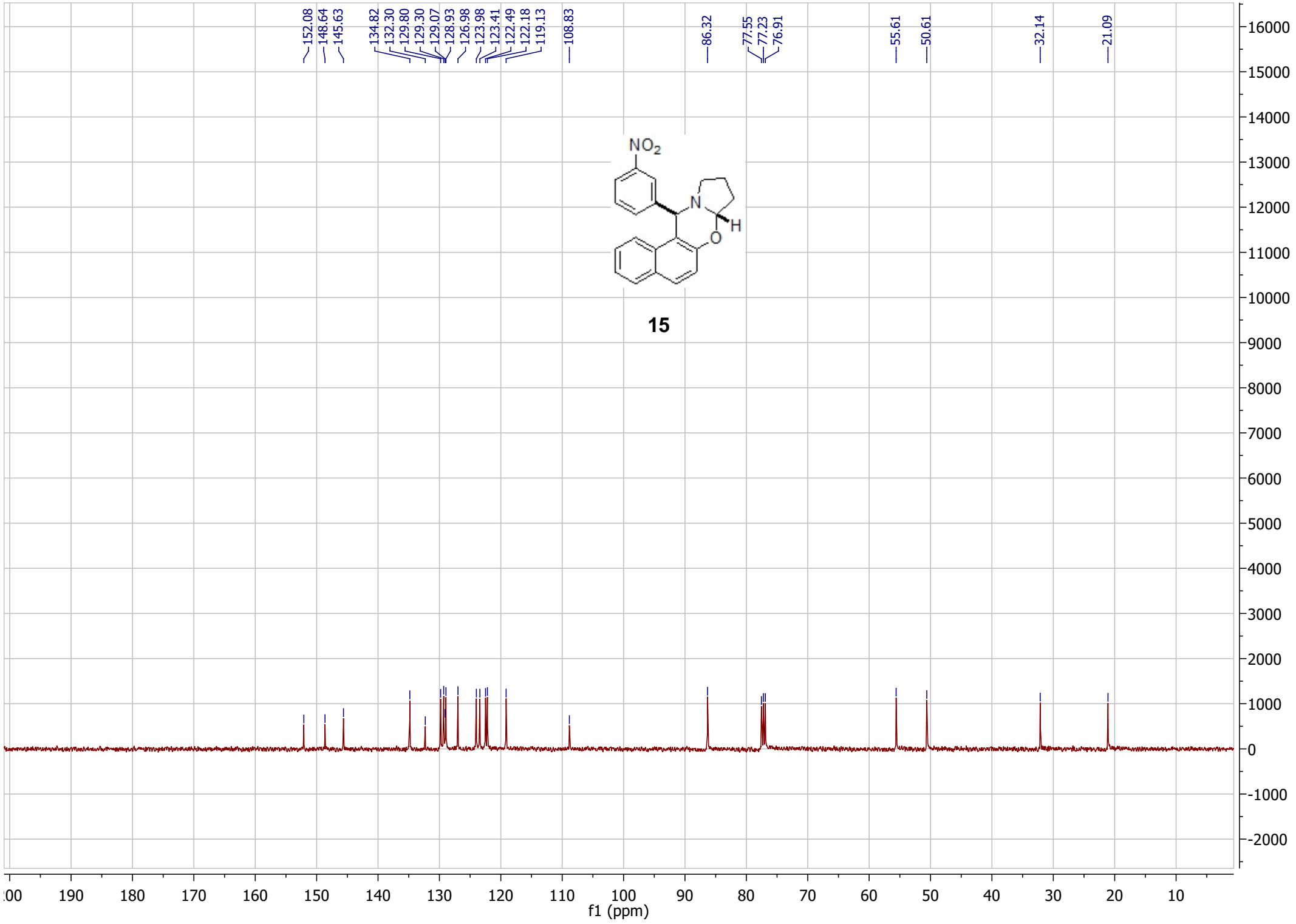
5k



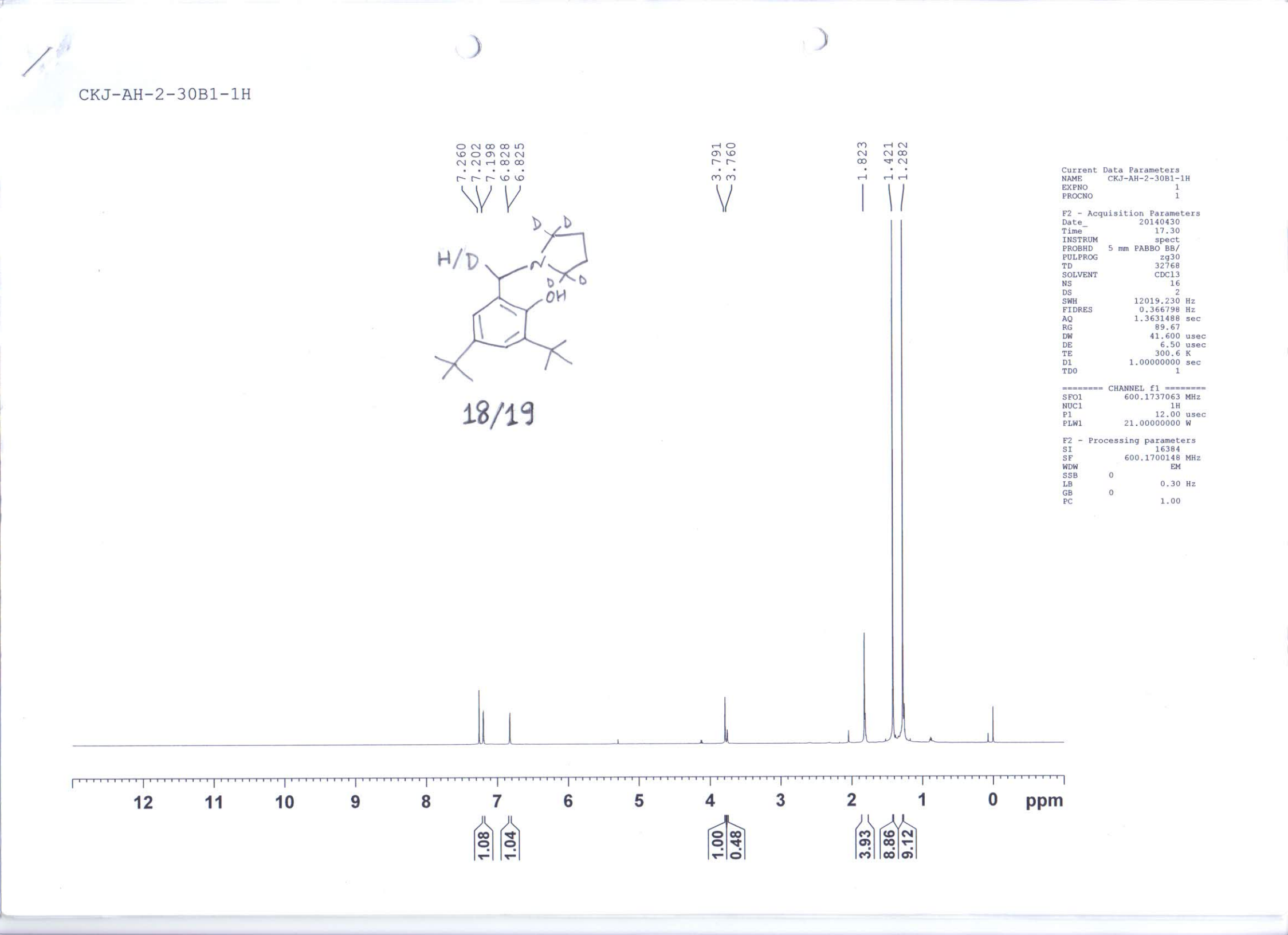
PULSE SEQUENCE Relax. delay 1.000 sec Pulse 45.0 degrees Acq. time 2.561 sec Width 8000.0 Hz 32 repetitions	OBSERVE H1, 399.8509633	DATA PROCESSING FT size 65536 Total time 1 minutes		CKJ-AH-2-20B Solvent: cdcl3 Temp. 25.0 C / 298.1 K Operator: chem File: CKJ-AH-2-20B Mercury-400 "IITG-NMR"


156.75
142.22
128.80
128.44
128.39
127.86
127.77
126.77
119.24
116.96
77.55
77.23
76.91
75.82
53.30
23.57
N
OH
5k
15000
14000
13000
12000
11000
10000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
-1000
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
f1 (ppm)


8.25
8.10
8.09
8.08
7.80
7.78
7.75
7.46
7.44
7.39
7.37
7.30
7.28
7.12
7.10
7.08
7.06
5.51
4.96
4.94
3.40
3.38
3.36
2.96
2.94
2.92
2.15
2.12
2.07
2.03
NO2
N
O
H
15
0.86
0.90
2.20
2.12
3.17
1.33
1.00
0.94
1.10
1.02
4.34
f1 (ppm)

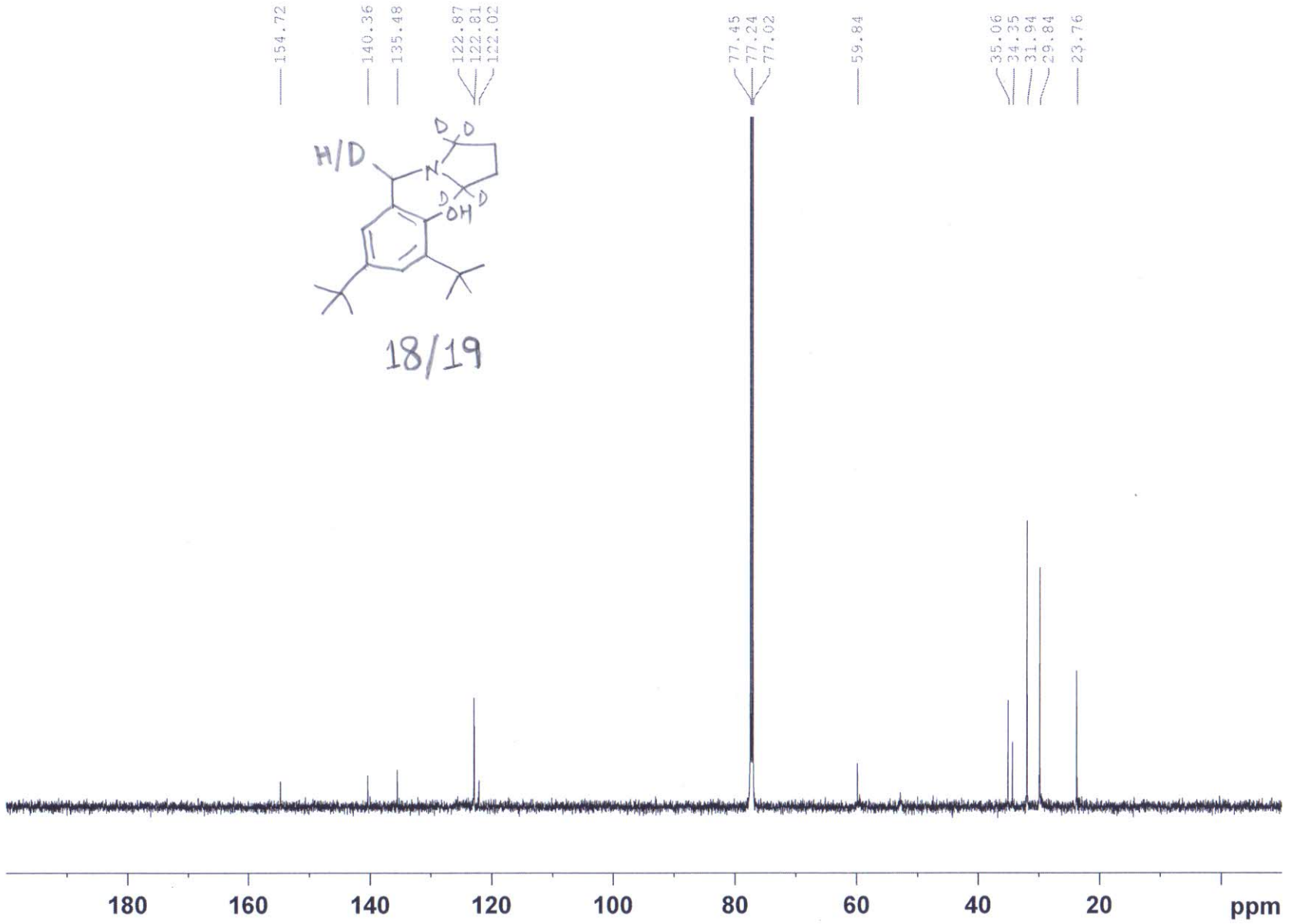

152.08
148.64
145.63
134.82
132.30
129.80
129.30
129.07
128.93
126.98
123.98
123.41
122.49
122.18
119.13
108.83
86.32
77.55
77.23
76.91
55.61
50.61
32.14
21.09
NO2
N
H
O
15
f1 (ppm)

CKJ-AH-2-30B1-1H




```
Current Data Parameters
NAME      CKJ-AH-2-30B1-1H
EXPNO                    1
PROCNO                   1

F2 - Acquisition Parameters
Date_             20140430
Time                 17.30
INSTRUM              spect
PROBHD    5 mm PABBO BB/
PULPROG              zg30
TD                  32768
SOLVENT             CDCl3
NS                     16
DS                      2
SWH              12019.230 Hz
FIDRES           0.366798 Hz
AQ               1.3631488 sec
RG                   89.67
DW                 41.600 usec
DE                  6.50 usec
TE                 300.6 K
D1             1.00000000 sec
TD0                     1

======== CHANNEL f1 ========
SFO1          600.1737063 MHz
NUC1                   1H
P1                  12.00 usec
PLW1          21.00000000 W

F2 - Processing parameters
SI                  16384
SF            600.1700148 MHz
WDW                    EM
SSB       0
LB                  0.30 Hz
GB        0
PC                  1.00
```

CKJ-AH-2-30B1-13C



BRUKER

Current Data Parameters

NAME	CKJ-AH-2-30B1-13C
EXPNO	1
PROCNO	1

F2 - Acquisition Parameters

Date_	20140430
Time	17.40
INSTRUM	spect
PROBHD	5 mm PABBO BB/
PULPROG	zgpg30
TD	32768
SOLVENT	CDCl3
NS	633
DS	2
SWH	36057.691 Hz
FIDRES	1.100393 Hz
AQ	0.4543829 sec
RG	65.24
DW	13.867 usec
DE	6.50 usec
TE	301.4 K
D1	2.00000000 sec
D11	0.03000000 sec
TD0	1

======== CHANNEL f1 ========

SFO1	150.9279571 MHz
NUC1	13C
P1	10.50 usec
PLW1	95.00000000 W

======== CHANNEL f2 ========

SFO2	600.1724007 MHz
NUC2	1H
CPDPRG[2	waltz16
PCPD2	70.00 usec
PLW2	21.00000000 W
PLW12	0.61714000 W
PLW13	0.30239999 W

F2 - Processing parameters

SI	16384
SF	150.9128315 MHz
WDW	EM
SSB	0
LB	1.00 Hz
GB	0
PC	1.40