Supporting Information for

Acceptorless Dehydrogenative Coupling of *o*-Aminobenzamides with the Activation of Methanol as a C1 Source for the Construction of Quinazolinones

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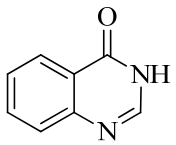
Experimental Details. Proton nuclear magnetic resonance (^{1}H NMR) spectra were recorded at 500 MHz. Chemical shifts are reported in delta (δ) units, parts per million (ppm) downfield from tetramethylsilane or ppm relative to the center of the singlet at 2.5 ppm for DMSO-d_6. Coupling constants *J* values are reported in Hertz (Hz), and the splitting patterns were designated as follows: s, singlet; d, doublet; t, triplet; m, multiplet; b, broad. Carbon-13 nuclear magnetic resonance (^{13}C NMR) spectra were recorded at 125 MHz. Chemical shifts are reported in delta (δ) units, ppm relative to the center of the triplet at 39.5 ppm for DMSO-d_6. ^{13}C NMR spectra were routinely run with broadband decoupling.

[Cp*IrCl$_2$]$_2$ (cat. **1**),[1] [Ir(cod)Cl]$_2$ (cat. **2**),[2] [Cp*Ir(NH$_3$)$_3$][Cl]$_2$ (cat. **3**),[3] [Cp*Ir(2-phenylpyridine-*k*C,N)]Cl (cat. **4**),[4] [Cp*Ir(bpy)Cl)][Cl] (cat. **5**),[5] [Cp*Ir(bpy)(H$_2$O)][OTf]$_2$ (cat. **6**),[6] [Cp*(2-(OH)py)]Cl$_2$ (cat. **7**),[7] [Cp*(6,6'-(OH)$_2$bpy)][OTf]$_2$ (cat. **8**)[8] and [Cp*Ir(2,2′-bpyO)(H$_2$O)] (cat. **9**)[9] were synthesized according to previous reports.

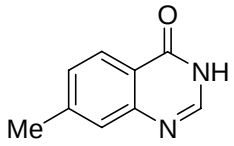
General procedure for acceptorless dehydrogenative coupling of o-aminobenzamides with methanol for the construction of quinazolinones catalyzed by [Cp*Ir(2,2′-bpyO)(H$_2$O)]: To an oven-dried, 5 mL microwave vial containing a stirrer bar were added *o*-aminobenzamides **1** (0.5 mmol), methanol **2** (0.5 mL), cat. **9** (2.7 mg, 0.005 mmol, 1 mol %) and Cs_2CO_3 (49 mg, 0.15 mmol, 0.3 equiv). The vial was placed in a focused, single-mode microwave synthesizer (Discover CEM, USA) at 130 ^{o}C for 2 h (300 W, sealed reaction vessel). Then, the mixture was allowed to cool to ambient temperature, concentrated in *vacuo* and purified by flash column chromatography with hexanes/ethyl acetate to afford the corresponding products. All experiments outlined in the paper are safe under current reaction conditions.

quinazolin-4(*3H*)-one (3a)[10]



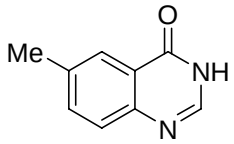
White solid; 88% yield (64 mg); mp 215-217 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 12.26 (br s, 1H, NH), 8.12 (d, 1H, J = 7.9 Hz, ArH), 8.09 (s, 1H, ArH), 7.81 (t, 1H, J = 7.1 Hz, ArH), 7.67 (t, 1H, J = 8.1 Hz, ArH), 7.52 (t, 1H, J = 7.7 Hz, ArH); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d_6) δ 160.7, 148.7, 145.3, 134.2, 127.2, 126.7, 125.8, 122.6.

7-methylquinazolin-4(*3H*)-one (3b)[11]



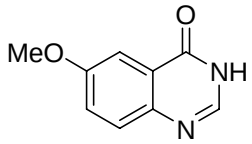
White solid, 87% yield (70 mg); mp 237-238 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 12.13 (br s, 1H, NH), 8.05 (s, 1H, ArH), 8.00 (d, 1H, J = 7.8 Hz, ArH), 7.46 (s, 1H, ArH), 7.33 (d, 1H, J = 7.5 Hz, ArH), 2.45 (s, 3H, CH$_3$); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 160.8, 148.9, 145.7, 144.6, 128.0, 126.7, 125.6, 120.2, 21.3.

6-methylquinazolin-4(*3H*)-one (3c)[12]



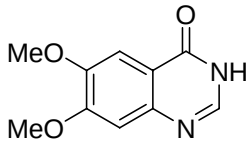
Light yellow solid; 90% yield (72 mg); mp 249-251 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 12.13 (br s, 1H, NH), 8.02 (s, 1H, ArH), 7.91 (s, 1H, ArH), 7.63 (dd, 1H, J = 8.3 Hz and 1.9 Hz, ArH), 7.56 (d, 1H, J = 8.3 Hz, ArH), 2.44 (s, 3H, CH$_3$); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 160.9, 146.8, 144.8, 136.4, 135.6, 127.1, 125.2, 122.4, 20.8.

6-methoxyquinazolin-4(*3H*)-one (3d)[13]



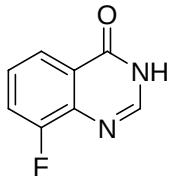
White solid; 91% yield (80 mg); mp 243-244 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 8.01 (s, 1H, ArH), 7.57 (d, 1H, J = 8.9 Hz, ArH), 7.49 (d, 1H, J = 2.3 Hz, ArH), 7.36 (dd, 1H, J = 8.9 Hz and 3.3 Hz, ArH), 3.85 (s, 3H, CH$_3$); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$): δ 162.0, 157.4, 144.9, 143.6, 128.6, 123.5, 123.3, 105.7, 55.5.

6,7-dimethoxyquinazolin-4(*3H*)-one (3e)[14]



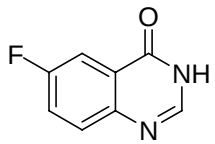
Gray solid, 86% yield (89 mg); mp 296-299 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 12.07 (br s, 1H, NH), 7.98 (s, 1H, ArH), 7.44 (s, 1H, ArH), 7.12 (s, 1H, ArH), 3.90 (s, 3H, OCH$_3$), 3.86 (s, 3H, OCH$_3$); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 160.1, 154.4, 148.5, 144.8, 143.8, 115.6, 108.0, 104.9, 55.9, 55.7.

8-fluoroquinazolin-4(*3H*)-one (3f)[11]



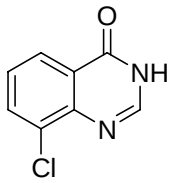
Gray solid, 72% yield (59 mg); mp 270-272 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 12.47 (br s, 1H, NH), 8.14 (s, 1H, ArH), 7.93 (d, 1H, J = 8.0 Hz, ArH), 7.68 (t, 1H, J = 9.3 Hz, ArH), 7.52-7.48 (m, 1H, ArH); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 162.2, 156.4 (d, $J_{C\text{-}F}$ = 251.5 Hz), 148.7, 138.7 (d, $J_{C\text{-}F}$ = 10.4 Hz), 125.8, 124.8, 121.6, 118.7 (d, $J_{C\text{-}F}$ = 18.3 Hz).

6-fluoroquinazolin-4(*3H*)-one (3g)[15]



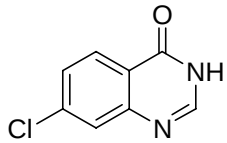
White solid, 77% yield (63 mg); mp 252-255 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 8.09 (s, 1H, ArH), 7.77 (dd, 1H, J = 8.7 Hz and 2.9 Hz, ArH), 7.73 (dd, 1H, J = 8.9 Hz and 5.2 Hz, ArH), 7.67 (td, 1H, J = 8.6 Hz and 3.0 Hz, ArH); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 160.6, 159.9 (d, $J_{C\text{-}F}$ = 243.8 Hz), 145.7, 145.4, 129.9 (d, $J_{C\text{-}F}$ = 7.5 Hz), 123.9 (d, $J_{C\text{-}F}$ = 8.8 Hz), 122.6 (d, $J_{C\text{-}F}$ = 23.8 Hz), 110.4 (d, $J_{C\text{-}F}$ = 22.5 Hz).

8-chloroquinazolin-4(*3H*)-one (3h)[12]



Light yellow solid, 68% yield (61mg); mp 296-298 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 12.39 (br s, 1H, NH), 8.21 (s, 1H, ArH), 8.08 (s, 1H, ArH), 7.97 (s, 1H, ArH), 7.49 (s, 1H, ArH); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 160.3, 146.5, 145.2, 134.4, 130.7, 127.0, 125.0, 124.3.

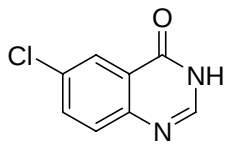
7-chloroquinazolin-4(*3H*)-one (3i)[16]



White solid, 70% yield (63 mg); mp 252-254 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 12.44 (br s,

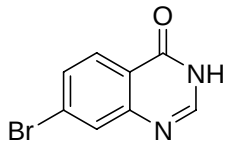
1H, NH), 8.13 (s, 2H, ArH), 7.72 (s, 1H, ArH), 7.55 (s, 1H, ArH); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 160.4, 149.9, 147.2, 138.8, 127.9, 126.9, 126.3, 121.4.

6-chloroquinazolin-4(*3H*)-one (3j)[15]



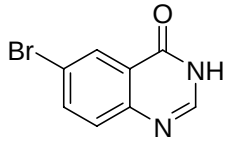
White solid, 77% yield (70 mg); mp 263-265 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 8.13 (s, 1H, ArH), 8.05 (d, 1H, J= 2.4 Hz, ArH), 7.83 (dd, 1H, J = 8.7 Hz and 2.4 Hz, ArH), 7.69 (d, 1H, J = 8.7 Hz, ArH); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 160.0, 147.5, 146.2, 134.3, 130.9, 129.4, 124.8, 123.9.

7-bromoquinazolin-4(*3H*)-one (3k)[17]



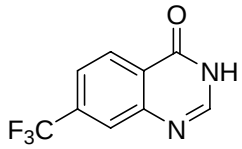
Gray solid; 80% yield (90 mg); mp 227-228 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 8.16 (d, 1H, J= 1.6 Hz, ArH), 8.14 (s, 1H, ArH), 7.87 (dd, 1H, J = 8.6 Hz and 1.6 Hz, ArH), 7.56 (d, 1H, J = 8.6 Hz, ArH); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 161.7, 148.6, 148.2, 136.2, 129.2, 127.9, 124.3, 118.1.

6-bromoquinazolin-4(*3H*)-one (3l)[18]



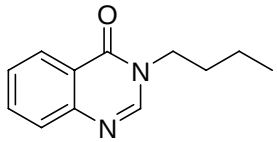
White solid; 82% yield (92 mg); mp 272-275 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 12.39 (br s, 1H, NH), 8.19 (s, 1H, ArH), 8.14 (s, 1H, ArH), 7.95 (d, 1H, J = 7.4 Hz, ArH), 7.62 (d, 1H, J = 8.6 Hz, ArH); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 159.7, 147.7, 146.1, 137.0, 129.6, 127.9, 124.2, 119.1.

7-(trifluoromethyl)quinazolin-4(*3H*)-one (3m)[11]



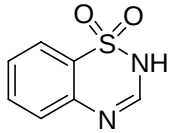
White solid, 78% yield (84 mg); mp 226-228 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 12.55 (br s, 1H, NH), 8.31 (d, 1H, J = 8.5 Hz, ArH), 8.23 (s, 1H, ArH), 7.98 (s, 1H, ArH), 7.82 (d, 1H, J = 7.7 Hz, ArH); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 160.1, 148.8, 147.2, 133.9 (q, $J_{C\text{-}F}$ = 32.2 Hz), 127.6, 125.5, 124.3, 123.5 (q, $J_{C\text{-}F}$ = 269.7 Hz), 122.4.

3-butylquinazolin-4(*3H*)-one (3n)[19]



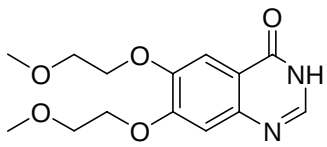
White solid; 66% yield (67 mg); mp 70-72 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 8.32 (d, 1H, J = 7.5 Hz, ArH), 8.03 (s, 1H, ArH), 7.76 (t, 1H, J = 7.3 Hz, ArH), 7.71 (d, 1H, J = 8.0 Hz, ArH), 7.51 (t, 1H, J = 7.3 Hz, ArH), 4.01 (t, 2H, J = 7.3 Hz, CH_2), 1.79 (quint, 2H, J = 7.5 Hz, CH_2), 1.42 (sext, 2H, J = 7.4 Hz, CH_2), 0.98 (t, 3H, J = 7.3 Hz, CH_3); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 161.1, 148.1, 146.6, 134.1, 127.4, 127.2, 126.7, 122.2, 46.8, 31.4, 19.9, 13.6.

***2H*-benzo[e][1,2,4]thiadiazine 1,1-dioxide (3p)[20]**



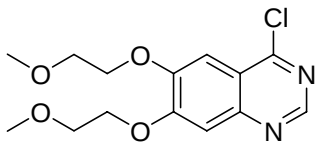
White solid; 76% yield (69 mg); mp 225-226 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 12.15 (br s, 1H, NH), 7.97 (s, 1H, ArH), 7.81 (d, 1H, J = 7.9 Hz, ArH), 7.67 (t, 1H, J = 7.7 Hz, ArH), 7.46 (t, 1H, J = 7.6 Hz, ArH), 7.32 (d, 1H, J = 8.3 Hz, ArH); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 147.7, 134.8, 133.0, 126.6, 123.6, 122.5, 117.5.

Procedure for the synthesis of 6,7-bis(2-methoxyethoxy)quinazolin-4(3H)-one (5).[21] To an oven-dried, 5 mL microwave vial containing a stirrer bar were added **4** (142 mg, 0.5 mmol), methanol **2** (0.5 mL), cat. **9** (2.7 mg, 0.005 mmol, 1 mol %) and Cs_2CO_3 (49 mg, 0.15 mmol, 0.3 equiv). The vial was placed in a focused, single-mode microwave synthesizer (Discover CEM, USA) at 130 ^{o}C for 2 h (300 W, sealed reaction vessel). Then, the mixture was allowed to cool to ambient temperature, concentrated in *vacuo* and purified by flash column chromatography with hexanes/ethyl acetate to afford the corresponding product **5**.



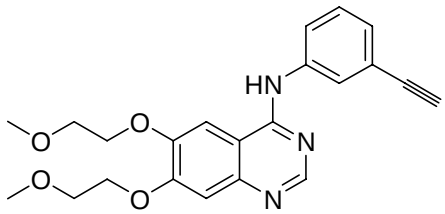
Light yellow solid; 85% yield (125 mg); mp 180-182 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 12.05 (br s, 1H, NH), 7.97 (s, 1H, ArH), 7.46 (s, 1H, ArH), 7.15 (s, 1H, ArH), 4.25 (t, 2H, J = 4.3 Hz, CH_2), 4.19 (t, 2H, J = 4.4 Hz, CH_2), 3.71 (m, 4H, 2xCH_2), 3.33 (s, 6H, 2xCH_3); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 160.1, 153.8, 147.8, 144.8, 143.9, 115.7, 109.0, 106.5, 70.2, 70.1, 68.1, 68.1, 58.3.

Procedure for the synthesis of 4-chloro-6,7-bis(2-methoxyethoxy)quinazoline (6).[21] 5 (328 mg, 1.1 mmol) was slowly added to a solution of phosphoryl chloride (1 mL) and *N*,*N*-diethylaniline (164 mg, 1.1 mmol). This mixture was heated at 90 ^{o}C for 2 h and was then cooled to room temperature. Most of the excess of phosphoryl chloride was removed under reduced pressure and the dark oil was purified by flash column chromatography with hexanes/ethyl acetate to afford the corresponding product.



White Solid, 83% yield (286 mg), mp 106-107 ^{o}C: ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 8.86 (s, 1H, ArH), 7.44 (s, 1H, ArH), 7.33 (s, 1H, ArH), 4.35-4.32 (m, 4H, 2xCH_2), 3.90-3.88 (m, 4H, 2xCH_2), 3.51 (s, 3H, CH_3), 3.49 (s, 3H, CH_3); ^{13}C {^{1}H} NMR (125 MHz, DMSO-d$_6$) δ 158.9, 156.2, 152.4, 150.8, 148.9, 119.4, 107.6, 104.0, 70.4, 70.2, 68.8, 68.7, 59.3 (2xOCH_3).

Procedure for the synthesis of *N*-(3-ethynylphenyl)-6,7-Bis-(2-methoxyethoxy)-quinazolin-4-amine (7, Erlotinib).[21] A solution of **6** (286 mg, 0.91 mmol) in *i*-PrOH (5 mL) was added drop wise to a solution of pyridine (79 mg, 1.0 mmol) and 3-ethynylphenylamine (117 mg, 1.0 mmol) in *i*-PrOH (5 mL). This mixture was heated at reflux for 4 h under nitrogen and was then cooled to room temperature. The precipitate was filtered and crystallized from methanol to afford a **7**·HCl product. **7**·HCl product was dissolved in water, basified using conc. aq. ammonia, and extracted with ethyl acetate and concentrated in *vacuo* to give free **7**.

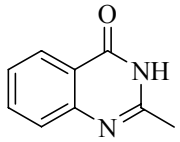


Yellowish Solid, 90% yield (323 mg); mp 156-157 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ 8.61 (s, 1H, ArH), 7.99 (s, 1H, ArH), 7.85 (s, 1H, ArH), 7.74 (dd, 1H, J = 8.1 Hz and 1.0 Hz, ArH), 7.30 (t,

1H, J = 7.9 Hz, ArH), 7.23 (d, 1H, J = 7.7 Hz, ArH), 4.18-4.14 (m, 4H, 2xCH_2), 3.77-3.73 (m, 4H, 2xCH_2), 3.40 (s, 3H, CH_3), 3.39 (s, 3H, CH_3), 3.08 (s, 1H, CH); ^{13}C {^1H} NMR (125 MHz, DMSO-d_6) δ 156.4, 154.2, 153.5, 148.5, 147.3, 138.9, 128.9, 127.6, 125.2, 122.6, 122.4, 109.2, 108.4, 102.5, 83.4, 77.4, 70.8, 70.3, 68.8, 68.1, 59.1, 59.1.

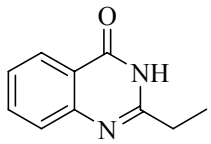
Procedure for acceptorless dehydrogenative coupling of *o*-aminobenzamides with other alcohols catalyzed by metal-organic bifunctional iridium complex: To an oven-dried, 5 mL microwave vial containing a stirrer bar were added *o*-aminobenzamide **1a** (68 mg, 0.5 mmol), alcohols **8-10** (0.5 mL), cat. **9** (2.7 mg, 0.005 mmol, 1 mol %) and Cs_2CO_3 (49 mg, 0.15 mmol, 0.3 equiv). The vial was placed in a focused, single-mode microwave synthesizer (Discover CEM, USA) at 130 ^oC for 2 h (300 W, sealed reaction vessel). Then, the mixture was allowed to cool to ambient temperature, concentrated in *vacuo* and purified by flash column chromatography with hexanes/ethyl acetate to afford the corresponding products.

2-methylquinazolin-4(3H)-one (11)[22]



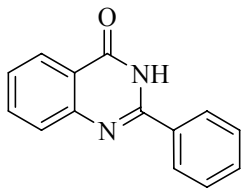
White solid; 90% yield (72 mg); mp 242-243 ^oC; ^1H NMR (500 MHz, $CDCl_3$) δ 11.85 (br s, 1H, NH), 8.28 (d, 1H, J = 7.5 Hz, ArH), 7.78 (t, 1H, J = 7.1 Hz, ArH), 7.68 (d, 1H, J = 8.0 Hz, ArH), 7.48 (t,1H, J = 7.5 Hz, ArH), 2.60 (s, 3H, CH_3); ^{13}C {^1H} NMR (125 MHz, $CDCl_3$) δ 164.5, 153.4, 149.5, 134.8, 127.0, 126.3, 126.2, 120.2, 22.0.

2-ethylquinazolin-4(*3H*)-one (12)[23]



White solid; 86% yield (75 mg); mp 232-233 ^oC; ^1H NMR (500 MHz, $CDCl_3$) δ 11.67 (br s, 1H, NH), 8.29 (d, 1H, J = 7.1 Hz, ArH), 7.77 (t, 1H, J = 8.3 Hz, ArH), 7.72 (d, 1H, J = 8.0 Hz, ArH), 7.47(t, 1H, J = 7.8 Hz, ArH), 2.85 (q, 2H, J = 7.6 Hz, CH_2) , 1.46 (t, 3H, J = 7.6 Hz, CH_3); ^{13}C {^1H} NMR (125 MHz, $CDCl_3$) δ 164.4, 157.8, 149.4, 134.7, 127.2, 126.3, 126.2, 120.4, 29.0, 11.5.

2-phenyl-4(*3H*)-quinazolinone (13)[24]



White solid; 91% yield (111 mg); mp 220-221 ^{o}C; ^{1}H NMR (500 MHz, $CDCl_3$) δ 10.27 (br s, 1H, NH), 8.33 (d, 1H, J = 8.1 Hz, ArH), 8.14-8.12 (m, 2H, ArH), 7.85-7.79 (m, 2H, ArH), 7.59(d,3H, J = 7.0 Hz, ArH), 7.52 (t, 1H, J = 7.9 Hz, ArH); ^{13}C {^{1}H} NMR (125 MHz, $CDCl_3$) δ 164.0, 151.8, 149.5, 134.8, 132.8, 131.6, 129.0, 128.0, 127.5, 126.7, 126.3, 120.8.

Procedure for the hydrogen evolution experiment.[25] **1a** (68 mg, 0.5 mmol), **2** (0.5 mL), cat. **9** (2.7 mg, 1 mol %) and Cs_2CO_3 (49 mg, 0.15 mmol, 0.3 equiv) were added into a 5 mL thick walled glass vessel fitted with a side arm and a rubber septum through a valve. The vessel was previously degassed three times and placed under a N_2 atmosphere. The vessel was connected to the gas collection apparatus (standard water displacement apparatus, using a graduated cylinder to determine volume) through a valve and the entire system was flushed with N_2 for 5 min and allowed to equilibrate for 5 min. The valve was closed and the reaction mixture was stirred vigorously at 150 ^{o}C for 12 h under sealed conditions. When reaction mixture was cooled to room temperature and the valve was turned on and the gas was collected by water displacement until gas evolution ceased.

The GC analysis was performed on a gas chromatograph with TCD detector. Injector temprature = 150 ^{o}C, column temprature = 80 ^{o}C, detector temperature (TCD) = 80 ^{o}C, carrier gas = N_2, column flow = 20 mL/min, t = 0.525 min.

The volume of 1 mol of H_2 at 283.15 K, 101810 Pa was calculated according to the van der Waals equation as shown below

$$\left(p+\frac{n^2a}{V^2}\right)(V-nb) = nRT$$

where R = 8.3145 m^3 Pa·mol^{-1}·K^{-1}; T = 283.15 K; p = 102600 Pa; a = 0.02476 m^6·Pa·mol^{-2}; b = 0.02661 × 10^{-3} m^3·mol^{-1}; thus, V (H_2, 283.15 K, 102600 Pa) = 23 L·mol^{-1}.

The collected volume of gas in this experiment above was 14 mL, which corresponds to 0.61 mmol of H_2.

Procedure for acceptorless dehydrogenative coupling of o-aminobenzamides with paraformaldehyde catalyzed by [Cp*Ir(2,2′-bpyO)(H_2O)]: To an oven-dried, 5 mL microwave vial containing a stirrer bar were added *o*-aminobenzamide **1a** (0.5 mmol, 68 mg), paraformaldehyde (45 mg), toluene (1 mL), cat. **9** (2.7 mg, 0.005 mmol, 1 mol %) and Cs_2CO_3 (49 mg, 0.15 mmol, 0.3 equiv). The vial was placed in a focused, single-mode microwave synthesizer (Discover CEM, USA) at 130 ^oC for 2 h (300 W, sealed reaction vessel). Then, the mixture was allowed to cool to ambient temperature, concentrated in *vacuo* and purified by flash column chromatography with hexanes/ethyl acetate to afford the corresponding product **3a** (60 mg, 82% yield).

References

(1) Ball, R. G.; Graham, W. A. G.; Heinekey, D. M.; Hoyano, J. K.; McMaster, A. D.; Mattson, B. M.; Michel, S. T. *Inorg. Chem.* **1990**, *29*, 2023-2025.

(2) Choudhury, J.; Podder, S.; Roy, S. J. *Am. Chem. Soc.* **2005**, *127*, 6162-6163.

(3) Kawahara, R.; Fujita, K.; Yamaguchia, R. *Adv. Synth. Catal.* **2011**, *353*, 1161-1168.

(4) Scheeren, C.; Maasarani, F.; Hijazi, A.; Djukic, J. P.; Pfeffer, M.; Zaric, S. D.; Goff, X. F. L.; Ricard, L. *Organometallics* **2007**, *26*, 3336-3345.

(5) Ogo, S.; Makihara, N.; Kaneko, Y.; Watanabe, Y. *Organometallics* **2001**, *20*, 4903-4910.

(6) Ziessel, R. *J. Chem. Soc., Chem. Commun.* **1988**, 16-17.

(7) Fujita, K.; Tanino, N.; Yamaguchi, R. *Org. Lett.* **2007**, *9*, 109-111.

(8) Kawahara, R.; Fujita, K.; Yamaguchi, R. *J. Am. Chem. Soc.* **2012**, *134*, 3643-3646.

(9) Kawahara, R.; Fujita, K.; Yamaguchi, R. *Angew. Chem. Int. Ed.* **2012**, *51*, 12790-12794.

(10) Yang, X.; Cheng, G.; Shen, J.; Kuai, C.; Cui, X. *Org. Chem. Front.* **2015**, *2*, 366-368.

(11) Kulkarni, S. S.; Singh, S.; Shah, J. R.; Low, W.; Talele, T. T. *Eur. J. Med. Chem.* **2012**, *50*, 264-273.

(12) Nara, H.; Sato, K.; Naito, T.; Mototani, H.; Oki, H.; Yamamoto, Y.; Kuno, H.; Santou, T.; Kanzaki, N.; Terauchi, J.; Uchikawa, O.; Kori, M. *Bioorg. Med. Chem.* **2014**, *22*, 5487-5505.

(13) LeMahieu, R. A.; Carson, M.; Nason, W. C.; Parrish, D. R.; Welton, A. F.; Baruth, H. W.; Yaremko, B. *J. Med. Chem.* **1983**, *26*, 420-425.

(14) VanBrocklin, H. F.; Lim, J. K.; Coffing, S. L.; Hom, D. L.; Negash, K.; Ono, M. Y.; Gilmore, J. L.; Bryant, I.; Riese, D. J. *J. Med. Chem.* **2005**, *48*, 7445-7456.

(15) Pedgaonkar, G. S.; Sridevi, J. P.; Jeankumar, V. U.; Saxena, S.; Devi, P. B.; Renuka, J.; Yogeeswari, P.; Sriram, D. *Eur. J. Med. Chem.* **2014**, *86*, 613-627.

(16) Luo, H.; Yang, S.; Cai, Y.; Peng, Z.; Liu, T. *Eur. J. Med. Chem.* **2014**, *84*, 746-752.

(17) Hedou, D.; Guillon, R.; Lecointe, C.; Logec, C.; Chosson, E.; Besson, T. *Tetrahedron* **2013**, *69*, 3182-3191.

(18) Gao, L.; Kovackova, S.; Sala, M.; Ramadori, A. T.; Jonghe, S. D.; Herdewijn, P. *J. Med. Chem.* **2014**, *57*, 7624-7643.

(19) Bao, Y.; Yan, Y.; Xu, K.; Su, J.; Zha, Z.; Wang, Z. *J. Org. Chem.* **2015**, *80*, 4736-4742.

(20) Chen, X.; Zhu, C.; Guo, F.; Qiu, X.; Yang, Y.; Zhang, S.; He, M.; Parveen, S.; Jing, C.; Li, Y.; Ma, B. *J. Med. Chem.* **2010**, *53*, 8330-8344.

(21) Knesl, P.; Röseling, D.; Jordis, U. *Molecules* **2006**, *11*, 286-297.

(22) Ding, P.; Gao, M.; Mao, B.; Cao, S.; Liu, C.; Yang, C.; Li, Z.; Liao, J.; Zhao, H.; Li, Z.; Li, J.; Wang, H.; Xu, X. *Eur. J. Med. Chem.* **2016**, *108*, 364-373.

(23) Xu, W.; Fu, H. *J. Org. Chem.* **2011**, *76*, 3846-3852.

(24) Zhao, W.; Liu, P.; Li, F. *ChemCatChem* **2016**, *8*, 1523-1530.

(25) Wu, J.; Talwar, D.; Johnston, S.; Yan, M.; Xiao, J. *Angew. Chem. Int. Ed.* **2013**, *52*, 6983-6987.

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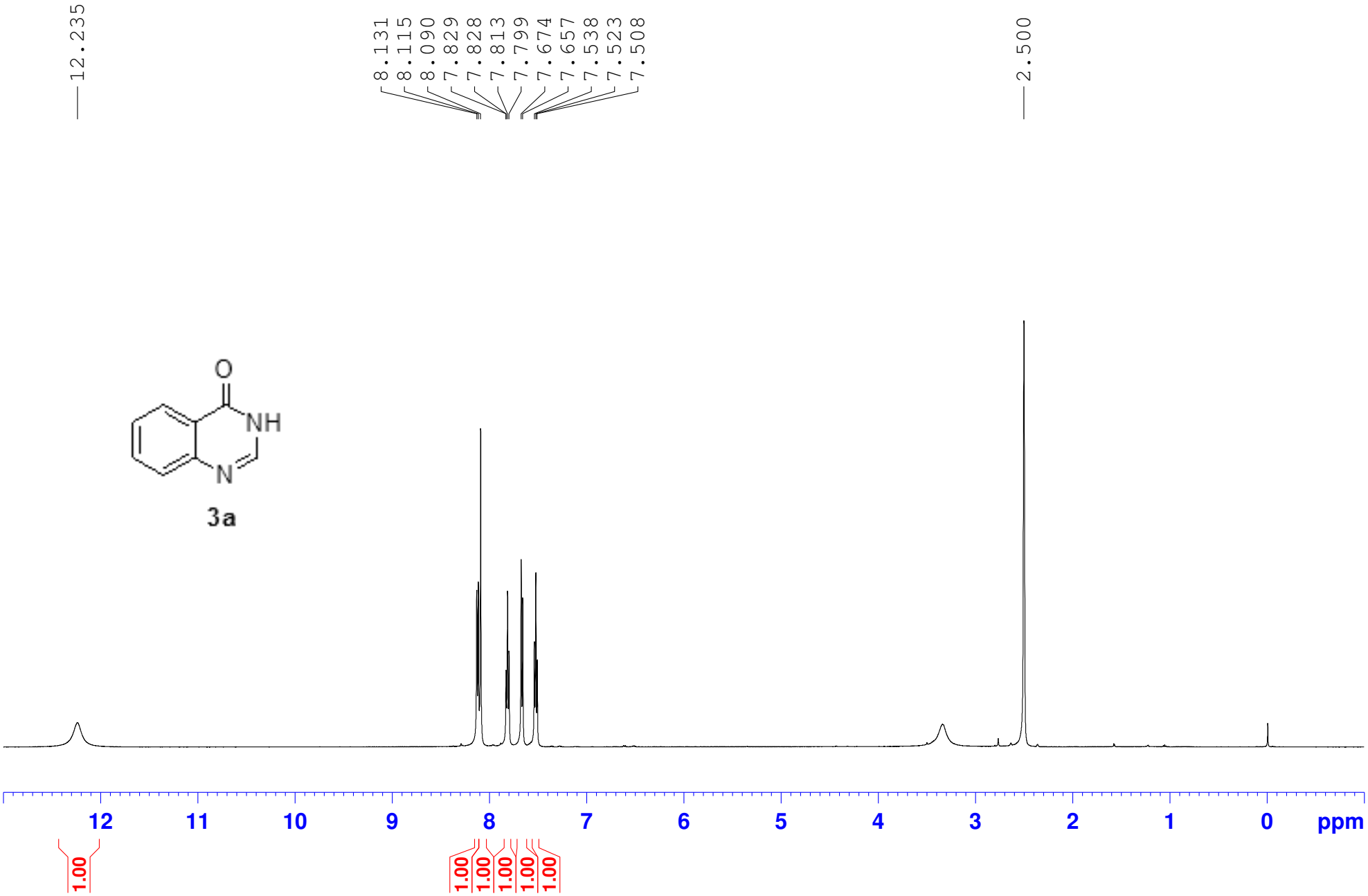

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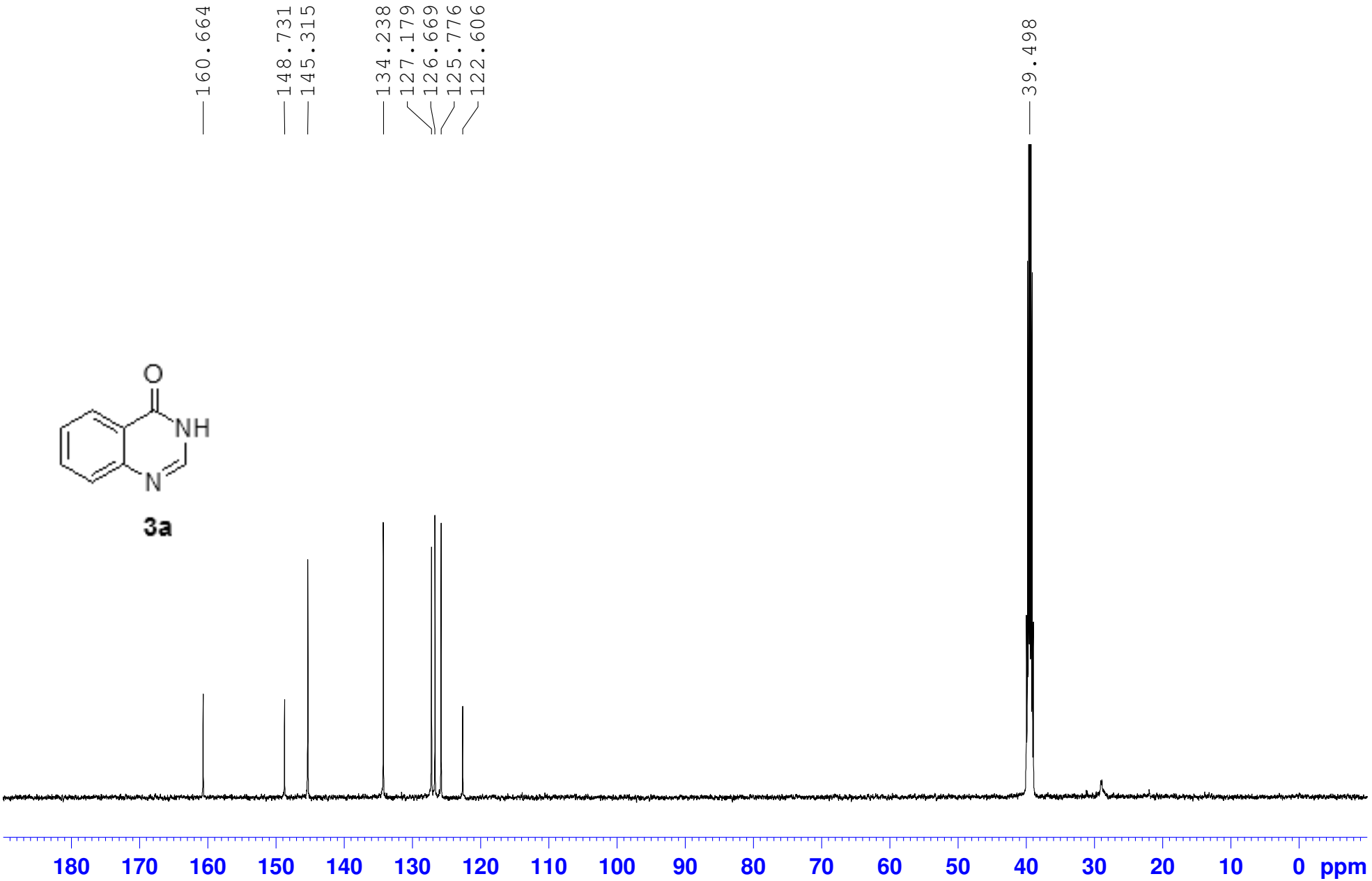

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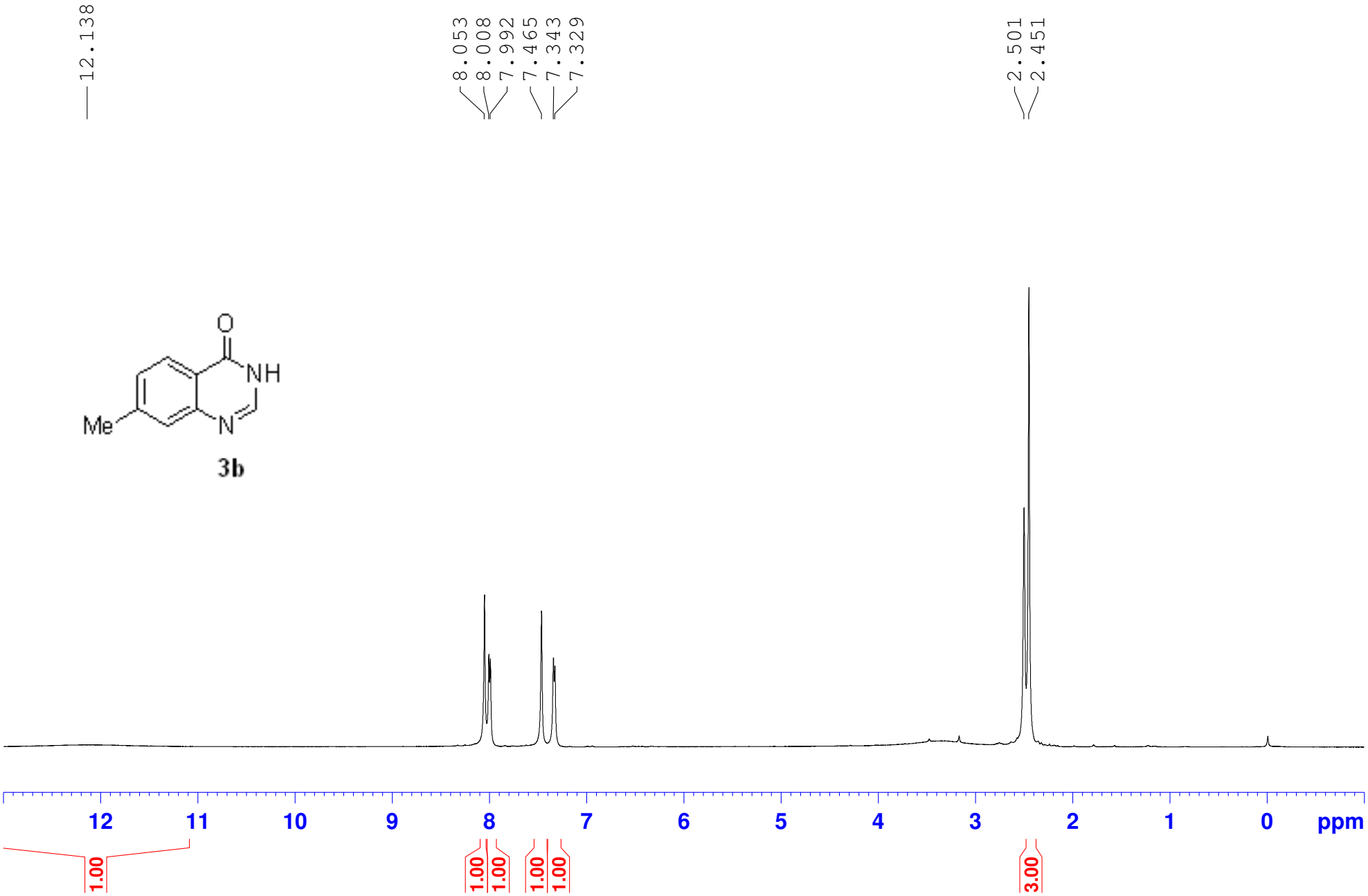

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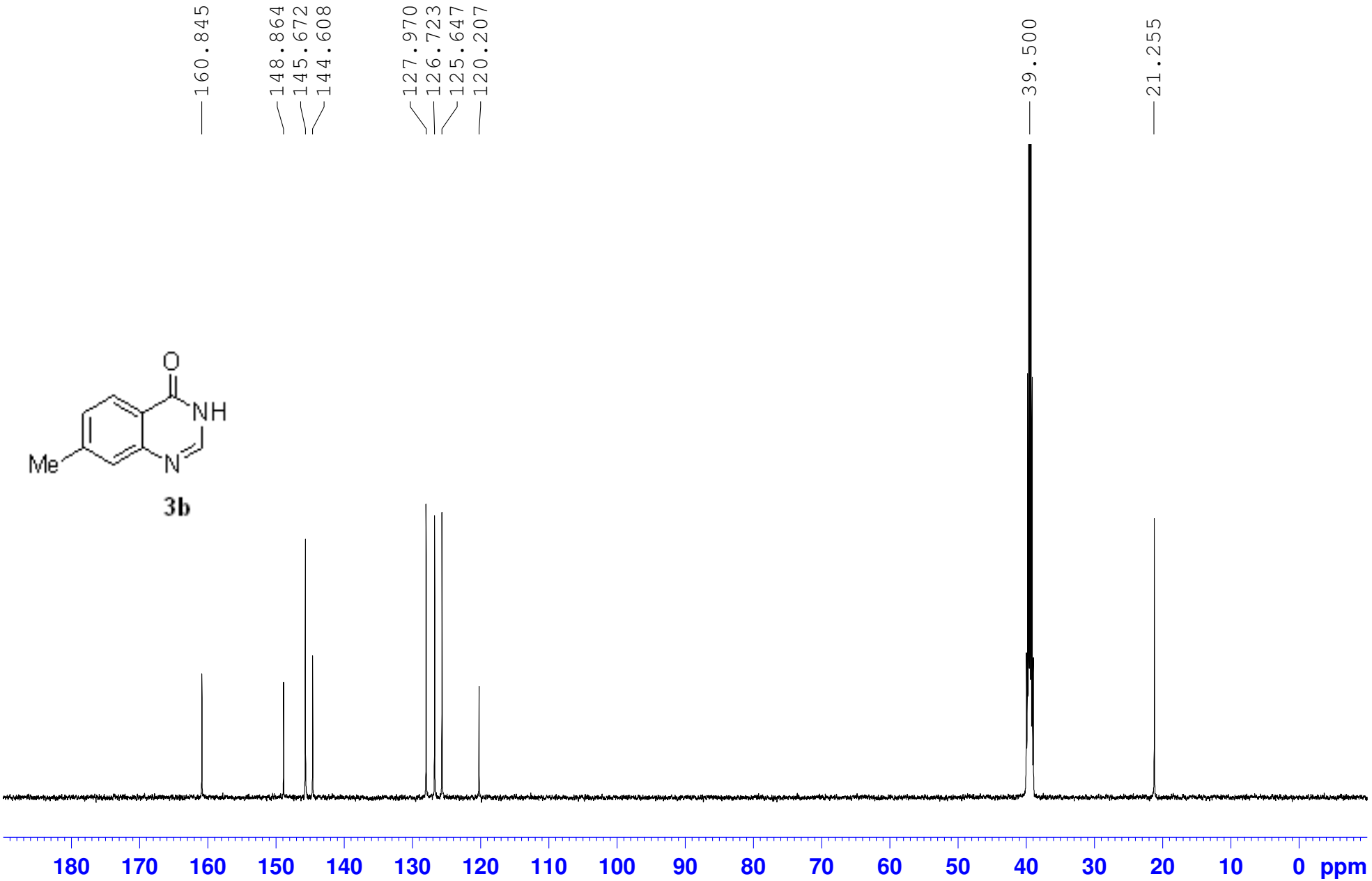

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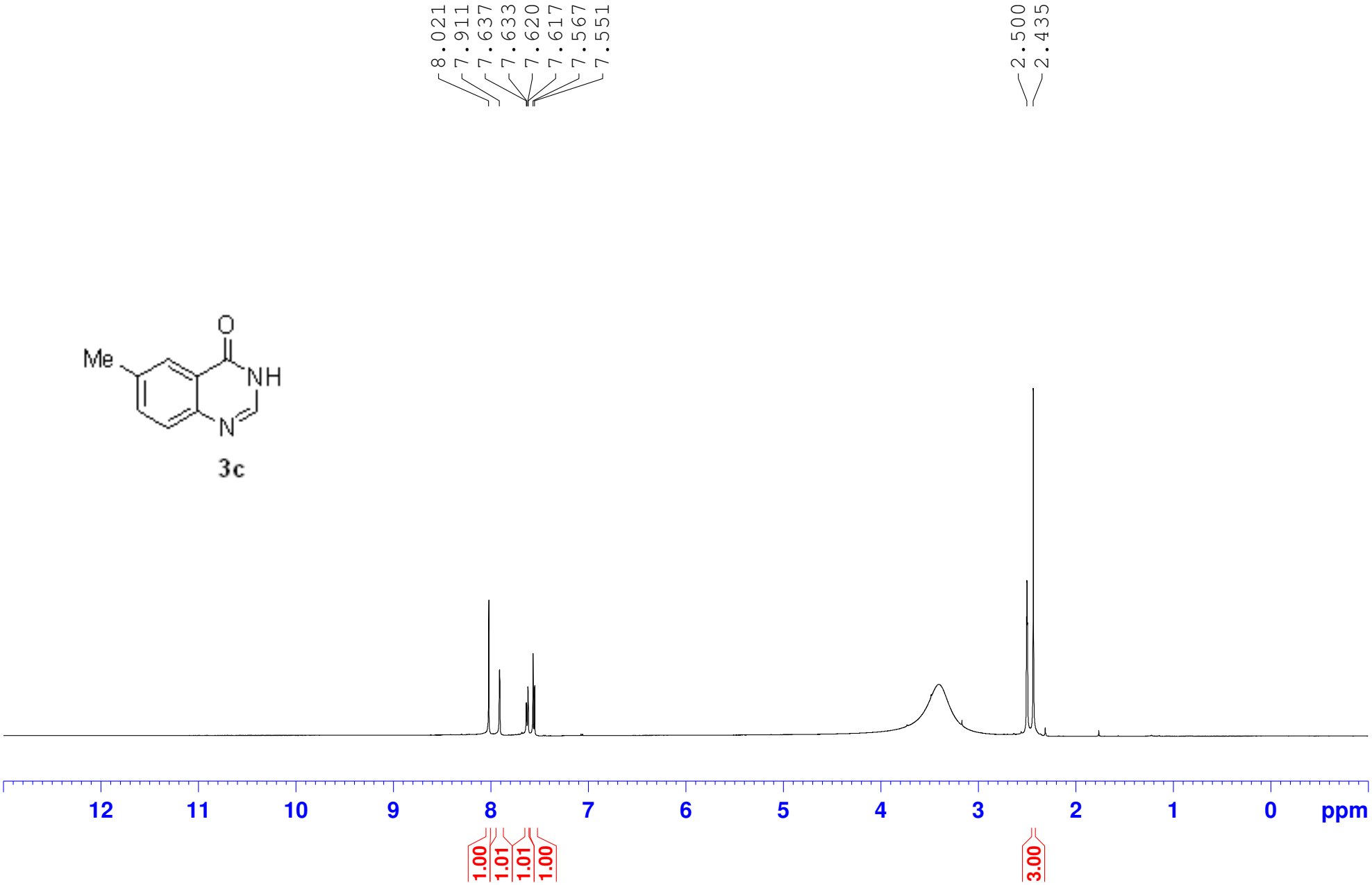

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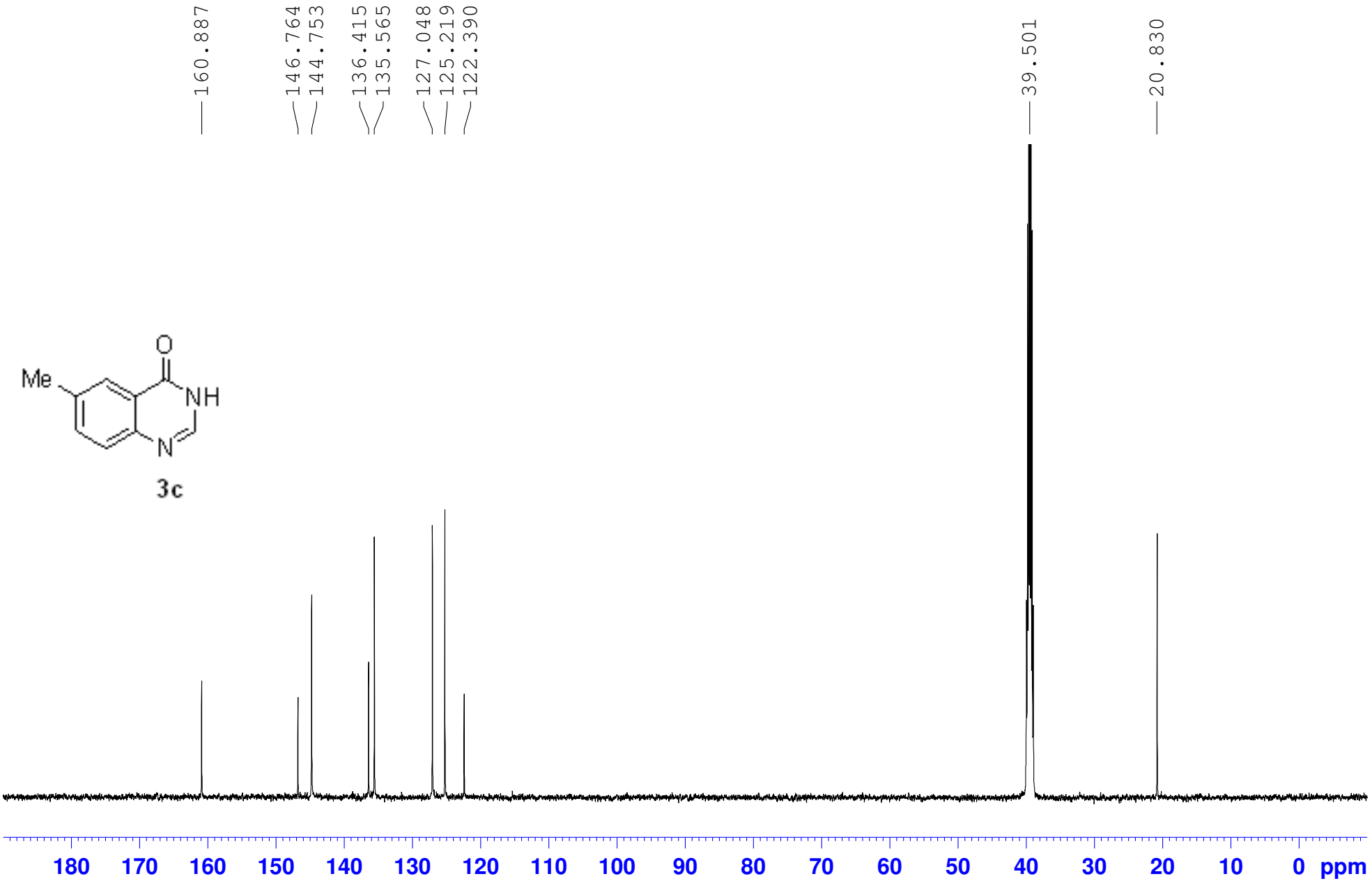

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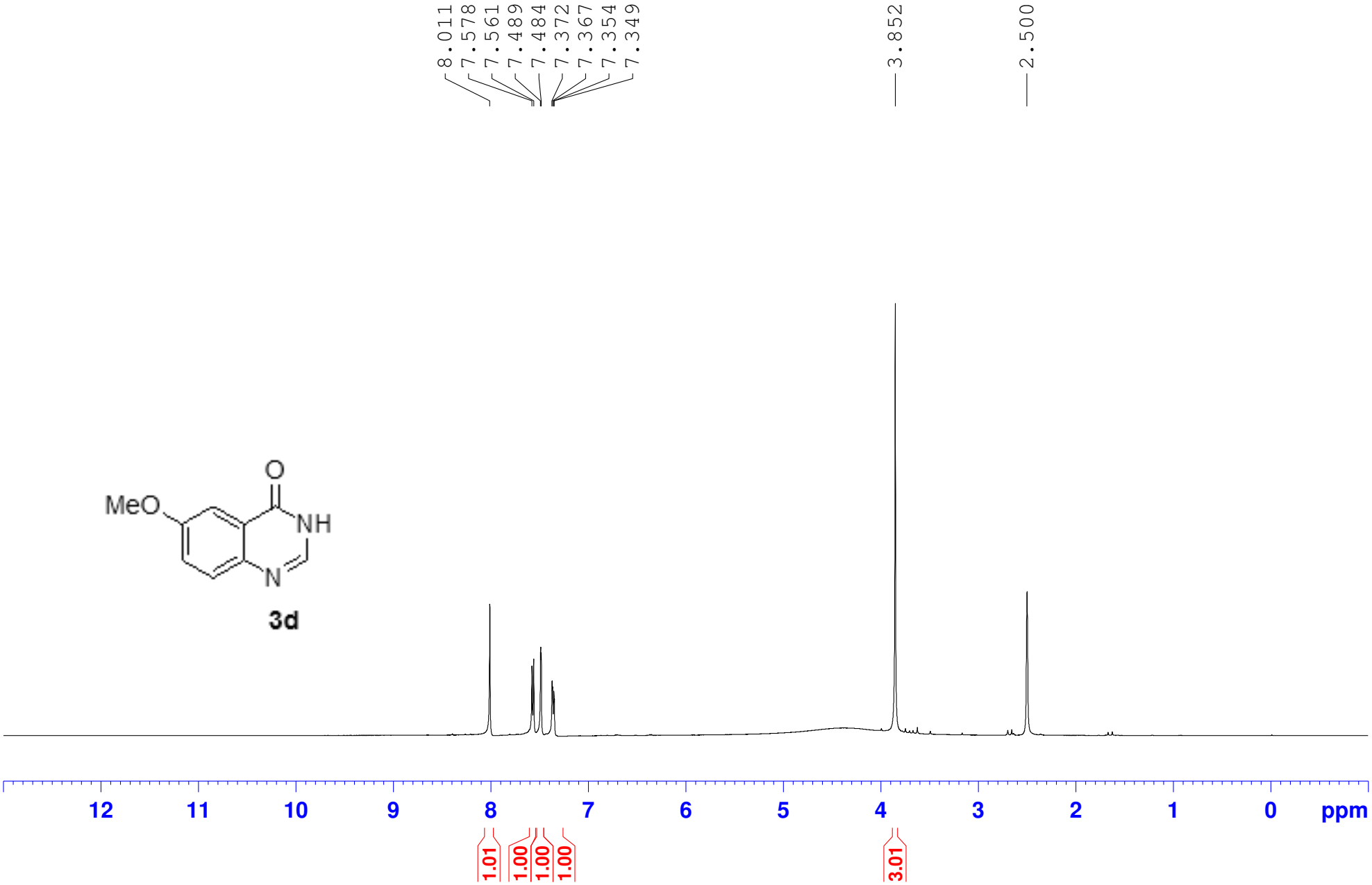

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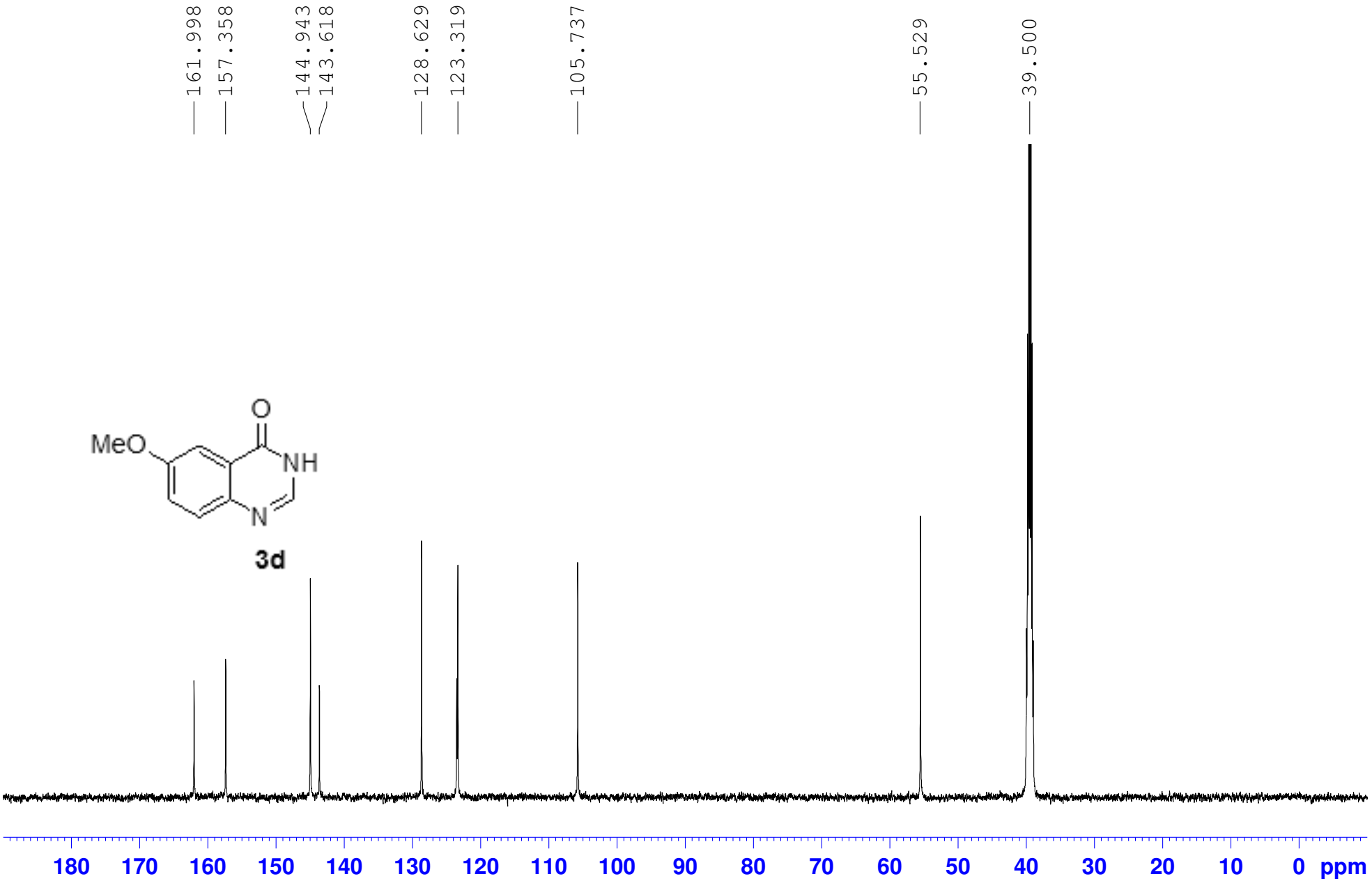

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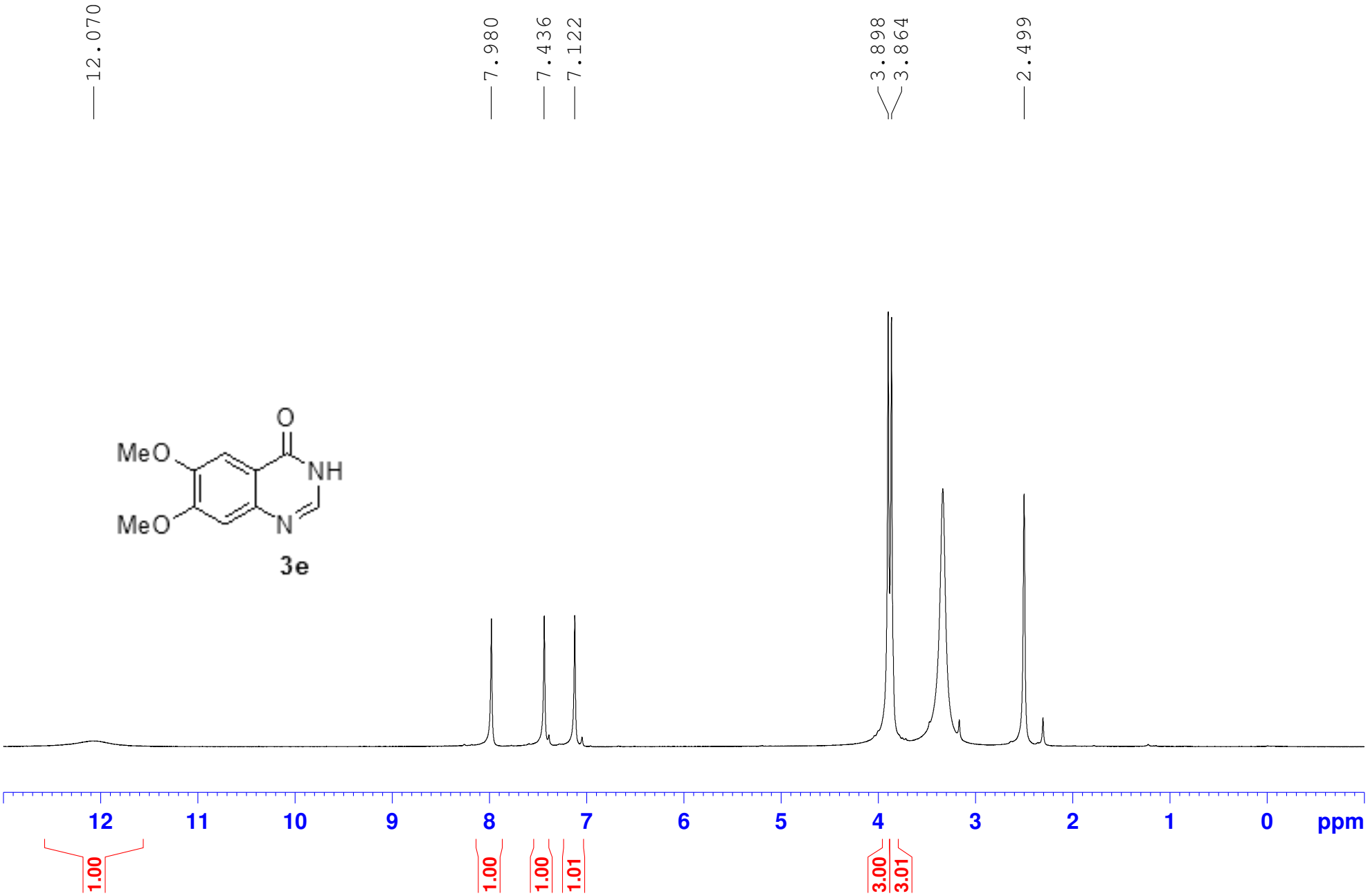

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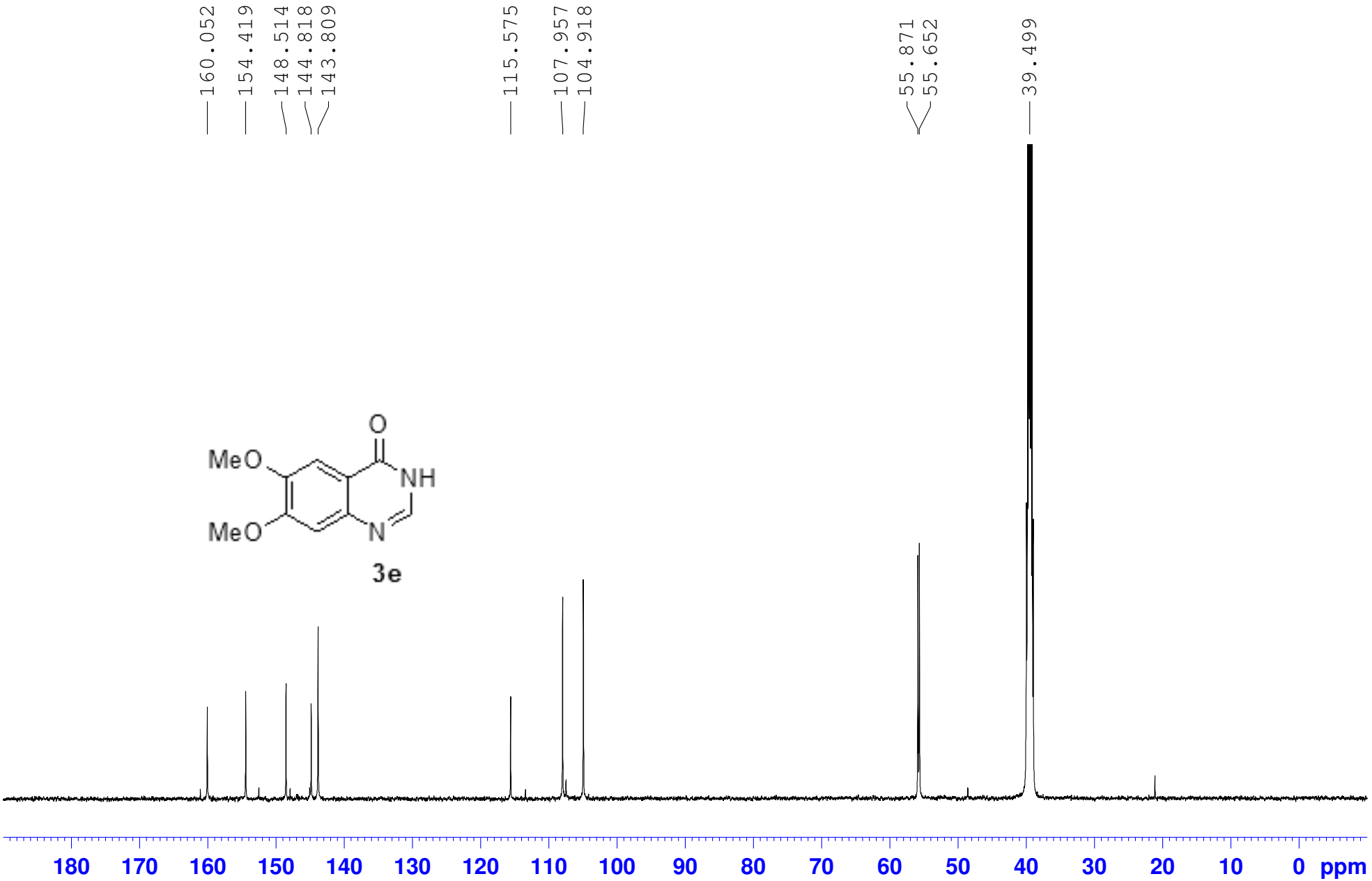

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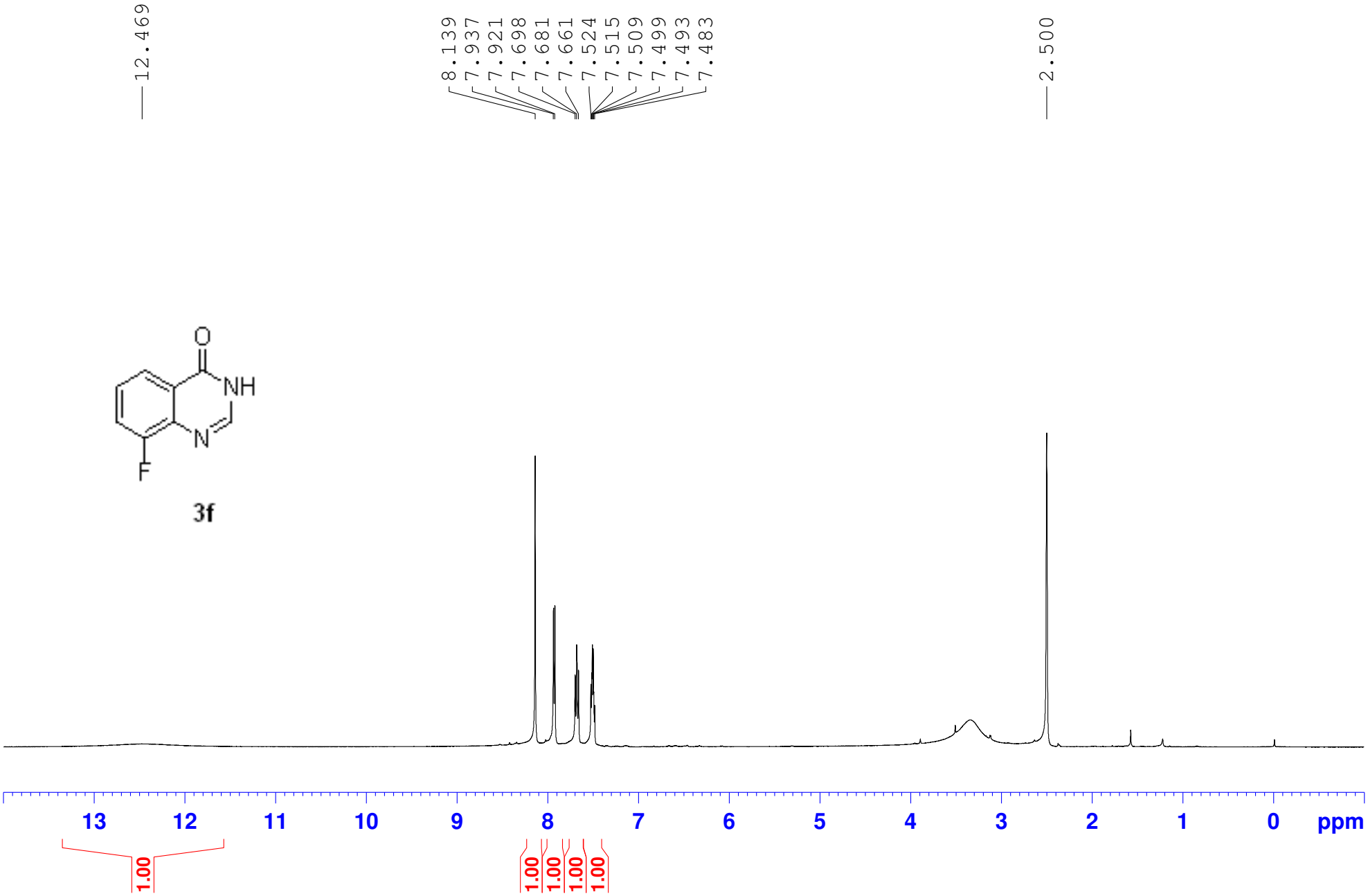

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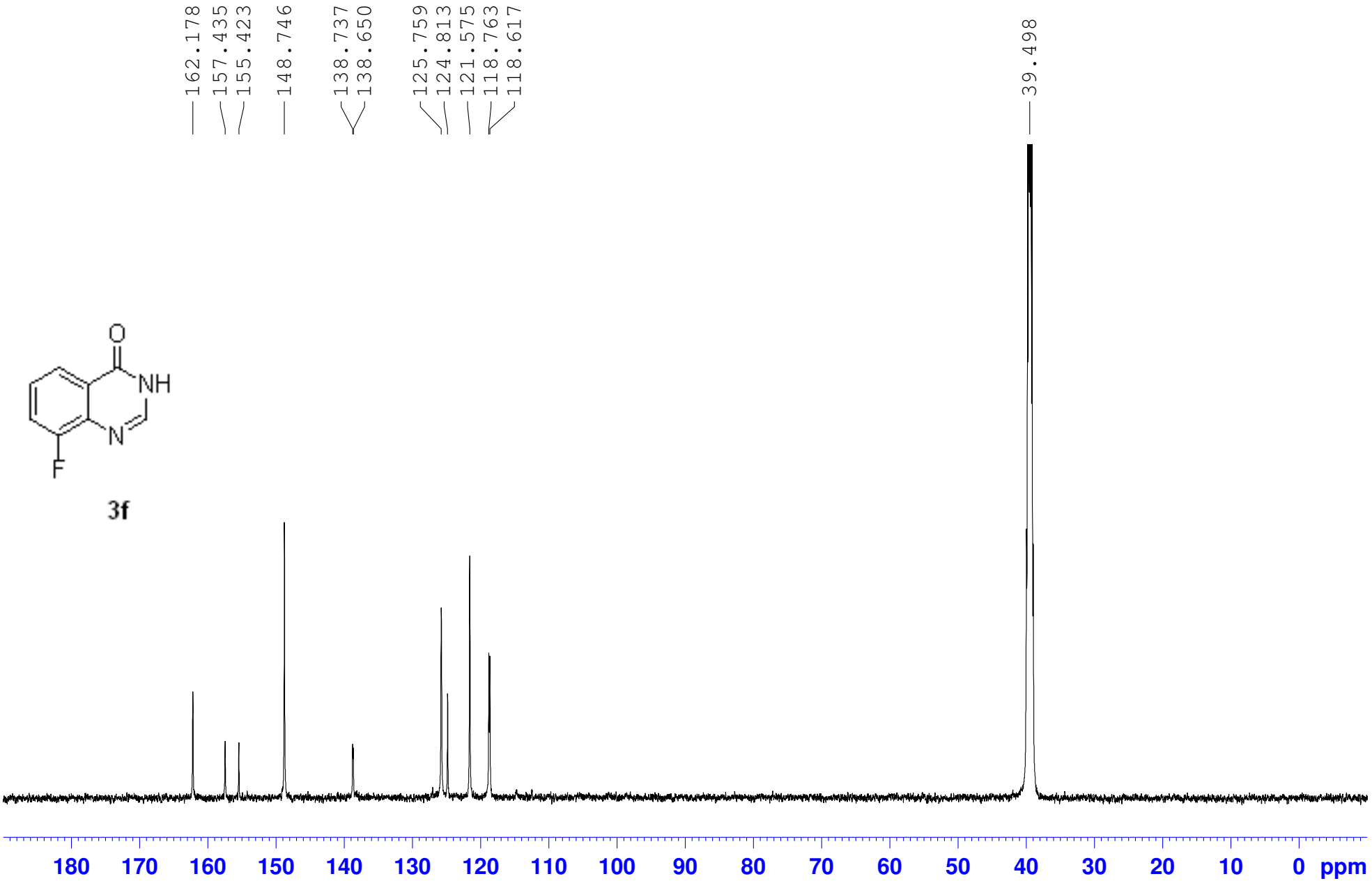

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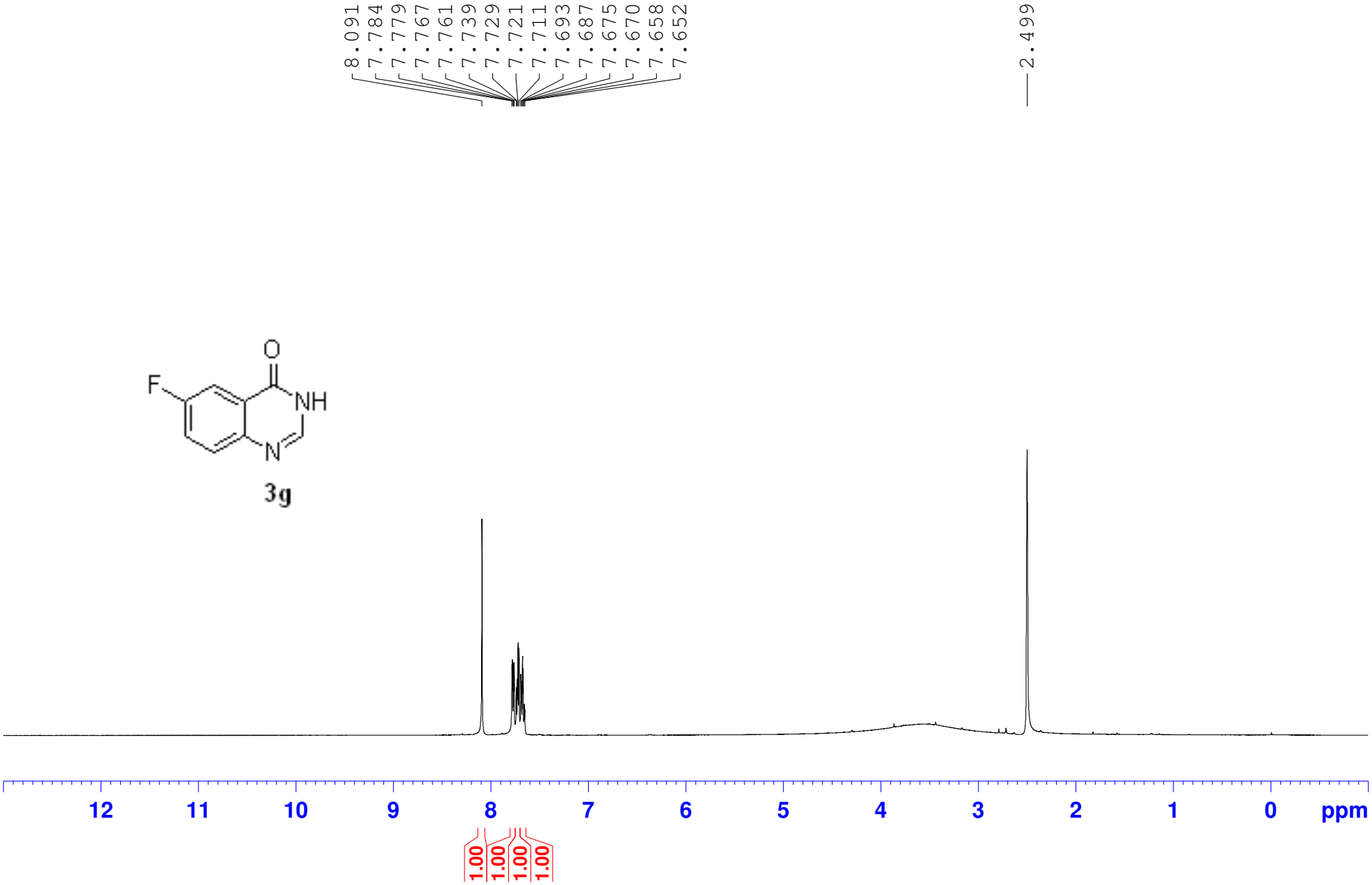

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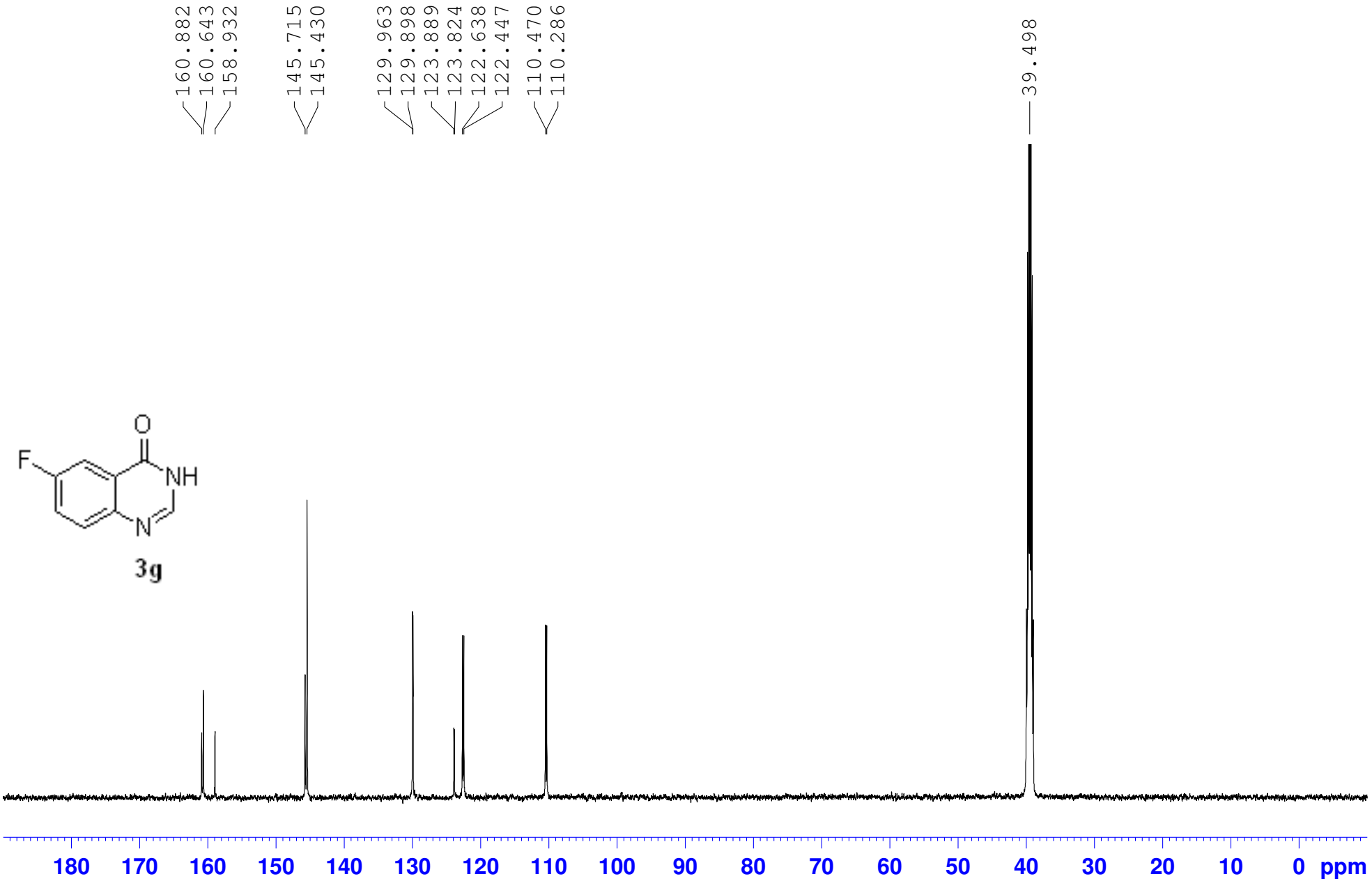

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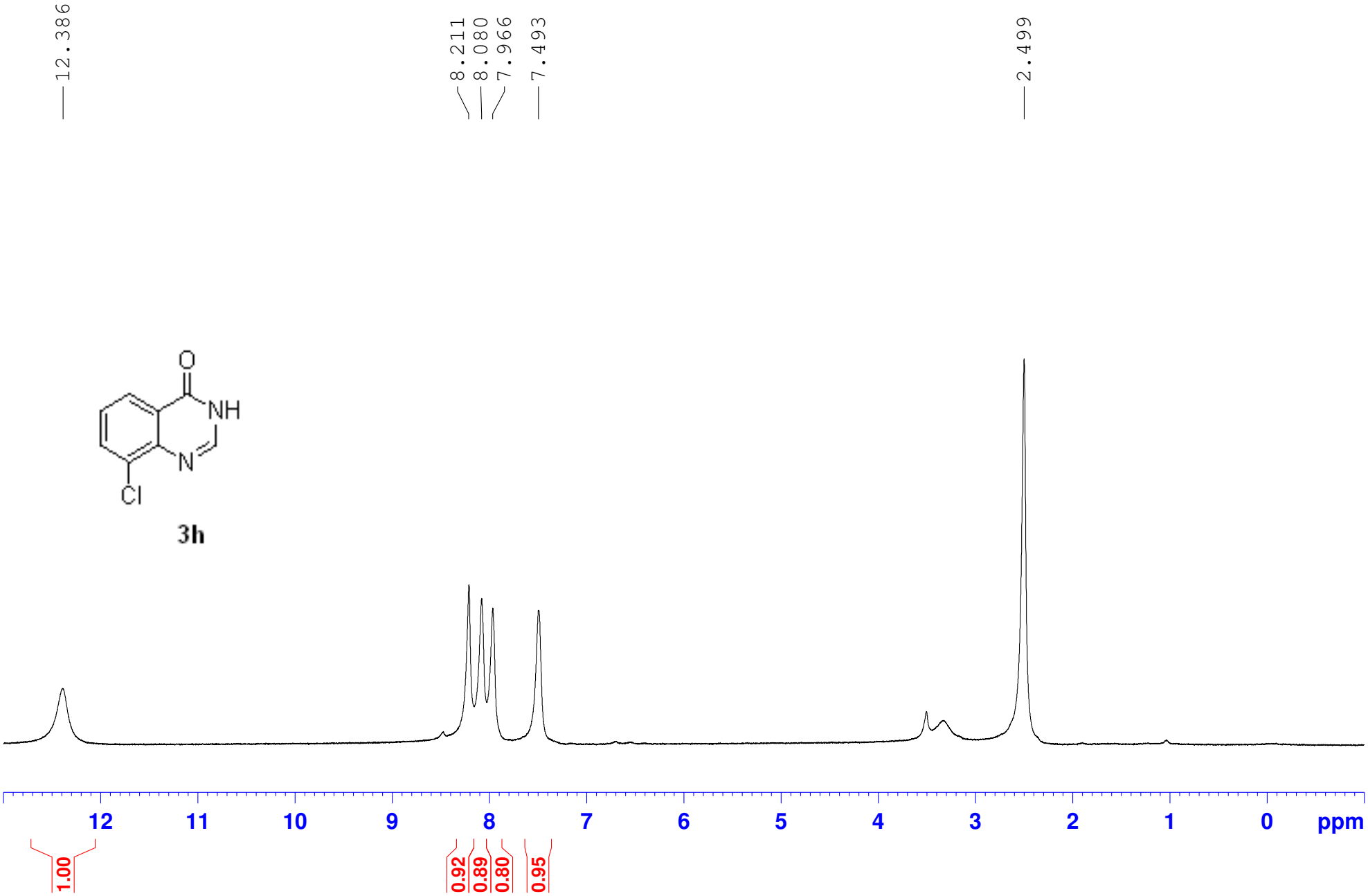

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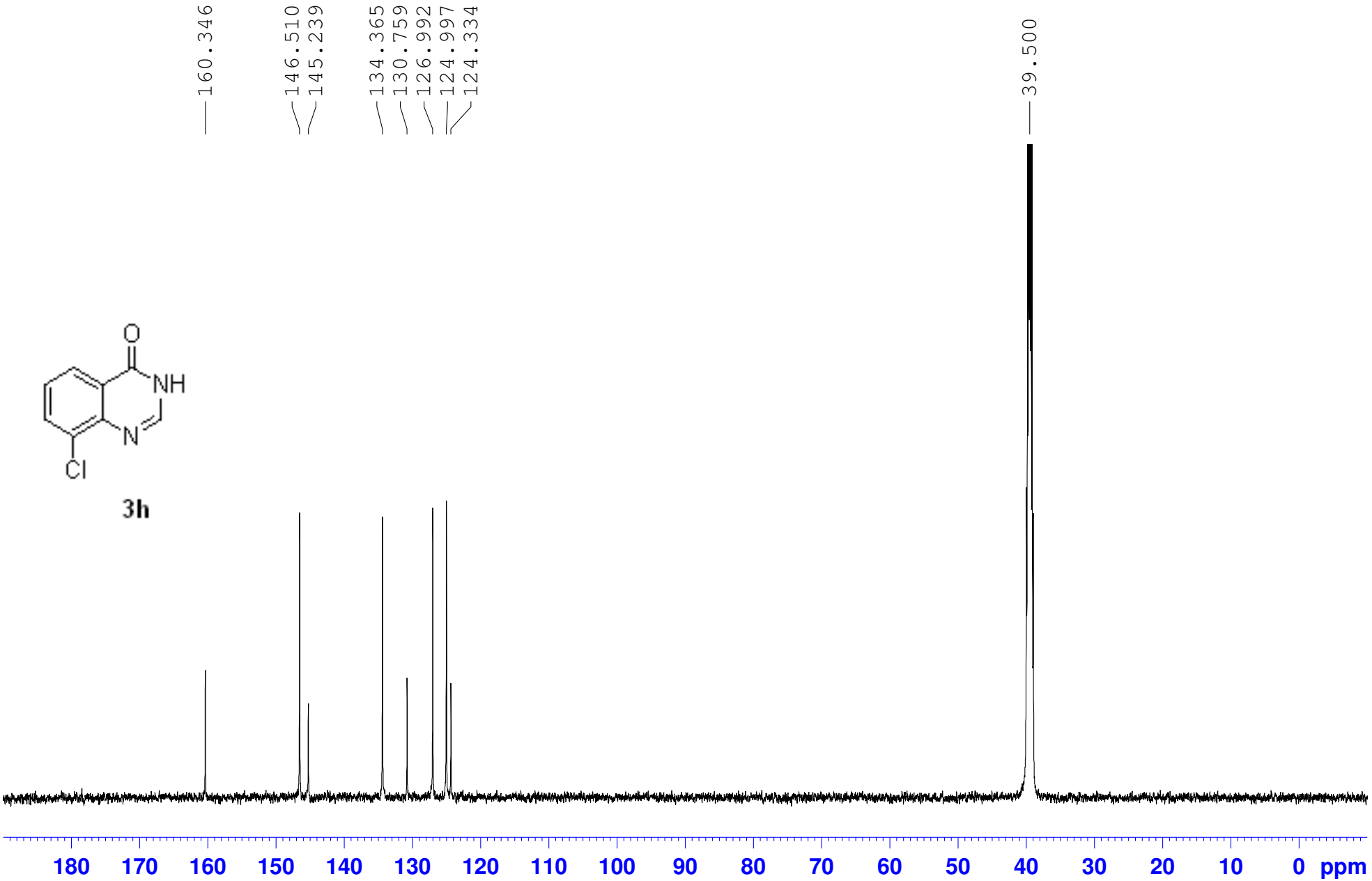

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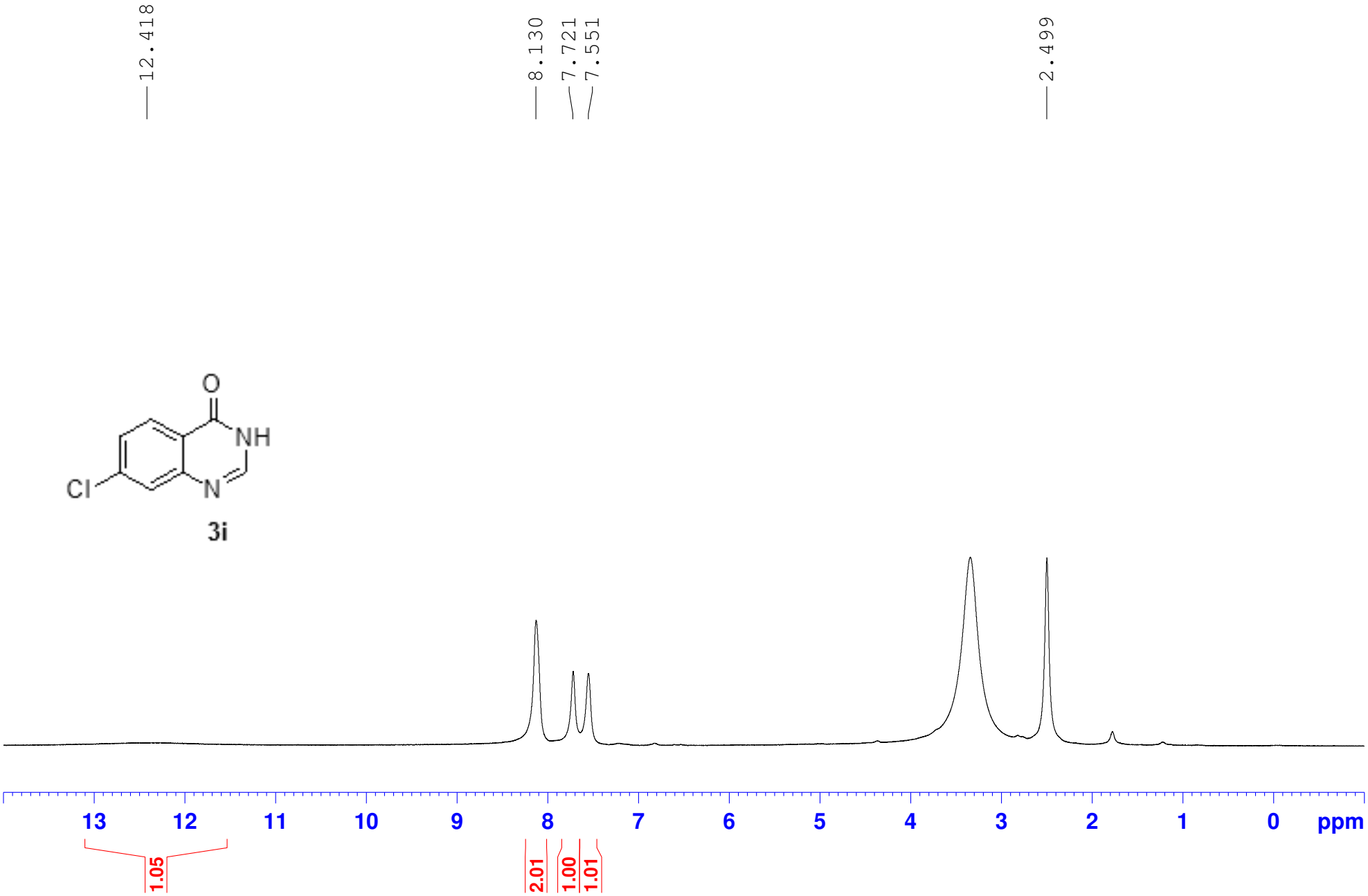

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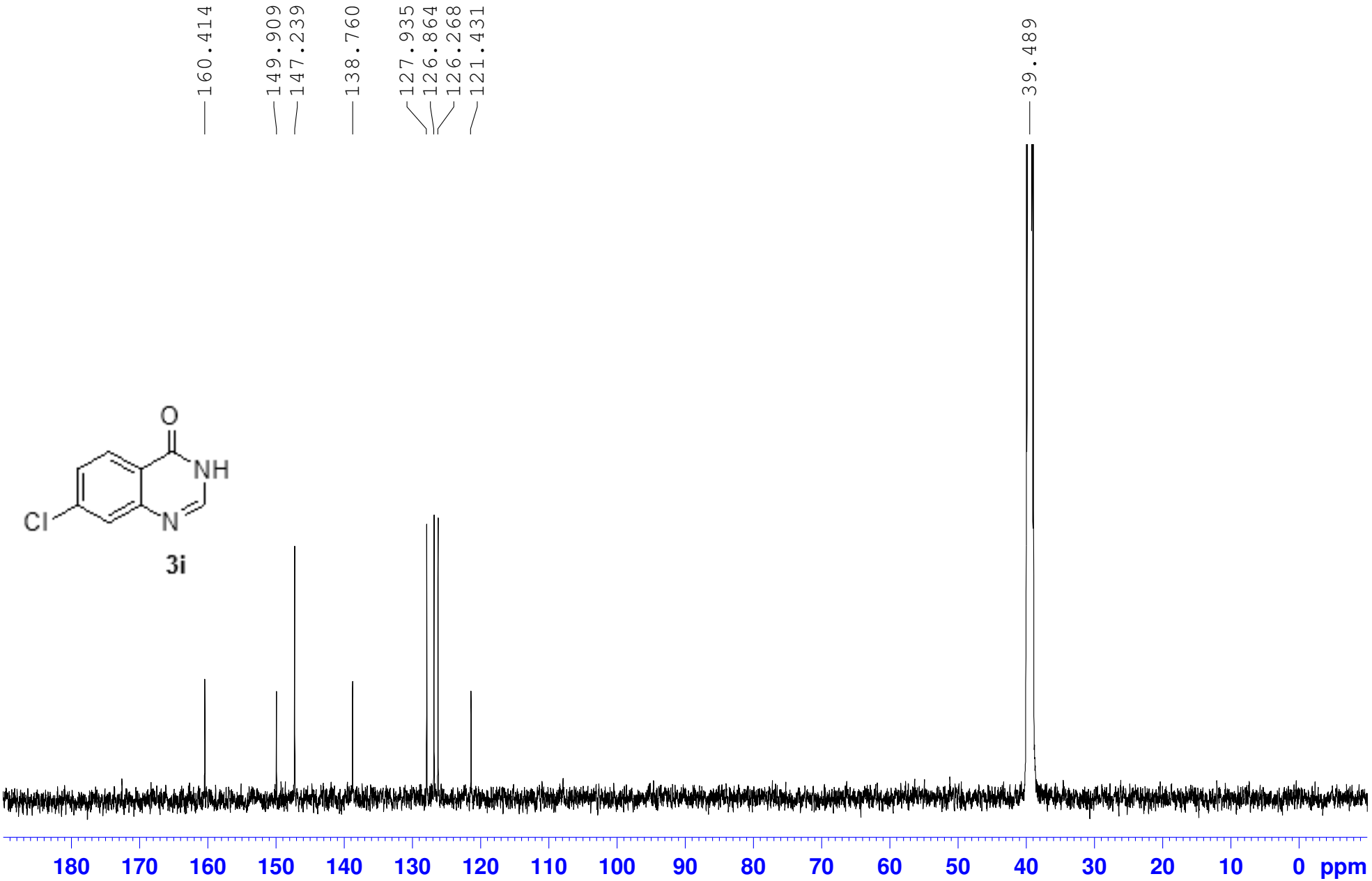

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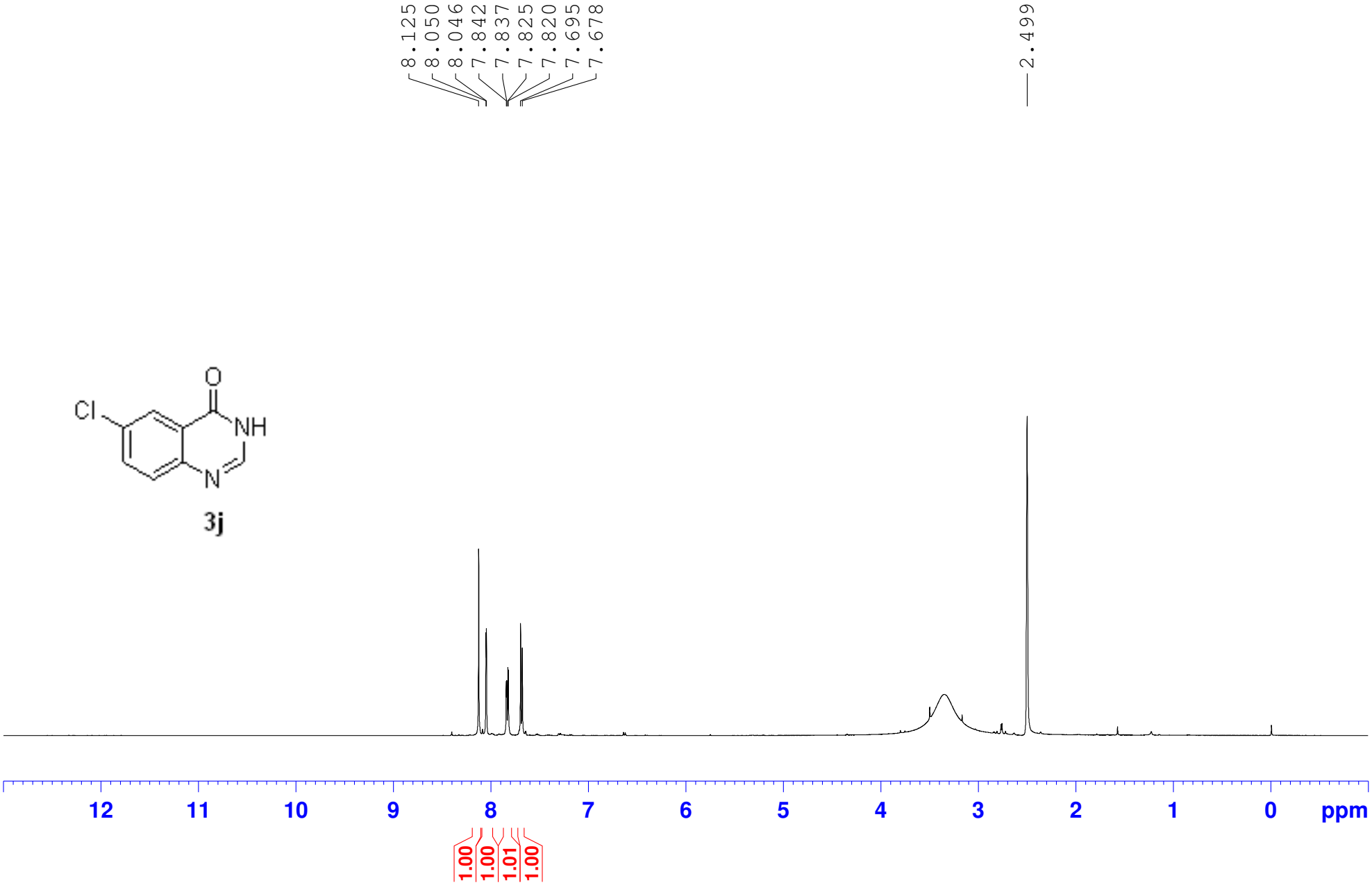

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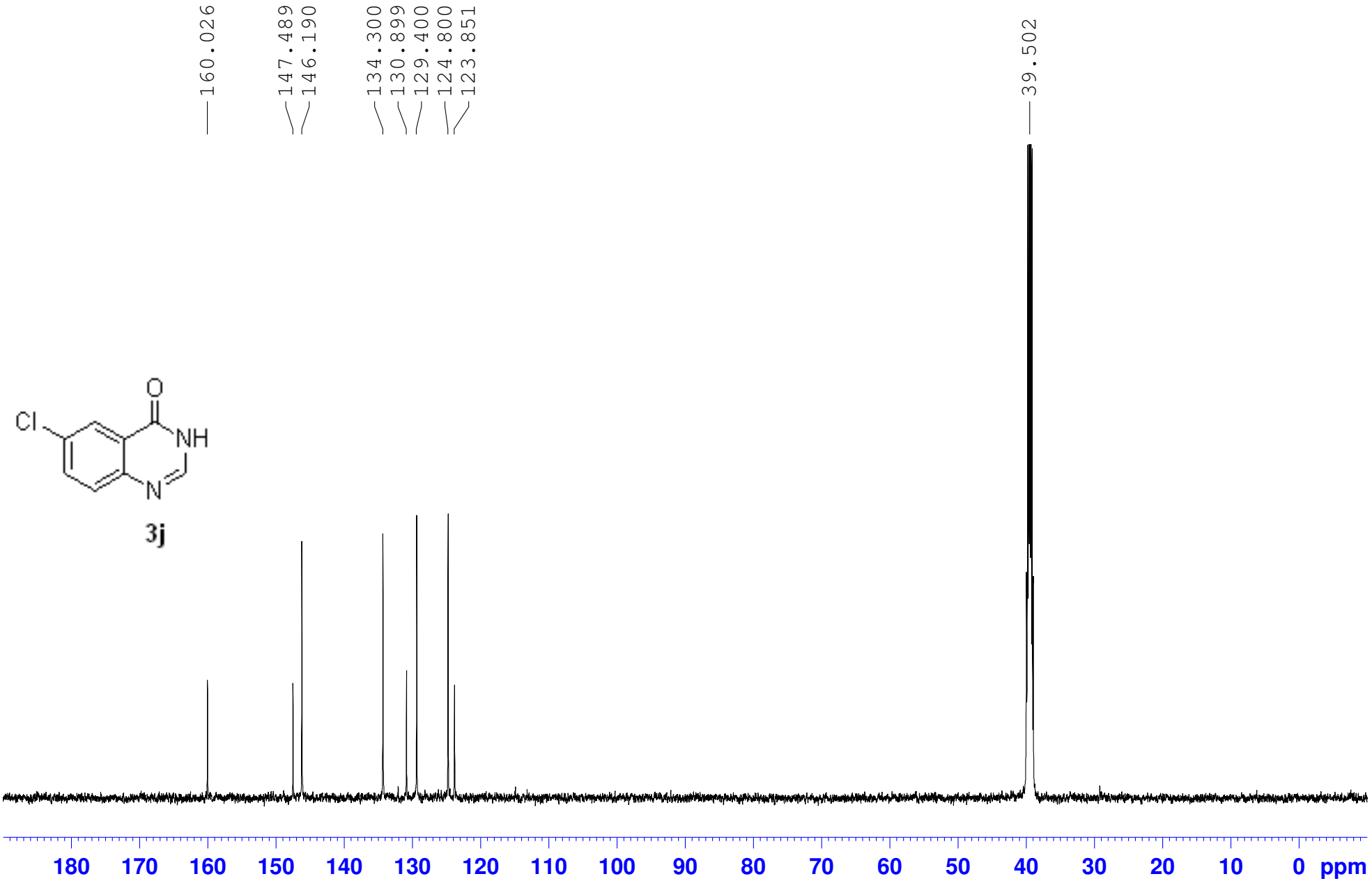

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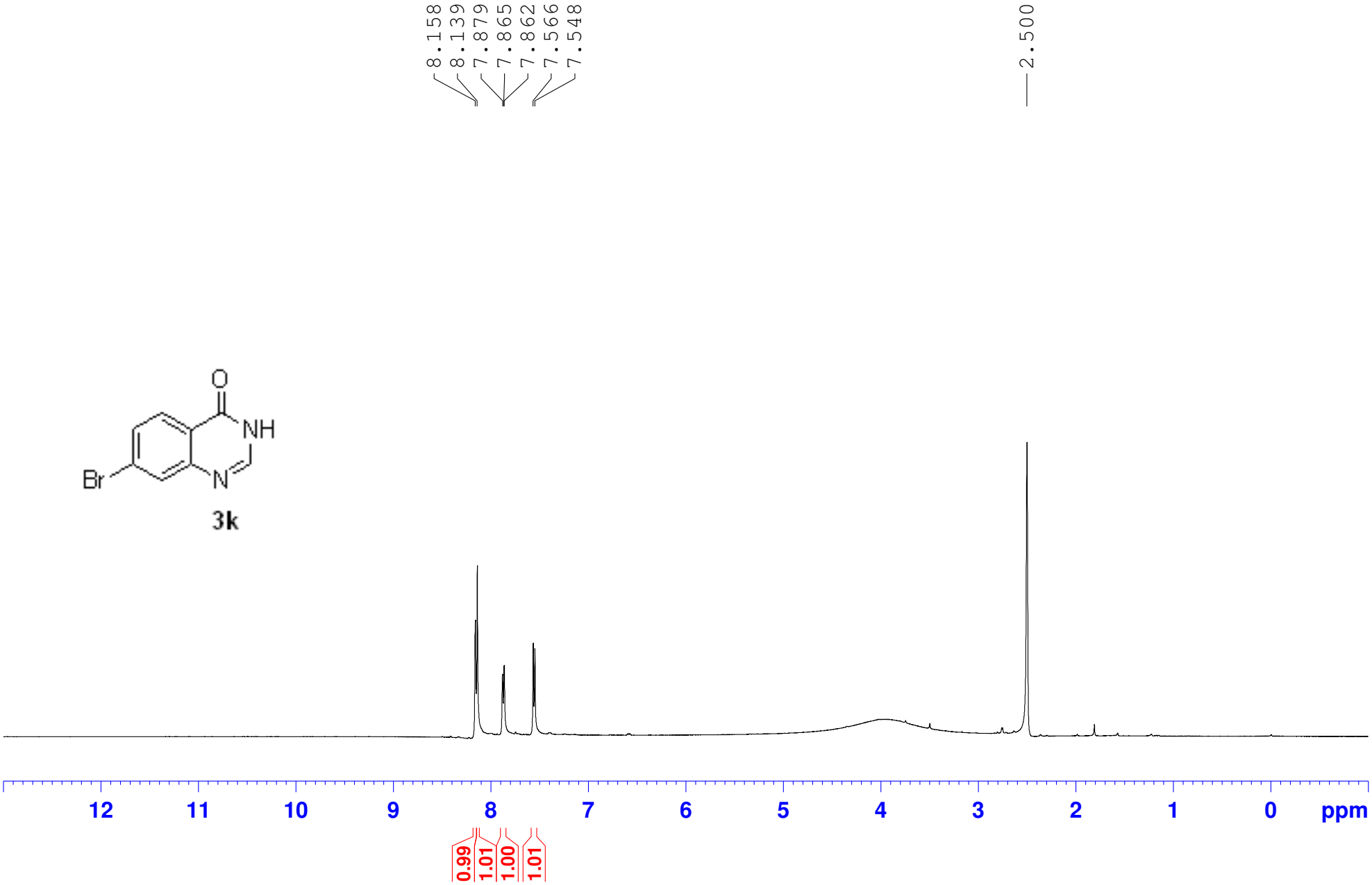

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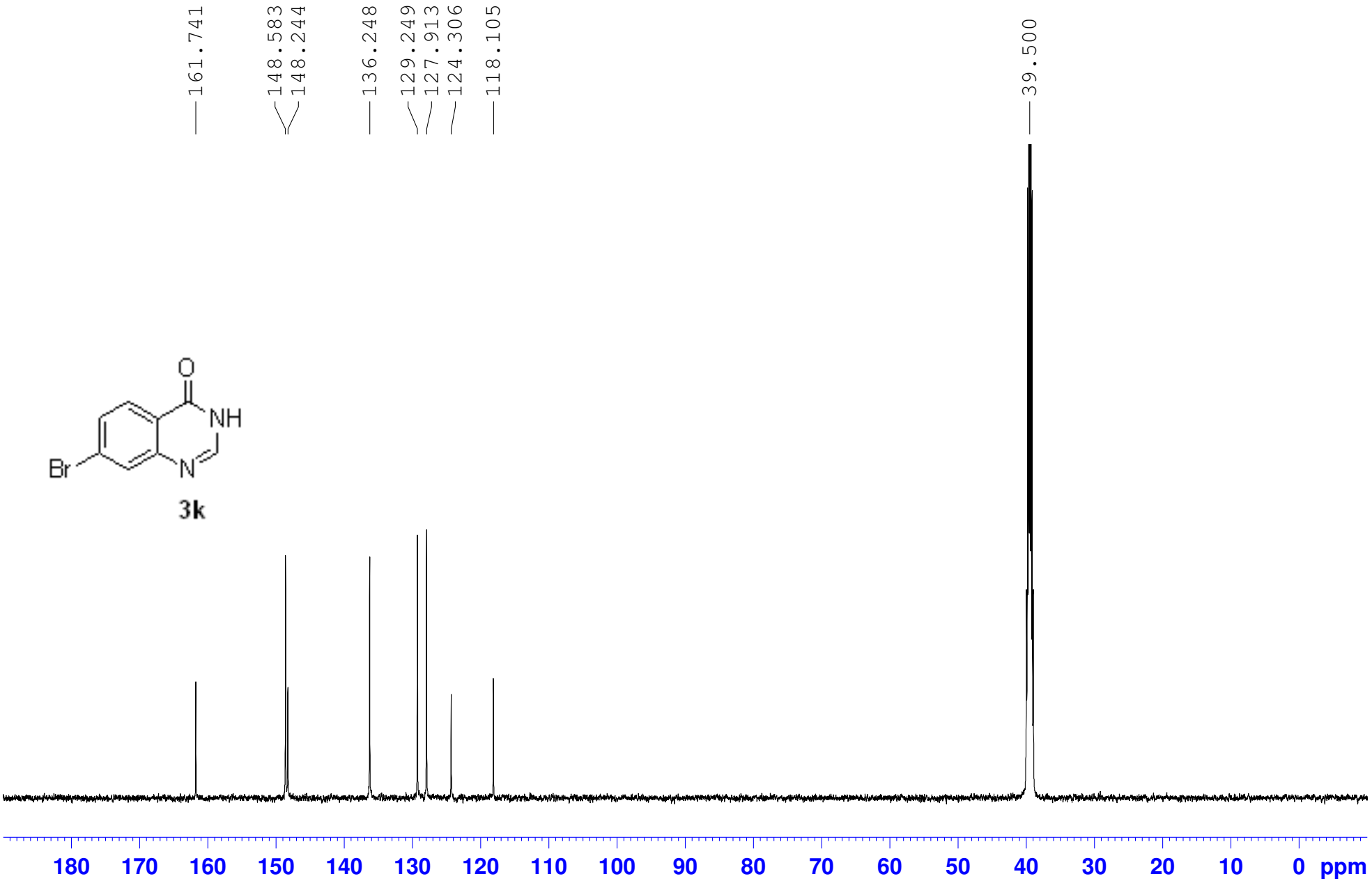

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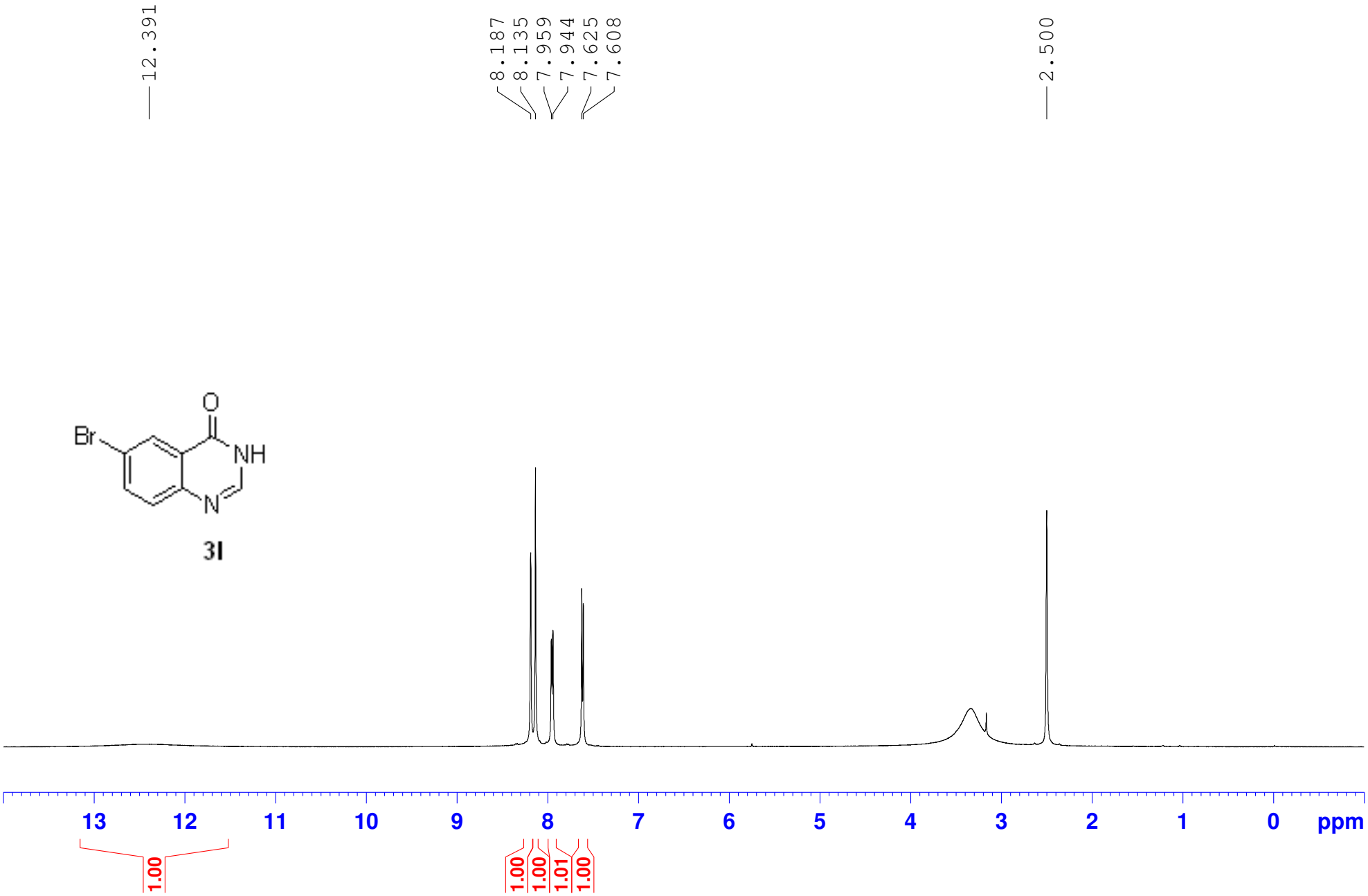

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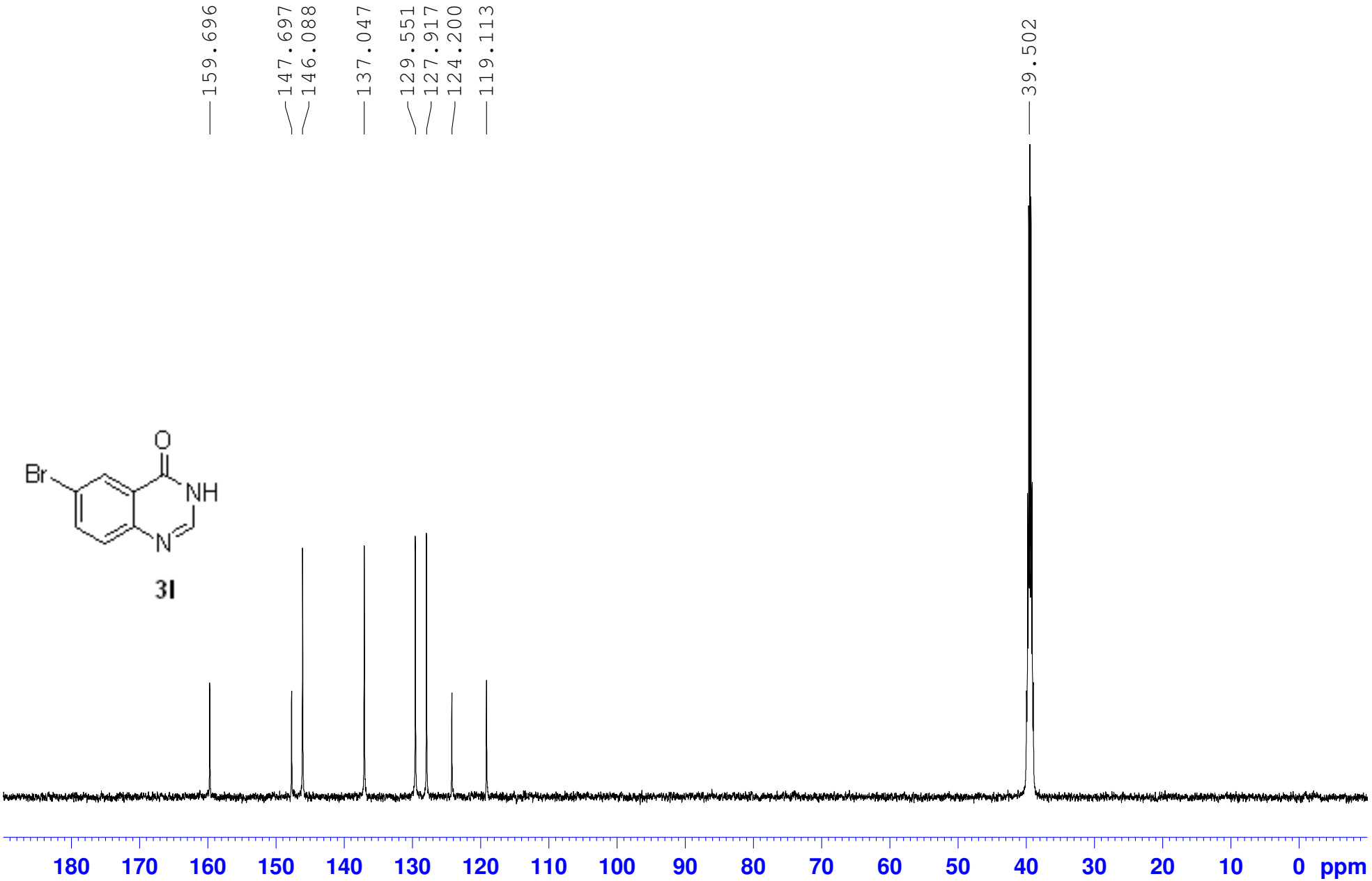

7-(trifluoromethyl)quinazolin-4(3H)-one
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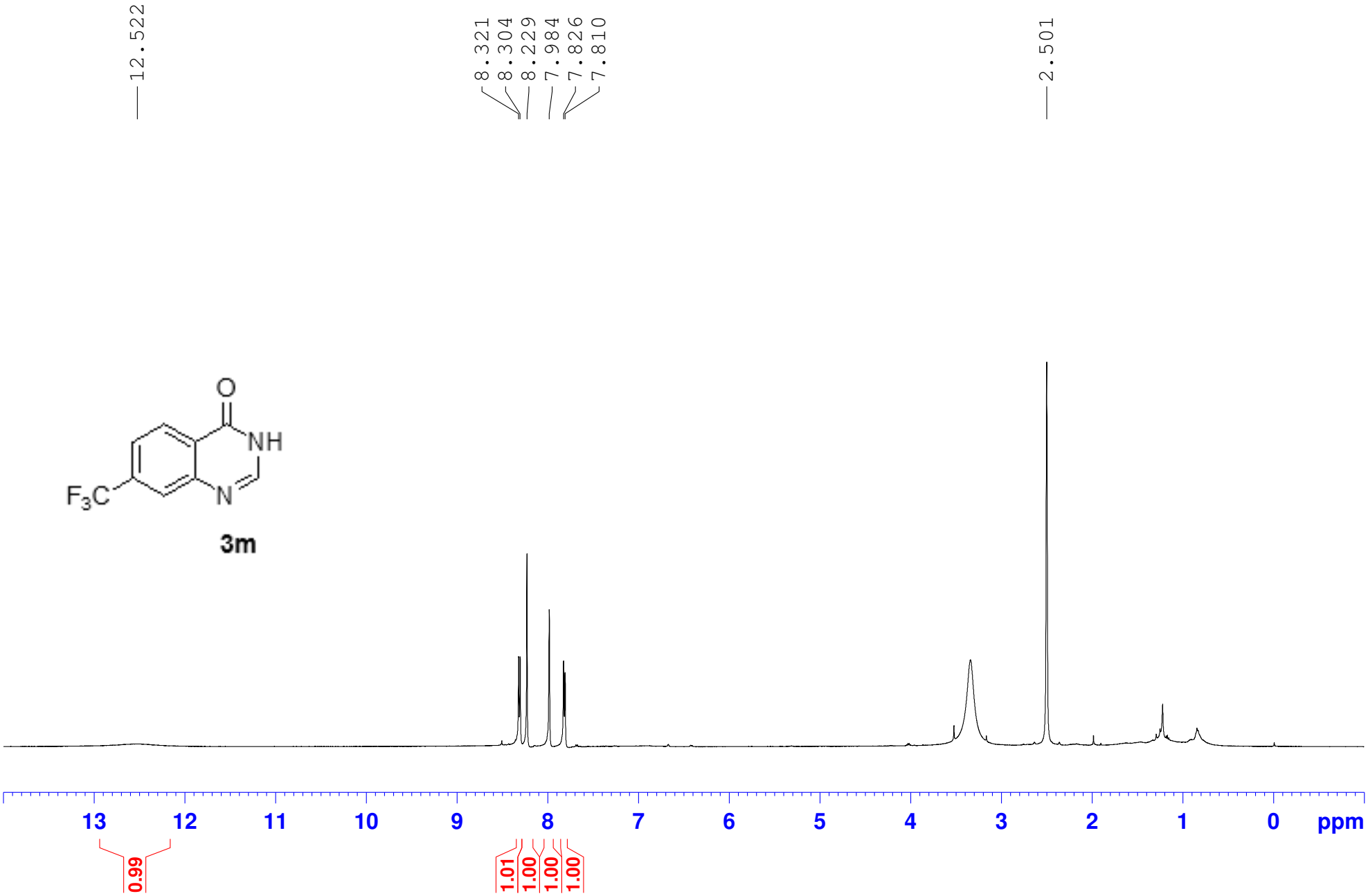

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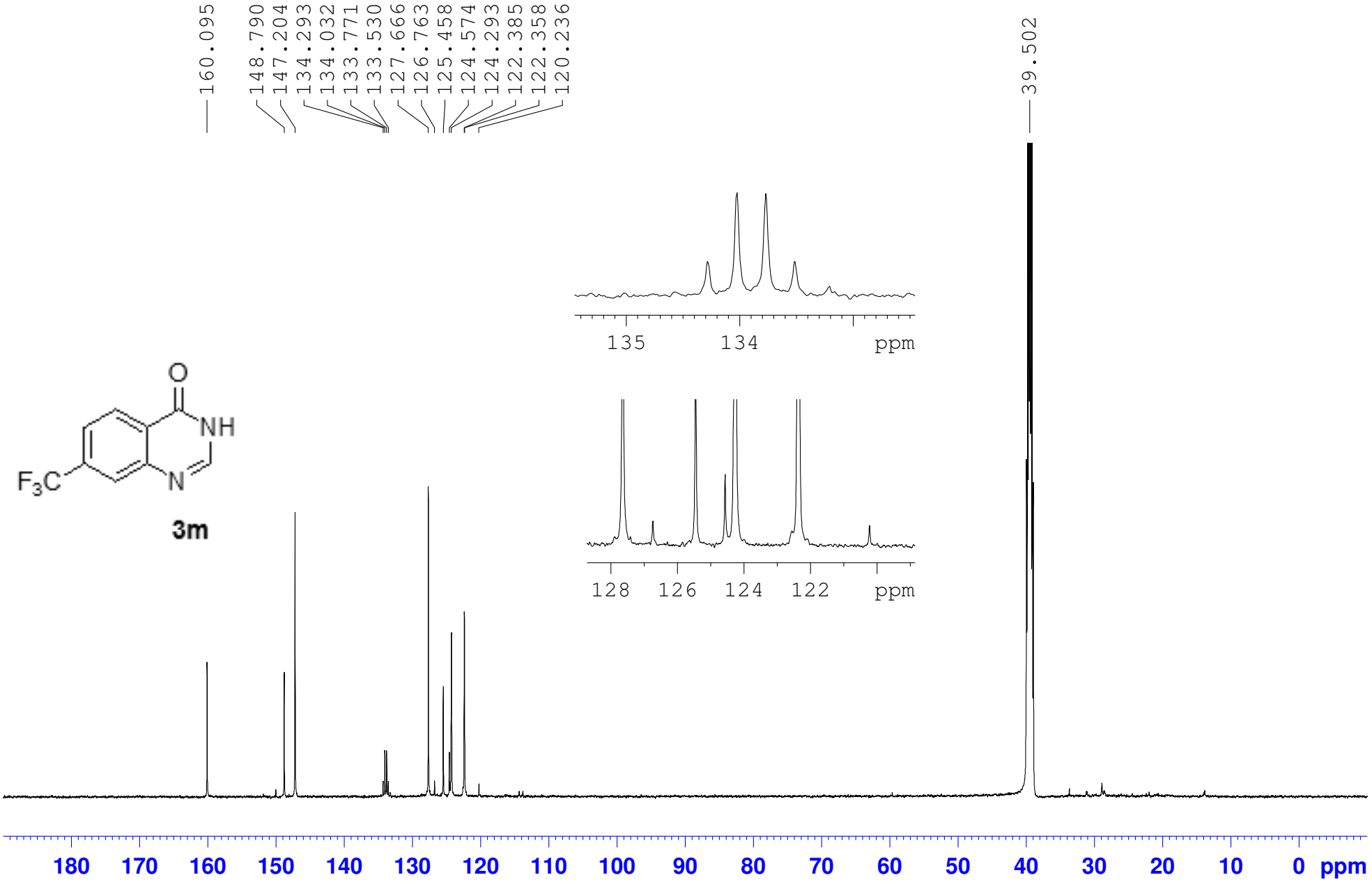

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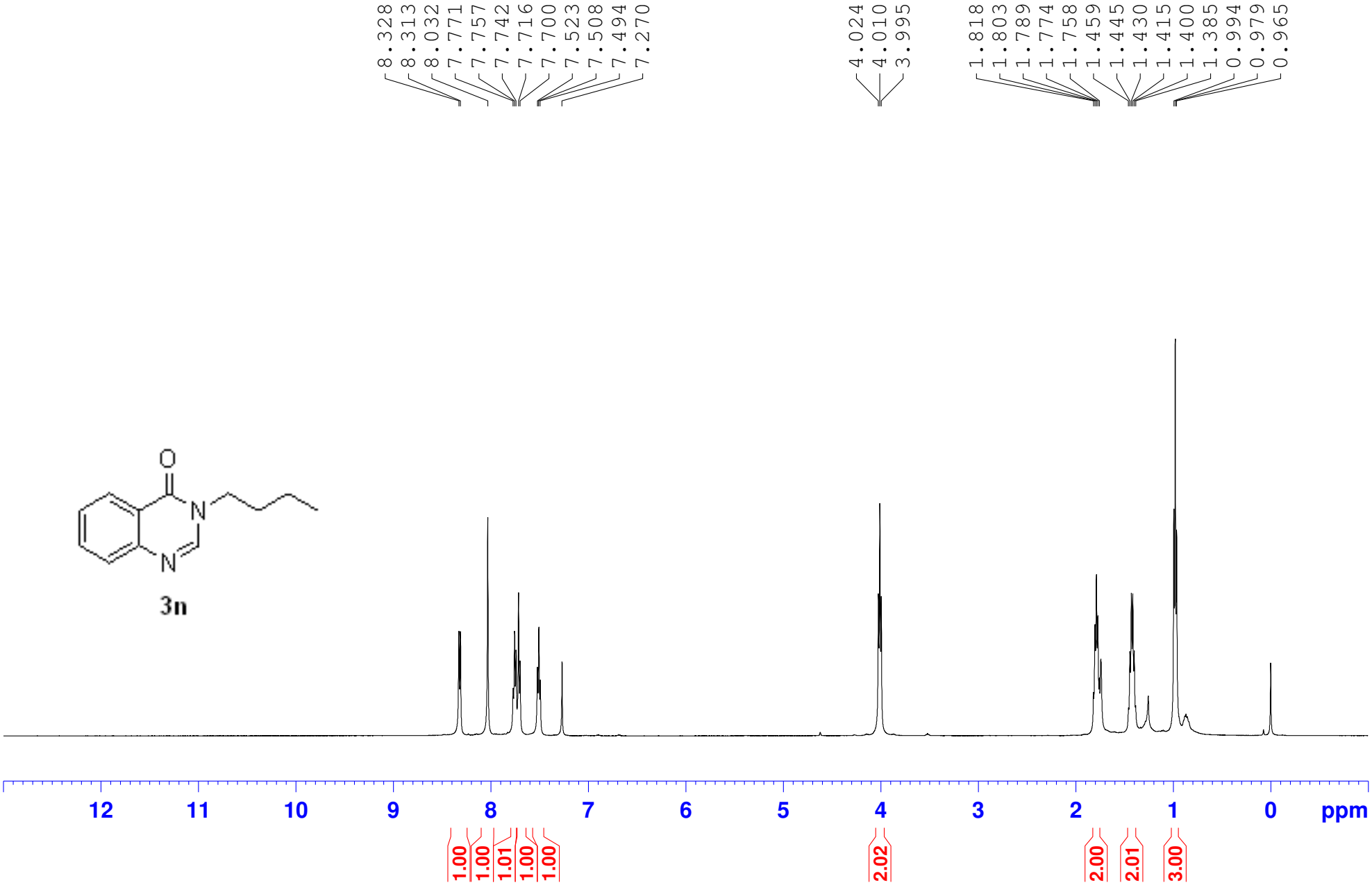
PROTON CDCl3



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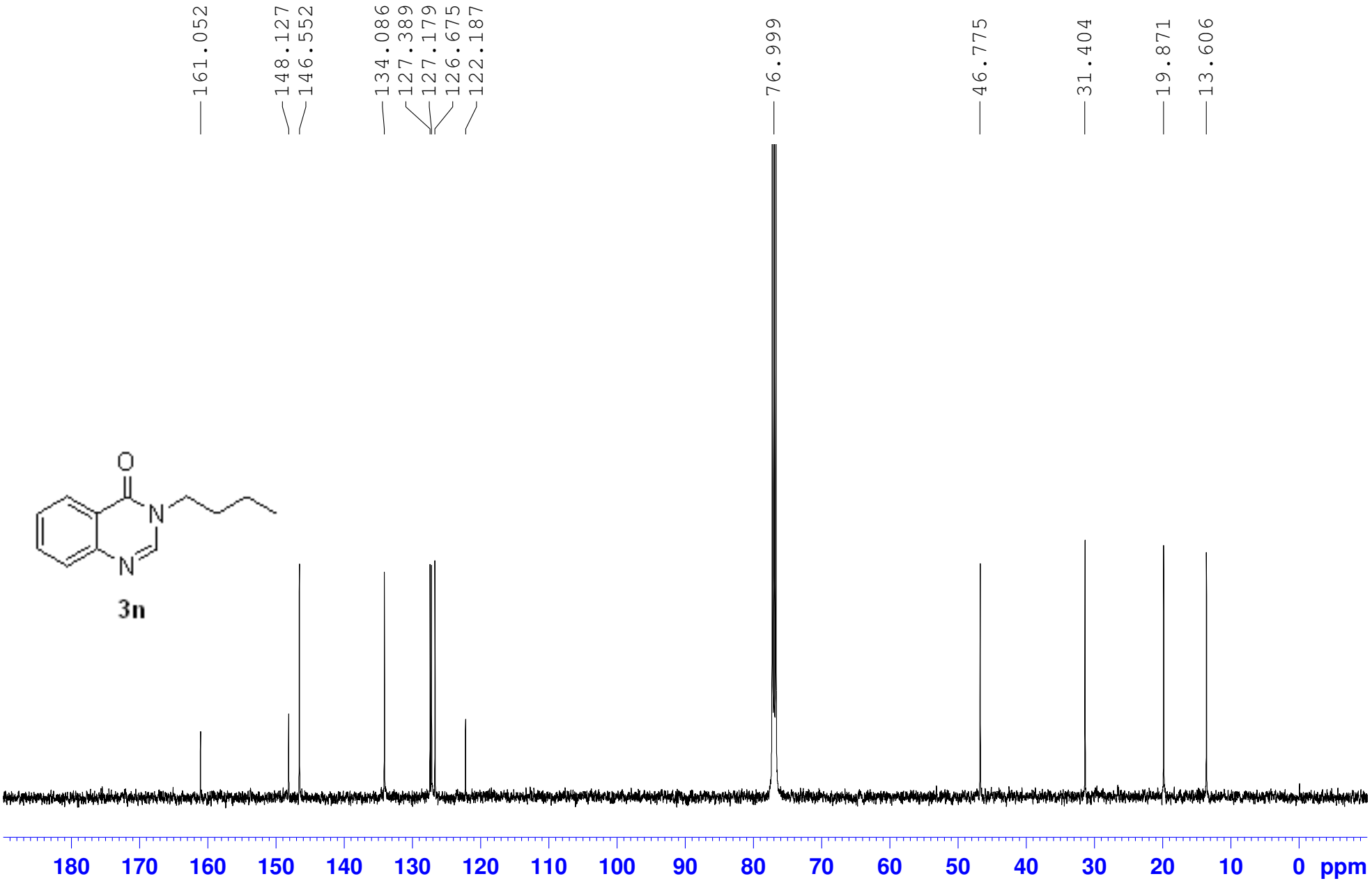
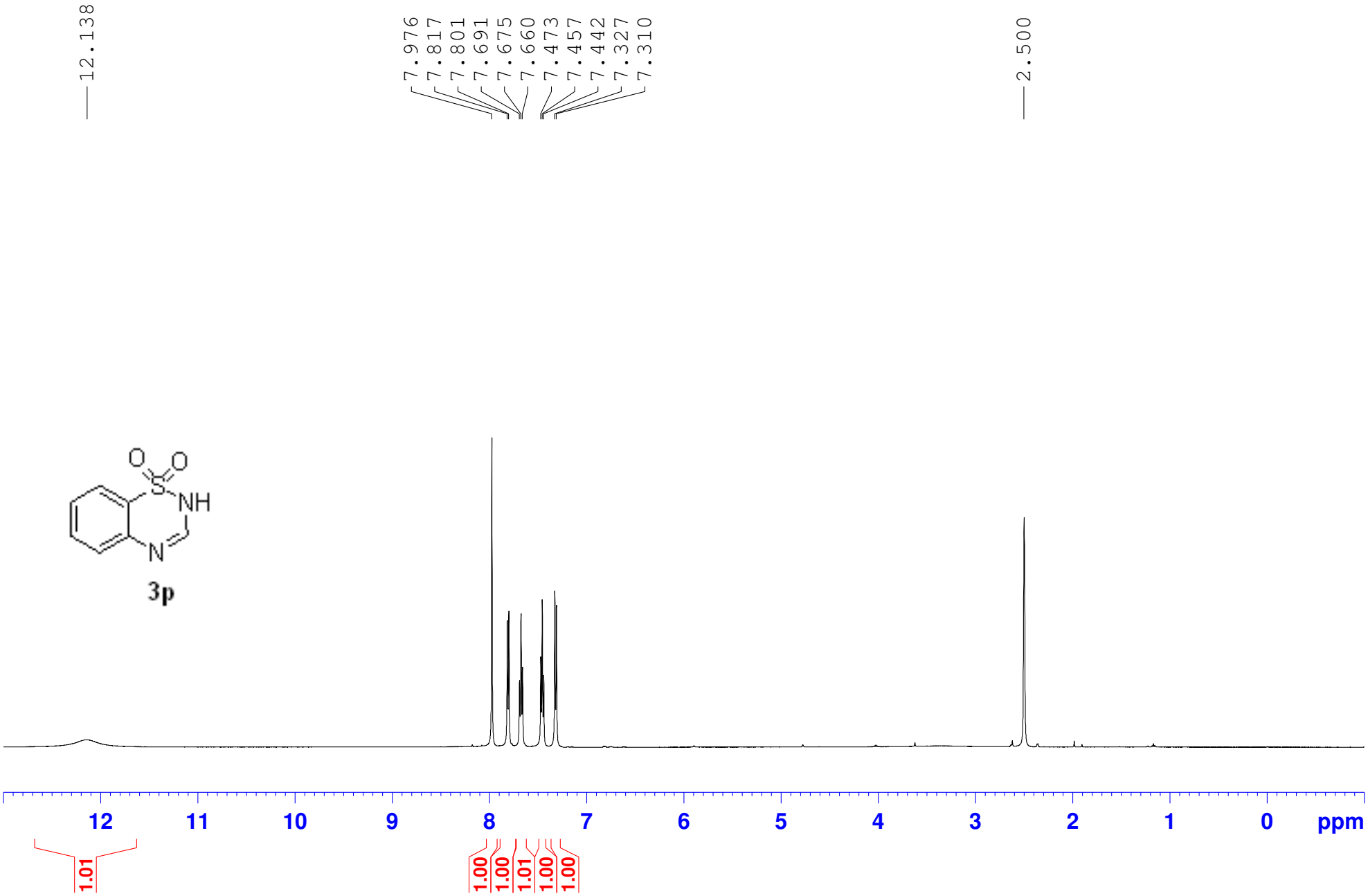
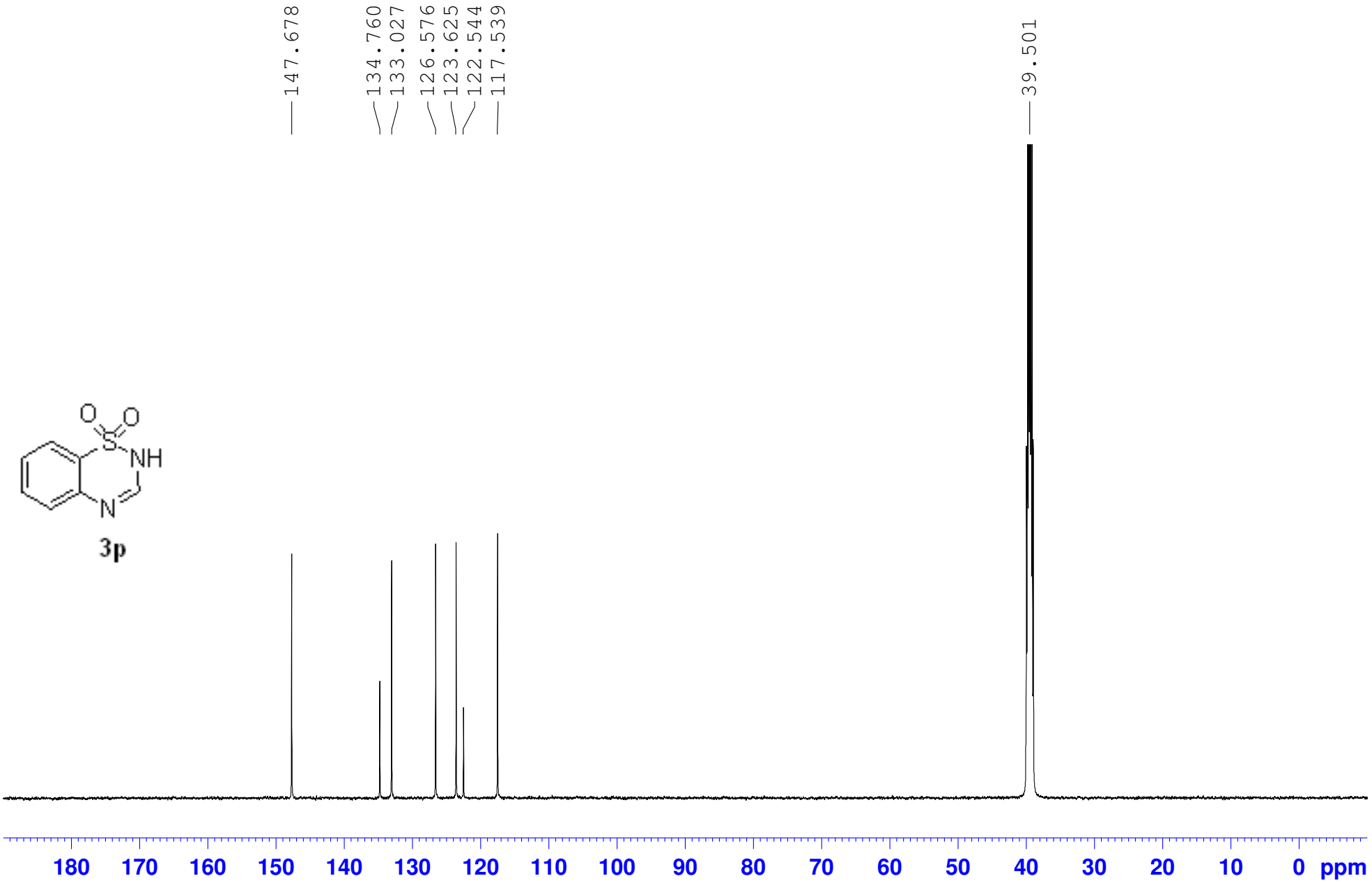
C13CPD CDCl3



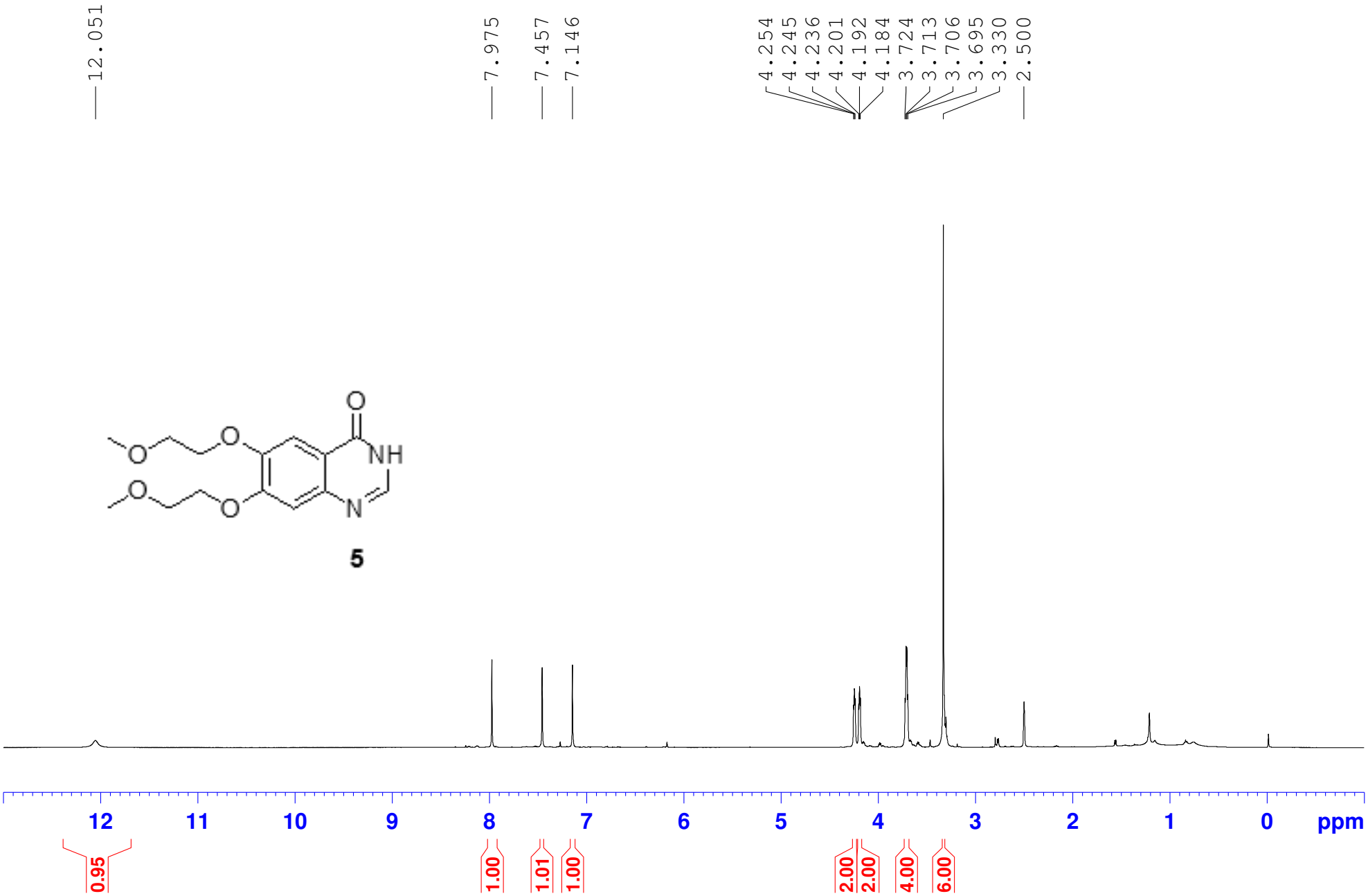
2H-benzo-1,2,4-thiadiazine 1,1-dioxide
PROTON DMSO-d6



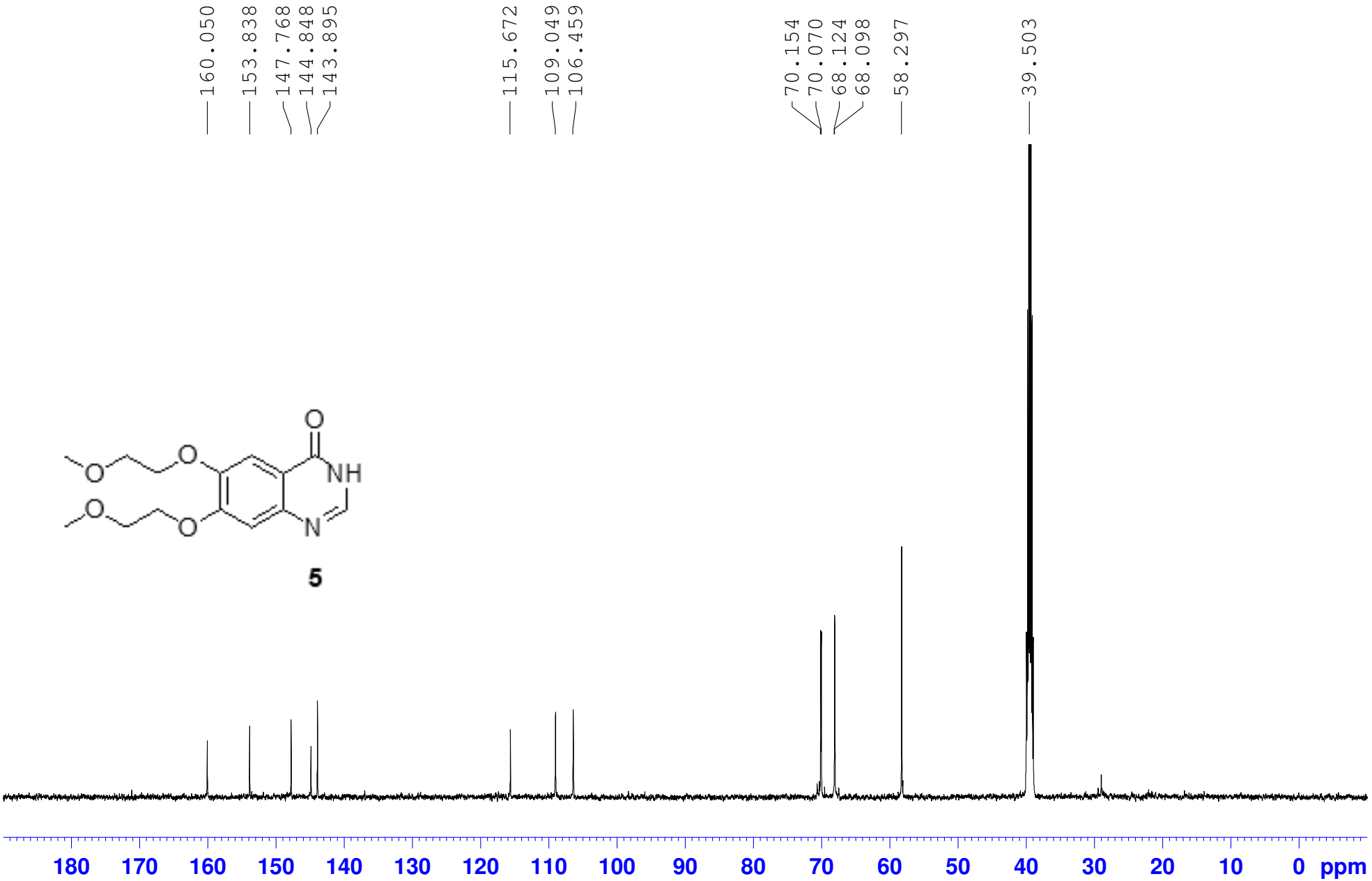
2H-benzo-1,2,4-thiadiazine 1,1-dioxide
C13CPD DMSO-d6



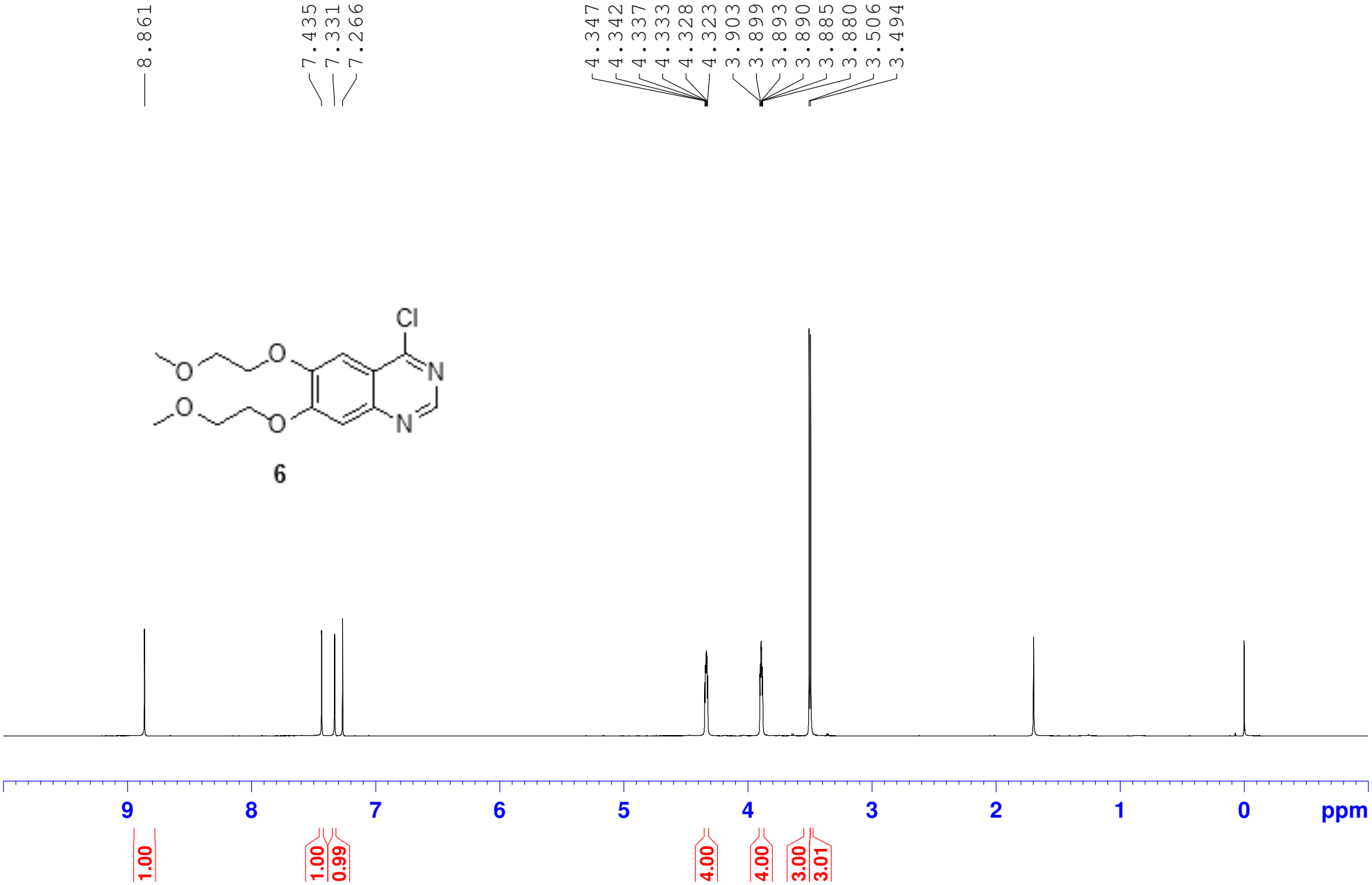
6,7-bis(2-methoxyethoxy)quinazolin-4(3H)-one
PROTON DMSO-d6



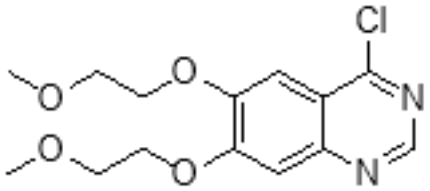
6,7-bis(2-methoxyethoxy)quinazolin-4(3H)-one
C13CPD DMSO-d6



4-chloro-6,7-bis(2-methoxyethoxy)quinazoline
Proton CDCl3



4-chloro-6,7-bis(2-methoxyethoxy)quinazoline
C13CPD CDCl3



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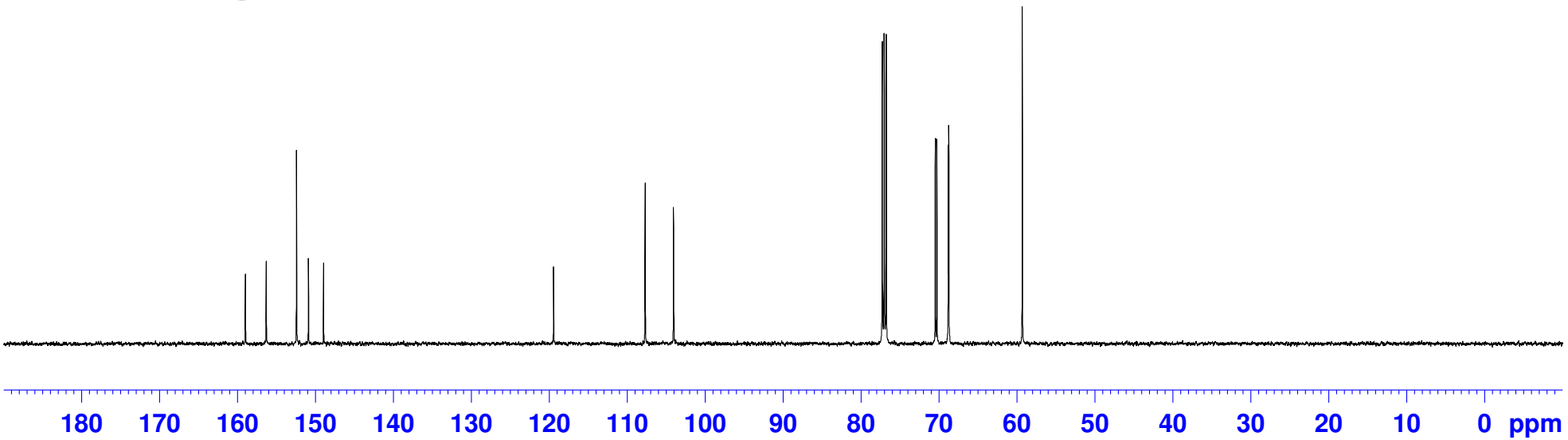
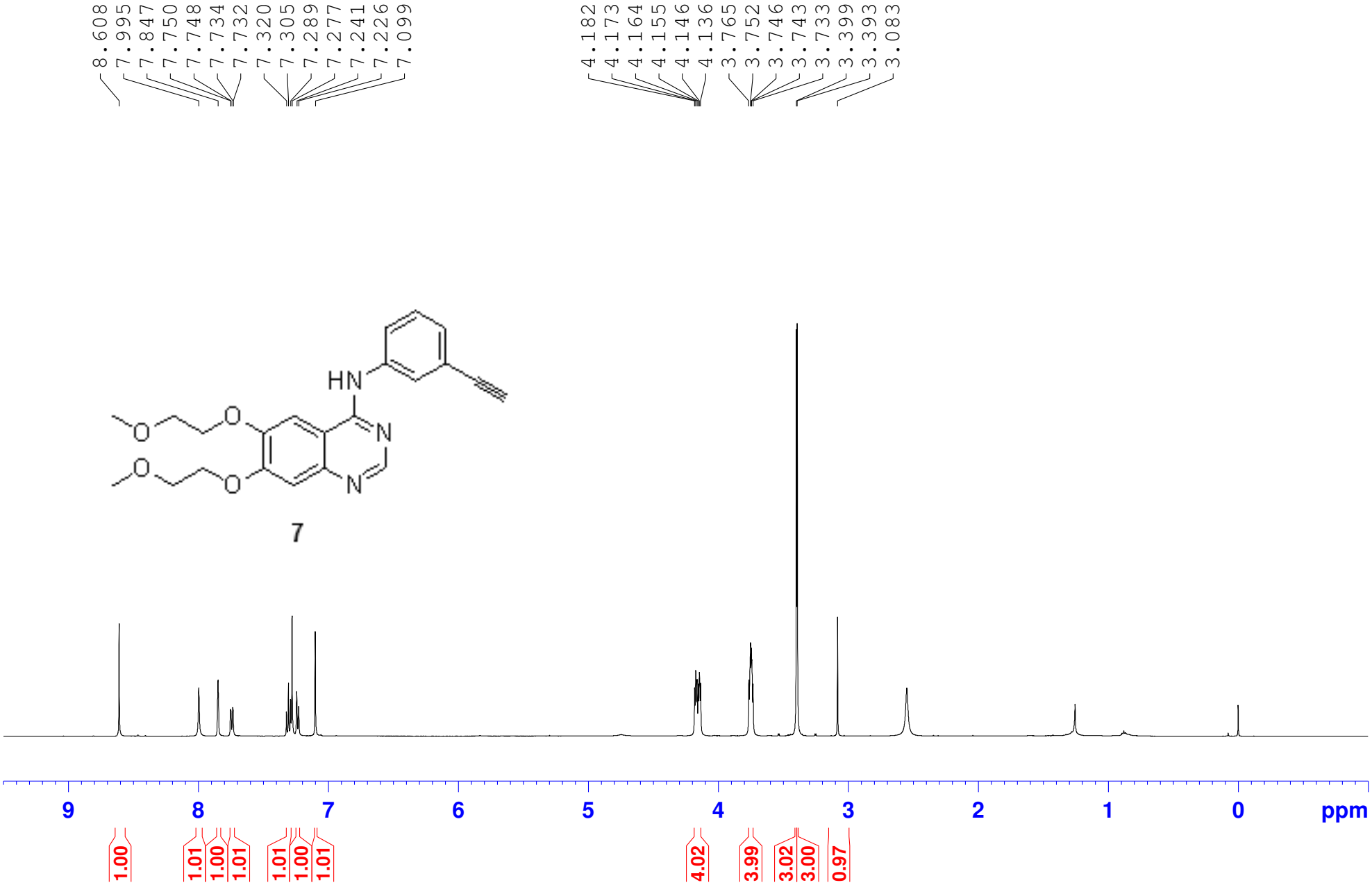


N-(3-ethynylphenyl)-6,7-bis(2-methoxyethoxy)quinazolin-4-amine
Proton CDCl3



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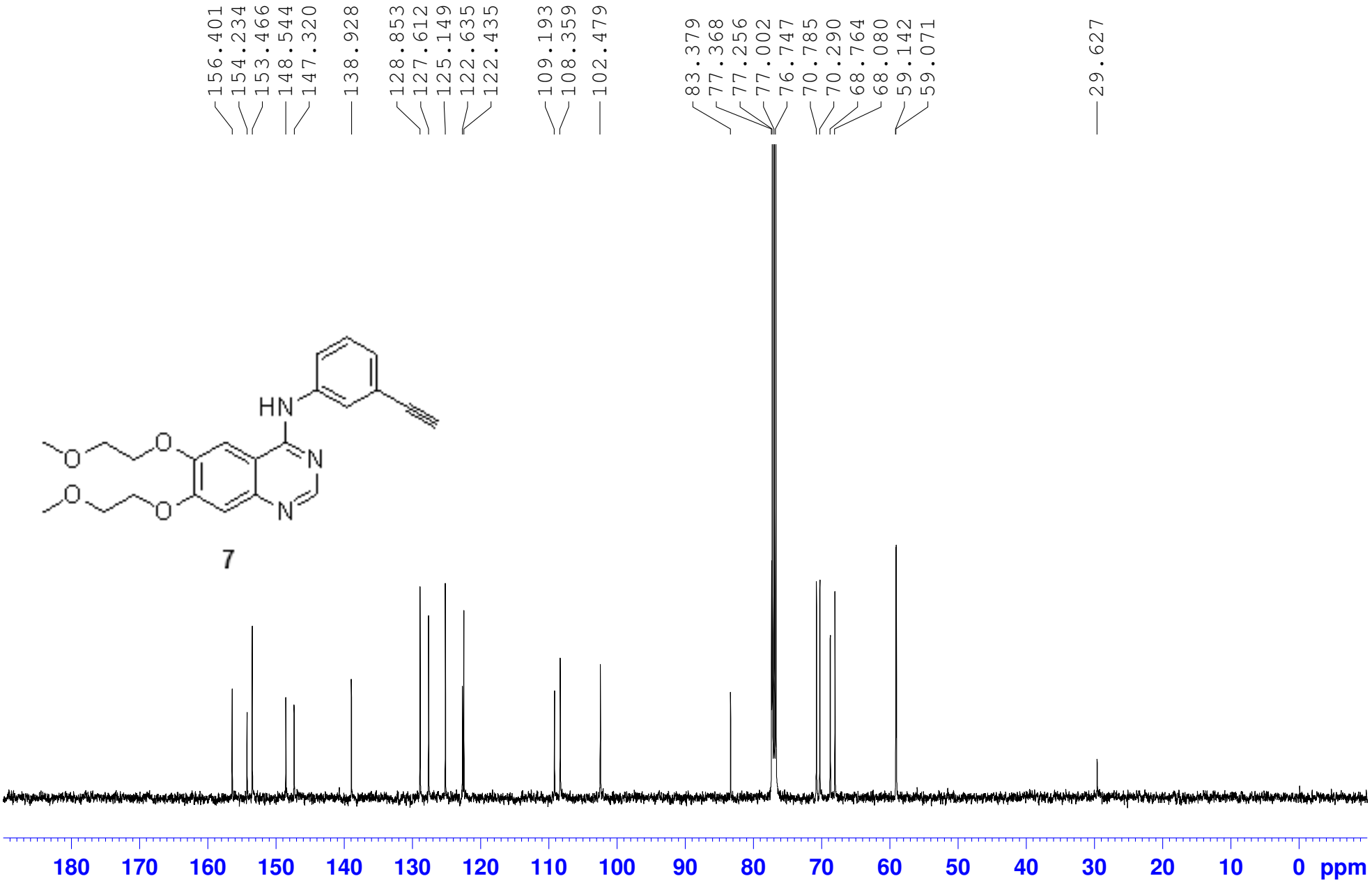
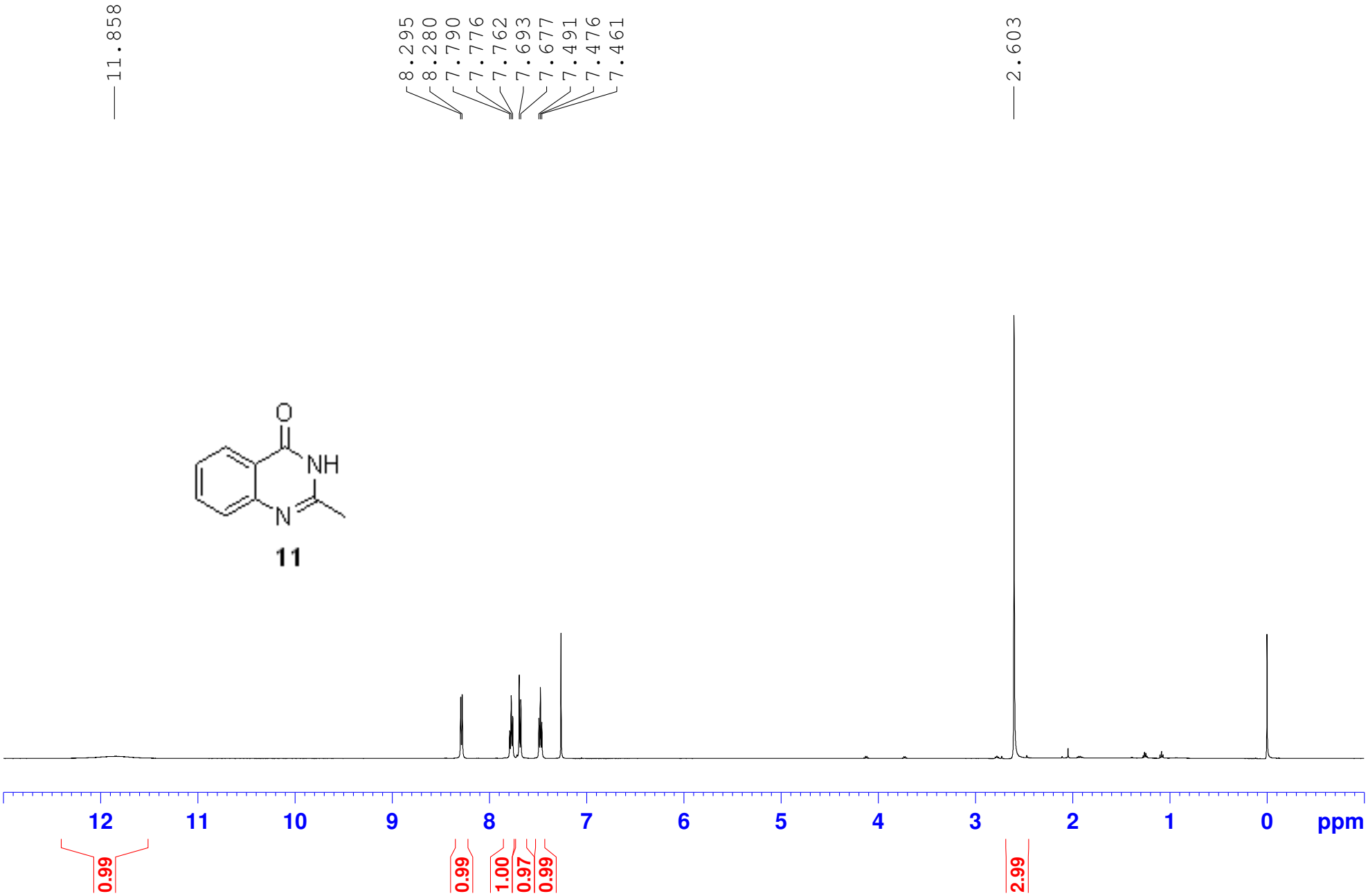
C13CPD CDCl3



2-methylquinazolin-4(3H)-one
PROTON CDCl3



2-methylquinazolin-4(3H)-one
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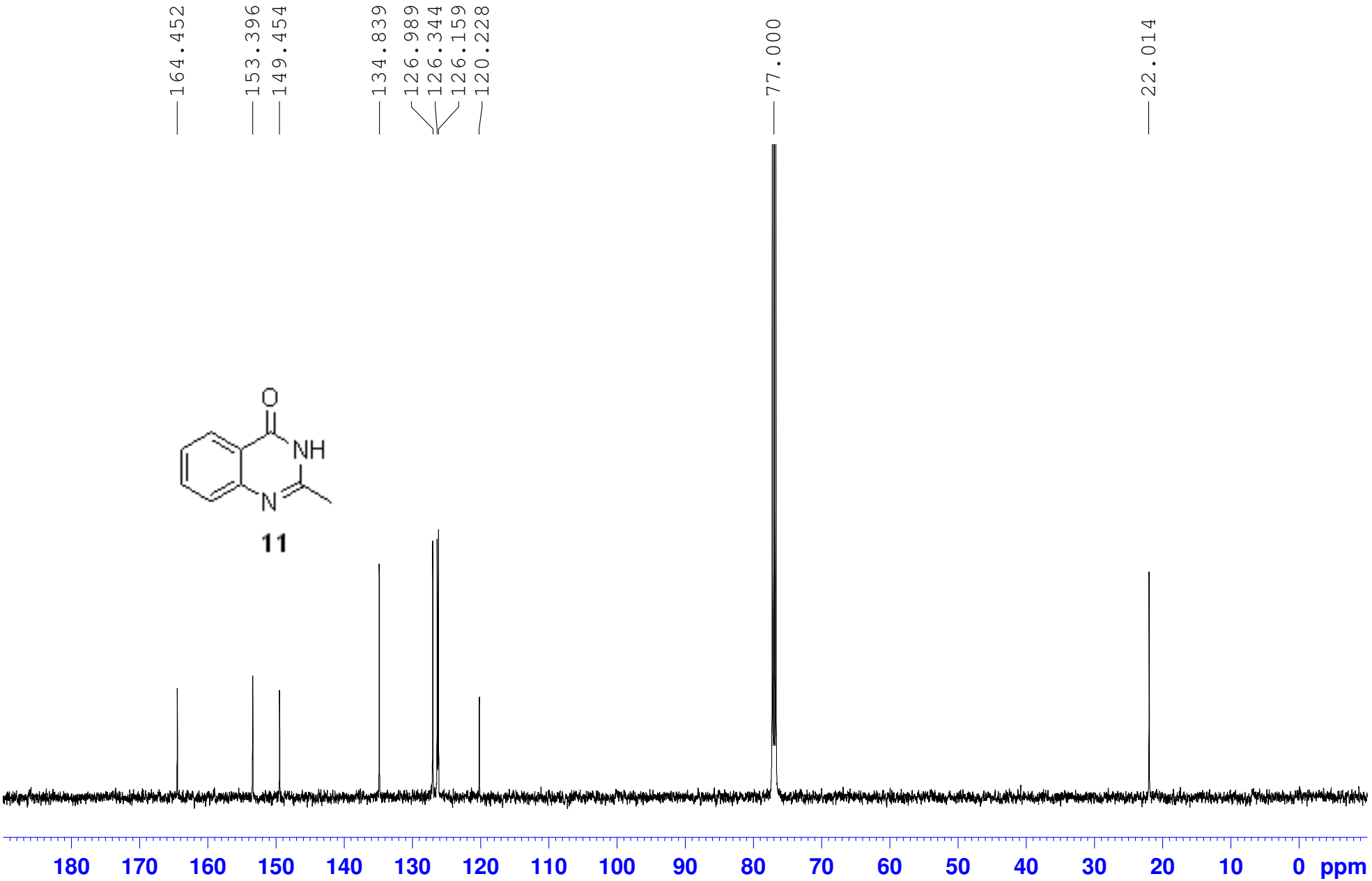

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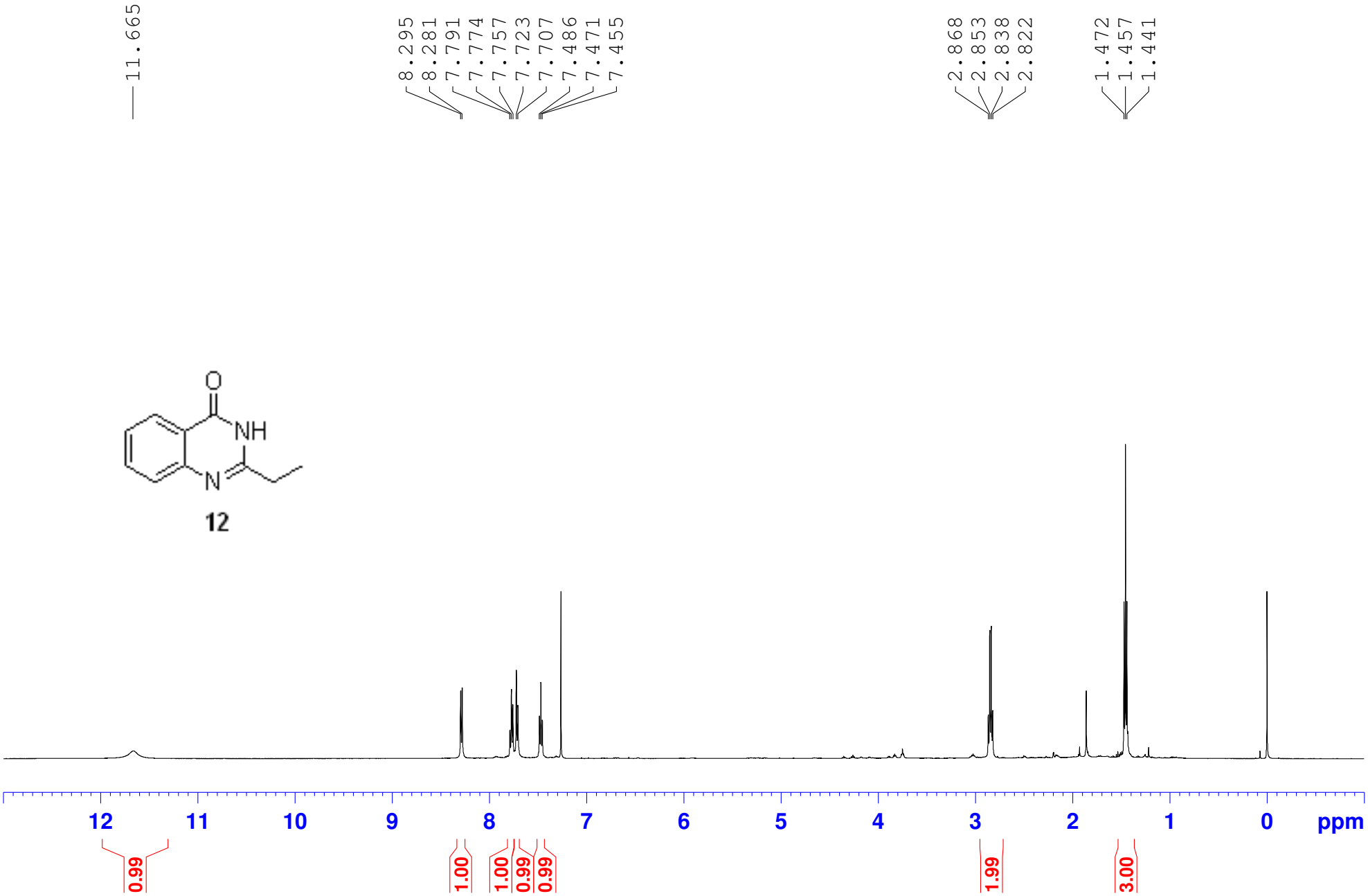

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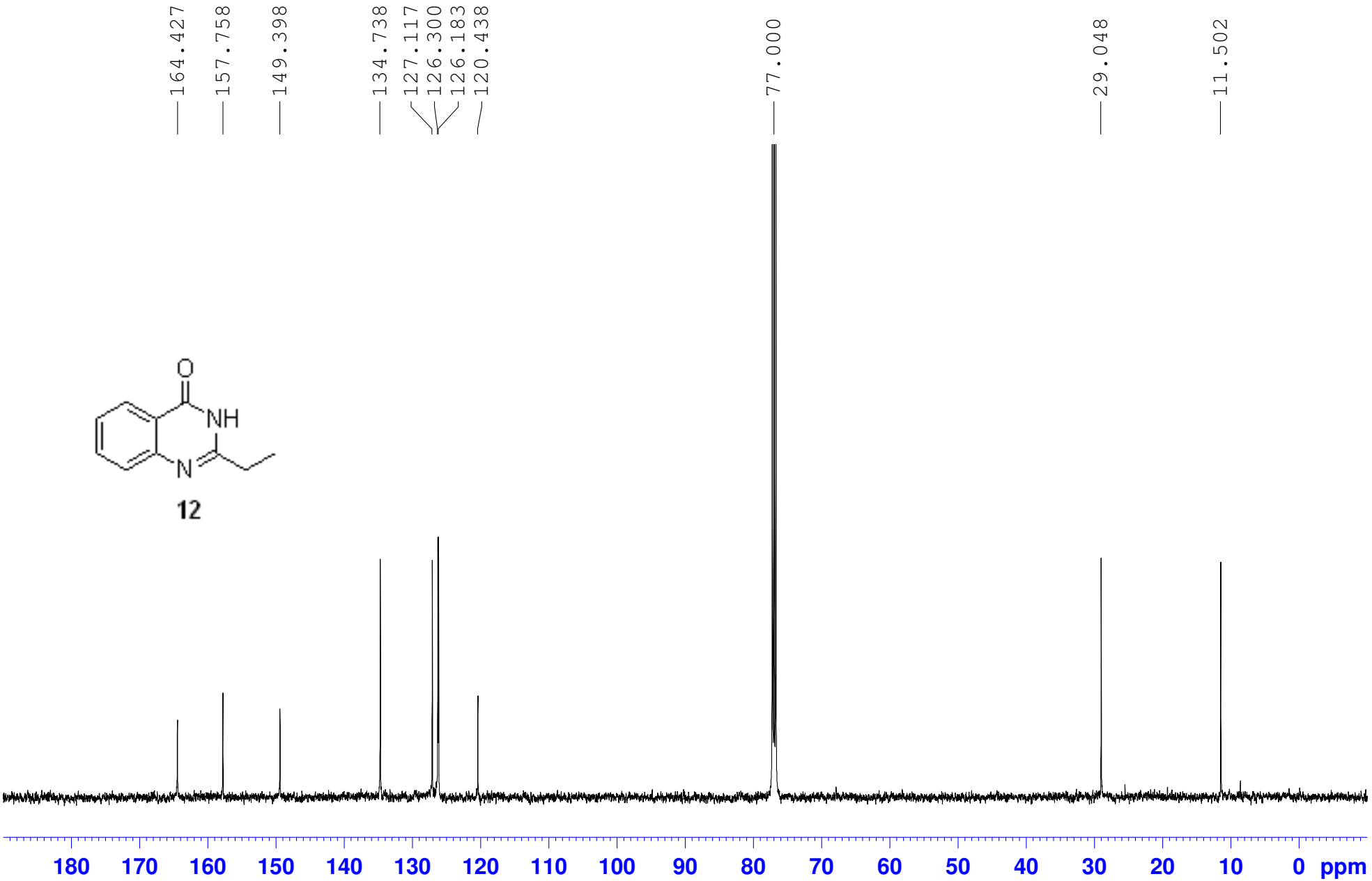
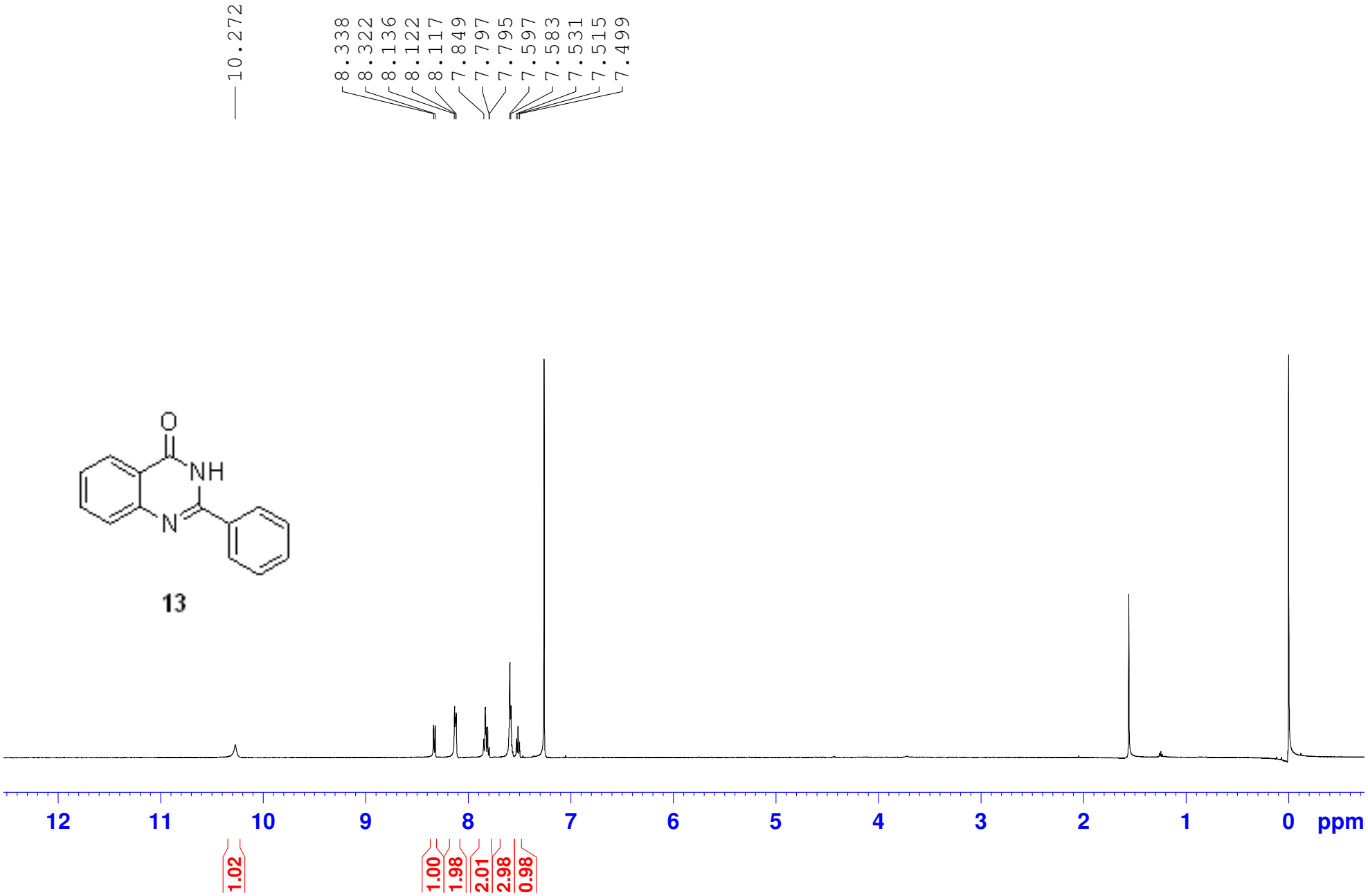
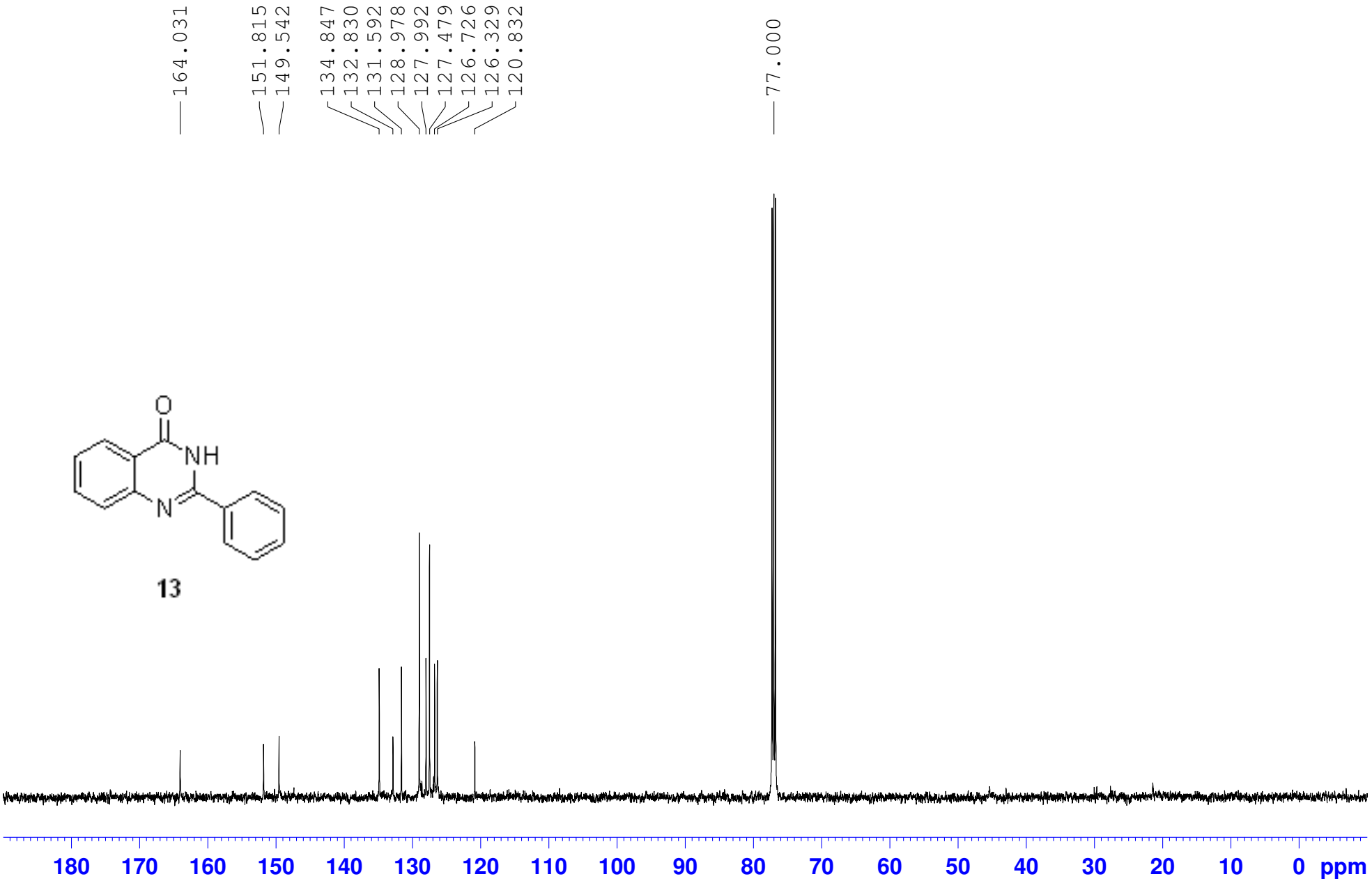


2-phenyl-4(3H)-quinazolinone
PROTON CDCl3



2-phenyl-4(3H)-quinazolinone
C13CPD CDCl3