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Supporting Information for Publication

Leveraging cascade alkynyl Prins cyclization towards the stereoselective synthesis of spiro-furan quinazolinone scaffolds

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Experimental section:

General information:

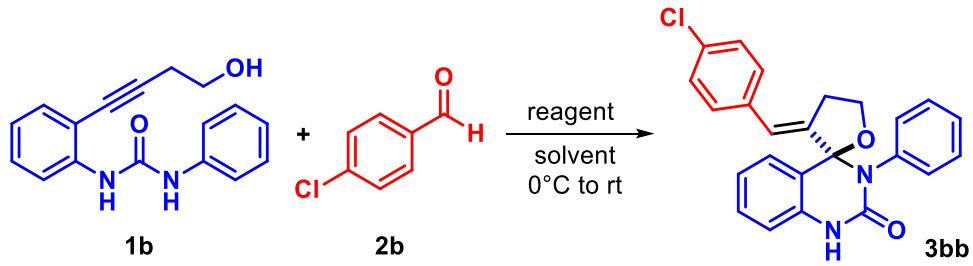
All the reagents were of reagent grade (AR grade) and were used as purchased without further purification. Silica gel (60-120 mesh size) was used for column chromatography. Reactions were monitored by TLC on silica gel GF254 (0.25 mm). Melting points were recorded in an open capillary tube and are uncorrected. Fourier transform-infra red (FT-IR) spectra were recorded as neat liquid or KBr pellets. NMR spectra were recorded in $CDCl_3$ with tetramethylsilane as the internal standard for ^{1}H (500 MHz and 400 MHz) or ^{13}C (125 MHz and 100 MHz) NMR. Chemical shifts (δ) are reported in ppm with abbreviations, s = singlet, d = doublet, dd = doublet of doublets, dt = doublet of triplets, t = triplet, td = triplet of doublets, tt = triplet of triplets, q = quartet, qd = quartet of doublets, p = quintet, m = multiplet and spin-spin coupling constants (*J*) are given in Hz. HRMS spectra were recorded using Q-TOF mass spectrometer.

Trifluoromethanesulfonic acid (reagent grade, 100 mL bottle) was purchased from Spectrochem and used without further purification. It is highly advisable to handle trifluoromethanesulfonic acid using glass(micro) syringes and steel needles. Disposable plastic syringes and needles should be avoided, as they may dissolve/melt in the presence of TfOH. After using triflic acid, two layers of PTFE tape were applied, followed by a layer of parafilm, to seal the bottle cap, and it was stored in the freezer at -5°C.

To synthesize the starting materials **1j** and **1k**, homopropargyl alcohol derivative (5-hexyn-3-ol) used was directly purchased from Sigma-Aldrich. The starting material, 4-(2-aminophenyl)but-3-yn-1-ol (**4**) was synthesized according to a literature report, and the spectroscopic data of the compound is in good agreement with the literature data.[1]

Optimization studies: To synchronize our assumption, a preliminary reaction was carried out between 1-(2-(4-hydroxybut-1-yn-1-yl)phenyl)-3-phenylurea (**1b**) and *p*-chlorobenzaldehyde (**2b**) in the presence of $BF_3{\cdot}OEt_2$ in DCM at 0 °C to room temperature (Table S3, entry 1). Gratifyingly, a new product was isolated in 86% yield in an *E*:*Z* ratio of 7:3. Spectroscopic evidences (^1H and ^{13}C NMR) and HRMS values revealed the structure of the product to be 3-(4-chlorobenzylidene)-3'-phenyl-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (**3bb**). However, the exact structure of the product was fully ascertained by single-crystal

Table S3 Optimization of the reaction



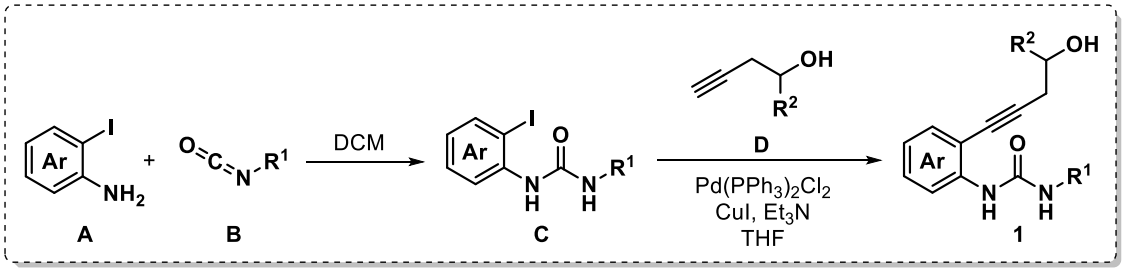
entry	reagent	equivalent	solvent	time (h)	yield (%)[b]	*E*/*Z* ratio[d]
01	$BF_3{\cdot}OEt_2$	2.0	DCM	2.0	86	7:3
02	TMSOTf	2.0	DCM	3.0	80	100:0
03	$FeCl_3$	2.0	DCM	24	33	9:1
04	$InCl_3$	0.3	DCM	24	20	100:0
05	$Bi(OTf)_3$	0.3	DCM	24	32	100:0
06	$In(OTf)_3$	0.3	DCM	24	24	100:0
07	$Cu(OTf)_2$	0.3	DCM	24	18	100:0
08	*p*-TSOH	2.0	DCM	24	–[c]	–
09	CSA	2.0	DCM	24	–[c]	–
10	TfOH	2.0	DCM	0.5	92	100:0
11	TfOH	1.5	DCM	4.0	89	100:0
12	TfOH	3.0	DCM	0.5	90	100:0
13	TfOH	2.0	DCE	0.5	88	100:0
14	TfOH	2.0	toluene	8.0	29	100:0
15	TfOH	2.0	CH_3CN	2.0	43	100:0

[a]Reaction conditions: **1b** (57mg, 0.2 mmol), **2b** (34 mg, 0.24 mmol), solvent (2 mL), 0 °C to rt, N_2 atmosphere, [b]Isolated yields, [c]Not detected, [d]*E*/*Z* ratio was analyzed by crude ^1H NMR analysis.

X-ray diffraction of one of its derivatives **3ai**. Albeit the reaction endowed the desired spiro-furan quinazolinone product **3bb** within 2 h in a good yield of 86%, it gave a mixture of geometrical isomers in the ratio *E*:*Z* = 7:3. Hence, further optimization of the reaction parameters was tuned to enhance the selectivity and productivity of the reaction. Initially, we began our investigation by varying different reagents in place of $BF_3{\cdot}OEt_2$. To our delight, the reaction, when performed with 2.0 equiv. of TMSOTf, exclusively delivered (*E*)-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one within 3 h in 80% yield (Table S3, entry 2). Under the influence of metal halides like $FeCl_3$ and $InCl_3$ in DCM at 0 °C to room temperature, the reaction furnished the desired *E*-selective product in significantly low yields (Table S3, entries 3 and 4). Metal triflates, too, failed to show any remarkable effect on yield and time (Table S3, entries 5-7). We screened the reaction in the presence of different Brønsted acids as well. *p*-TsOH and CSA were found to be ineffective for the reaction (Table S3, entries 8 and 9). Nonetheless, the use of 2.0 equiv. of TfOH acid has resulted in an escalated yield of 92% of the product with *E*-selectivity, exclusively within 0.5 h of the reaction (Table S3, entry 10). Altering the stoichiometry of TfOH to 1.5 equiv. and 3.0 equiv. resulted in the *E*-selective product in 89% and 90% yield, respectively (Table S3, entries 11 and 12). Furthermore, we explored the influence of solvents on the reaction outcome. In contrast to less efficacious solvents like toluene and acetonitrile (Table S3, entries 14 and 15), DCE delivered *E*-selective product with an admirable yield of 88% (Table S3, entry 13). Finally, the use of 2.0 equiv. of TfOH in DCM at 0 °C to room temperature is found to be the optimized condition for this transformation.

General experimental procedure and characterization data of the compounds 1a–1k:

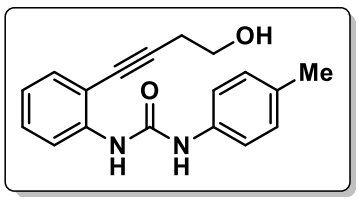
Schematic representation of starting materials **1a**–**1k**:



To a solution of substituted 2-iodoaniline **A** (2.0 mmol, 1.0 equiv.) in DCM under N_2 atmosphere, substituted phenyl isocyanate **B** (2.2 mmol, 1.1 equiv.) was added dropwise. The reaction mixture was stirred overnight at room temperature to obtain a white precipitate, which was then filtered and washed with n-hexane to obtain a substituted 1,3-diphenyl urea **C**.

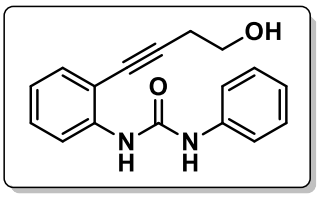
To a mixture of $Pd(PPh_3)_2Cl_2$ (0.04 mmol, 0.04 equiv.), CuI (0.02 mmol, 0.02 equiv.) and iodo substituted 1,3-diphenyl urea derivatives **C** (1.0 mmol, 1.0 equiv.) in dry THF under N_2 atmosphere, triethylamine (5.0 mmol, 5.0 equiv.) was added. The reaction mixture was stirred for 10 minutes at room temperature, after which substituted homo propargylic alcohol **D** (1.2 mmol, 1.2 equiv.) was added dropwise over a period of 5 minutes. The reaction mixture was stirred for 3.0 hours (monitored by TLC analysis) before filtering through a pad of celite. The solids were washed with ethyl acetate, and the combined filtrates were concentrated in a rotary evaporator. The crude product was then purified by column chromatography to provide the corresponding product **1**.

1-(2-(4-Hydroxybut-1-yn-1-yl)phenyl)-3-(*p*-tolyl)urea (1a):



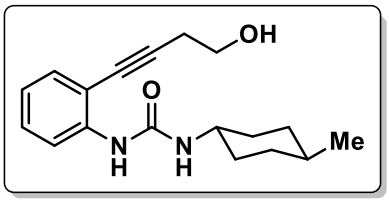
White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 160–162 °C. Yield 277 mg, 94%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.25 (dd, *J* = 8.7, 1.1 Hz, 1H), 7.98 (s, 1H), 7.58 (s, 1H), 7.25–7.22 (m, 4H), 7.03 (d, *J* = 8.0 Hz, 2H), 6.89 (td, *J* = 7.6, 1.2 Hz, 1H), 3.84 (t, *J* = 5.6 Hz, 2H), 2.98 (s, 1H), 2.61 (t, *J* = 5.7 Hz, 2H), 2.25 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) δ 153.2, 140.9, 135.9, 133.2, 130.1, 129.6, 129.3, 121.7, 120.5, 118.0, 111.6, 94.9, 78.9, 61.5, 23.9, 20.8; IR (KBr, neat) 3317, 3050, 2922, 1661, 1511, 1445, 1293, 1201, 1040, 751, 735, 507 cm^{-1}; HRMS (ESI) calcd. for $C_{18}H_{19}N_2O_2$ $(M + H)^+$ 295.1441, found 295.1440.

1-(2-(4-Hydroxybut-1-yn-1-yl)phenyl)-3-phenylurea (1b):



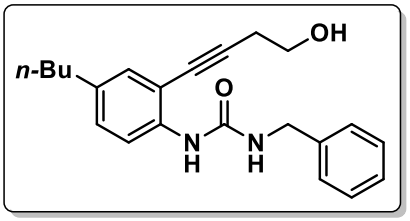
White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 136–138 °C. Yield 258 mg, 92%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.24 (dd, *J* = 8.7, 1.5 Hz, 1H), 8.07 (s, 1H), 7.83 (d, *J* = 5.6 Hz, 1H), 7.37 (d, *J* = 7.9 Hz, 2H), 7.25–7.20 (m, 4H), 6.99 (t, *J* = 7.3 Hz, 1H), 6.88 (t, *J* = 7.5 Hz, 1H), 3.89–3.84 (m, 2H), 3.22–3.13 (m, 1H), 2.63 (td, *J* = 5.7, 1.7 Hz, 2H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) δ 153.0, 140.8, 138.7, 130.0, 129.2, 129.0, 123.2, 121.7, 119.8, 117.9, 111.7, 95.2, 78.9, 61.5, 23.9; IR (KBr, neat) 3330, 3057, 2887, 1677, 1539, 1447, 1305, 1249, 1044, 752, 593 cm^{-1}; HRMS (ESI) calcd. for $C_{17}H_{17}N_2O_2$ $(M + H)^+$ 281.1285, found 281.1282.

1-(2-(4-Hydroxybut-1-yn-1-yl)phenyl)-3-(4-methylcyclohexyl)urea (1c):



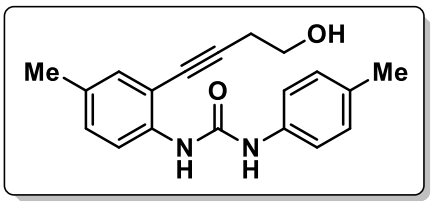
White solid; R_f (Hexane:EtOAc, 7:3) 0.5; mp 139–141 °C. Yield 280 mg, 93%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.13 (d, *J* = 8.3 Hz, 1H), 7.78 (s, 1H), 7.26–7.19 (m, 2H), 6.86 (t, *J* = 7.5 Hz, 1H), 5.65 (s, 1H), 3.86 (q, *J* = 5.6 Hz, 2H), 3.72 (s, 1H), 3.55–3.45 (m, 1H), 2.68 (t, *J* = 5.7 Hz, 2H), 1.93 (d, *J* = 10.4 Hz, 2H), 1.60 (d, *J* = 12.3 Hz, 2H), 1.25–1.17 (m, 1H), 1.06 (qd, *J* = 12.4, 3.1 Hz, 2H), 0.95 (t, *J* = 12.0 Hz, 2H), 0.81 (d, *J* = 6.2 Hz, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 155.1, 141.4, 130.4, 129.1, 121.3, 118.1, 111.6, 94.9, 78.9, 61.3, 49.1, 33.9, 33.6, 31.9, 24.0, 22.2.; IR (KBr, neat) 3332, 2926, 2850, 1659, 1547, 1448, 1306, 1217, 1042, 752, 542 cm^{-1}; HRMS (ESI) calcd. for $C_{18}H_{25}N_2O_2$ $(M + H)^+$ 301.1911, found 301.1916.

1-Benzyl-3-(4-butyl-2-(4-hydroxybut-1-yn-1-yl)phenyl)urea (1d):



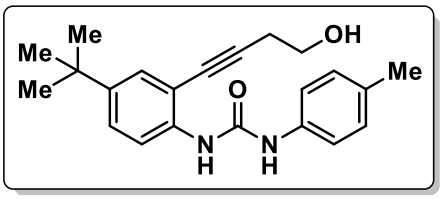
White solid; R_f (Hexane:EtOAc, 7:3) 0.5; mp 100–102 °C. Yield 316 mg, 90%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.98 (d, *J* = 8.4 Hz, 1H), 7.64 (s, 1H), 7.29–7.18 (m, 5H), 7.09 (d, *J* = 2.1 Hz, 1H), 7.04 (dd, *J* = 8.5, 2.2 Hz, 1H), 5.87 (t, *J* = 6.0 Hz, 1H), 4.36 (d, *J* = 5.7 Hz, 2H), 3.78 (q, *J* = 5.7 Hz, 2H), 3.11 (t, *J* = 6.2 Hz, 1H), 2.62 (t, *J* = 5.7 Hz, 2H), 2.48 (t, *J* = 7.7 Hz, 2H), 1.56–1.49 (m, 2H), 1.36–1.26 (m, 2H), 0.90 (t, *J* = 7.3 Hz, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 156.0, 139.2, 138.7, 136.5, 130.3, 129.3, 128.5, 127.3, 127.1, 118.8, 112.3, 94.4, 79.0, 61.2, 43.9, 34.7, 33.5, 23.9, 22.2, 13.9; IR (KBr, neat) 3329, 3032, 2956, 2931, 2860, 1661, 1550, 1522, 1420, 1301, 1050, 697 cm^{-1}; HRMS (ESI) calcd. for $C_{22}H_{27}N_2O_2$ $(M + H)^+$ 351.2067, found 351.2071.

1-(2-(4-Hydroxybut-1-yn-1-yl)-5-methylphenyl)-3-(*p*-tolyl)urea (1e):



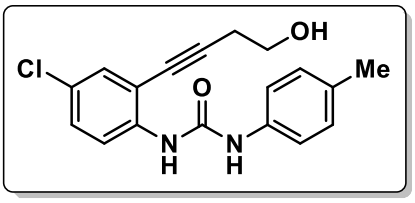
White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 97–99 °C. Yield 299 mg, 97%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.08 (d, *J* = 9.1 Hz, 1H), 7.84 (s, 1H), 7.49 (s, 1H), 7.24 (d, *J* = 8.4 Hz, 2H), 7.05–7.03 (m, 4H), 3.83 (q, *J* = 5.8 Hz, 2H), 3.01 (t, *J* = 6.6 Hz, 1H), 2.60 (t, *J* = 5.8 Hz, 2H), 2.26 (s, 3H), 2.21 (s, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 153.2, 138.4, 136.0, 133.1, 131.3, 130.6 130.0, 129.6, 120.5, 118.3, 111.8, 94.4, 78.9, 61.5, 23.9, 20.8, 20.5; IR (KBr, neat) 3317, 2921, 1660, 1509, 1291, 1200, 1040, 815, 734, 507 cm^{-1}; HRMS (ESI) calcd. for $C_{19}H_{21}N_2O_2$ $(M + H)^+$ 309.1598, found 309.1598.

1-(4-(tert-Butyl)-2-(4-hydroxybut-1-yn-1-yl)phenyl)-3-(*p*-tolyl)urea (1f):



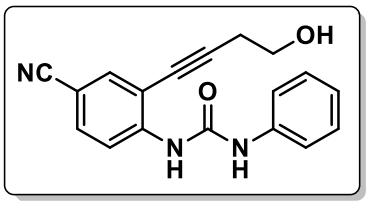
White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 117–119 °C. Yield 308 mg, 88%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.00 (d, *J* = 8.6 Hz, 1H), 7.84 (s, 1H), 7.61 (s, 1H), 7.19–7.13 (m, 4H), 6.91 (d, *J* = 7.9 Hz, 2H), 3.72 (t, *J* = 5.8 Hz, 2H), 3.41(s, 1H), 2.50 (t, *J* = 5.8 Hz, 2H), 2.14 (s, 3H), 1.16 (s, 9H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 153.5, 144.7, 138.2, 135.9, 133.0, 129.5, 127.1, 126.3, 120.4, 118.1, 111.7, 94.3, 79.2, 61.3, 34.1, 31.3, 23.9, 20.8; IR (KBr, neat) 3323, 2958, 1664, 1509, 1289, 1202, 1042, 817, 734, 507 cm^{-1}; HRMS (ESI) calcd. for $C_{22}H_{27}N_2O_2$ $(M + H)^+$ 351.2067, found 351.2075.

1-(5-Chloro-2-(4-hydroxybut-1-yn-1-yl)phenyl)-3-(*p*-tolyl)urea (1g):



White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 176–178 °C. Yield 312 mg, 95%; ^{1}H NMR (400 MHz, $CDCl_3$/DMSO-d_6) δ 8.27–8.21 (m, 3H), 7.31 (d, *J* = 8.4 Hz, 2H), 7.22–7.16 (m, 2H), 7.04 (d, *J* = 8.3 Hz, 2H), 4.84 (s, 1H), 3.88–3.83 (m, 2H), 2.69–2.65 (m, 2H), 2.26 (s, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$/DMSO-d_6) δ 152.7, 139.8, 136.3, 132.3, 129.7, 129.3, 128.9, 125.9, 119.3, 119.1, 113.0, 96.8, 77.5, 60.7, 23.9, 20.7; IR (KBr, neat) 3300, 2917, 1639, 1598, 1546, 1511, 1402, 1292, 1210, 1040, 816, 505 cm^{-1}; HRMS (ESI) calcd. for $C_{18}H_{18}ClN_2O_2$ $(M + H)^+$ 329.1051, found 329.1054.

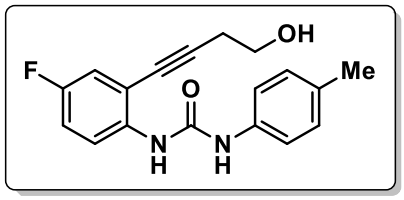
1-(5-Cyano-2-(4-hydroxybut-1-yn-1-yl)phenyl)-3-(*p*-tolyl)urea (1h):



White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 175–177 °C. Yield 259 mg, 85%; ^{1}H NMR (500 MHz, $CDCl_3$/DMSO-d_6) δ 8.58 (dd, *J* = 20.5, 5.5 Hz, 2H), 8.44 (t, *J* = 7.5 Hz, 1H), 7.48–7.41 (m, 4H), 7.23 (q, *J* = 7.2 Hz, 2H), 6.97 (q, *J* = 6.9 Hz, 1H),

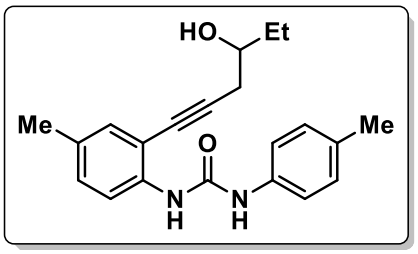
3.86 (q, *J* = 5.9 Hz, 2H), 3.72 (s, 1H), 2.68 (q, *J* = 5.8 Hz, 2H); ^{13}C NMR (125 MHz, $CDCl_3$/DMSO-d_6) δ 152.0, 145.0, 138.6, 133.5, 132.6, 128.8, 123.0, 119.1, 118.7, 117.6, 112.1, 104.0, 98.3, 60.6, 24.0; IR (KBr, neat) 3325, 2944, 2887, 2226, 1713, 1553, 1520, 1307, 1250, 1197, 1041, 751, 694 cm^{-1}; HRMS (ESI) calcd. for $C_{18}H_{16}N_3O_2$ $(M + H)^+$ 306.1237, found 306.1237.

1-(4-Fluoro-2-(4-hydroxybut-1-yn-1-yl)phenyl)-3-(*p*-tolyl)urea (1i):



White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 179–181 °C. Yield 281 mg, 90%; ^{1}H NMR (400 MHz, $CDCl_3$/DMSO-d_6) δ 8.32–8.26 (m, 1H), 8.19 (dd, *J* = 8.8, 5.2 Hz, 1H), 8.12 (s, 1H), 7.29 (d, *J* = 8.2 Hz, 2H), 7.01 (d, *J* = 8.1 Hz, 2H), 6.93–6.87 (m, 2H), 5.09 (s, 1H), 3.81 (q, *J* = 5.8 Hz, 2H), 2.64 (t, *J* = 5.5, 2H), 2.22 (s, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$/DMSO-d_6) δ 156.9 (d, *J* = 239.3), 152.8, 137.5, 136.7, 132.0, 129.2, 119.44 (d, *J* = 8.2 Hz), 119.1, 116.4 (d, *J* = 3.4 Hz), 116.2, 115.6 (d, *J* = 25.3 Hz), 112.8 (d, *J* = 8.8 Hz), 96.77, 60.6, 24.0, 20.6; ^{19}F NMR (470 MHz, $CDCl_3$/C_6F_6) δ -124.51 (s, -CF-); IR (KBr, neat) 3307, 2924, 1674, 1605, 1537, 1427, 1167, 806, 688, 507 cm^{-1}; HRMS (ESI) calcd. for $C_{18}H_{18}FN_2O_2$ $(M + H)^+$ 313.1347, found 313.1356.

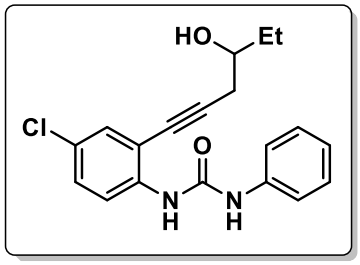
1-(2-(4-Hydroxyhex-1-yn-1-yl)-5-methylphenyl)-3-(*p*-tolyl)urea (1j):



White solid; R_f (Hexane:EtOAc, 7:3) 0.50; mp 134–136 °C. Yield 303 mg, 90%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.01 (d, *J* = 8.3 Hz, 1H), 7.90 (s, 1H), 7.62 (s, 1H), 7.14 (d, *J* = 7.8 Hz, 2H), 6.90 (d, *J* = 6.7 Hz, 4H), 3.67 (s, 1H), 2.84 (s, 1H), 2.52 (dd, *J* = 17.1, 4.0 Hz, 1H), 2.33 (dd, *J* = 17.0, 6.7 Hz, 1H), 2.13 (s, 3H), 2.08 (s, 3H), 1.57–1.43 (m, 2H), 0.85 (t, *J* = 7.4 Hz, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 153.1, 138.7, 136.3, 132.4, 130.9, 130.1, 129.8, 129.5, 119.6, 117.7, 111.6, 94.3, 79.3, 72.7, 29.3, 27.9, 20.7, 20.5, 10.3; IR (KBr, neat) 3316, 2969, 2923, 1662, 1509, 1292, 1198, 816, 508 cm^{-1}; HRMS (ESI) calcd. for $C_{21}H_{25}N_2O_2$ $(M + H)^+$ 337.1911, found 337.1912.

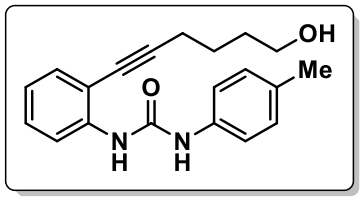
1-(5-Chloro-2-(4-hydroxyhex-1-yn-1-yl)phenyl)-3-phenylurea (1k):

White solid; R_f (Hexane:EtOAc, 7:3) 0.50; mp 163–165 °C. Yield 329 mg, 96%; ^{1}H NMR (500 MHz, $CDCl_3$/DMSO-d_6) δ 8.53 (s, 1H), 8.41 (s, 1H), 8.24 (dd, *J* = 8.8, 3.5 Hz, 1H), 7.44–7.42 (m, 2H), 7.23–7.19 (m, 2H), 7.18–7.13 (m, 2H), 6.95–6.92 (m, 1H), 5.19–5.17 (m, 1H), 3.84–



3.78 (m, 1H), 2.72 (dt, *J* = 17.1, 3.9 Hz, 1H), 2.54–2.48 (m, 1H), 1.71–1.63 (m, 1H), 1.62–1.53 (m, 1H), 0.95 (td, *J* = 7.1, 3.5 Hz, 3H), $^{13}C\{^{1}H\}$ NMR (125 MHz, $CDCl_3$/DMSO-d_6) δ 152.4, 140.0, 139.2, 129.2, 128.75, 128.73, 125.7, 122.4, 118.7, 118.6, 112.9, 96.8, 77.8, 71.9, 29.1, 27.8, 10.3; IR (KBr, neat) 3328, 2960, 2928, 1692, 1601, 1549, 1524, 1499, 1306, 1209, 749, 692 cm^{-1}; HRMS (ESI) calcd. for $C_{19}H_{20}ClN_2O_2$ $(M + H)^+$ 343.1208, found 343.1211.

1-(2-(6-hydroxyhex-1-yn-1-yl)phenyl)-3-(*p*-tolyl)urea (1l):

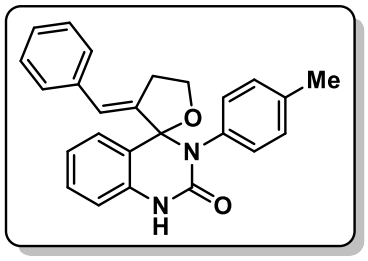


White semi-solid; R_f (Hexane:EtOAc, 3:2) 0.40. Yield 319 mg, 99%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.60–8.53 (m, 1H), 8.23 (d, *J* = 8.3 Hz, 1H), 7.76 (d, *J* = 6.7 Hz, 1H), 7.36 (d, *J* = 8.4 Hz, 2H), 7.30 (dd, *J* = 7.7, 1.5 Hz, 1H), 7.23 (t, *J* = 7.8 Hz, 1H), 7.08 (d, *J* = 8.1 Hz, 2H), 6.90 (td, *J* = 7.6, 1.2 Hz, 1H), 3.93 (s, 1H), 3.75 (t, *J* = 5.6 Hz, 2H), 2.51 (t, *J* = 6.3 Hz, 2H), 2.28 (s, 3H), 1.81–1.69 (m, 4H); $^{13}C\{^{1}H\}$ NMR (125 MHz, $CDCl_3$/DMSO-d_6) δ 153.0, 140.4, 136.4, 132.4, 131.4, 129.3, 128.6, 121.6, 120.0, 118.7, 112.1, 96.9, 61.6, 31.2, 24.8, 20.7, 19.2; IR (KBr, neat) 3330, 2934, 1665, 1533, 1513, 1446, 1297, 1249, 1208, 1055, 817, 752 cm^{-1}; HRMS (ESI) calcd. for $C_{20}H_{23}N_2O_2$ $(M + H)^+$ 323.1754, found 323.1754.

General experimental procedure and characterization data of the compounds 3aa-3aw, 3ba, 3bb, 3bi, 3bx, 3by, 3bm, 3cv, 3di, 3ei, 3er, 3fa, 3gi, 3hq, 3ii, 3jm, 3kv and 3li:

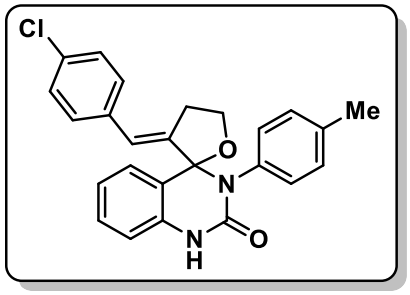
To an ice-cold suspension of alkynol substituted diphenyl urea **1** (0.4 mmol, 1.0 equiv.) and aldehyde or ketone (0.48 mmol, 1.2 equiv.) in DCM (3.0 mL), triflic acid (0.8 mmol, 2.0 equiv.) was added under N_2 atmosphere. The reaction was continued to stir at room temperature till all the starting material was consumed, as evident by TLC. It was then treated with saturated $NaHCO_3$ solution, and the organic layer was extracted with DCM (2 x 10 mL). The combined organic layers were further washed with brine, followed by drying over anhydrous Na_2SO_4. The organic phase was concentrated in a rotary evaporator to give the crude product, which was then subjected to column chromatography over silica gel to provide the final product.

(*E*)-3-Benzylidene-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3aa):



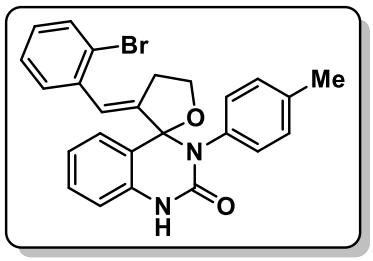
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 136–138 °C. Yield 150 mg, 98%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.77–8.56 (m, 1H), 7.36 (s, 1H), 7.25 (t, *J* = 7.6 Hz, 2H), 7.18–7.13 (m, 3H), 7.07 (d, *J* = 7.6 Hz, 2H), 7.04 (d, *J* = 7.8 Hz, 1H), 7.01–7.00 (m, 1H), 6.96 (s, 1H), 6.89 (t, *J* = 7.5 Hz, 1H), 6.71 (d, *J* = 8.0 Hz, 1H), 6.42 (t, *J* = 2.6 Hz, 1H), 3.81–3.74 (m, 2H), 2.73–2.67 (m, 1H), 2.29 (s, 3H), 2.18–2.11 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.6, 143.8, 137.5, 136.6, 135.7, 135.5, 131.5, 129.4, 129.2, 128.5, 128.47, 128.41, 127.5, 127.3, 123.1, 122.0, 114.3, 99.2, 67.1, 31.3, 21.3; IR (KBr, neat) 3212, 3055, 2980, 1671, 1601, 1402, 1264, 1038, 731, 509 cm^{-1}; HRMS (ESI) calcd. for $C_{25}H_{23}N_2O_2$ $(M + H)^+$ 383.1754, found 383.1759.

(*E*)-3-(4-Chlorobenzylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3ab):



White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 136–138 °C. Yield 165 mg, 99%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.60 (s, 1H), 7.43 (s, 1H), 7.29 (d, *J* = 8.0 Hz, 2H), 7.23 (d, *J* = 7.2 Hz, 2H), 7.11–7.06 (m, 4H), 6.97 (t, *J* = 7.6 Hz, 2H), 6.80 (d, *J* = 7.9 Hz, 1H), 6.45 (s, 1H), 3.91–3.82 (m, 2H), 2.77–2.71 (m, 1H), 2.37 (s, 3H), 2.24–2.15 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.4, 144.6, 137.6, 135.6, 135.5, 135.0, 133.3, 131.4, 129.7, 129.6, 129.2, 128.7, 127.3, 127.2, 122.9, 122.1, 114.3, 99.2, 67.1, 31.3, 21.3; IR (KBr, neat) 3237, 3060, 2919, 1675, 1602, 1493, 1403, 1040, 754, 514 cm^{-1}; HRMS (ESI) calcd. for $C_{25}H_{22}ClN_2O_2$ $(M + H)^+$ 417.1364, found 417.1360.

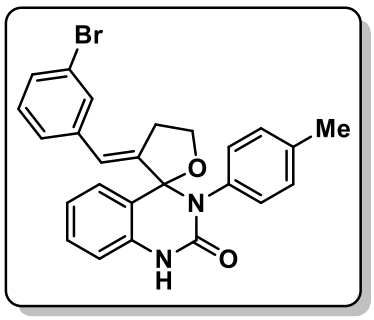
(*E*)-3-(2-Bromobenzylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3ac):



White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 114–116 °C. Yield 170 mg, 92%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.43 (s, 1H), 7.59 (dd, *J* = 8.0, 1.2 Hz, 1H), 7.51 (d, *J* = 8.0 Hz, 1H), 7.33–7.27 (m, 4H), 7.20–7.12 (m, 3H), 7.06 (t, *J* = 7.6 Hz, 1H), 6.94 (dd, *J* = 7.6, 1.7 Hz, 1H), 6.85 (d, *J* = 8.1 Hz, 1H), 6.72 (t, *J* = 2.6 Hz, 1H), 3.94–3.90 (m, 1H), 3.84–3.79 (m, 1H), 2.60–2.55 (m, 1H), 2.44 (s, 3H), 2.14–2.06 (m, 1H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) δ 152.5, 145.5, 137.7, 136.9, 135.7, 135.6, 132.7, 131.9, 131.4, 129.6, 129.5, 129.4, 129.2, 128.9, 128.1, 127.7, 127.2, 124.4, 122.6, 122.0,

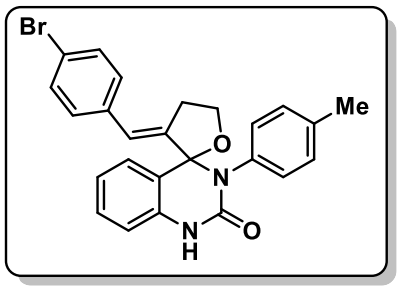
114.2, 98.5, 66.6, 31.0, 21.3; IR (KBr, neat) 3214, 3057, 2917, 1671, 1600, 1398, 1263, 1033, 731, 508 cm^{-1}; HRMS (ESI) calcd. for $C_{25}H_{22}BrN_2O_2$ $(M + H)^+$ 461.0859, found 461.0858.

(*E*)-3-(3-Bromobenzylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3ad):



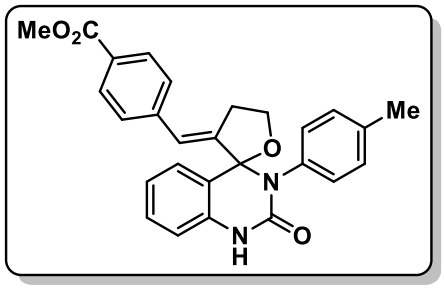
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 209–211 °C. Yield 172 mg, 93%; ^1H NMR (400 MHz, $CDCl_3$) δ 8.46 (s, 1H), 7.43 (s, 1H), 7.37 (d, *J* = 8.0 Hz, 1H), 7.29–7.23 (m, 3H), 7.19 (t, *J* = 7.8 Hz, 1H), 7.10–7.05 (m, 3H), 7.00–6.96 (m, 2H), 6.80 (d, *J* = 8.0 Hz, 1H), 6.43 (t, *J* = 2.7 Hz, 1H), 3.9–3.82 (m, 2H), 2.81–2.73 (m, 1H), 2.38 (s, 3H), 2.28–2.19 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$) δ 152.4, 145.6, 138.7, 137.7, 135.6, 135.4, 131.4, 131.2, 130.4, 130.0, 129.6, 129.2, 127.3, 127.1, 127.0, 122.9, 122.7, 122.1, 114.3, 99.2, 67.0, 31.3, 21.3; IR (KBr, neat) 3214, 3058, 2919, 1668, 1600, 1400, 1267, 1033, 751, 558, 507 cm^{-1}; HRMS (ESI) calcd. for $C_{25}H_{22}BrN_2O_2$ $(M + H)^+$ 461.0859, found 461.0857.

(*E*)-3-(4-Bromobenzylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3ae):



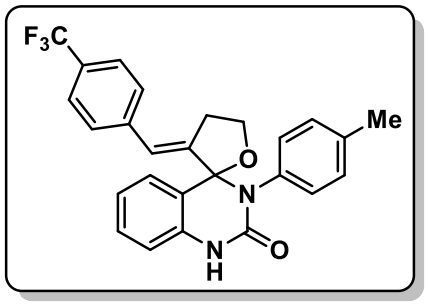
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 152–154 °C. Yield 183 mg, 99%; ^1H NMR (500 MHz, $CDCl_3$) δ 8.46 (s, 1H), 7.44 (d, *J* = 8.2 Hz, 2H), 7.42 (s, 1H), 7.26–7.21 (m, 2H), 7.09 (d, *J* = 7.8 Hz, 1H), 7.07 (s, 1H), 7.01–6.96 (m, 4H), 6.79 (d, *J* = 8.0 Hz, 1H), 6.43 (t, *J* = 2.8 Hz, 1H), 3.90–3.83 (m, 2H), 2.76–2.70 (m, 1H), 2.36 (s, 3H), 2.22–2.15 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$) δ 152.3, 144.7, 137.7, 135.6, 135.5, 135.4, 131.7, 131.4, 130.0, 129.6, 129.2, 127.3, 122.9, 122.2, 121.5, 114.3, 99.2, 67.1, 31.3, 21.3; IR (KBr, neat) 3224, 3059, 2916, 1672, 1602, 1495, 1401, 1039, 754, 512 cm^{-1}; HRMS (ESI) calcd. for $C_{25}H_{22}BrN_2O_2$ $(M + H)^+$ 461.0859, found 461.0862.

Methyl (*E*)-4-((2'-oxo-3'-(*p*-tolyl)-2',3',4,5-tetrahydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-3-ylidene)methyl)benzoate (3af):



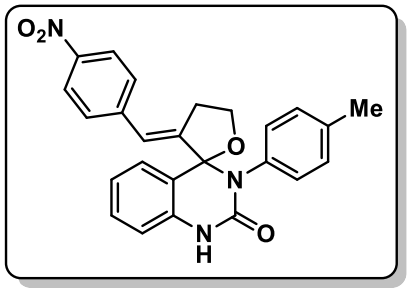
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 248–250 °C. Yield 155 mg, 88%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.52 (s, 1H), 7.91 (d, *J* = 8.0 Hz, 2H), 7.36 (s, 1H), 7.19–7.16 (m, 2H), 7.13 (d, *J* = 8.2 Hz, 2H), 7.03 (d, *J* = 7.9 Hz, 1H), 7.00 (s, 1H), 6.93 (s, 1H), 6.90 (t, *J* = 7.6 Hz, 1H), 6.73 (d, *J* = 8.0 Hz, 1H), 6.46 (t, *J* = 2.7 Hz, 1H), 3.83 (s, 3H), 3.82–3.76 (m, 2H), 2.75–2.69 (m, 1H), 2.29 (s, 3H), 2.22–2.15 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 166.7, 152.5, 146.6, 141.0, 137.7, 135.6, 135.5, 131.4, 129.7, 129.6, 129.2, 128.9, 128.3, 127.5, 127.2, 122.8, 122.1, 114.4, 99.2, 67.0, 52.2, 31.5, 21.2; IR (KBr, neat) 3224, 3058, 2919, 1717, 1671, 1601, 1399, 1275, 1108, 1037, 733, 510 cm^{-1}; HRMS (ESI) calcd. for $C_{27}H_{25}N_2O_4$ $(M + H)^+$ 441.1809, found 441.1808.

(*E*)-3'-(*p*-Tolyl)-3-(4-(trifluoromethyl)benzylidene)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'H)-one (3ag):



White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 246–248 °C. Yield 137 mg, 76%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.11 (s, 1H), 7.63 (d, *J* = 8.0 Hz, 2H), 7.51 (d, *J* = 7.9 Hz, 1H), 7.29 (d, *J* = 8.3 Hz, 4H), 7.14 (d, *J* = 7.6 Hz, 2H), 7.07–7.00 (m, 2H), 6.84 (d, *J* = 8.1 Hz, 1H), 6.59 (s, 1H), 3.95–3.91 (m, 2H), 2.86–2.79 (m, 1H), 2.43 (s, 3H), 2.35–2.25 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.4, 146.7, 140.0, 137.8, 135.6, 135.5, 131.4, 129.7, 129.5, 129.2, 128.6, 127.3, 127.1, 125.4 (q, *J* = 3.7 Hz), 124.1 (q, *J* = 270.4 Hz), 122.8, 122.2, 114.4, 99.2, 67.0, 31.4, 21.3; ^{19}F NMR (470 MHz, $CDCl_3$/C_6F_6) δ -65.78 (s, -CF_3-); IR (KBr, neat) 3220, 2955, 2923, 2855, 1676, 1604, 1407, 1324, 1122, 1069, 754 cm^{-1}; HRMS (ESI) calcd. for $C_{26}H_{22}F_3N_2O_2$ $(M + H)^+$ 451.1628, found 451.1631.

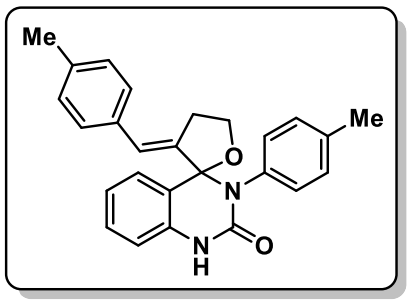
(*E*)-3-(4-Nitrobenzylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3ah):



Yellow solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 135–137 °C. Yield 111 mg, 65%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.90 (s, 1H), 8.20 (d, *J* = 8.8 Hz, 2H), 7.46 (s, 1H), 7.31–7.25 (m, 4H), 7.10 (dd, *J* = 7.9, 1.4 Hz, 2H), 7.01 (t, *J* = 7.4 Hz, 2H), 6.82 (d, *J* = 8.0 Hz, 1H), 6.59 (t, *J* = 2.7 Hz, 1H), 3.97–3.88 (m, 2H), 2.86–2.78 (m, 1H), 2.39 (s, 3H), 2.34–2.25 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.5, 148.9, 146.6, 142.9, 137.9, 135.5, 131.3, 129.7,

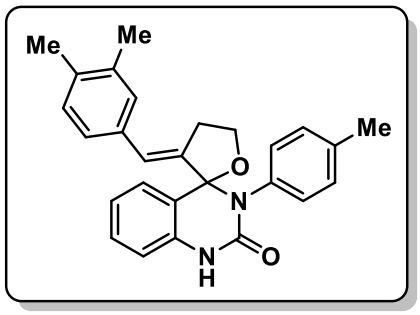
129.2, 129.0, 127.1, 126.5, 123.8, 122.5, 122.2, 114.6, 99.2, 67.0, 31.5, 21.2; IR (KBr, neat) 3238, 3062, 2922, 1672, 1599, 1513, 1342, 1040, 754, 510 cm^{-1}; HRMS (ESI) calcd. for $C_{25}H_{22}N_3O_4$ $(M + H)^+$ 428.1605, found 428.1605.

(*E*)-3-(4-Methylbenzylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3ai):



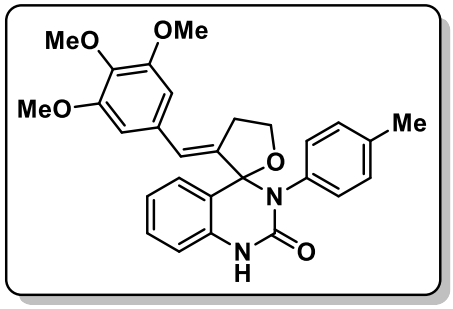
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 133–135 °C. Yield 157 mg, 99%; ^1H NMR (500 MHz, $CDCl_3$) δ 8.46–8.26 (m, 1H), 7.43 (s, 1H), 7.27–7.21 (m, 2H), 7.13 (t, *J* = 9.0 Hz, 3H), 7.08 (s, 1H), 7.06 (d, *J* = 8.0 Hz, 2H), 7.02 (s, 1H), 6.97 (td, *J* = 7.6, 1.3 Hz, 1H), 6.80 (d, *J* = 8.0 Hz, 1H), 6.47 (t, *J* = 2.7 Hz, 1H), 3.89–3.82 (m, 2H), 2.81–2.75 (m, 1H), 2.36 (s, 3H), 2.34 (s, 3H), 2.25–2.18 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$) δ 152.4, 142.7, 137.52, 137.49, 135.7, 135.4, 133.8, 131.5, 131.4, 129.4, 129.2, 128.5, 128.3, 127.4, 123.2, 122.1, 114.2, 99.3, 67.2, 31.3, 21.3, 21.2; IR (KBr, neat) 3220, 3056, 2919, 1670, 1601, 1399, 1263, 1038, 752, 734, 505 cm^{-1}; HRMS (ESI) calcd. for $C_{26}H_{25}N_2O_2$ $(M + H)^+$ 397.1911, found 397.1914.

(*E*)-3-(3,4-Dimethylbenzylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3aj):



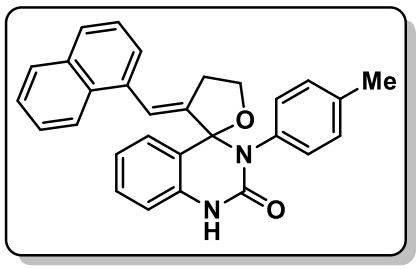
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 230–232 °C. Yield 158 mg, 96%; ^1H NMR (400 MHz, $CDCl_3$) δ 9.28 (s, 1H), 7.47 (s, 1H), 7.30–7.25 (m, 1H), 7.22 (t, *J* = 7.7 Hz, 1H), 7.13 (d, *J* = 7.7 Hz, 3H), 7.08 (s, 1H), 6.98 (s, 1H), 6.95 (t, *J* = 7.5 Hz, 2H), 6.81 (d, *J* = 8.0 Hz, 1H), 6.48 (t, *J* = 2.6 Hz, 1H), 3.92–3.83 (m, 2H), 2.86–2.78 (m, 1H), 2.40 (s, 3H), 2.28 (s, 3H), 2.26 (s, 3H), 2.24–2.20 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$) δ 152.7, 142.7, 137.3, 136.6, 136.1, 135.8, 135.5, 134.3, 131.5, 130.0, 129.7, 129.3, 129.1, 128.3, 127.3, 125.8, 123.2, 121.9, 114.4, 99.3, 67.1, 31.3, 21.2, 19.8, 19.5; IR (KBr, neat) 3210, 3055, 2923, 1669, 1602, 1399, 1264, 1039, 732, 502 cm^{-1}; HRMS (ESI) calcd. for $C_{27}H_{27}N_2O_2$ $(M + H)^+$ 411.2067, found 411.2071.

(*E*)-3'-(*p*-Tolyl)-3-(3,4,5-trimethoxybenzylidene)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3ak):



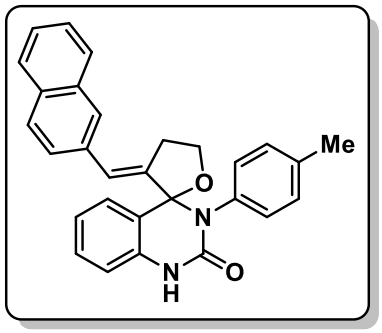
White solid; R_f (Hexane:EtOAc, 1:4) 0.50; mp 142–144 °C. Yield 148 mg, 78%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.89 (s, 1H), 7.00 (t, *J* = 7.7 Hz, 1H), 6.92–6.86 (m, 5H), 6.68 (d, *J* = 7.5 Hz, 1H), 6.65 (d, *J* = 7.5 Hz, 1H), 6.47 (d, *J* = 7.8 Hz, 1H), 6.33 (s, 1H), 6.16 (s, 1H), 3.76 (s, 6H), 3.67–3.60 (m, 2H), 3.42 (s, 3H), 2.70–2.62 (m, 1H), 2.41–2.33 (m, 1H), 2.17 (s, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 154.7, 154.3, 152.6, 149.7, 140.2, 137.5, 137.4, 137.0, 135.5, 134.5, 128.7, 128.5, 126.8, 124.8, 121.6, 118.6, 114.6, 100.6, 60.7, 60.5, 59.6, 56.0, 30.4, 21.0; IR (KBr, neat) 3223, 3057, 2932, 1663, 1599, 1406, 1109, 732, 504 cm^{-1}; HRMS (ESI) calcd. for $C_{28}H_{29}N_2O_5$ $(M + H)^+$ 473.2071, found 473.2074.

(*E*)-3-(Naphthalen-1-ylmethylene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3al):



Yellow solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 180–182 °C. Yield 135 mg, 78%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.06 (s, 1H), 7.85 (d, *J* = 8.1 Hz, 1H), 7.77 (d, *J* = 8.2 Hz, 1H), 7.56–7.51 (m, 2H), 7.48 (t, *J* = 7.5 Hz, 1H), 7.42 (q, *J* = 7.4 Hz, 2H), 7.34–7.28 (m, 3H), 7.20 (s, 1H), 7.13 (s, 1H), 7.11–7.05 (m, 3H), 6.90 (d, *J* = 8.0 Hz, 1H), 3.96 (td, *J* = 8.7, 2.7 Hz, 1H), 3.76 (q, *J* = 8.3 Hz, 1H), 2.53–2.48 (m, 1H), 2.45 (s, 3H), 2.20–2.12 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.8, 145.6, 137.6, 136.0, 135.9, 133.9, 133.4, 131.8, 131.5, 131.1, 129.6, 128.5, 128.0, 127.2, 126.5, 126.2, 126.0, 125.6, 125.2, 124.3, 123.1, 121.9, 114.7, 98.4, 66.4, 31.4, 21.3; IR (KBr, neat) 3301, 3055, 2922, 1665, 1601, 1509, 1401, 1237, 1033, 753, 507 cm^{-1}; HRMS (ESI) calcd. for $C_{29}H_{25}N_2O_2$ $(M + H)^+$ 433.1911, found 433.1909.

(*E*)-3-(Naphthalen-2-ylmethylene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quin-azolin]-2'(3'*H*)-one (3am):

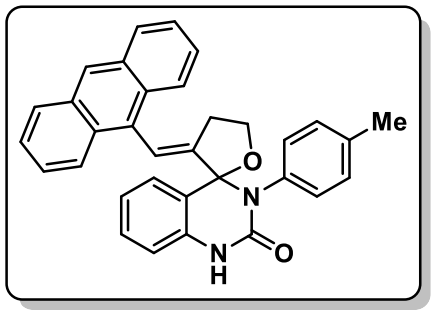


White solid; R_f (Hexane:EtOAc, 1:1) 0.60; mp 124–126 °C. Yield 166 mg, 96%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.94 (s, 1H), 7.71–7.66 (m, 3H), 7.50 (s, 1H), 7.40–7.35 (m, 3H), 7.21 (d, *J* = 8.5 Hz, 1H), 7.17–7.12 (m, 2H), 7.06 (d, *J* = 7.7 Hz, 1H), 6.99 (s, 2H), 6.89 (t, *J* = 7.5 Hz, 1H), 6.72 (d, *J* = 8.0 Hz, 1H), 6.57 (t, *J* = 2.7 Hz, 1H), 3.84–3.76 (m, 2H), 2.83–2.77 (m, 1H), 2.28 (s, 3H), 2.27–2.23 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.7, 144.3, 137.5, 135.8, 135.6, 134.1, 133.3, 132.5, 131.5, 129.4, 129.2,

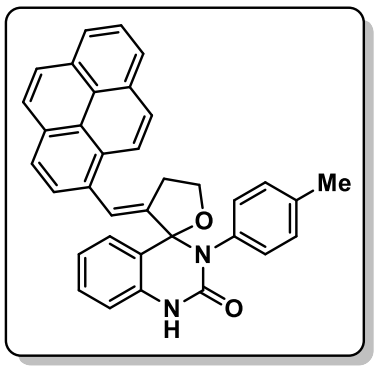
128.4, 128.1, 128.0, 127.8, 127.6, 127.3, 126.4, 126.3, 126.2, 123.1, 122.0, 114.5, 99.3, 67.1, 31.4, 21.2; IR (KBr, neat) 3217, 3055, 2921, 1670, 1600, 1400, 1264, 1038, 732, 479 cm^{-1}; HRMS (ESI) calcd. for $C_{29}H_{25}N_2O_2$ $(M + H)^+$ 433.1911, found 433.1914.

(*E*)-3-(Anthracen-9-ylmethylene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quina-zolin]-2'(3'*H*)-one (3an):

Yellow solid; R_f (Hexane:EtOAc, 1:1) 0.60; mp 290–292 °C. Yield 156 mg, 81%; ^1H NMR (400 MHz, $CDCl_3$/DMSO-d_6) δ 9.42 (s, 1H), 8.35 (s, 1H), 7.96 (s, 2H), 7.83 (s, 1H), 7.73 (s, 1H), 7.43–7.36 (m, 5H), 7.35 (s, 1H), 7.30 (s, 2H), 7.22 (s, 1H), 7.14 (td, *J* = 7.6, 1.3 Hz, 1H), 7.10 (d, *J* = 8.1 Hz, 1H), 7.04 (s, 1H), 6.48 (s, 1H), 4.05 (td, *J* = 9.0, 2.2 Hz, 1H), 3.69–3.63 (m, 1H), 2.51 (s, 3H), 2.29–2.19 (m, 1H), 1.97–1.91 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$/DMSO-d_6) δ 152.8, 146.6, 137.3, 137.1, 135.9, 131.8, 130.8, 130.5, 129.7, 129.4, 129.1, 128.4, 126.7, 126.6, 126.3, 126.1, 125.4, 124.9, 123.2, 121.2, 114.5, 97.9, 64.5, 31.9, 21.1; IR (KBr, neat) 3243, 3057, 2923, 1676, 1601, 1401, 1034, 754, 510 cm^{-1}; HRMS (ESI) calcd. for $C_{33}H_{27}N_2O_2$ $(M + H)^+$ 483.2067, found 483.2067.

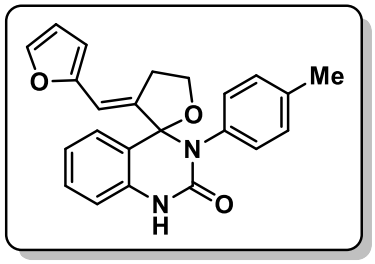


(*E*)-3-(Pyren-1-ylmethylene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazoli-n]-2'(3'*H*)-one (3ao):



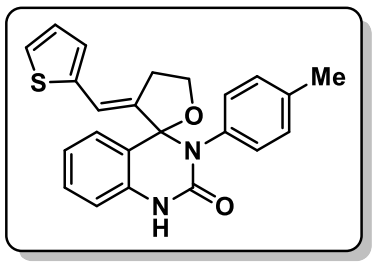
Yellow solid; R_f (Hexane:EtOAc, 1:1) 0.40; mp 268–270 °C. Yield 174 mg, 86%; ^1H NMR (400 MHz, $CDCl_3$) δ 8.91 (s, 1H), 8.15 (t, *J* = 8.3 Hz, 2H), 8.07–7.97 (m, 4H), 7.94 (d, *J* = 9.2 Hz, 1H), 7.69 (d, *J* = 9.3 Hz, 1H), 7.61 (s, 1H), 7.57 (d, *J* = 8.0 Hz, 1H), 7.40–7.35 (m, 4H), 7.25 (s, 2H), 7.15 (td, *J* = 7.6, 1.2 Hz, 1H), 6.94 (d, *J* = 7.9 Hz, 1H), 3.97 (td, *J* = 8.7, 3.1 Hz, 1H), 3.81 (td, *J* = 8.9, 6.9 Hz, 1H), 2.59–2.52 (m, 1H), 2.49 (s, 3H), 2.33–2.24 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$) δ 152.3, 146.0, 137.7, 136.1, 136.0, 131.9, 131.3, 131.2, 130.8, 130.7, 129.7, 129.4, 128.8, 127.7, 127.6, 127.30, 127.27, 127.0, 126.1, 125.7, 125.4, 125.2, 124.7, 124.4, 123.6, 123.3, 122.0, 114.7, 98.7, 66.5, 31.5, 21.4; IR (KBr, neat) 3225, 3043, 2923, 2853, 1672, 1601, 1404, 1262, 1034, 845, 734, 509 cm^{-1}; HRMS (ESI) calcd. for $C_{35}H_{27}N_2O_2$ $(M + H)^+$ 507.2067, found 507.2067.

(*E*)-3-(Furan-2-ylmethylene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazoli-n]-2'(3'*H*)-one (3ap):



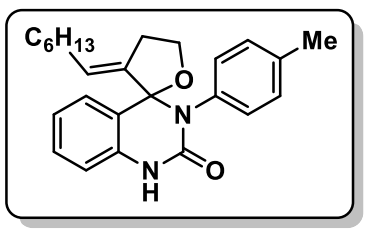
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 137–139 °C. Yield 101 mg, 68%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.90 (s, 1H), 7.40 (s, 2H), 7.20 (t, *J* = 7.5 Hz, 2H), 7.10 (d, *J* = 7.8 Hz, 2H), 7.02 (d, *J* = 7.4 Hz, 1H), 6.95 (t, *J* = 7.6 Hz, 1H), 6.76 (d, *J* = 8.0 Hz, 1H), 6.40 (dd, *J* = 3.3, 1.8 Hz, 1H), 6.32 (t, *J* = 2.7 Hz, 1H), 6.21 (d, *J* = 3.4 Hz, 1H), 3.95 (q, *J* = 7.8 Hz, 1H), 3.76 (td, *J* = 8.5, 4.9 Hz, 1H), 3.00–2.92 (m, 1H), 2.36 (s, 3H), 2.34–2.29 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.6, 152.3, 142.6, 142.1, 137.5, 135.6, 135.2, 131.2, 129.4, 127.2, 123.0, 122.0, 116.2, 114.4, 111.5, 110.2, 99.0, 67.4, 31.5, 21.2; IR (KBr, neat) 3216, 3057, 2919, 1668, 1600, 1399, 1264, 1037, 731, 508 cm^{-1}; HRMS (ESI) calcd. for $C_{23}H_{21}N_2O_3$ $(M + H)^+$ 373.1547, found 373.1548.

(*E*)-3-(Thiophen-2-ylmethylene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinaz-olin]-2'(3'*H*)-one (3aq):



White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 210–212 °C. Yield 136 mg, 87%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.77 (s, 1H), 7.41 (s, 1H), 7.33 (d, *J* = 5.1 Hz, 1H), 7.22 (t, *J* = 7.7 Hz, 2H), 7.10 (d, *J* = 7.8 Hz, 2H), 7.04 (t, *J* = 4.5 Hz, 1H), 7.00–6.94 (m, 3H), 6.78 (d, *J* = 8.0 Hz, 1H), 6.69 (s, 1H), 3.98 (q, *J* = 7.8 Hz, 1H), 3.81 (td, *J* = 8.6, 4.7 Hz, 1H), 2.90–2.82 (m, 1H), 2.36 (s, 3H), 2.27–2.18 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.5, 142.0, 140.5, 137.6, 135.6, 135.3, 131.3, 129.5, 127.9, 127.4, 127.3, 126.4, 123.0, 122.1, 121.5, 114.3, 99.3, 67.4, 31.5, 21.2; IR (KBr, neat) 3212, 3056, 2921, 1671, 1601, 1401, 1264, 1038, 732, 701, 508 cm^{-1}; HRMS (ESI) calcd. for $C_{23}H_{21}N_2O_2S$ $(M + H)^+$ 389.1318, found 389.1316.

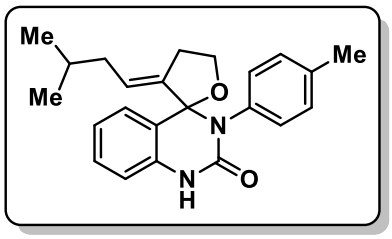
(*E*)-3-Heptylidene-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3ar):



White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 145–147 °C. Yield 108 mg, 69%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.16 (s, 1H), 7.38 (s, 1H), 7.22 (t, *J* = 7.7 Hz, 1H), 7.18 (s, 1H), 7.13 (s, 1H), 7.08 (d, *J* = 7.7 Hz, 1H), 6.99 (s, 1H), 6.95 (t, *J* = 7.6 Hz, 1H), 6.75 (d, *J* = 8.0 Hz, 1H), 5.50–5.46 (m, 1H), 3.83–3.77 (m, 2H), 2.54–2.48 (m, 1H), 2.37 (s, 3H), 2.01–1.90 (m, 3H), 1.31–1.16 (m, 8H), 0.88 (t, *J* = 7.1 Hz, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.7, 141.4, 137.3, 136.0, 135.3, 131.6, 130.8, 129.8, 129.4, 129.2, 129.0, 127.2, 123.7, 121.8, 114.0, 97.7, 66.4, 31.7, 30.2, 29.4, 29.1, 28.7,

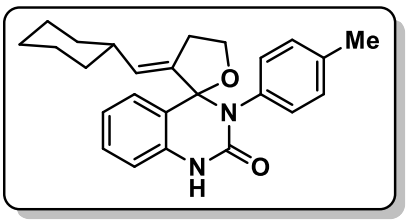
22.6, 21.2, 14.1; IR (KBr, neat) 3227, 3059, 2923, 1672, 1601, 1395, 1035, 751, 655, 508 cm^{-1}; HRMS (ESI) calcd. for $C_{25}H_{31}N_2O_2$ $(M + H)^+$ 391.2380, found 391.2380.

(*E*)-3-(3-Methylbutylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3as):



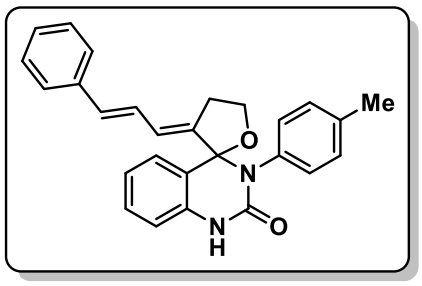
White semi-solid; R_f (Hexane:EtOAc, 3:2) 0.50. Yield 90 mg, 62%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.38 (s, 1H), 7.39 (s, 1H), 7.21 (td, *J* = 7.7, 1.4 Hz, 1H), 7.17 (s, 1H), 7.13 (s, 1H), 7.08 (d, *J* = 7.8 Hz, 1H), 7.03–6.98 (m, 1H), 6.95 (td, *J* = 7.7, 1.2 Hz, 1H), 6.76 (d, *J* = 7.9 Hz, 1H), 5.50 (tt, *J* = 7.5, 2.6 Hz, 1H), 3.84–3.77 (m, 2H), 2.54–2.49 (m, 1H), 2.36 (s, 3H), 1.99–1.91 (m, 1H), 1.85 (t, *J* = 7.2 Hz, 2H), 1.60–1.52 (m, 1H), 0.81 (d, *J* = 6.6 Hz, 3H), 0.78 (d, *J* = 6.6 Hz, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.8, 142.2, 137.3, 136.0, 135.3, 131.5, 130.7, 129.2, 128.7, 127.2, 123.7, 121.7, 114.1, 97.8, 66.3, 39.5, 29.6, 28.4, 22.6, 22.4, 21.2; IR (KBr, neat) 3224, 3057, 2918, 1717, 1671, 1601, 1398, 1275, 1108, 752, 512 cm^{-1}; HRMS (ESI) calcd. for $C_{23}H_{27}N_2O_2$ $(M + H)^+$ 363.2067, found 363.2067.

(*E*)-3-(Cyclohexylmethylene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3at):



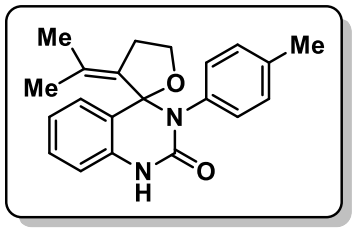
White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 104–106 °C. Yield 120 mg, 77%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.44 (s, 1H), 7.39 (d, *J* = 7.9 Hz, 1H), 7.20 (td, *J* = 7.7, 1.5 Hz, 1H), 7.17 (s, 1H), 7.12 (d, *J* = 8.0 Hz, 1H), 7.07 (d, *J* = 7.7 Hz, 1H), 7.01–6.98 (m, 1H), 6.94 (t, *J* = 7.5 Hz, 1H), 6.75 (d, *J* = 8.0 Hz, 1H), 5.31 (dt, *J* = 9.5, 2.6 Hz, 1H), 3.84 (td, *J* = 8.7, 3.9 Hz, 1H), 3.77 (q, *J* = 8.2 Hz, 1H), 2.55–2.49 (m, 1H), 2.37 (s, 3H), 2.01–1.90 (m, 2H), 1.69–1.64 (m, 2H), 1.63–1.57 (m, 2H), 1.42 (d, *J* = 12.9 Hz, 1H), 1.24–1.11 (m, 3H), 1.05–0.93 (m, 2H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.8, 139.4, 137.3, 136.0, 135.5, 135.2, 132.0, 130.8, 129.3, 129.1, 128.9, 127.2, 123.6, 121.7, 114.1, 97.7, 66.3, 39.7, 32.3, 31.6, 29.3, 25.9, 25.8, 25.7, 21.2; IR (KBr, neat) 3221, 3058, 2922, 2851, 1671, 1600, 1398, 1256, 1035, 732, 509 cm^{-1}; HRMS (ESI) calcd. for $C_{25}H_{29}N_2O_2$ $(M + H)^+$ 389.2224, found 389.2225.

(*E*)-3-((*E*)-3-Phenylallylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3au):



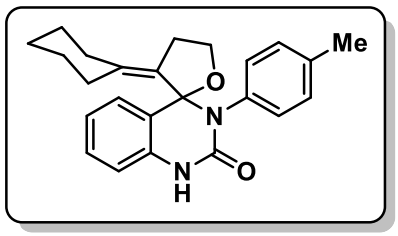
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 152–154 °C. Yield 155 mg, 95%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.60 (s, 1H), 7.33 (s, 1H), 7.30 (d, *J* = 7.6 Hz, 2H), 7.23 (t, *J* = 7.5 Hz, 2H), 7.16 (d, *J* = 7.8 Hz, 1H), 7.13 (d, *J* = 7.3 Hz, 2H), 7.05 (s, 1H), 7.01 (d, *J* = 8.0, 1H), 6.94 (s, 1H), 6.88 (t, *J* = 7.5 Hz, 1H), 6.70 (d, *J* = 8.0 Hz, 1H), 6.61 (dd, *J* = 15.6, 10.8 Hz, 1H), 6.45 (d, *J* = 15.6 Hz, 1H), 6.15 (d, *J* = 10.8 Hz, 1H), 3.83 (q, *J* = 7.9 Hz, 1H), 3.70 (td, *J* = 8.5, 4.5 Hz, 1H), 2.73–2.66 (m, 1H), 2.28 (s, 3H), 2.13–2.04 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.7, 144.4, 137.5, 136.9, 135.7, 135.3, 134.4, 131.3, 129.4, 128.7, 128.0, 127.7, 127.2, 126.5, 125.3, 123.0, 122.0, 114.3, 98.3, 66.9, 29.9, 21.2; IR (KBr, neat) 3209, 3057, 2919, 1670, 1600, 1398, 1264, 1031, 749, 509 cm^{-1}; HRMS (ESI) calcd. for $C_{27}H_{25}N_2O_2$ $(M + H)^+$ 409.1911, found 409.1911.

3-(Propan-2-ylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3av):



White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 238–240 °C. Yield 119 mg, 89%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.49 (s, 1H), 7.44 (s, 1H), 7.20 (td, *J* = 7.6, 1.4 Hz, 1H), 7.17 (s, 1H), 7.12 (s, 1H), 7.07 (d, *J* = 7.8 Hz, 1H), 6.94 (t, *J* = 7.4 Hz, 2H), 6.75 (d, *J* = 7.8 Hz, 1H), 3.86 (td, *J* = 8.7, 3.4 Hz, 1H), 3.75 (q, *J* = 8.3, 1H), 2.56–2.51 (m, 1H), 2.37 (s, 3H), 2.02–1.94 (m, 1H), 1.59 (s, 3H), 1.46 (s, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.9, 137.3, 136.2, 134.9, 134.2, 130.5, 130.0, 129.2, 129.0, 126.3, 122.7, 121.9, 114.2, 96.7, 65.7, 31.9, 22.9, 21.2, 21.1; IR (KBr, neat) 3211, 3058, 2915, 1670, 1600, 1397, 1264, 1038, 753, 734, 510 cm^{-1}; HRMS (ESI) calcd. for $C_{21}H_{23}N_2O_2$ $(M + H)^+$ 335.1754, found 335.1754.

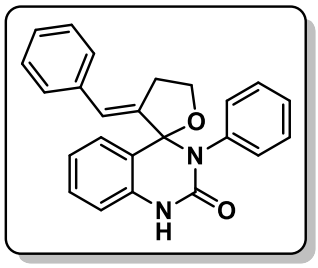
3-Cyclohexylidene-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3aw):



White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 204–206 °C. Yield 126 mg, 84%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.74 (s, 1H), 7.45 (d, *J* = 8.1 Hz, 1H), 7.18 (t, *J* = 7.8 Hz, 2H), 7.13 (d, *J* = 7.7 Hz, 1H), 7.11 (s, 1H), 6.99 (d, *J* = 8.0 Hz, 1H), 6.94 (t, *J* = 7.5 Hz, 1H), 6.74 (d, *J* = 8.0 Hz, 1H), 3.87 (td, *J* = 8.7, 3.2 Hz, 1H), 3.74 (q, *J* = 8.4 Hz, 1H), 2.61–2.56 (m, 1H), 2.37 (s, 3H), 2.15–2.10 (m, 1H), 2.07–1.98 (m, 2H), 1.86–1.79 (m, 2H), 1.59–1.54 (m, 1H), 1.50–1.46 (m, 1H), 1.43–1.33 (m,

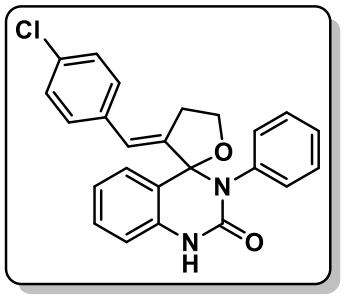
3H), 1.04–0.96 (m, 1H); $^{13}C\{^{1}H\}$ NMR (125 MHz, $CDCl_3$) δ 153.0, 137.2, 137.1, 136.2, 134.7, 131.1, 130.3, 129.9, 129.1, 129.0, 128.7, 126.4, 123.1, 121.6, 114.3, 96.3, 65.5, 33.1, 31.2, 31.0, 27.2, 26.0, 25.9, 21.2; IR (KBr, neat) 3212, 3055, 2927, 1672, 1600, 1400, 1265, 1037, 732, 512 cm^{-1}; HRMS (ESI) calcd. for $C_{24}H_{26}N_2O_2$ $(M + H)^+$ 375.2067, found 375.2067.

(*E*)-3-Benzylidene-3'-phenyl-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3ba):



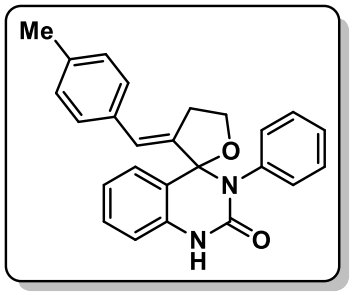
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 187–189 °C. Yield 131 mg, 89%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.09 (s, 1H), 7.59 (s, 1H), 7.46–7.41 (m, 1H), 7.38–7.29 (m, 4H), 7.27–7.21 (m, 2H), 7.19 (s, 1H), 7.13 (t, *J* = 6.4 Hz, 3H), 6.97 (t, *J* = 7.6 Hz, 1H), 6.81 (d, *J* = 8.0 Hz, 1H), 6.53 (t, *J* = 2.6 Hz, 1H), 3.86 (dd, *J* = 8.6, 5.6 Hz, 2H), 2.82–2.74 (m, 1H), 2.23–2.14 (m, 1H); $^{13}C\{^{1}H\}$ NMR (125 MHz, $CDCl_3$) δ 152.7, 143.6, 138.5, 136.6, 135.6, 132.0, 131.7, 129.5, 128.6, 128.47, 128.46, 127.8, 127.5, 127.2, 122.9, 122.0, 114.5, 99.2, 67.0, 31.3; IR (KBr, neat) 3227, 3057, 2922, 1669, 1600, 1399, 1264, 1034, 734, 696, 549 cm^{-1}; HRMS (ESI) calcd. for $C_{24}H_{21}N_2O_2$ $(M + H)^+$ 281.1285, found 281.1285.

(*E*)-3-(4-Chlorobenzylidene)-3'-phenyl-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3bb):



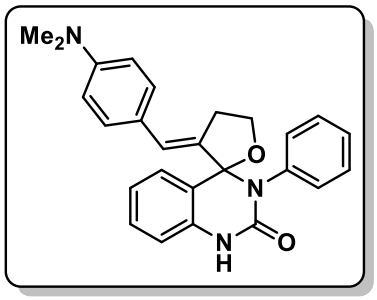
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 208–210 °C. Yield 148 mg, 92%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.03 (s, 1H), 7.58 (s, 1H), 7.44 (s, 1H), 7.35 (t, *J* = 7.2 Hz, 1H), 7.32–7.27 (m, 3H), 7.23 (td, *J* = 7.6, 1.4 Hz, 1H), 7.13 (m, 1H), 7.10 (d, *J* = 7.8 Hz, 1H), 7.05 (d, *J* = 8.5 Hz, 2H), 6.98 (d, *J* = 7.5 Hz, 1H), 6.80 (d, *J* = 8.0 Hz, 1H), 6.46 (t, *J* = 2.6 Hz, 1H), 3.86 (dd, *J* = 8.5, 5.6 Hz, 2H), 2.76–2.69 (m, 1H), 2.19–2.10 (m, 1H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) δ 152.5, 144.4, 138.4, 135.5, 134.5, 133.4, 131.8, 129.7, 129.6, 128.7, 128.5, 127.9, 127.4, 127.2, 122.8, 122.1, 114.5, 99.2, 67.0, 31.3; IR (KBr, neat) 3206, 3052, 2919, 1673, 1599, 1395, 1032, 756, 702, 555 cm^{-1}; HRMS (ESI) calcd. for $C_{24}H_{20}ClN_2O_2$ $(M + H)^+$ 403.1208, found 403.1213.

(*E*)-3-(4-Methylbenzylidene)-3'-phenyl-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3bi):



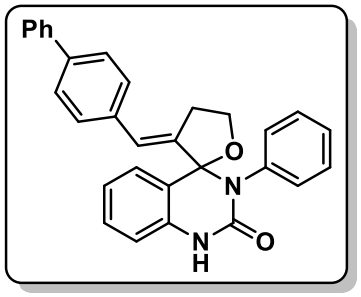
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 190–192 °C. Yield 136 mg, 89%; ^{1}H NMR (500 MHz, $CDCl_3$) 9.58 (s, 1H), 7.64 (s, 1H), 7.48 (s, 1H), 7.38 (t, *J* = 7.2 Hz, 1H), 7.33 (s, 1H), 7.21 (t, *J* = 7.7 Hz, 2H), 7.17 (d, *J* = 7.8 Hz, 2H), 7.14 (d, *J* = 7.7 Hz, 1H), 7.08 (d, *J* = 7.8 Hz, 2H), 6.98 (t, *J* = 7.6 Hz, 1H), 6.81 (d, *J* = 8.0 Hz, 1H), 6.52 (t, *J* = 2.6 Hz, 1H), 3.88 (dd, *J* = 8.9, 5.6 Hz, 2H), 2.82–2.76 (m, 1H), 2.37 (s, 3H), 2.23–2.16 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.8, 142.6, 138.5, 137.4, 135.6, 133.7, 131.9, 131.7, 129.3, 129.1, 128.5, 128.4, 127.6, 127.1, 122.9, 121.8, 114.6, 99.2, 66.9, 31.2, 21.1; IR (KBr, neat) 3227, 3057, 2920, 1669, 1601, 1398, 1265, 1038, 753, 704, 551 cm^{-1}; HRMS (ESI) calcd. for $C_{25}H_{23}N_2O_2$ $(M + H)^+$ 383.1754, found 383.1756.

(*E*)-3-(4-(Dimethylamino)benzylidene)-3'-phenyl-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3bx):



Yellow solid; R_f (Hexane:EtOAc, 1:1) 0.40; mp 212–214°C. Yield 112 mg, 68%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.62 (s, 1H), 7.57 (s, 1H), 7.42 (s, 1H), 7.33 (t, *J* = 7.2 Hz, 1H), 7.28 (s, 1H), 7.23 (t, *J* =7.8 Hz, 1H), 7.17 (s, 1H), 7.14 (d, *J* = 7.7 Hz, 1H), 7.06 (d, *J* = 8.4 Hz, 2H), 6.96 (t, *J* = 7.6 Hz, 1H), 6.79 (d, *J* = 8.0 Hz, 1H), 6.68 (d, *J* = 8.4 Hz, 2H), 6.40 (t, *J* = 2.6 Hz, 1H), 3.89–3.80 (m, 2H), 2.97 (s, 6H), 2.81–2.74 (m, 1H), 2.19–2.10 (m, 1H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) δ 152.5, 149.6, 138.8, 138.6, 135.5, 132.0, 131.7, 129.7, 129.3, 128.5, 127.6, 127.5, 123.4, 122.0, 114.2, 112.1, 99.5, 67.2, 40.4, 31.3; IR (KBr, neat) 3214, 3055, 2906, 1669, 1602, 1400, 1164, 1036, 732, 701, 550 cm^{-1}; HRMS (ESI) calcd. for $C_{26}H_{26}N_3O_2$ $(M + H)^+$ 412.2020, found 412.2025.

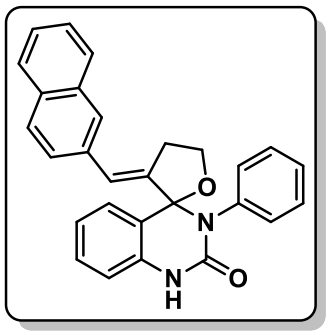
(*E*)-3-([1,1'-Biphenyl]-4-ylmethylene)-3'-phenyl-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3by):



White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 239–241 °C. Yield 160 mg, 90%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.92 (s, 1H), 7.52 (s, 1H), 7.48 (t, *J* = 9.1 Hz, 4H), 7.35 (t, *J* = 7.6 Hz, 3H), 7.27–7.22 (m, 3H), 7.14 (t, *J* = 8.8 Hz, 3H), 7.10 (s, 1H), 7.04 (d, *J* = 7.8 Hz, 1H), 6.89 (t, *J* = 7.5 Hz, 1H), 6.72 (d, *J* = 8.0 Hz, 1H), 6.46 (s, 1H), 3.79 (t, *J* = 6.9 Hz, 2H), 2.76–2.70 (m, 1H), 2.17–2.10 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.6, 143.7, 140.4, 140.3, 138.5,

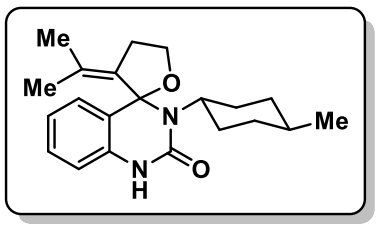
135.5, 131.7, 129.5, 129.0, 128.9, 128.5, 128.2, 127.8, 127.5, 127.3, 127.2, 127.0, 123.0, 122.1, 114.5, 99.3, 67.0, 31.4; IR (KBr, neat) 3220, 3057, 2922, 1670, 1600, 1401, 1263, 1036, 734, 698, 551cm^{-1}; HRMS (ESI) calcd. for $C_{30}H_{25}N_2O_2$ $(M + H)^+$ 445.1911, found 445.1908.

(*E*)-3-(Naphthalen-2-ylmethylene)-3'-phenyl-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3bm):



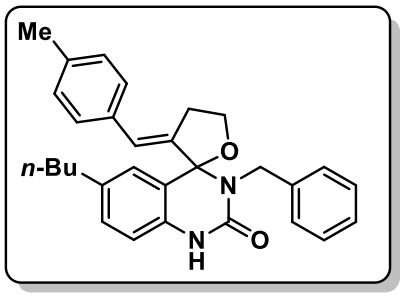
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 134–136 °C. Yield 144 mg, 86%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.14 (s, 1H), 7.81 (d, *J* = 5.1 Hz, 1H), 7.80–7.77 (m, 2H), 7.63 (s, 1H), 7.59 (s, 1H), 7.49–7.46 (m, 3H), 7.37 (t, *J* = 7.2 Hz, 1H), 7.32 (s, 1H), 7.30 (d, *J* = 8.5 Hz, 1H), 7.25 (t, *J* = 7.6 Hz, 1H), 7.23 (s, 1H), 7.18 (d, *J* = 7.8 Hz, 1H), 7.00 (t, *J* = 7.6 Hz, 1H), 6.84 (d, *J* = 8.0 Hz, 1H), 6.69 (t, *J* = 2.6 Hz, 1H), 3.94–3.87 (m, 2H), 2.92–2.86 (m, 1H), 2.36–2.28 (m, 1H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) δ 152.7, 144.1, 138.5, 135.6, 134.1, 133.2, 132.5, 131.8, 129.5, 128.6, 128.1, 128.0, 127.8, 127.7, 127.6, 127.2, 126.4, 126.3, 126.1, 123.0, 122.0, 114.6, 99.3, 67.0, 31.4.; IR (KBr, neat) 3218, 3056, 2903, 1669, 1600, 1399, 1264, 1037, 731, 702, 550 cm^{-1}; HRMS (ESI) calcd. for $C_{28}H_{23}N_2O_2$ $(M + H)^+$ 419.1754, found 419.1757.

3'-(4-Methylcyclohexyl)-3-(propan-2-ylidene)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3cv):



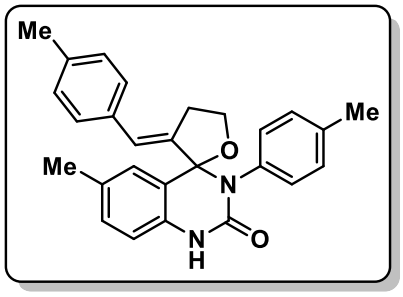
White solid; R_f (Hexane:EtOAc, 4:1) 0.5; mp 224–226 °C. Yield 112 mg, 82%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.81–8.44 (m, 1H), 7.17 (td, *J* = 7.7, 1.5 Hz, 1H), 6.98 (d, *J* = 7.8 Hz, 1H), 6.87 (t, *J* = 7.6 Hz, 1H), 6.73 (dd, *J* = 8.2, 4.4 Hz, 1H), 4.15–4.09 (m, 1H), 3.94 (q, *J* = 8.1 Hz, 1H), 3.00–2.88 (m, 3H), 2.68–2.51 (m, 2H), 1.86–1.72 (m, 7H), 1.47 (s, 1H), 1.37 (s, 3H), 0.98–0.79 (m, 5H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.2, 135.0, 134.7, 130.0, 128.8, 126.4, 122.2, 121.3, 113.3, 97.7, 65.3, 59.3, 36.0, 35.9, 32.8, 31.9, 30.2, 29.7, 23.1, 22.5, 20.5; IR (KBr, neat) 3192, 3063, 2918, 1666, 1606, 1441, 1372, 1042, 750, 582 cm^{-1}; HRMS (ESI) calcd. for $C_{21}H_{29}N_2O_2$ $(M + H)^+$ 341.2224, found 341.2229.

(*E*)-3'-Benzyl-6'-butyl-3-(4-methylbenzylidene)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3di):



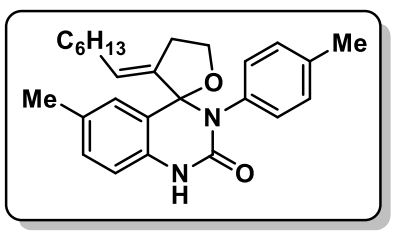
White solid; R_f (Hexane:EtOAc, 7:3) 0.6; mp 110–112 °C. Yield 170 mg, 94%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.33 (s, 1H), 7.36 (d, *J* = 7.5 Hz, 2H), 7.21 (t, *J* = 7.4 Hz, 2H), 7.16–7.13 (m, 3H), 7.04 (d, *J* = 8.2 Hz, 2H), 7.01 (s, 1H), 6.85 (s, 1H), 6.68 (d, *J* = 8.1 Hz, 1H), 6.30 (s, 1H), 4.81 (d, *J* = 15.9 Hz, 1H), 4.65 (d, *J* = 16.0 Hz, 1H), 4.16 (q, *J* = 7.4 Hz, 1H), 4.03 (q, *J* = 7.8 Hz, 1H), 3.15 (t, *J* = 7.4 Hz, 2H), 2.51 (t, *J* = 7.7 Hz, 2H), 2.35 (s, 3H), 1.51 (p, *J* = 8.0 Hz, 2H), 1.36–1.26 (m, 2H), 0.89 (t, *J* = 7.3 Hz, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 154.2, 140.1, 139.8, 137.4, 136.3, 133.6, 132.9, 129.4, 129.1, 128.4, 128.2, 128.0, 127.2, 126.3, 126.2, 122.9, 114.2, 99.1, 66.3, 47.6, 35.1, 33.8, 31.8, 22.2, 21.2, 13.9; IR (KBr, neat) 3200, 3030, 2954, 2928, 2860, 1669, 1514, 1443, 1052, 1042, 712, 519 cm^{-1}; HRMS (ESI) calcd. for $C_{30}H_{33}N_2O_2$ $(M + H)^+$ 453.2537, found 453.2540.

(*E*)-7'-Methyl-3-(4-methylbenzylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3ei):



White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 217–219 °C. Yield 156 mg, 95%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.65 (s, 1H), 7.43 (d, *J* = 8.0 Hz, 1H), 7.25–7.20 (m, 1H), 7.16 (d, *J* = 7.9 Hz, 2H), 7.09 (d, *J* = 7.9 Hz, 3H), 7.04 (dd, *J* = 8.1, 1.9 Hz, 2H), 6.90 (s, 1H), 6.70 (d, *J* = 8.1 Hz, 1H), 6.47 (t, *J* = 2.7 Hz, 1H), 3.89 (q, *J* = 8.0 Hz, 1H), 3.81 (td, *J* = 8.5, 4.3 Hz, 1H), 2.84–2.76 (m, 1H), 2.37 (s, 3H), 2.36 (s, 3H), 2.26 (s, 3H), 2.23–2.18 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.6, 142.9, 137.4, 137.3, 135.8, 133.9, 133.1, 131.5, 131.3, 130.2, 129.4, 129.2, 128.5, 128.1, 127.4, 123.0, 114.2, 99.4, 67.2, 31.3, 21.23, 21.20, 20.9; IR (KBr, neat) 3206, 3047, 2923, 1669, 1512, 1417, 1264, 1037, 732, 514 cm^{-1}; HRMS (ESI) calcd. for $C_{27}H_{27}N_2O_2$ $(M + H)^+$ 411.2067, found 411.2065.

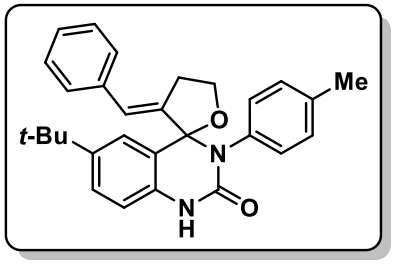
(*E*)-3-Heptylidene-7'-methyl-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3er):



White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 134–136 °C. Yield 120 mg, 74%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.95 (s, 1H), 7.37 (s, 1H), 7.17 (s, 1H), 7.12 (s, 1H), 7.03 (d, *J* = 7.7 Hz, 1H), 7.00 (s, 1H), 6.86 (s, 1H), 6.65 (d, *J* = 8.0 Hz, 1H), 5.47 (t, *J* = 6.4Hz, 1H), 3.84–3.75 (m, 2H), 2.54–2.49 (m, 1H), 2.36 (s, 3H), 2.27 (s, 3H), 2.01–1.91 (m, 3H), 1.31–1.17 (m, 8H), 0.88 (t, *J* = 7.0 Hz, 3H);

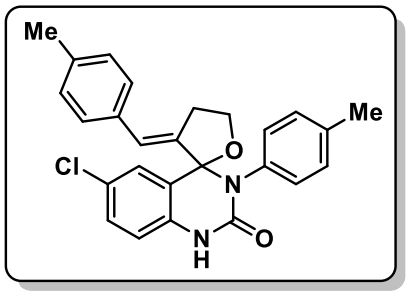
$^{13}C\{^{1}H\}$ NMR (125 MHz, $CDCl_3$) δ 153.0, 141.7, 137.0, 136.1, 133.0, 131.7, 130.8, 129.8, 129.5, 129.2, 128.8, 127.1, 123.3, 114.2, 97.8, 66.4, 31.7, 30.1, 29.3, 29.0, 28.6, 22.5, 21.2, 20.9, 14.0; IR (KBr, neat) 3217, 3039, 2924, 2857, 1671, 1512, 1414, 1033, 819, 733, 514 cm^{-1}; HRMS (ESI) calcd. for $C_{26}H_{33}N_2O_2$ $(M + H)^+$ 405.2537, found 405.2537.

(*E*)-3-Benzylidene-6'-(tert-butyl)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3fa):



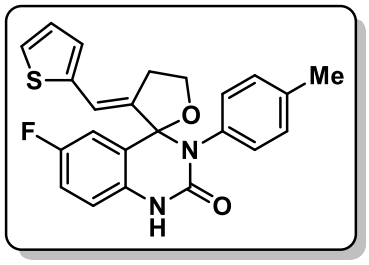
White solid; R_f (Hexane:EtOAc, 1:1) 0.60; mp 208–210 °C. Yield 170 mg, 97%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.40 (s, 1H), 7.46 (s, 1H), 7.34 (t, *J* = 7.6 Hz, 2H), 7.30 (dd, *J* = 8.4, 2.1 Hz, 1H), 7.26 (t, *J* = 7.5 Hz, 1H), 7.23 (s, 1H), 7.13 (d, *J* = 7.8 Hz, 3H), 7.10 (s, 1H), 7.03 (d, *J* = 7.6 Hz, 1H), 6.75 (d, *J* = 8.4 Hz, 1H), 6.53 (t, *J* = 2.7 Hz, 1H), 3.89–3.82 (m, 2H), 2.80–2.74 (m, 1H), 2.38 (s, 3H), 2.24–2.16 (m, 1H), 1.26 (s, 9H); $^{13}C\{^{1}H\}$ NMR (125 MHz, $CDCl_3$) δ 152.6, 144.8, 143.9, 137.4, 136.8, 135.9, 133.2, 131.6, 129.4, 128.5, 128.4, 128.3, 127.4, 126.6, 123.9, 122.4, 113.9, 99.3, 67.0, 34.3, 31.4, 31.3, 21.3; IR (KBr, neat) 3218, 3050, 2958, 1672, 1512, 1389, 1264, 1037, 732, 697, 514 cm^{-1}; HRMS (ESI) calcd. for $C_{29}H_{31}N_2O_2$ $(M + H)^+$ 439.2380, found 439.2380.

(*E*)-7'-Chloro-3-(4-methylbenzylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3gi):



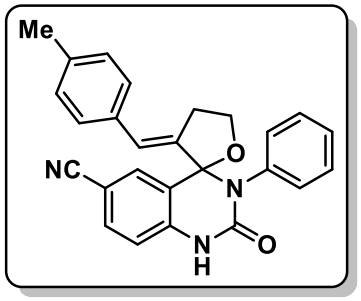
White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 226–228 °C. Yield 159 mg, 92%; ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.34 (s, 1H), 7.41 (d, *J* = 7.9 Hz, 1H), 7.23 (d, *J* = 7.6 Hz, 1H), 7.16 (d, *J* = 7.8 Hz, 2H), 7.13 (dd, *J* = 8.5, 2.3 Hz, 1H), 7.08 (d, *J* = 7.2 Hz, 4H), 7.01 (d, *J* = 8.1 Hz, 1H), 6.69 (d, *J* = 8.5 Hz, 1H), 6.46 (s, 1H), 3.88 (q, *J* = 7.9 Hz, 1H), 3.80 (td, *J* = 8.6, 4.3 Hz, 1H), 2.82–2.76 (m, 1H), 2.38 (s, 3H), 2.35 (s, 3H), 2.26–2.18 (m, 1H); $^{13}C\{^{1}H\}$ NMR (125 MHz, $CDCl_3$) δ 152.5, 142.5, 137.7, 137.6, 135.5, 134.3, 133.6, 131.4, 129.5, 129.2, 128.55, 128.52, 126.9, 126.7, 124.6, 116.0, 98.9, 67.4, 31.1, 21.3, 21.2; IR (KBr, neat) 3223, 3049, 2925, 1672, 1497, 1381, 1263, 1039, 731, 513 cm^{-1}; HRMS (ESI) calcd. for $C_{26}H_{24}ClN_2O_2$ $(M + H)^+$ 431.1521, found 431.1521.

(*E*)-6'-Fluoro-3-(thiophen-2-ylmethylene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3hq):



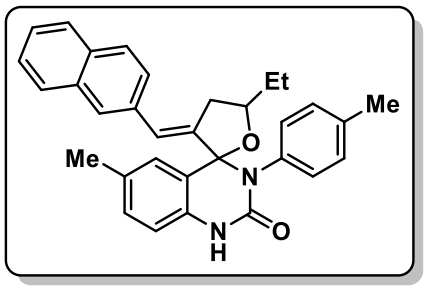
White solid; R_f (Hexane:EtOAc, 2:3) 0.50; mp 239–241 °C. Yield 146 mg, 90%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.05 (s, 1H), 7.38 (s, 1H), 7.34 (d, *J* = 5.0 Hz, 1H), 7.21 (s, 1H), 7.09–7.04 (m, 2H), 6.99 (s, 1H), 6.96 (d, *J* = 3.6 Hz, 1H), 6.92 (td, *J* = 8.4, 2.9 Hz, 1H), 6.79 (dd, *J* = 9.1, 2.8 Hz, 1H), 6.72 (dd, *J* = 8.8, 4.6 Hz, 1H), 6.68 (t, *J* = 2.7 Hz, 1H), 3.97 (q, *J* = 7.8 Hz, 1H), 3.77 (td, *J* = 8.6, 4.9 Hz, 1H), 2.89–2.81 (m, 1H), 2.35 (s, 3H), 2.27–2.18 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 157.9 (d, *J* = 238.5 Hz), 152.5, 141.5, 140.3, 137.7, 135.4, 131.68 (d, *J* = 2.1 Hz), 131.3, 131.1, 129.6, 129.4, 128.1, 127.4, 126.7, 124.1 (d, *J* = 6.9 Hz), 121.7, 116.8 (d, *J* = 23.2 Hz), 115.81 (d, *J* = 7.8 Hz), 113.5 (d, *J* = 24.1 Hz), 98.9, 67.6, 31.4, 21.2; ^{19}F NMR (470 MHz, $CDCl_3/C_6F_6$) δ -123.62 (s, -CF-); IR (KBr, neat) 3232, 3054, 2964, 1671, 1508, 1418, 1387, 1263, 1037, 732, 700, 513 cm^{-1}; HRMS (ESI) calcd. for $C_{23}H_{20}FN_2O_2S$ $(M + H)^+$ 407.1224, found 407.1224.

(*E*)-3-(4-Methylbenzylidene)-2'-oxo-3'-phenyl-2',3',4,5-tetrahydro-1'*H*,3*H*-spiro[furan-2,4'-quinazoline]-7'-carbonitrile (3ii):



White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 265–267 °C. Yield 137 mg, 84%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 10.10 (s, 1H), 7.56 (d, *J* = 7.8 Hz, 1H), 7.48 (t, *J* = 7.0 Hz, 1H), 7.43–7.39 (m, 2H), 7.34–7.29 (m, 2H), 7.16 (d, *J* = 7.8 Hz, 2H), 7.13 (s, 1H), 7.04 (d, *J* = 7.8 Hz, 2H), 6.58 (d, *J* = 8.4 Hz, 1H), 6.42 (t, *J* = 2.6 Hz, 1H), 3.90–3.79 (m, 2H), 2.84–2.76 (m, 1H), 2.35 (s, 3H), 2.22–2.13 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$) δ 152.1, 141.9, 139.4, 138.0, 137.9, 133.1, 133.0, 131.9, 131.8, 131.6, 129.3, 129.2, 128.9, 128.6, 128.5, 128.2, 123.9, 119.1, 115.6, 104.8, 98.5, 67.5, 31.1, 21.2; IR (KBr, neat) 3234, 3064, 2941, 2224, 1678, 1612, 1414, 1389, 1036, 709, 515 cm^{-1}; HRMS (ESI) calcd. for $C_{26}H_{22}N_3O_2$ $(M + H)^+$ 408.1707, found 408.1710.

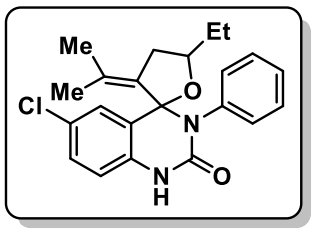
(*E*)-5-Ethyl-7'-methyl-3-(naphthalen-2-ylmethylene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3jm):



White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 133–135 °C. Yield 173 mg, 91% (d.r. = 2:1); ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.40 (s, 1H, minor), 8.30 (s, 1H, major), 7.72–7.68 (m, 4H, major), 7.59 (s, 4H, minor), 7.46–7.30 (m, 3H), 7.18–7.13 (m, 2H), 7.07 (d, *J* = 7.9 Hz, 1H), 7.00–6.87 (m, 3H), 6.66–6.59 (m, 2H, major), 6.52 (d, *J* = 3.2 Hz, 2H, minor), 3.84–3.78 (m, 1H, minor), 3.34–3.28 (m, 1H, major), 2.82 (dd, *J* = 16.0, 5.4

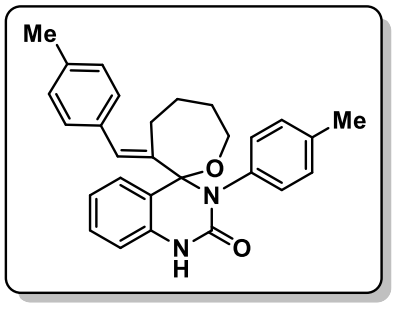
Hz, 2H, minor), 2.62–2.49 (m, 2H, major), 2.28 (d, *J* = 10.4 Hz, 3H), 2.20 (s, 3H), 1.67–1.58 (m, 1H), 1.47–1.34 (m, 1H), 0.84–0.80 (m, 3H); $^{13}C\{^{1}H\}$ NMR (125 MHz, $CDCl_3$) δ 152.8, 152.5, 146.6, 144.5, 137.6, 137.4, 135.7, 134.3, 134.2, 133.4, 133.33, 133.29, 132.52, 132.48, 132.1, 131.8, 131.5, 131.3, 130.2, 129.6, 129.3, 129.0, 128.6, 128.1, 128.02, 127.97, 127.83, 127.78, 127.6, 127.4, 127.3, 127.2, 126.4, 126.3, 126.2, 123.7, 123.2, 114.3, 114.2, 99.7, 99.0, 80.9, 79.4, 37.5, 36.9, 29.5, 28.3, 21.3, 21.2, 21.0, 10.3, 10.0; IR (KBr, neat) 3209, 3046, 2962, 2925, 1669, 1604, 1511, 1416, 1388, 1265, 1017, 817, 738, 476 cm^{-1}; HRMS (ESI) calcd. for $C_{32}H_{31}N_2O_2$ $(M + H)^+$ 475.2380, found 475.2380.

7'-Chloro-5-ethyl-3-(propan-2-ylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (3kv):



White solid; R_f (Hexane:EtOAc, 3:2) 0.50; mp 240–242 °C. Yield 150 mg, 98% (d.r. = 7:3); ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.00 (s, 1H), 7.58 (d, *J* = 7.8 Hz, 1H), 7.39 (t, *J* = 7.8 Hz, 1H), 7.36–7.30 (m, 2H), 7.14 (dd, *J* = 8.5, 2.3 Hz, 1H), 7.06 (dd, *J* = 2.3 Hz, 1H), 7.04–7.00 (m, 1H), 6.67 (d, *J* = 8.5 Hz, 1H), 3.77–3.72 (m, 1H), 2.57 (dd, *J* = 15.0, 5.7 Hz, 1H), 1.72–1.64 (m, 1H), 1.56 (s, 3H), 1.46 (d, *J* = 2.7 Hz, 3H), 1.44–1.38 (m, 2H), 0.86 (t, *J* = 7.6 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (125 MHz, $CDCl_3$) δ 153.0, 138.5, 133.8, 133.6, 131.3, 130.7, 129.4, 129.1, 128.1, 127.8, 126.6, 125.6, 124.4, 116.0, 96.0, 77.7, 37.6, 28.1, 22.7, 21.5, 10.2; IR (KBr, neat) 3245, 2960, 2928, 1681, 1599, 1491, 1409, 1379, 1257, 982, 707, 557 cm^{-1}; HRMS (ESI) calcd. for $C_{22}H_{24}ClN_2O_2$ $(M + H)^+$ 383.1521, found 383.1521.

(*E*)-3-(4-methylbenzylidene)-3'-(*p*-tolyl)-1'*H*-spiro[oxepane-2,4'-quinazolin]-2'(3'*H*)-one (3li):



White semi-solid; R_f (Hexane:EtOAc, 3:2) 0.50. Yield 34 mg, 20%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.38 (d, *J* = 8.0 Hz, 2H), 7.25 (s, 1H), 7.23–7.19 (m, 2H), 7.16 (s, 1H), 7.13–6.99 (m, 5H), 6.86 (td, *J* = 7.1, 2.0 Hz, 1H), 6.60 (d, *J* = 7.5 Hz, 1H), 6.44 (t, *J* = 1.9 Hz, 1H), 3.59 (t, *J* = 6.2 Hz, 2H), 2.32–2.26 (m, 4H), 2.23 (s, 3H), 2.20–2.15 (m, 1H), 1.71–1.62 (m, 2H), 1.61–1.54 (m, 2H); $^{13}C\{^{1}H\}$ NMR (125 MHz, $CDCl_3$) δ 151.6, 151.5, 148.2, 141.7, 138.8, 136.2, 135.9, 132.5, 130.0, 129.5, 129.2, 127.7, 124.4, 123.8, 123.4, 123.1, 122.7, 120.6, 119.4, 91.0, 62.6, 32.4, 26.5, 23.4, 21.4, 20.8; IR (KBr, neat) 3283, 2924, 2858, 1634, 1585, 1519, 1478, 1409, 1227, 1029, 908, 816, 760, 730, 648, 504 cm^{-1}; HRMS (ESI) calcd. for $C_{28}H_{29}N_2O_2$ $(M + H)^+$ 425.2224, found 425.2225.

Experimental procedure of one-pot strategy:

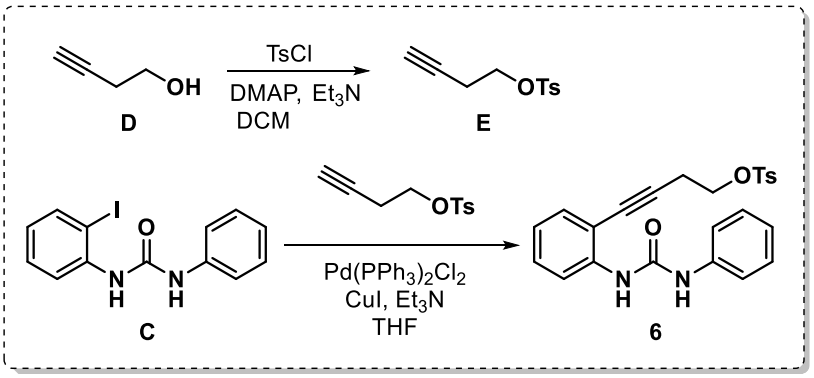
To a solution of 4-(2-aminophenyl)but-3-yn-1-ol (**4**) (161 mg, 1.0 mmol, 1.0 equiv.) in DCM (3 mL), phenyl isocyanate (0.12 mL, 1.1 mmol, 1.1 equiv.) was added under N_2 atmosphere. The reaction was stirred at room temperature for 3 h, and the progress of the reaction was monitored by TLC. Meanwhile, a white solid precipitate started to form in the reaction mixture, which indicated the formation of the desired diphenyl urea compound. When all the alkynol substituted aniline was consumed, *p*-tolualdehyde (**2i**) (0.14 mL, 1.2 mmol, 1.2 equiv.) was introduced into the system, along with some more DCM solvent (3 mL). The reaction was then cooled to 0 °C and triflic acid (0.18 mL, 2.0 mmol, 2.0 equiv.) was added slowly. The reaction mixture was further stirred for 8 h at room temperature before quenching with a saturated $NaHCO_3$ solution. The organic layer was extracted with DCM (2 x 20 mL), and the combined organic layers were further washed with brine and dried over anhydrous Na_2SO_4. The organic phase was concentrated in a rotary evaporator to give the crude product, which was then subjected to column chromatography over silica gel to provide the final product **3bi** in 62% yield (237 mg).

Experimental procedure for gram-scale synthesis of compound 3ai:

To an ice-cold suspension of 1-(2-(4-hydroxybut-1-yn-1-yl)phenyl)-3-(*p*-tolyl)urea (**1a**) (1.0 g, 3.4 mmol, 1.0 equiv.) and *p*-tolualdehyde (**2i**) (490 mg, 4.08 mmol, 1.2 equiv.) in DCM (15.0 mL), triflic acid (0.6 mL, 6.8 mmol, 2.0 equiv.) was added slowly under N_2 atmosphere. The reaction was continued to stir for 0.75 h at room temperature, and the progress of the reaction was monitored by TLC. It was then treated with saturated $NaHCO_3$ solution, and the organic layer was extracted with DCM (2 x 30 mL). The combined organic layers were further washed with brine, followed by drying over anhydrous Na_2SO_4. The organic phase was concentrated in a rotary evaporator to give the crude product, which was then subjected to column chromatography over silica gel to provide the final product **3ai** as a white solid in 97% yield (1.31 g).

Experimental procedure and characterization data of compound 6:

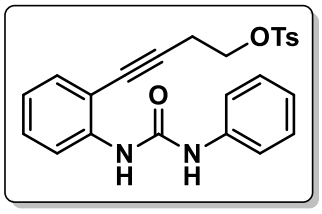
Schematic representation of starting material **6**:



To an ice-cold solution of 3-butyn-1-ol (**D**) (2.0 mmol, 1.0 equiv.), DMAP (0.2 mmol, 0.1 equiv.) and Et_3N (2.4 mmol, 1.2 equiv.) in DCM under N_2 atmosphere, tosyl chloride (2.4 mmol, 1.2 equiv.) in DCM was added dropwise over a period of 5 minutes. The reaction was allowed to stir at room temperature for 2 h and then quenched with saturated NH_4Cl solution. The organic layer was extracted with DCM (2 x 20 mL). The combined organic layers were further washed with brine, dried over anhydrous Na_2SO_4, and concentrated in a rotary evaporator to give the crude product, which was then subjected to column chromatography over silica gel to provide product **E** in 98% yield.

To a mixture of $Pd(PPh_3)_2Cl_2$ (0.04 mmol, 0.04 equiv.), CuI (0.02 mmol, 0.02 equiv.) and 1-(2-iodophenyl)-3-phenylurea (**C**) (1.0 mmol, 1.0 equiv.) in dry THF under N_2 atmosphere, triethylamine (5.0 mmol, 5.0 equiv.) was added. The reaction mixture was stirred for 10 minutes at room temperature, after which tosyl protected homo propargylic alcohol **E** (1.2 mmol, 1.2 equiv.) was added dropwise over a period of 5 minutes. The reaction mixture was stirred for 5.0 hours (monitored by TLC analysis) before filtering through a pad of celite. The solids were washed with ethyl acetate, and the combined filtrates were concentrated in a rotary evaporator. The crude product was then purified by column chromatography to provide the corresponding product **6** in 65% yield (282 mg).

4-(2-(3-Phenylureido)phenyl)but-3-yn-1-yl 4-methylbenzenesulfonate (6):



White semi-solid; R_f (Hexane:EtOAc, 7:3) 0.60. Yield 282 mg, 65%; ^1H NMR (500 MHz, $CDCl_3$) δ 8.22 (d, *J* = 8.3 Hz, 1H), 7.72 (d, *J* = 8.0 Hz, 2H), 7.62–7.58 (m, 1H), 7.47 (s, 1H), 7.39 (d, *J* = 8.0 Hz, 2H), 7.25–7.17 (m, 6H), 6.95 (t, *J* = 7.4 Hz, 1H), 6.85 (t, *J* = 7.5 Hz, 1H), 4.18 (t, *J* = 5.7 Hz, 2H), 2.77 (t, *J* = 5.6 Hz, 2H), 2.34 (s, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$) δ 152.6, 145.7, 140.4, 138.8, 132.3, 131.8,

130.2, 130.0, 129.4, 128.9, 128.0, 127.8, 123.3, 121.9, 120.1, 119.0, 111.2, 91.1, 78.7, 68.3, 21.7, 21.1; IR (KBr, neat) 3340, 2954, 2926, 2852, 1682, 1601, 1496, 1446, 1187, 1174, 1029, 757, 552 cm^{-1}; HRMS (ESI) calcd. for $C_{24}H_{23}N_2O_4S$ $(M + H)^+$ 435.1373, found 435.1381.

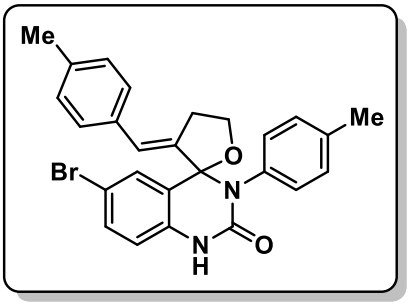
Experimental procedure of control experiment:

To an ice-cold suspension of 4-(2-(3-phenylureido)phenyl)but-3-yn-1-yl 4-methylbenzenesulfonate (**6**) (174 mg, 0.4 mmol, 1.0 equiv.) and *p*-tolualdehyde (**2i**) (58 mg, 0.48 mmol, 1.2 equiv.) in DCM (30.0 mL), triflic acid (0.07 mL, 0.8 mmol, 2.0 equiv.) was added slowly under N_2 atmosphere. The reaction was continued to stir for 5 h at room temperature, and the progress of the reaction was monitored by TLC. Although no new spot was formed, to collect the starting material, the reaction mixture was treated with saturated $NaHCO_3$ solution, and the organic layer was extracted with DCM (2 x 10 mL). The combined organic layers were further washed with brine, followed by drying over anhydrous Na_2SO_4. The organic phase was concentrated in a rotary evaporator which was then subjected to column chromatography over silica gel to get the unreacted starting material **6** in 87% yield.

Experimental procedure and characterization data of compound 8:[2]

To an ice-cold solution of spiro-furan quinazolinone derivative **3ai** (159 mg, 0.4 mmol, 1.0 equiv.) in DCM (3.0 mL), N-Bromosuccinimide (NBS) (85 mg, 0.48 mmol, 1.2 equiv.) was added portion wise over a period of 5 minutes. The reaction was stirred for 1.0 hour at room temperature, and the progress of the reaction was monitored by TLC. It was then treated with saturated $Na_2S_2O_3$ solution, and the organic layer was extracted with DCM (2 x 10 mL). The combined organic layers were further washed with brine, followed by drying over anhydrous Na_2SO_4. The organic phase was concentrated in a rotary evaporator to give the crude product, which was then subjected to column chromatography over silica gel to provide the final product **8** as a white solid in 80% yield (152 mg).

(*E*)-6'-Bromo-3-(4-methylbenzylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (8):



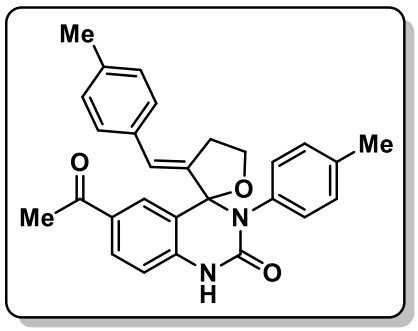
White solid; R_f (Hexane:EtOAc, 1:1) 0.50; mp 216–218 °C. Yield 152 mg, 80%; ^1H NMR (400 MHz, $CDCl_3$) δ 9.35 (s, 1H), 7.32 (d, *J* = 8.0 Hz, 1H), 7.17–7.12 (m, 2H), 7.11 (d, *J* = 1.9 Hz, 1H), 7.07 (d, *J* = 8.0 Hz, 2H), 6.99 (d, *J* = 7.9 Hz, 3H), 6.92 (d, *J* = 8.2 Hz, 1H), 6.53 (d, *J* = 8.5 Hz, 1H), 6.37 (t, *J* = 2.7 Hz, 1H), 3.79 (q, *J* = 8.0 Hz, 1H), 3.70 (td, *J* =

8.5, 4.4 Hz, 1H), 2.74–2.66 (m, 1H), 2.29 (s, 3H), 2.27 (s, 3H), 2.17–2.08 (m, 1H); $^{13}C\{^{1}H\}$ NMR (125 MHz, $CDCl_3$) δ 152.5, 142.5, 137.7, 137.6, 135.4, 134.7, 133.5, 132.3, 131.4, 129.8, 129.5, 129.2, 128.55, 128.53, 125.0, 116.4, 113.9, 98.8, 67.4, 31.1, 21.3, 21.2; IR (KBr, neat) 3235, 2923, 2870, 1676, 1599, 1411, 1379, 1259, 1039, 814, 512 cm^{-1}; HRMS (ESI) calcd. for $C_{26}H_{24}BrN_2O_2$ $(M + H)^+$ 475.1016, found 475.1037.

Experimental procedure and characterization data of compound 9:[3]

To an ice-cold mixture of aluminum trichloride (134 mg, 1.0 mmol, 2.5 equiv.) in DCM (3.0 mL), spiro-furan quinazolinone derivative **3ai** (159 mg, 0.4 mmol, 1.0 equiv.) was added slowly under N_2 atmosphere at 0 °C, followed by the dropwise addition of acetyl chloride (0.06 mL, 0.8 mmol, 2.0 equiv.). The reaction mixture was stirred at 0 °C for about an hour, and the progress of the reaction was monitored by TLC. Upon completion of the reaction, it was quenched with ice-cold saturated $NaHCO_3$ solution, and the organic layer was extracted with DCM (2 x 10 mL). The combined organic layers were further washed with brine, followed by drying over anhydrous Na_2SO_4. The organic phase was concentrated in a rotary evaporator to give the crude product, which was then subjected to column chromatography over silica gel to provide the final product **9** as a white solid in 87% yield (153 mg).

(*E*)-6'-Acetyl-3-(4-methylbenzylidene)-3'-(*p*-tolyl)-4,5-dihydro-1'*H*,3*H*-spiro[furan-2,4'-quinazolin]-2'(3'*H*)-one (9):

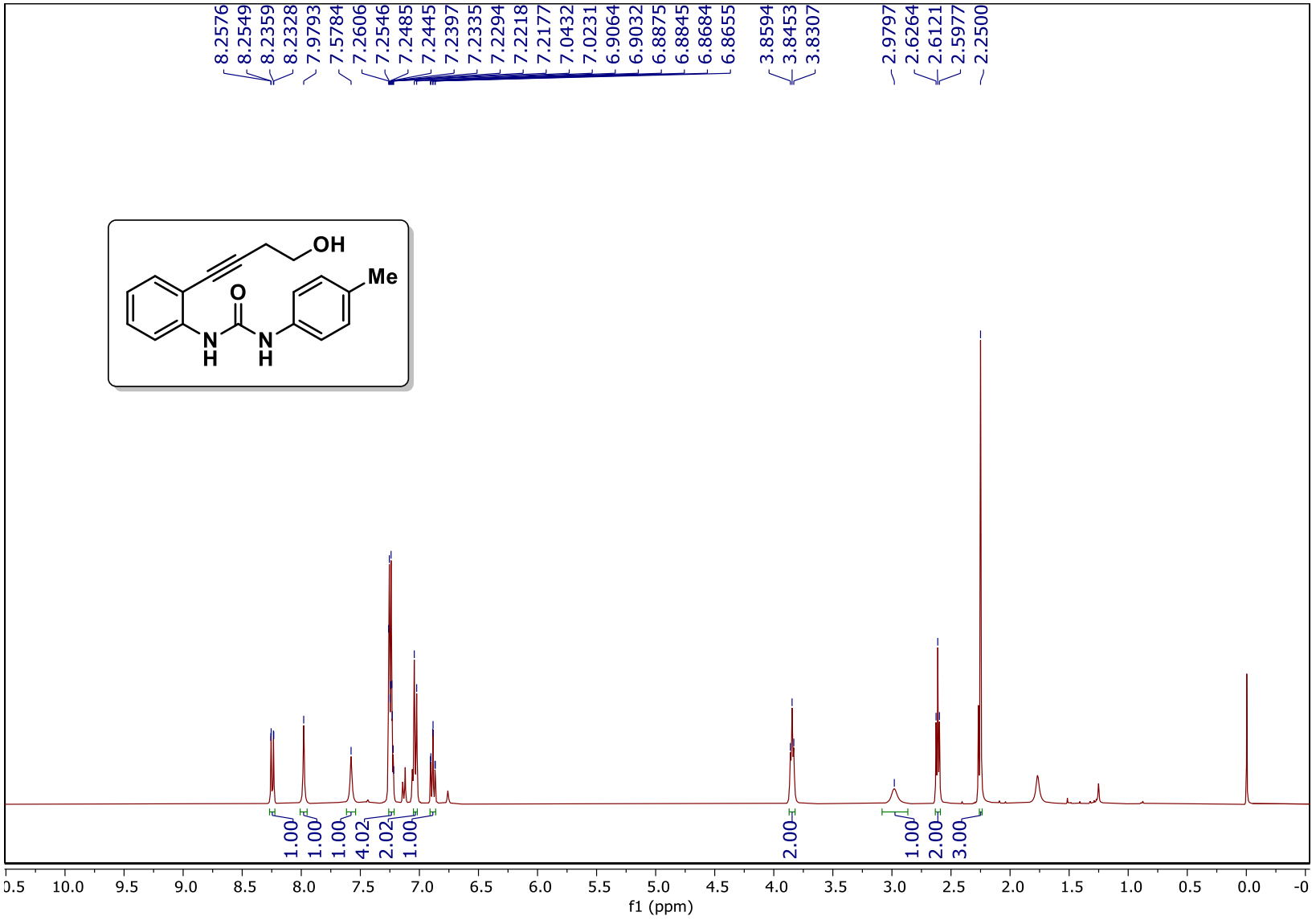


White solid; R_f (Hexane:EtOAc, 7:3) 0.50; mp 218–220 °C. Yield 153 mg, 87%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.34 (s, 1H), 8.23 (d, *J* = 8.4 Hz, 1H), 7.37 (dd, *J* = 7.8, 1.6 Hz, 1H), 7.31 (t, *J* = 7.9 Hz, 1H), 7.13 (d, *J* = 7.8 Hz, 3H), 7.03 (d, *J* = 8.0 Hz, 2H), 6.95 (d, *J* = 6.7 Hz, 2H), 6.92 (s, 1H), 6.84 (t, *J* = 7.5 Hz, 1H), 4.18 (t, *J* = 6.6 Hz, 2H), 2.89 (t, *J* = 6.7 Hz, 2H), 2.20 (s, 3H), 2.13 (s, 3H), 1.79 (s, 3H); $^{13}C\{^{1}H\}$ NMR (125 MHz, $CDCl_3$) δ 201.0, 171.4, 153.2, 144.7, 140.4, 139.5, 136.9, 135.6, 133.1, 131.9, 131.8, 129.6, 129.5, 129.4, 125.1, 121.6, 121.1, 120.9, 62.9, 27.3, 21.4, 20.9, 20.8; IR (KBr, neat) 3213, 2958, 2923, 1739, 1686, 1506, 1386, 1234, 1037, 814, 754 cm^{-1}; HRMS (ESI) calcd. for $C_{28}H_{27}N_2O_3$ $(M + H)^+$ 439.2016, found 439.2031.

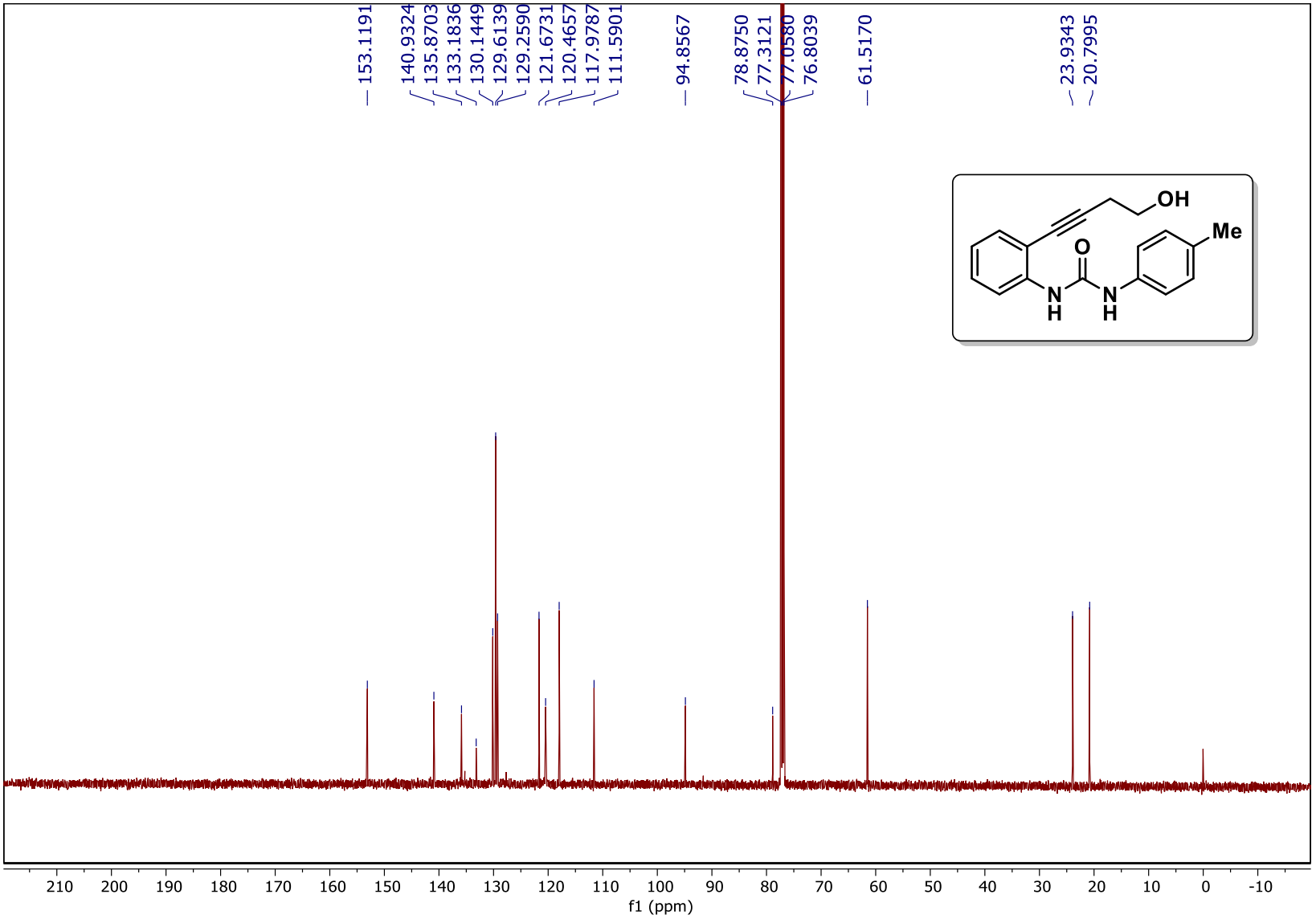
References:

1. A. Wetzel and F. Gagosz, *Angew. Chem. Int. Ed.*, 2011, **50**, 7354-7358.
2. B. Altenburg, M. Frings, J.-H. Schöbel, J. Goßen, K. Pannen, K. Vanderliek, G. Rossetti, S. Koschmieder, N. Chatain and C. Bolm, *ACS Med. Chem. Lett.*, 2020, **11**, 1928-1934.
3. L. Marais, A. Petzer, J. P. Petzer and L. J. Legoabe, *Mol. Divers.*, 2020, **24**, 391-406.

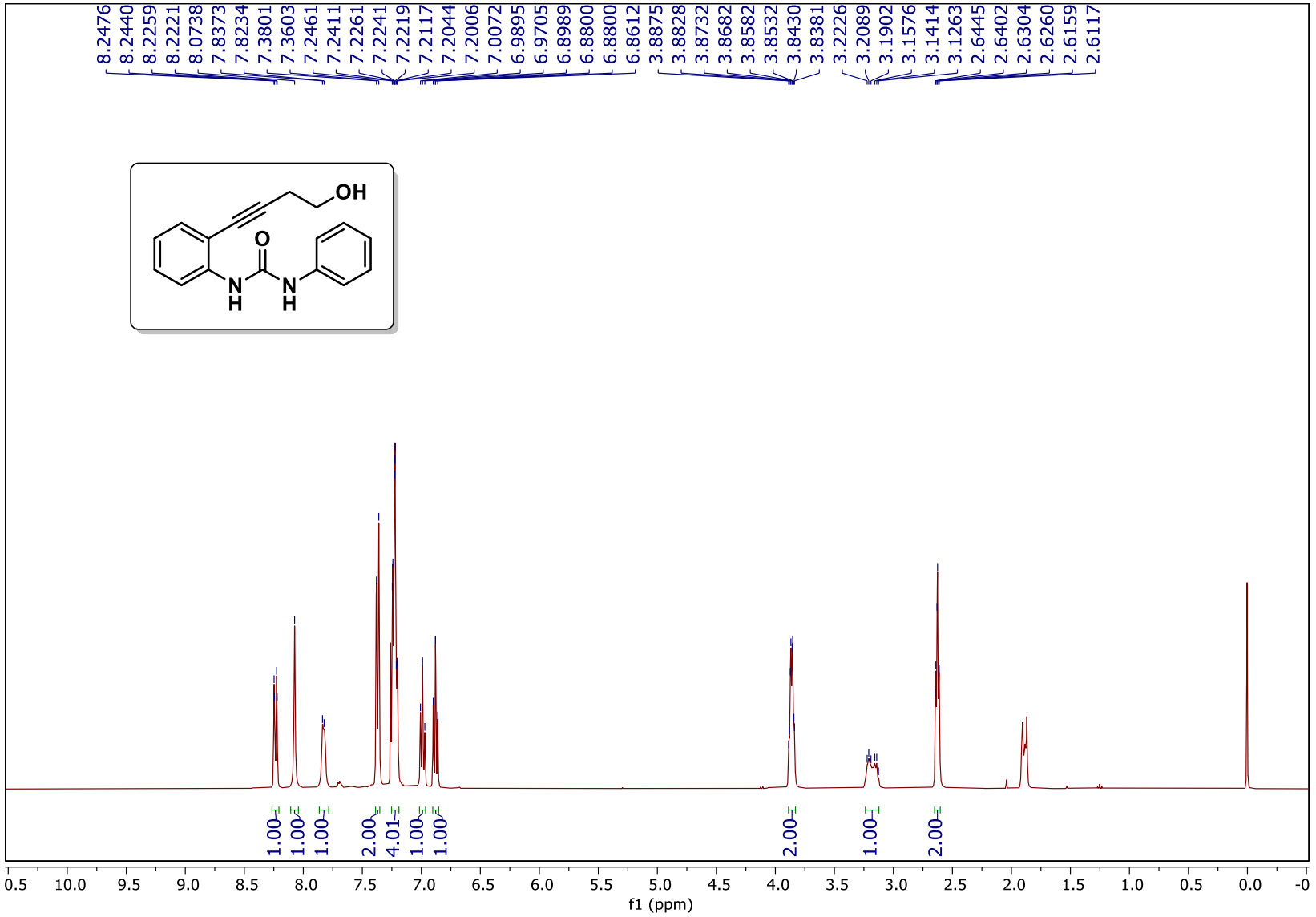
^{1}H spectrum of compound **1a** (400 MHz, $CDCl_3$)



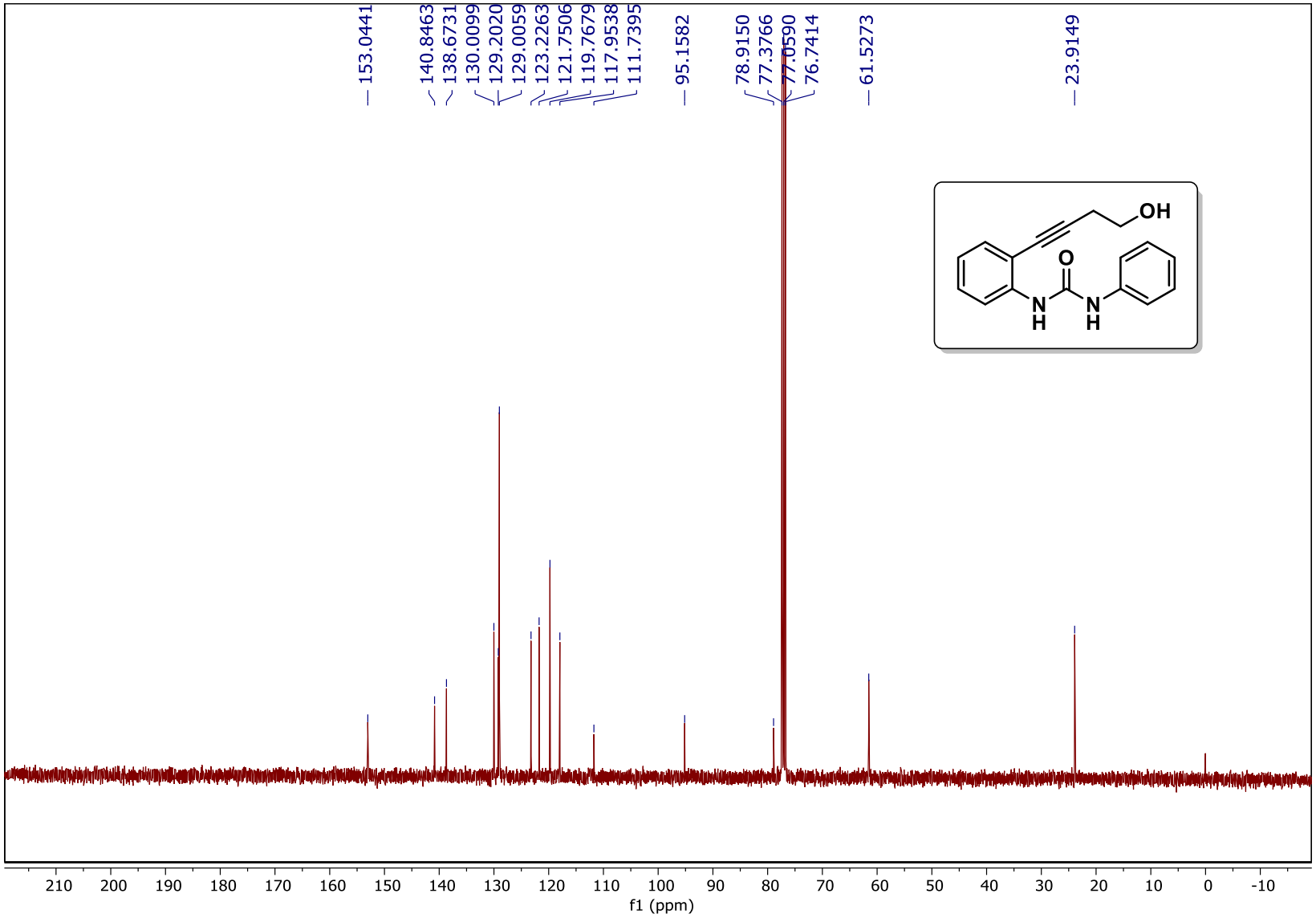
^{13}C spectrum of compound **1a** (125 MHz, $CDCl_3$)



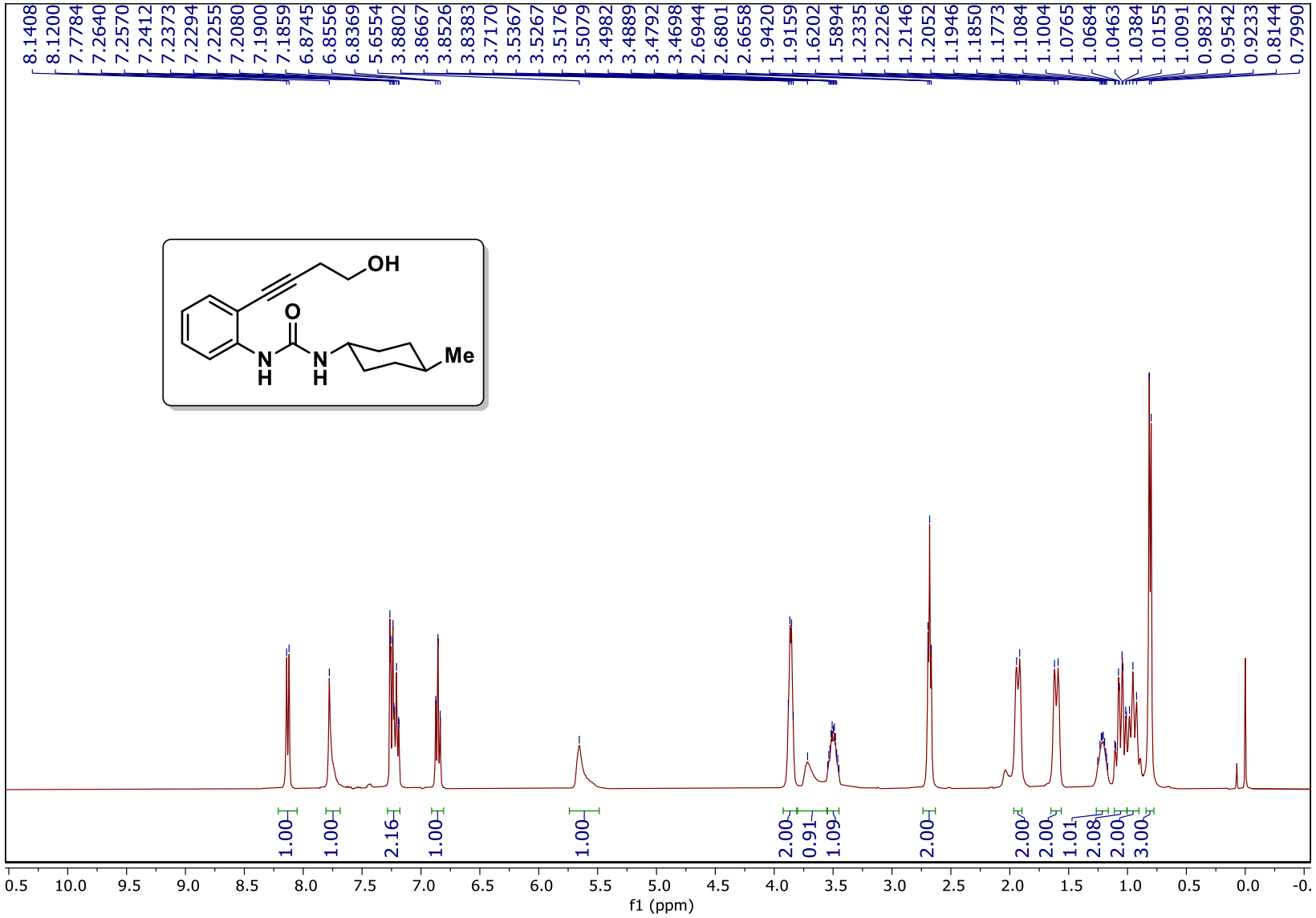
^{1}H spectrum of compound **1b** (400 MHz, $CDCl_3$)



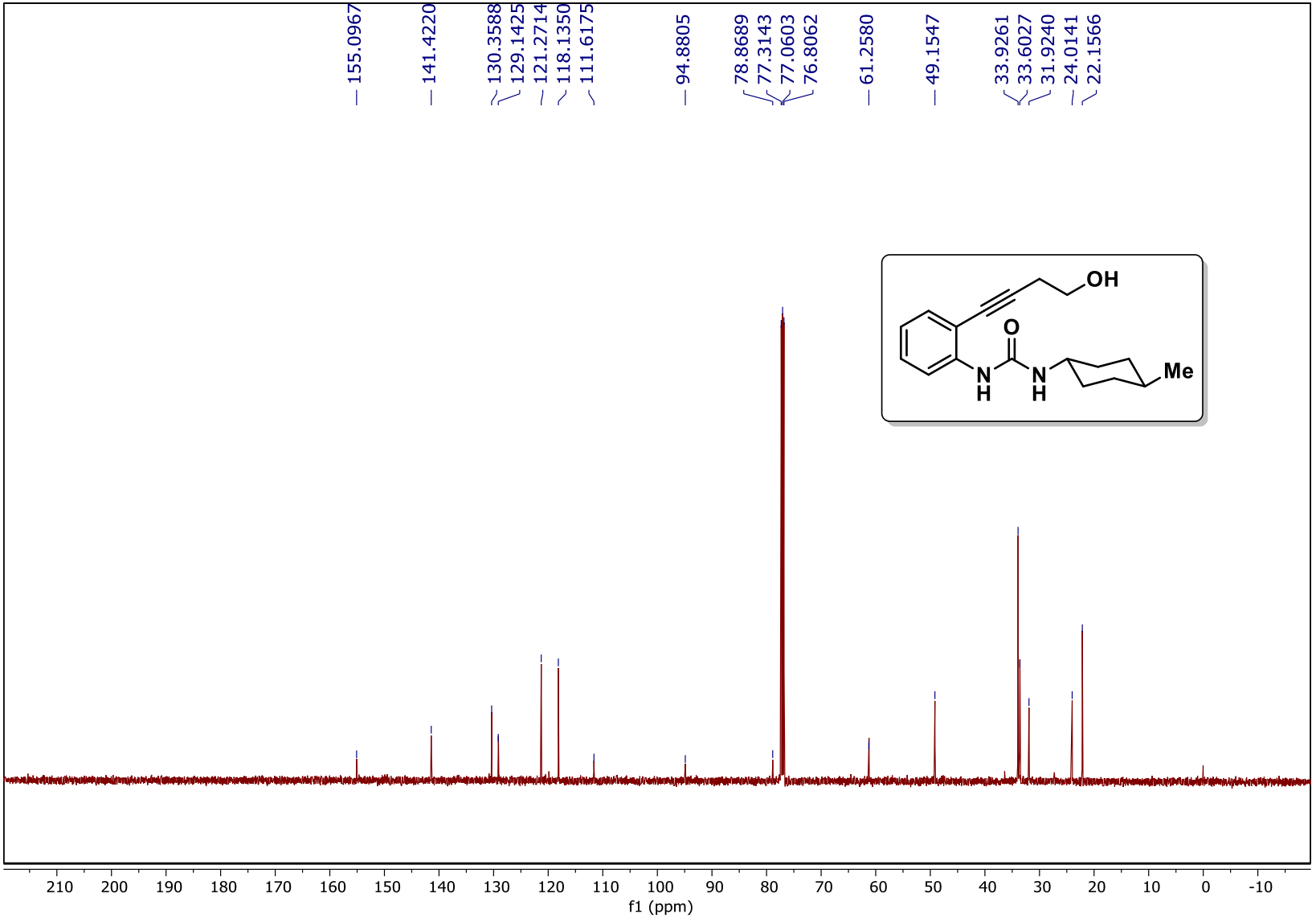
^{13}C spectrum of compound **1b** (100 MHz, $CDCl_3$)



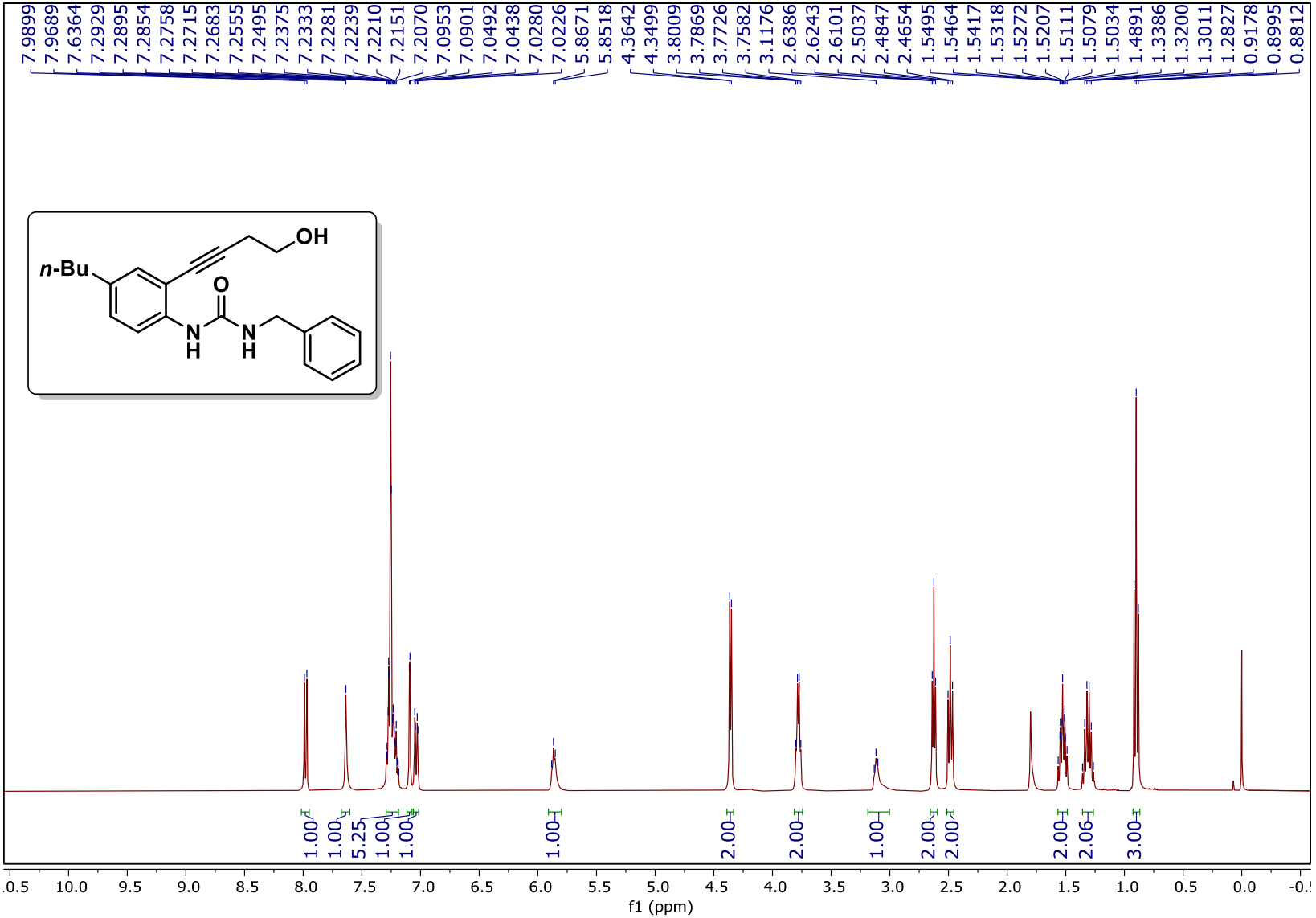
^{1}H spectrum of compound **1c** (400 MHz, $CDCl_3$)



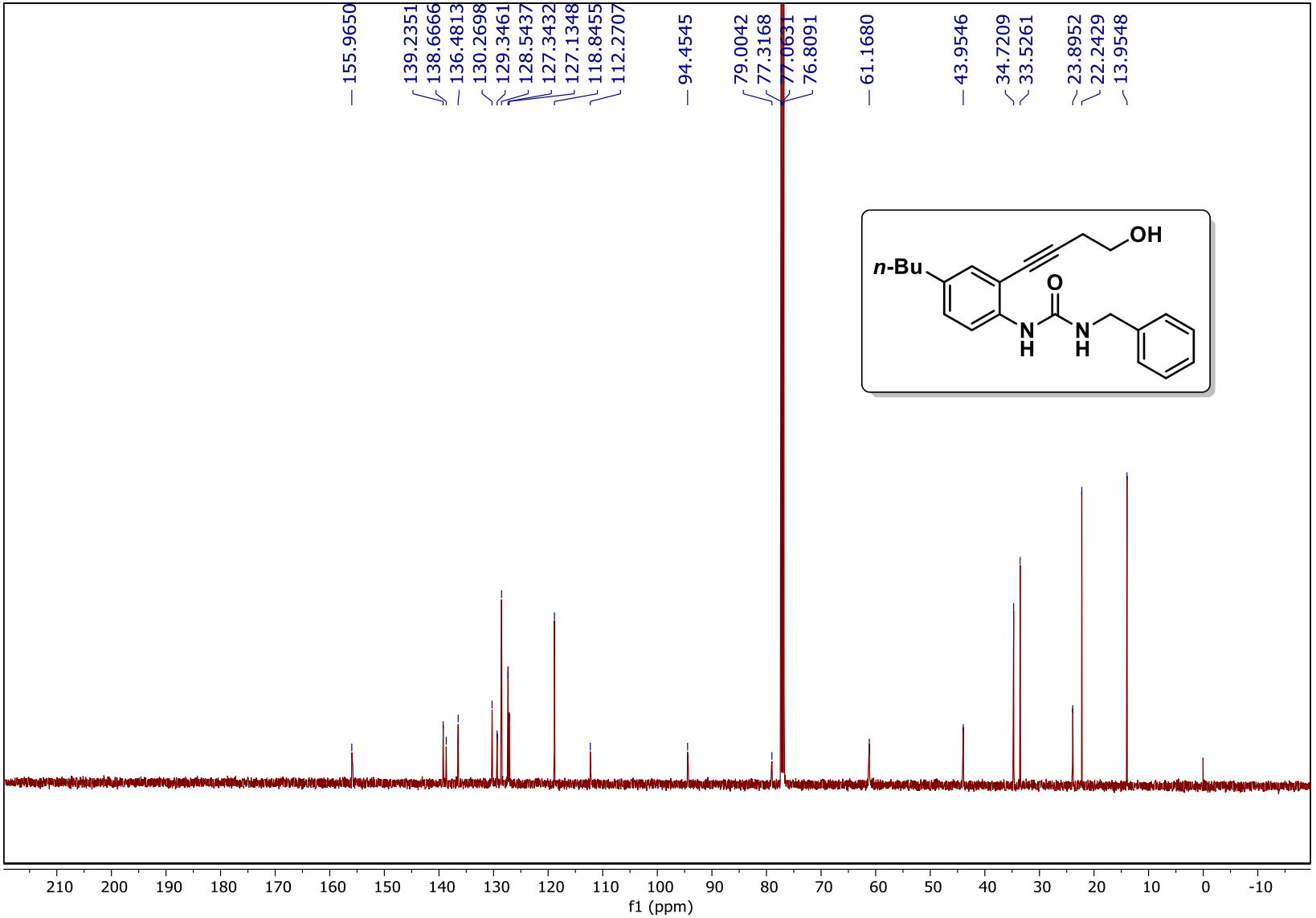
^{13}C spectrum of compound **1c** (125 MHz, $CDCl_3$)



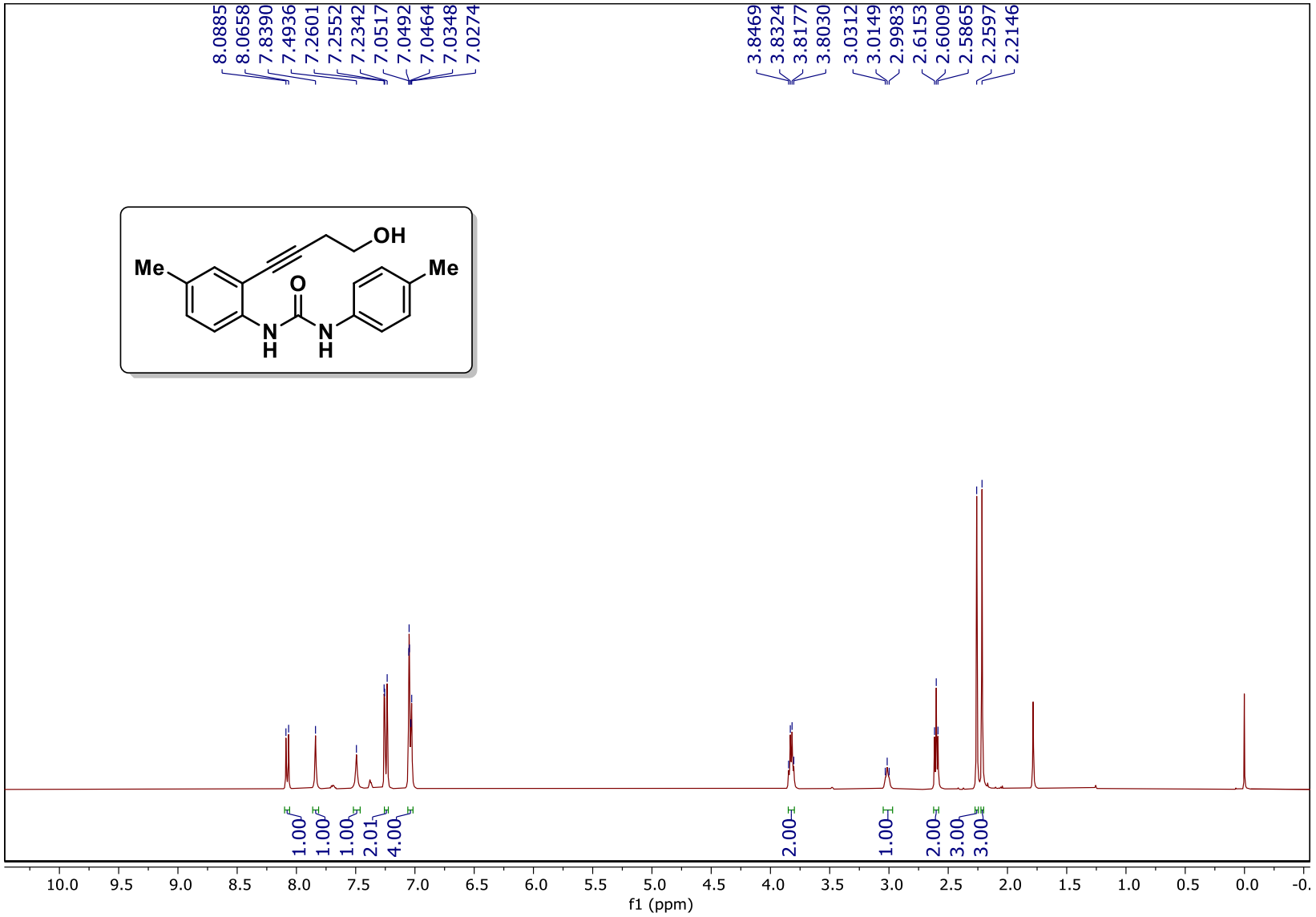
^{1}H spectrum of compound **1d** (400 MHz, $CDCl_3$)



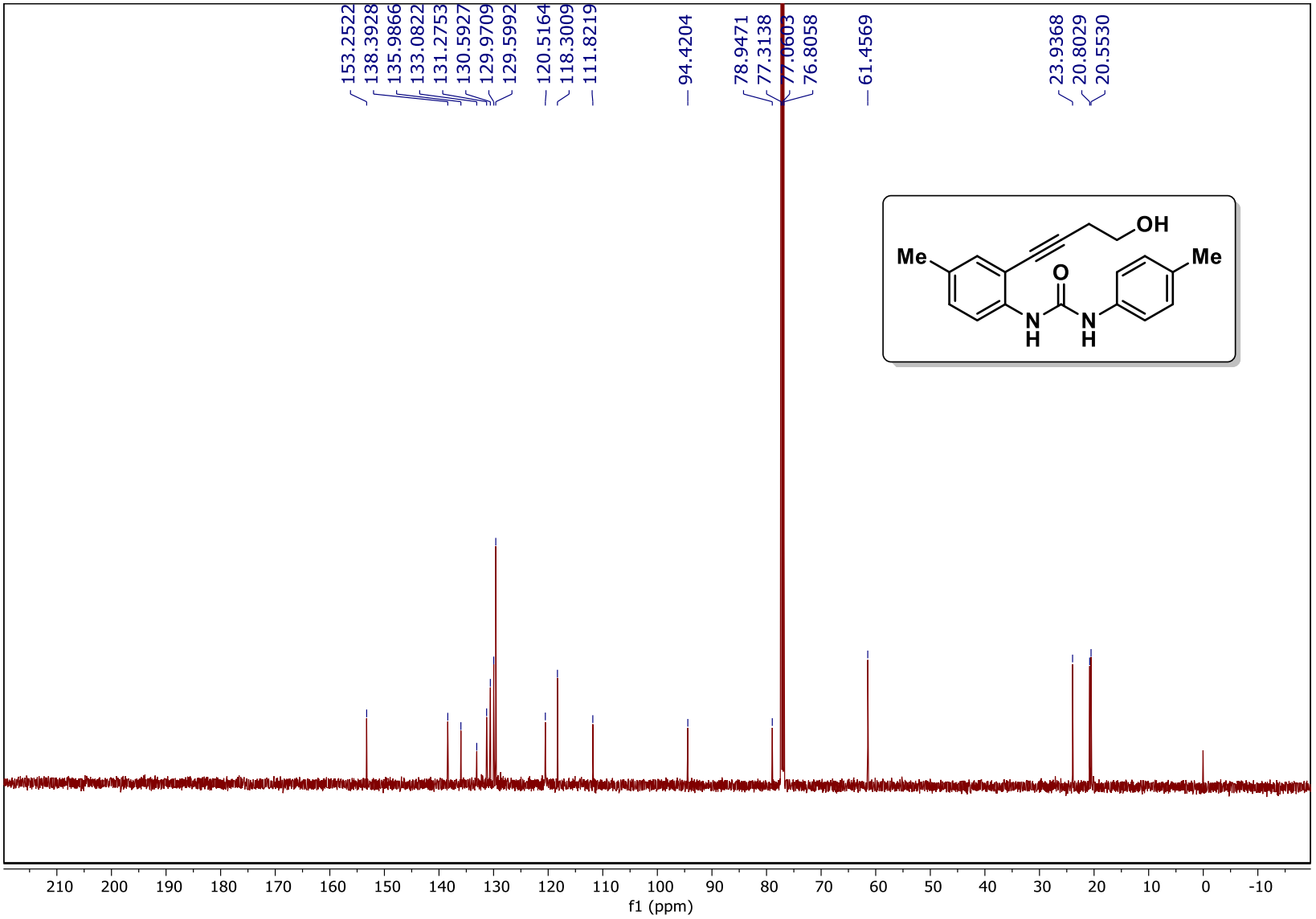
^{13}C spectrum of compound **1d** (125 MHz, $CDCl_3$)



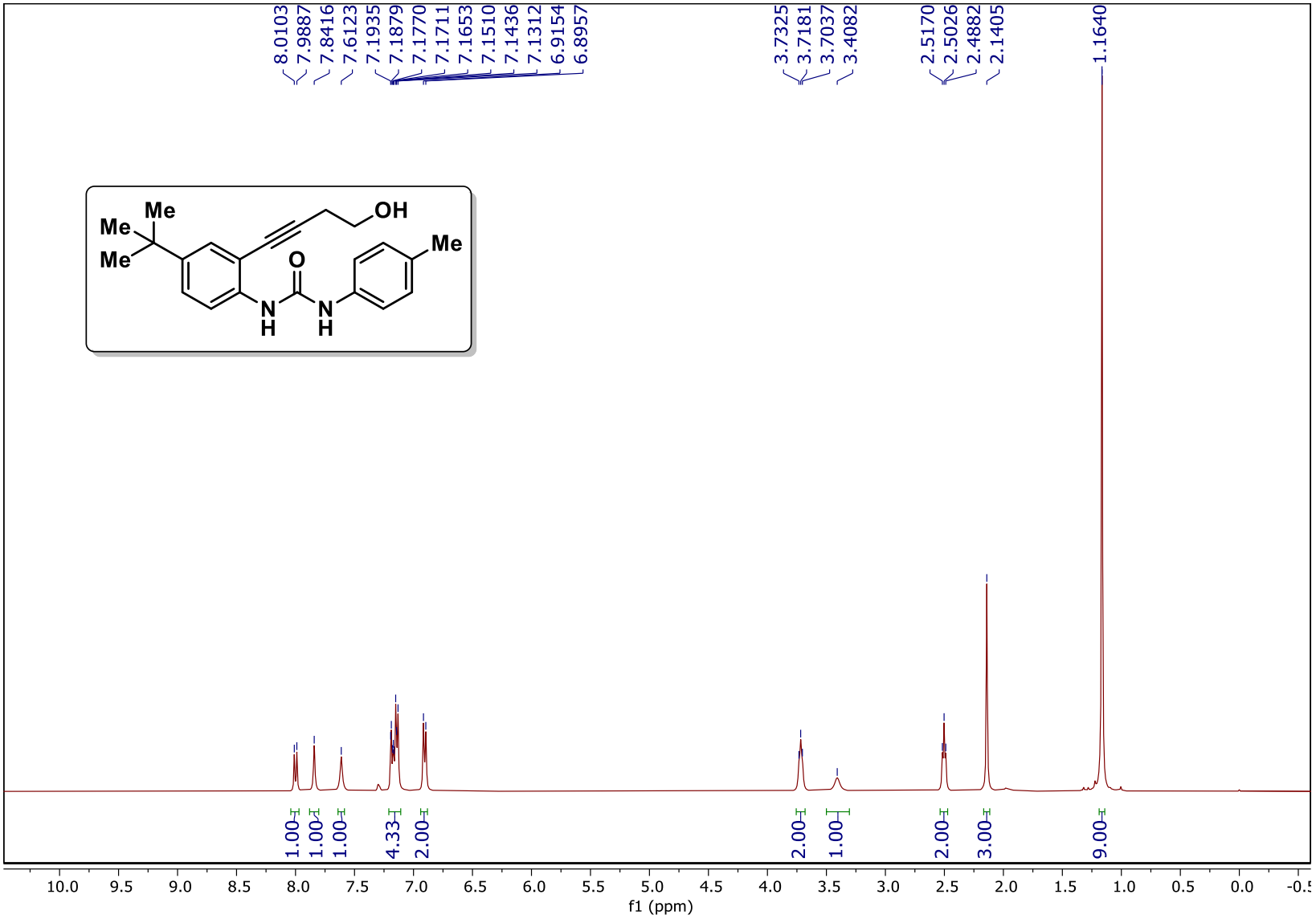
^{1}H spectrum of compound **1e** (400 MHz, $CDCl_3$)



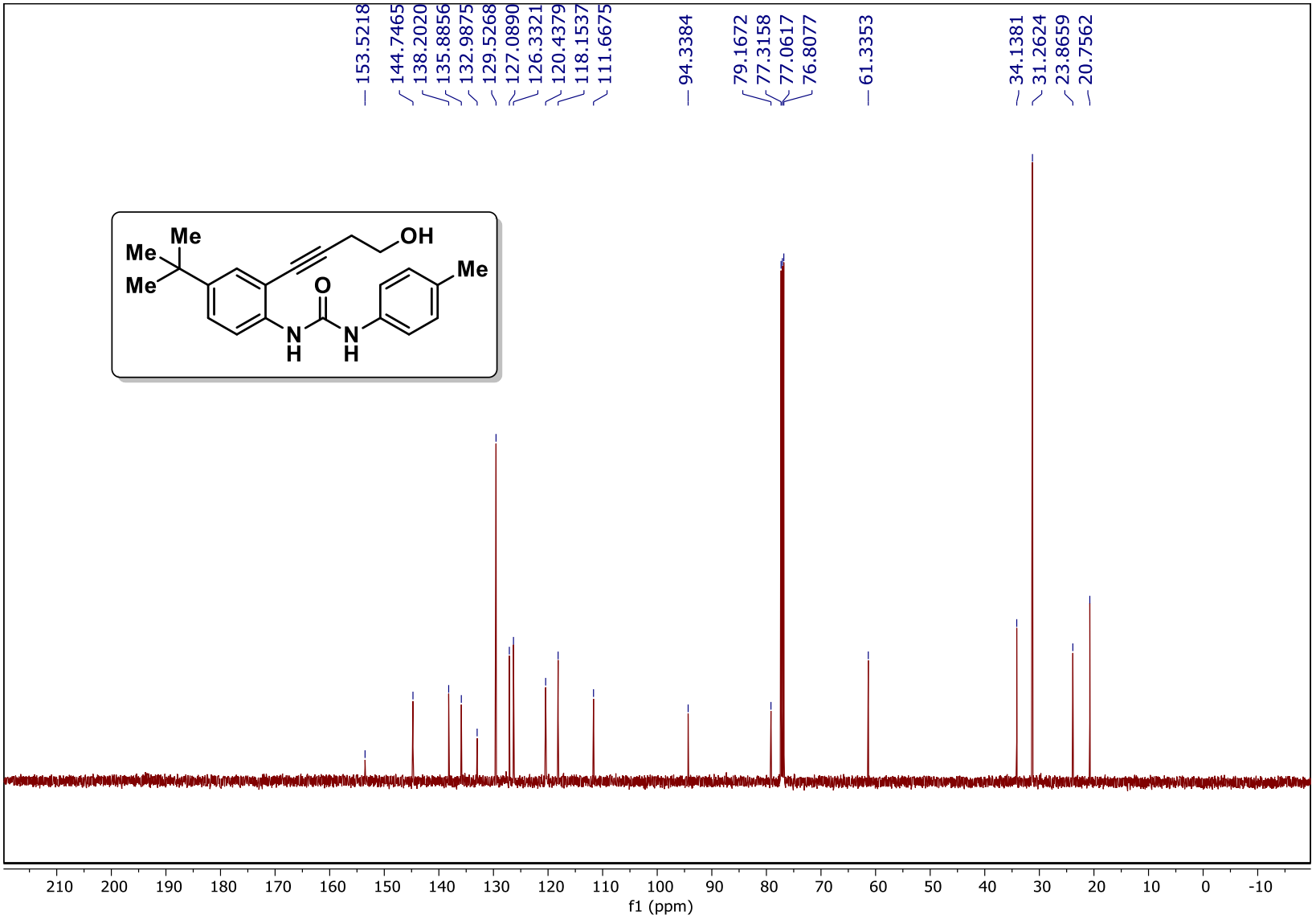
^{13}C spectrum of compound **1e** (125 MHz, $CDCl_3$)



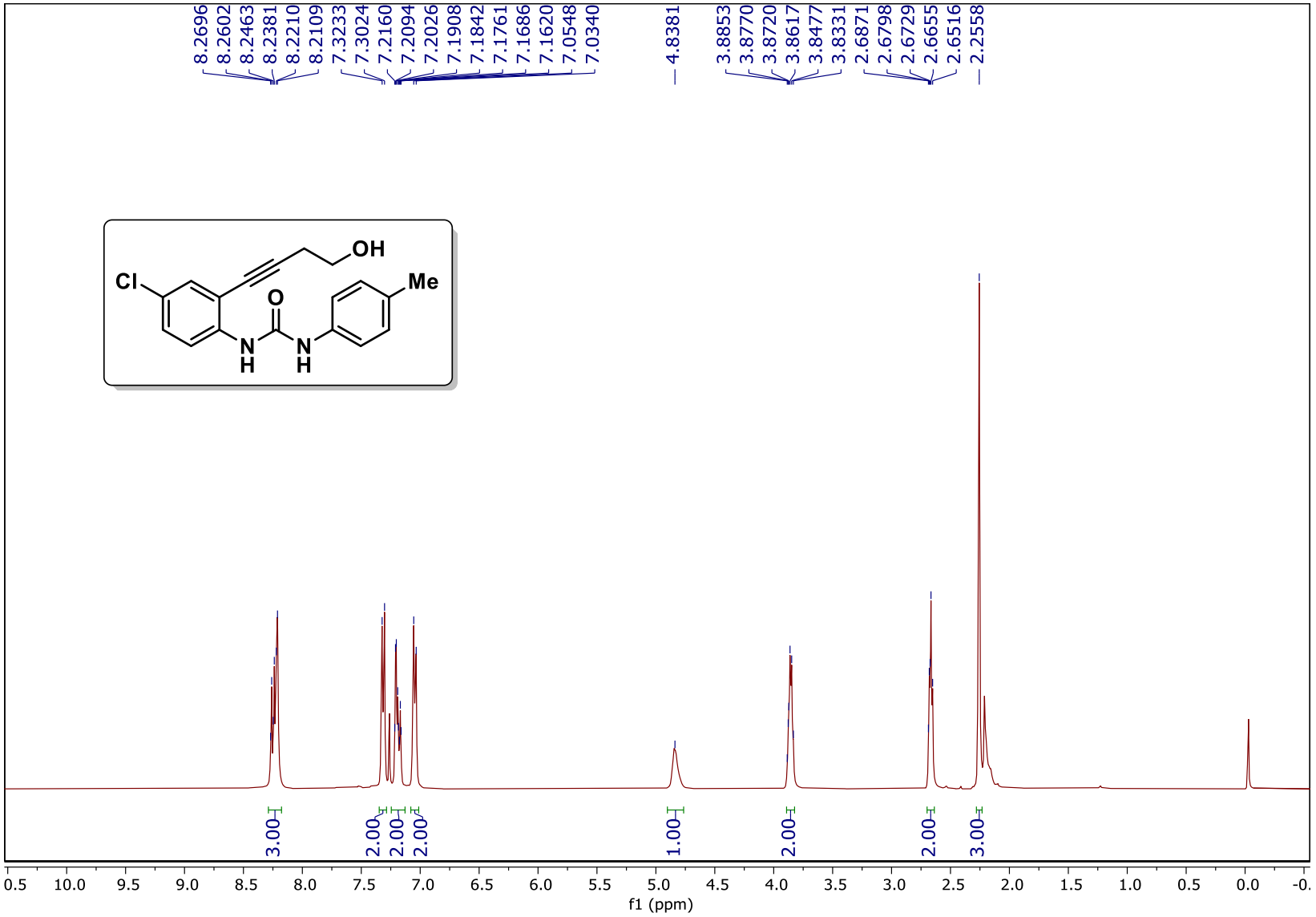
^{1}H spectrum of compound **1f** (400 MHz, $CDCl_3$)



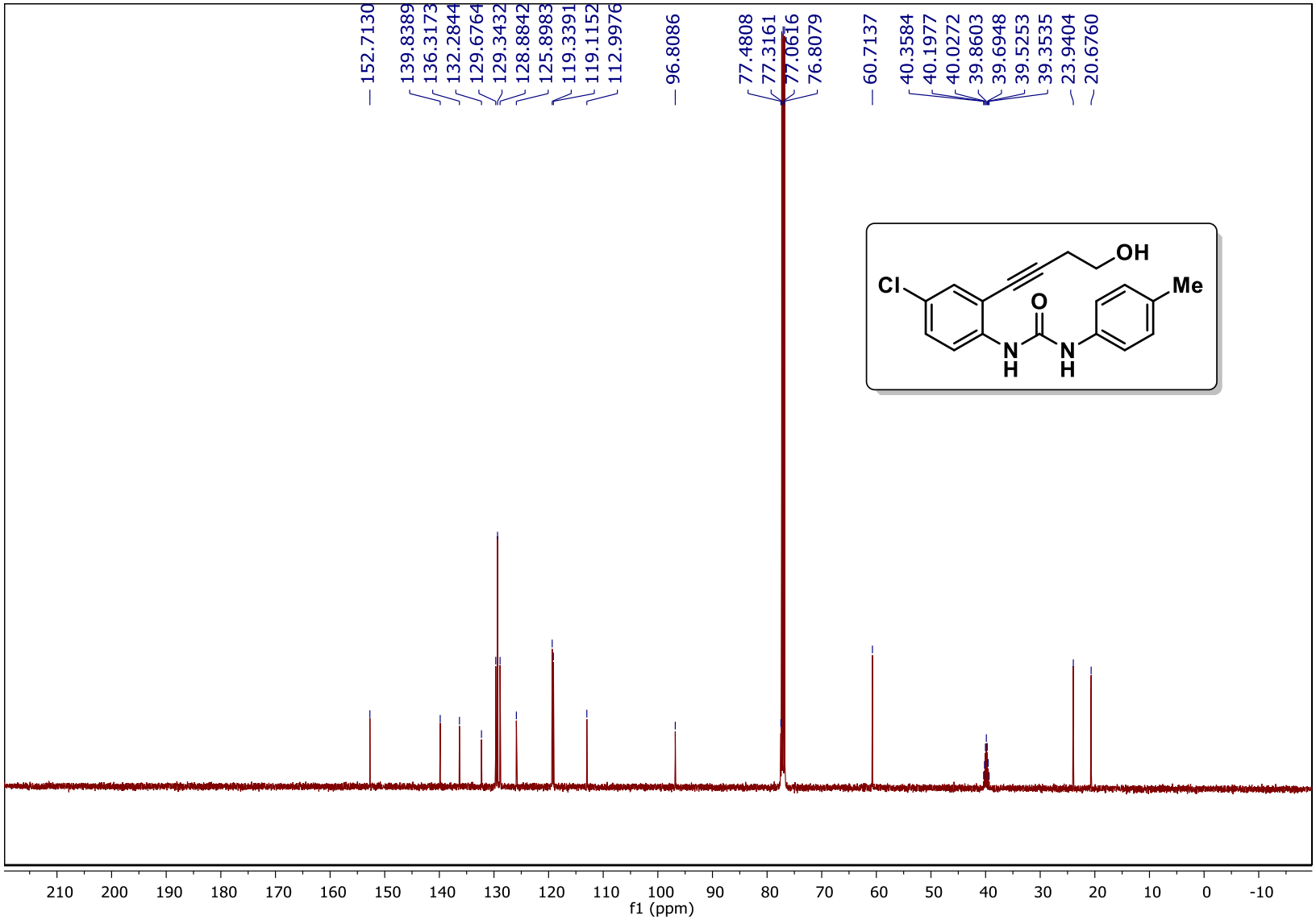
^{13}C spectrum of compound **1f** (125 MHz, $CDCl_3$)



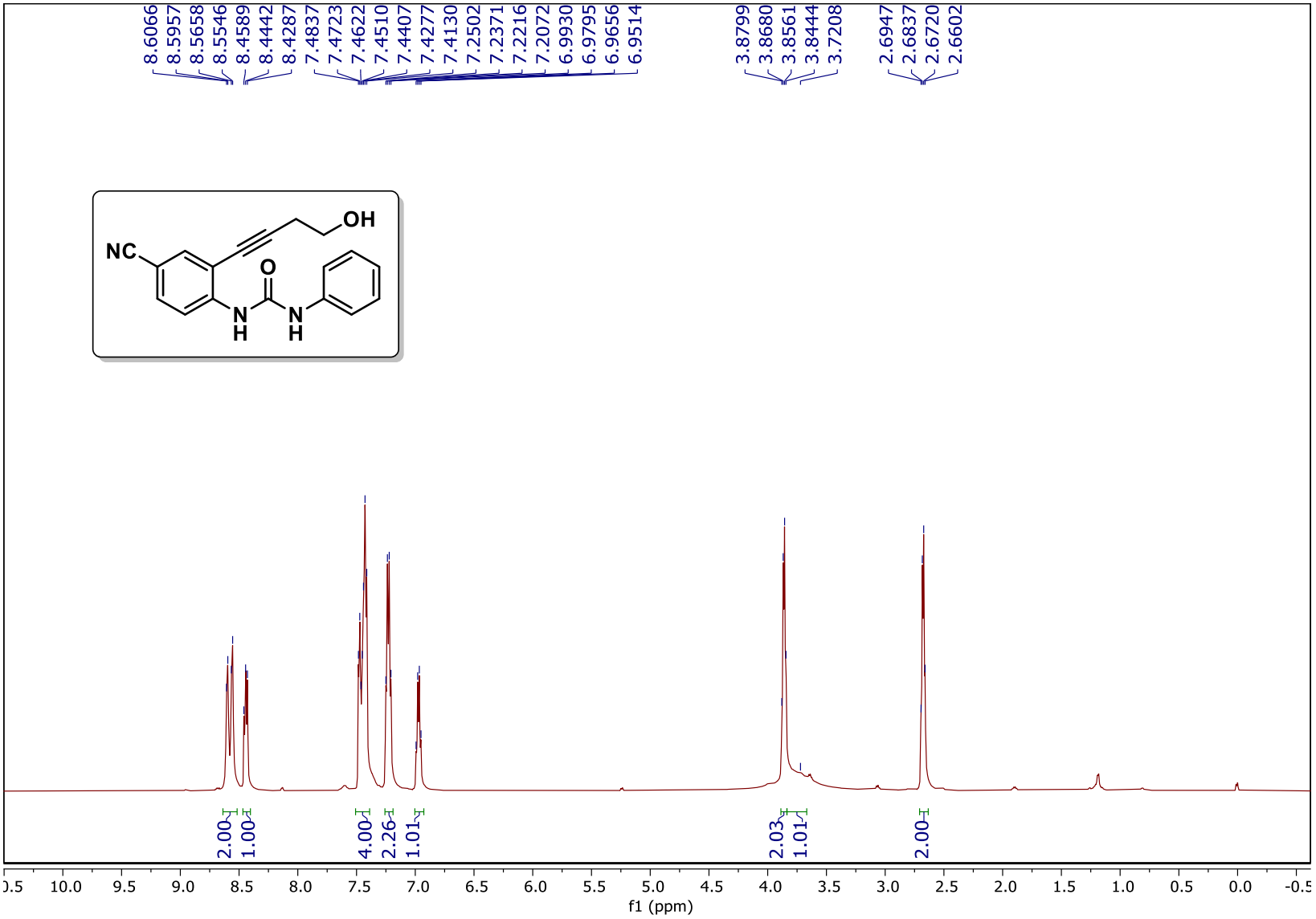
^{1}H spectrum of compound **1g** (400 MHz, $CDCl_3$/DMSO-d_6)



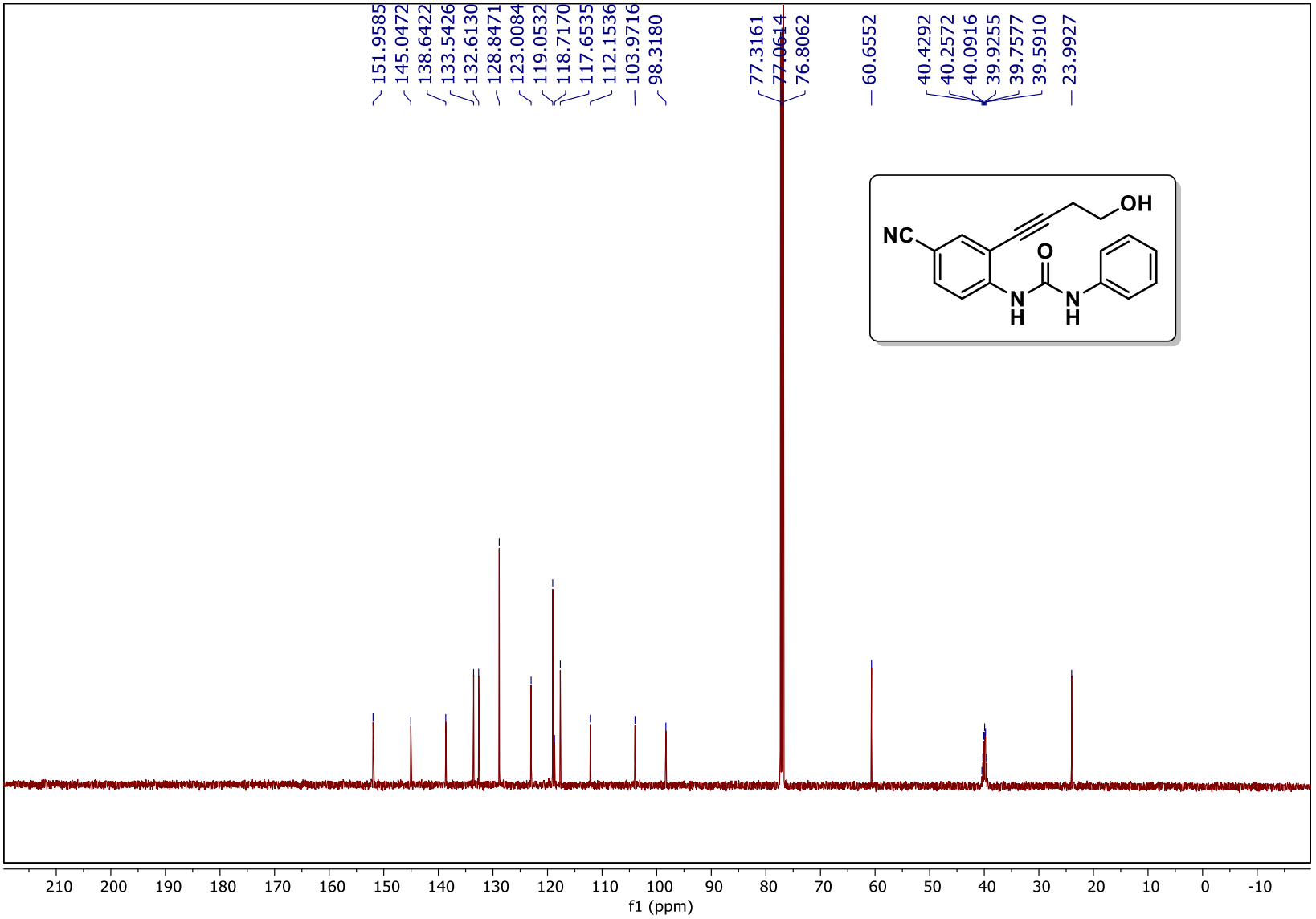
^{13}C spectrum of compound **1g** (125 MHz, $CDCl_3$/DMSO-d_6)



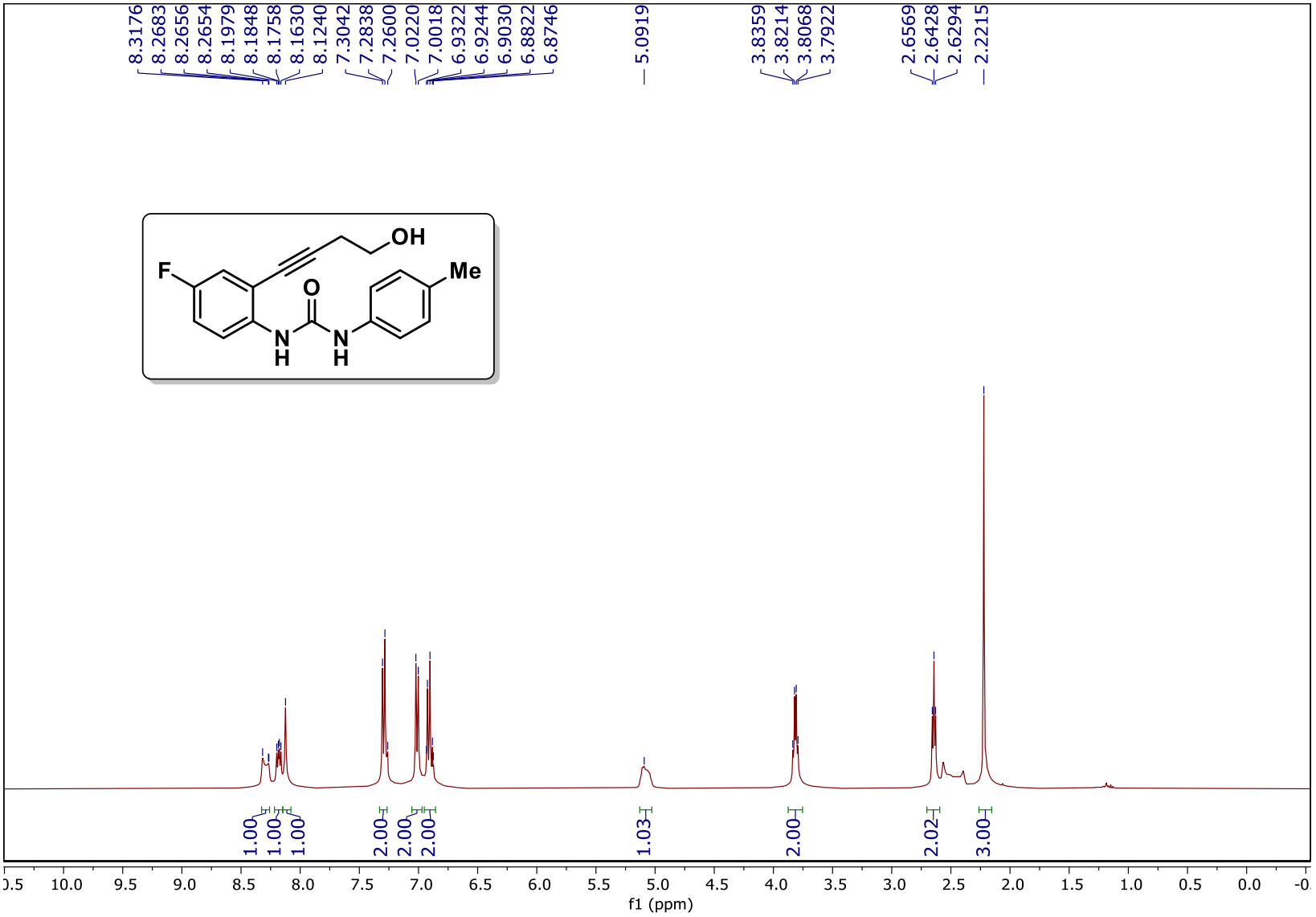
^{1}H spectrum of compound **1h** (500 MHz, $CDCl_3$/DMSO-d_6)



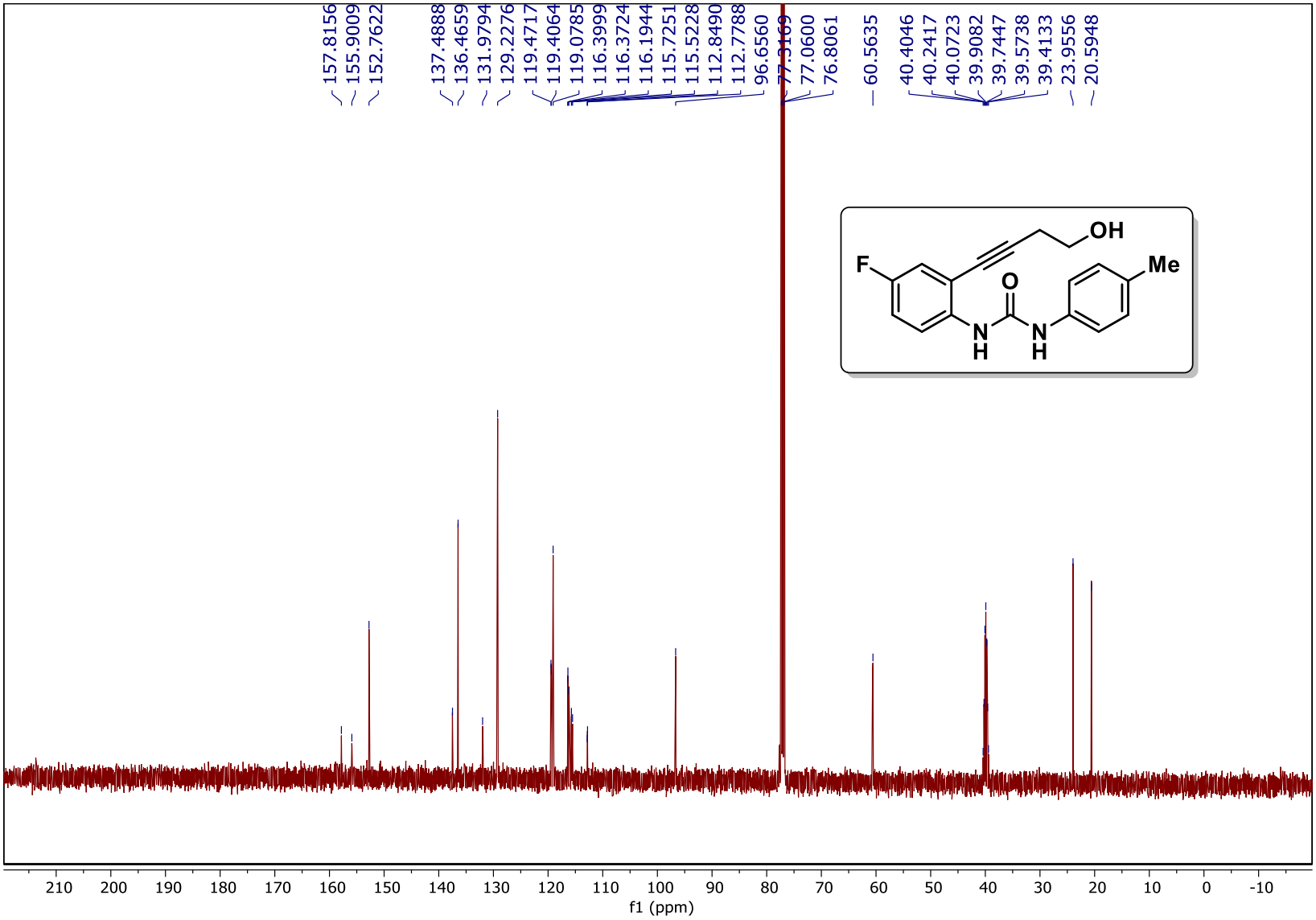
^{13}C spectrum of compound **1h** (125 MHz, $CDCl_3$/DMSO-d_6)



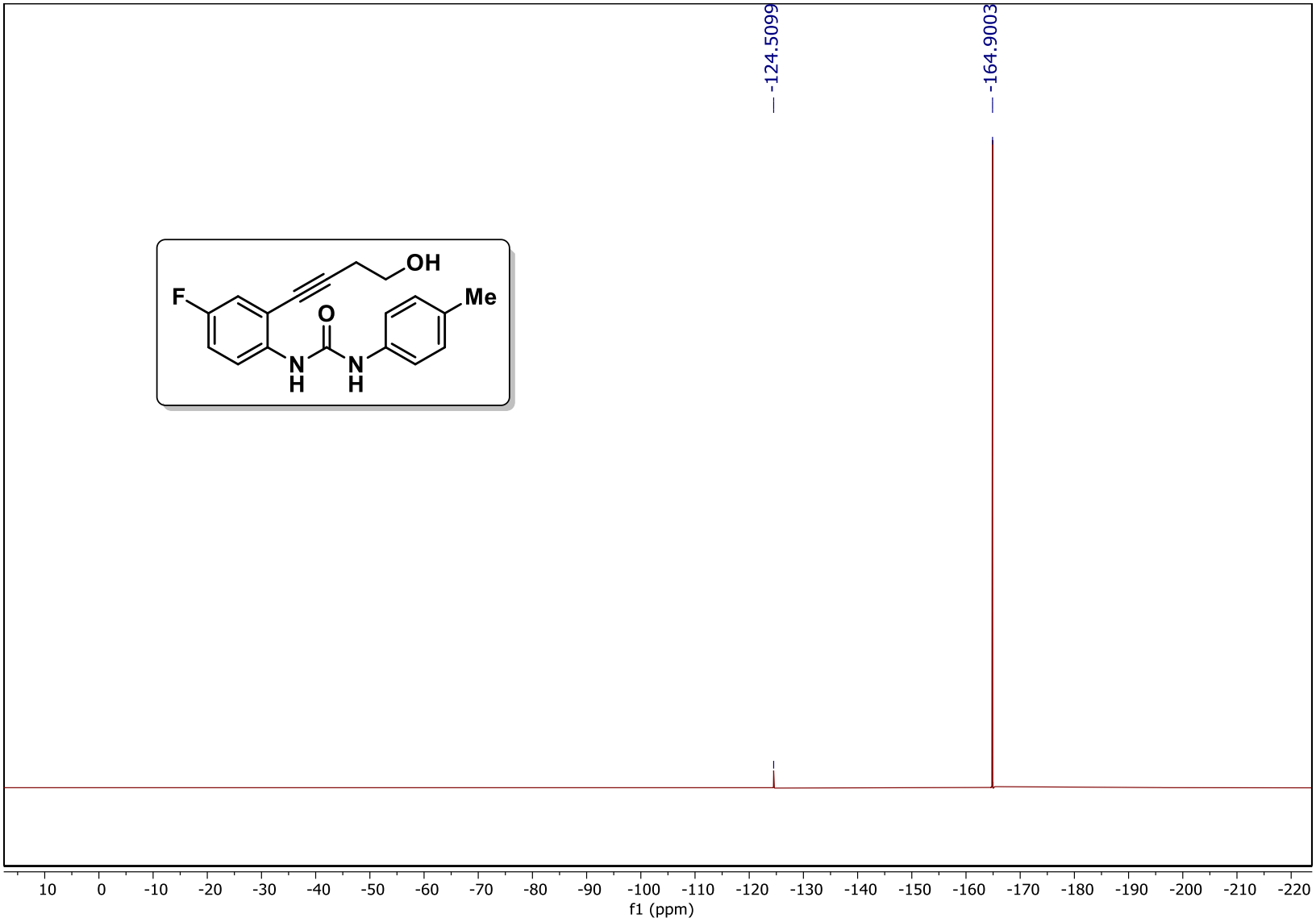
^{1}H spectrum of compound **1i** (400 MHz, $CDCl_3$/DMSO-d_6)



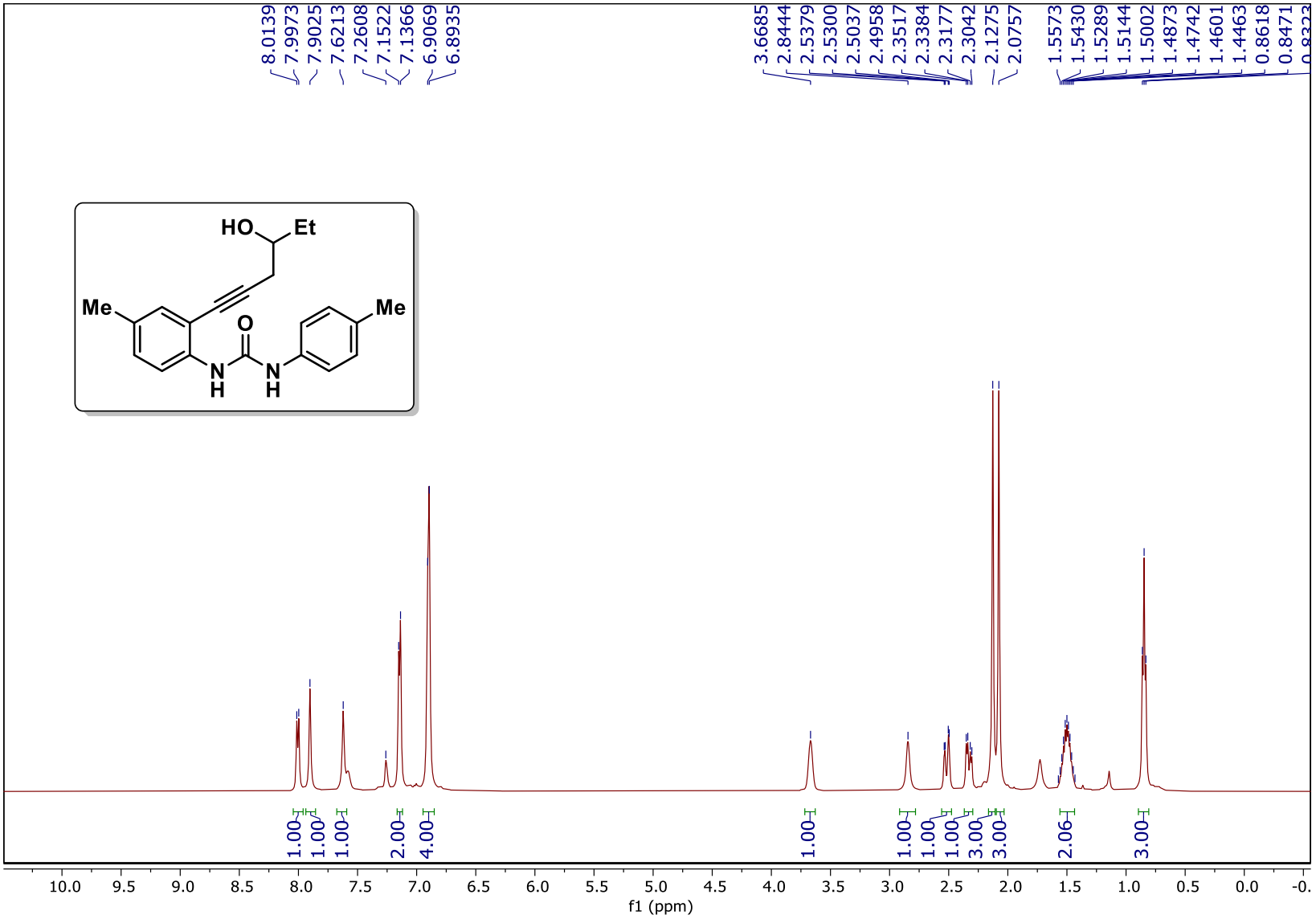
^{13}C spectrum of compound **1i** (125 MHz, $CDCl_3$/DMSO-d_6)



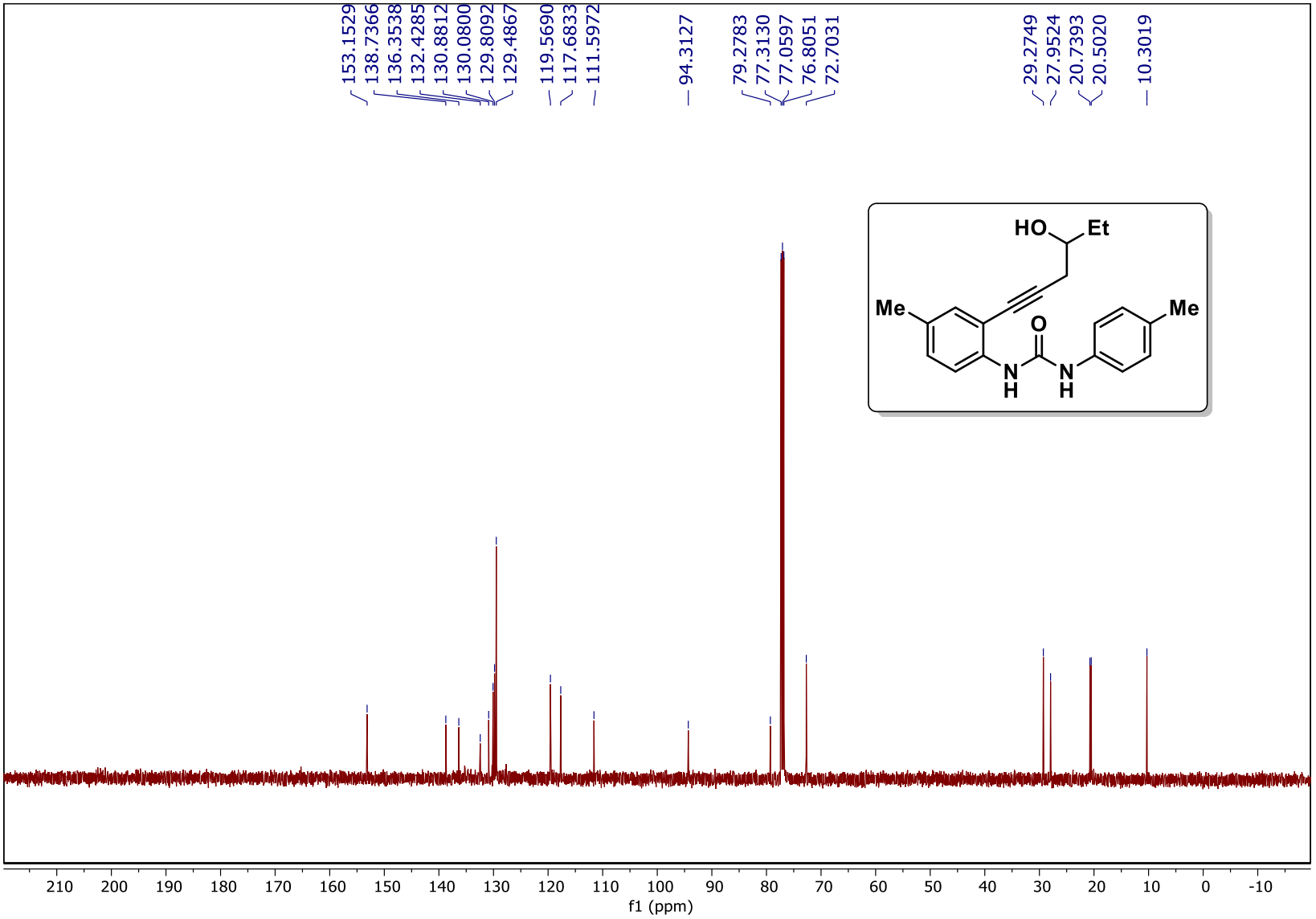
^{19}F spectrum of compound **1i** (470 MHz, $CDCl_3/C_6F_6$)



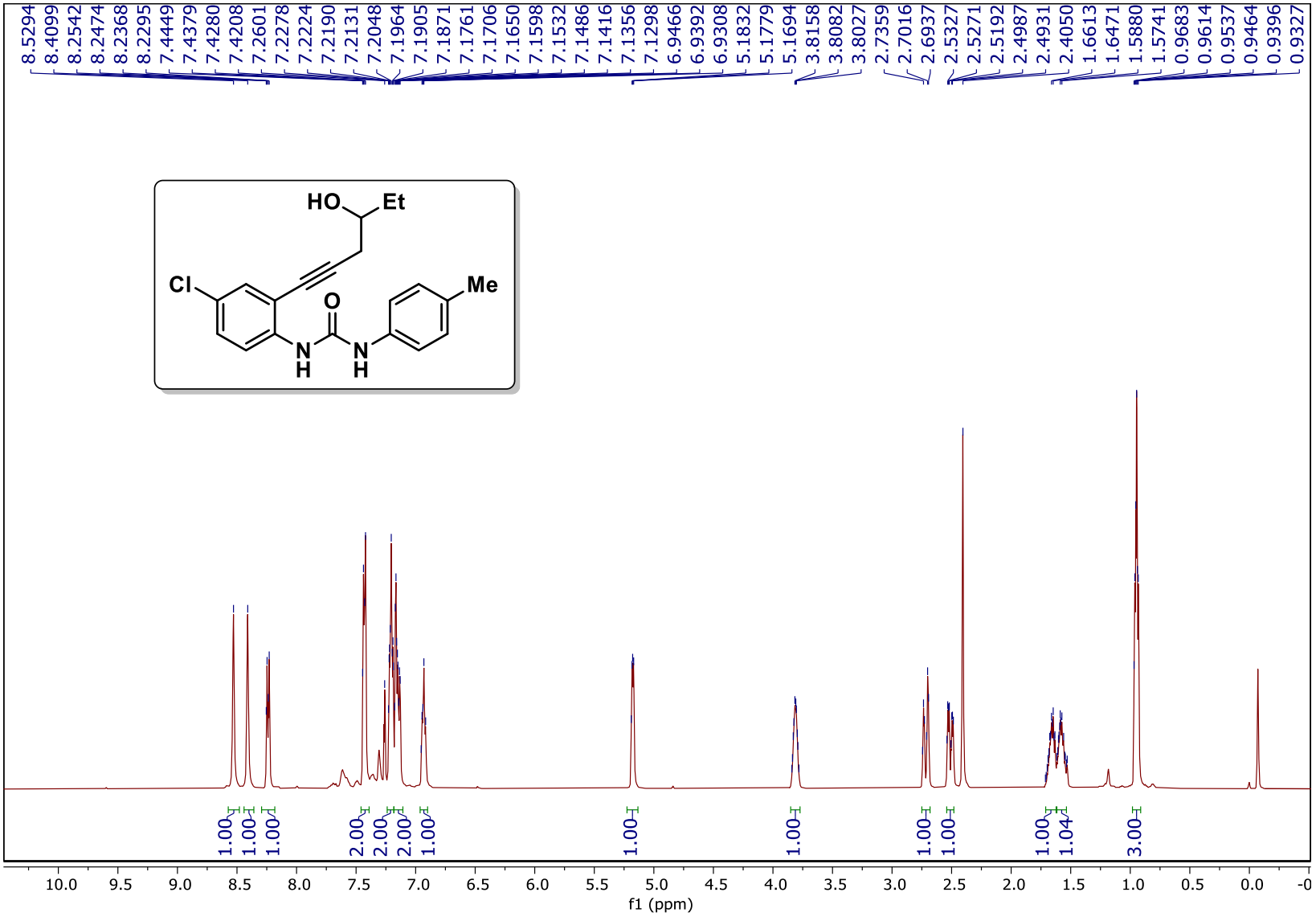
^{1}H spectrum of compound **1j** (500 MHz, $CDCl_3$)



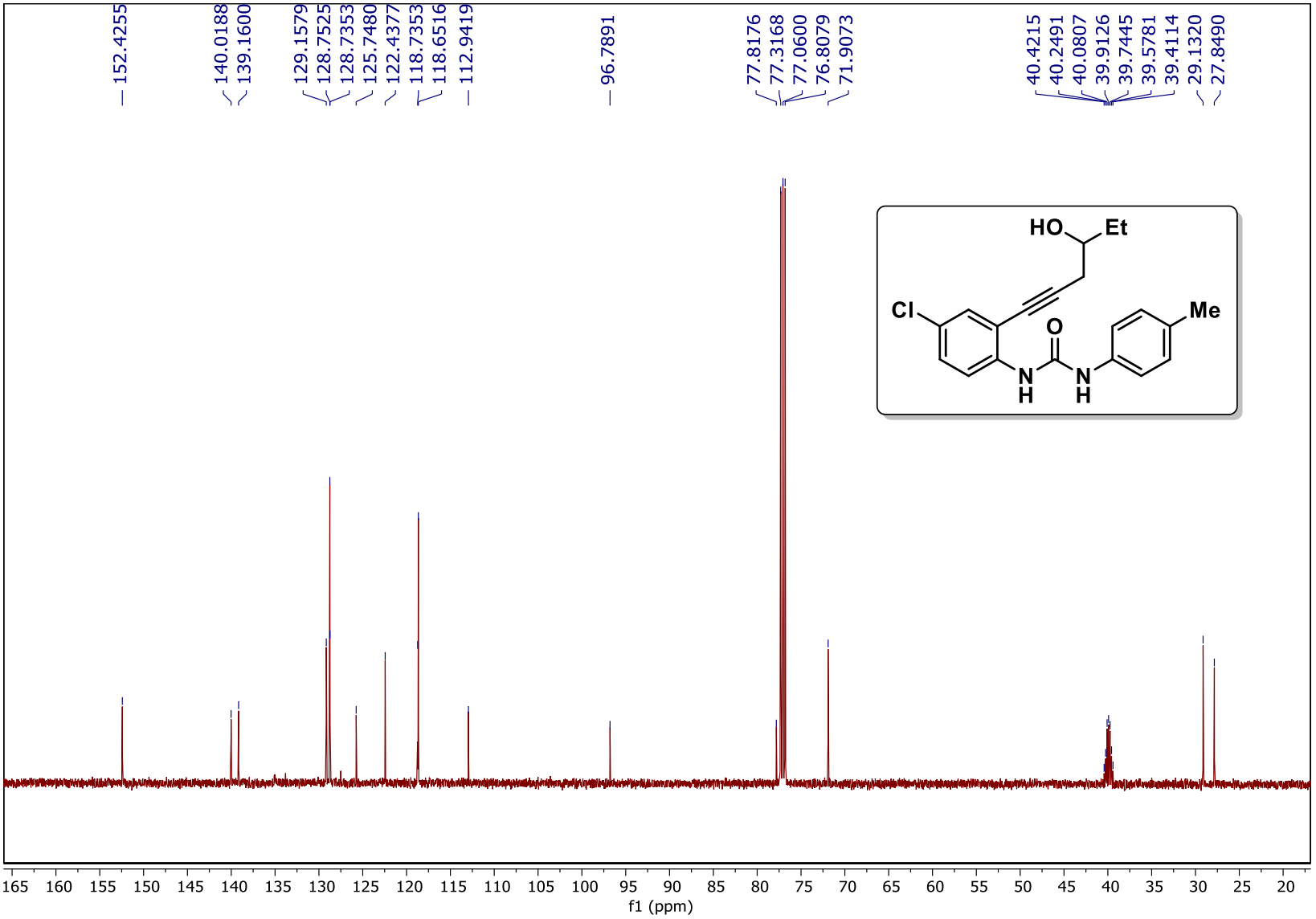
^{13}C spectrum of compound **1j** (125 MHz, $CDCl_3$)



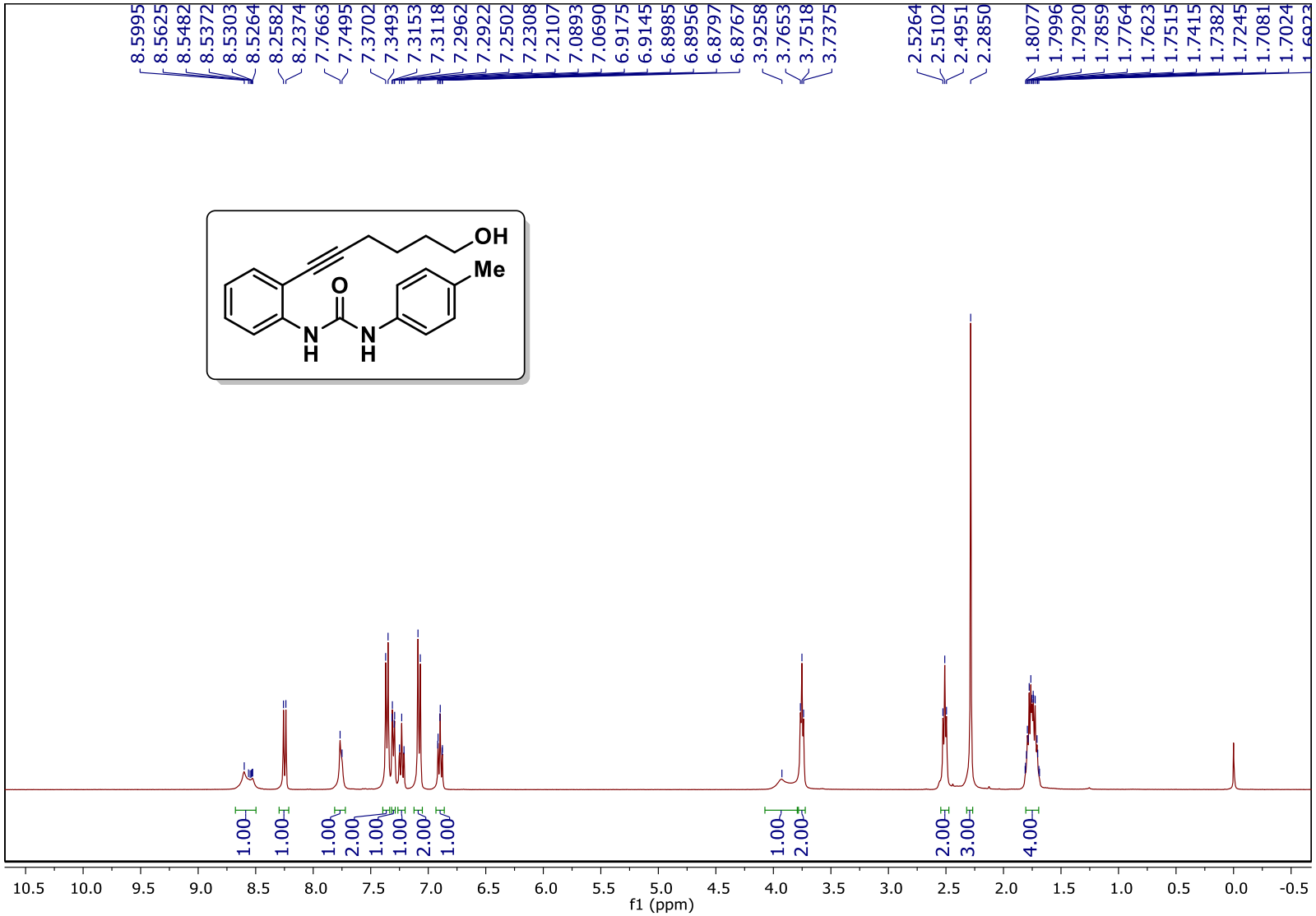
^{1}H spectrum of compound **1k** (500 MHz, $CDCl_3$/DMSO-d_6)



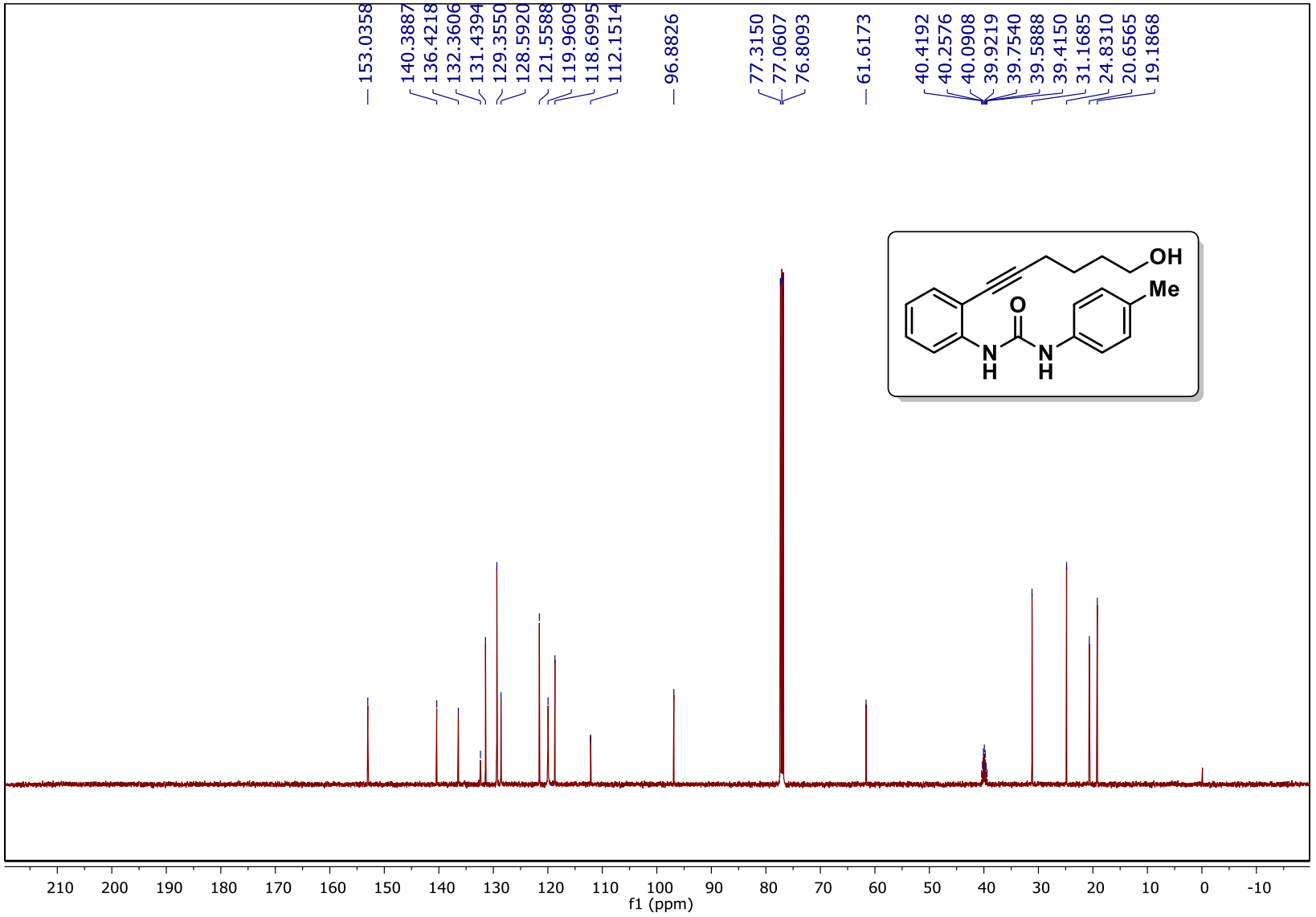
^{13}C spectrum of compound **1k** (125 MHz, $CDCl_3$/DMSO-d_6)



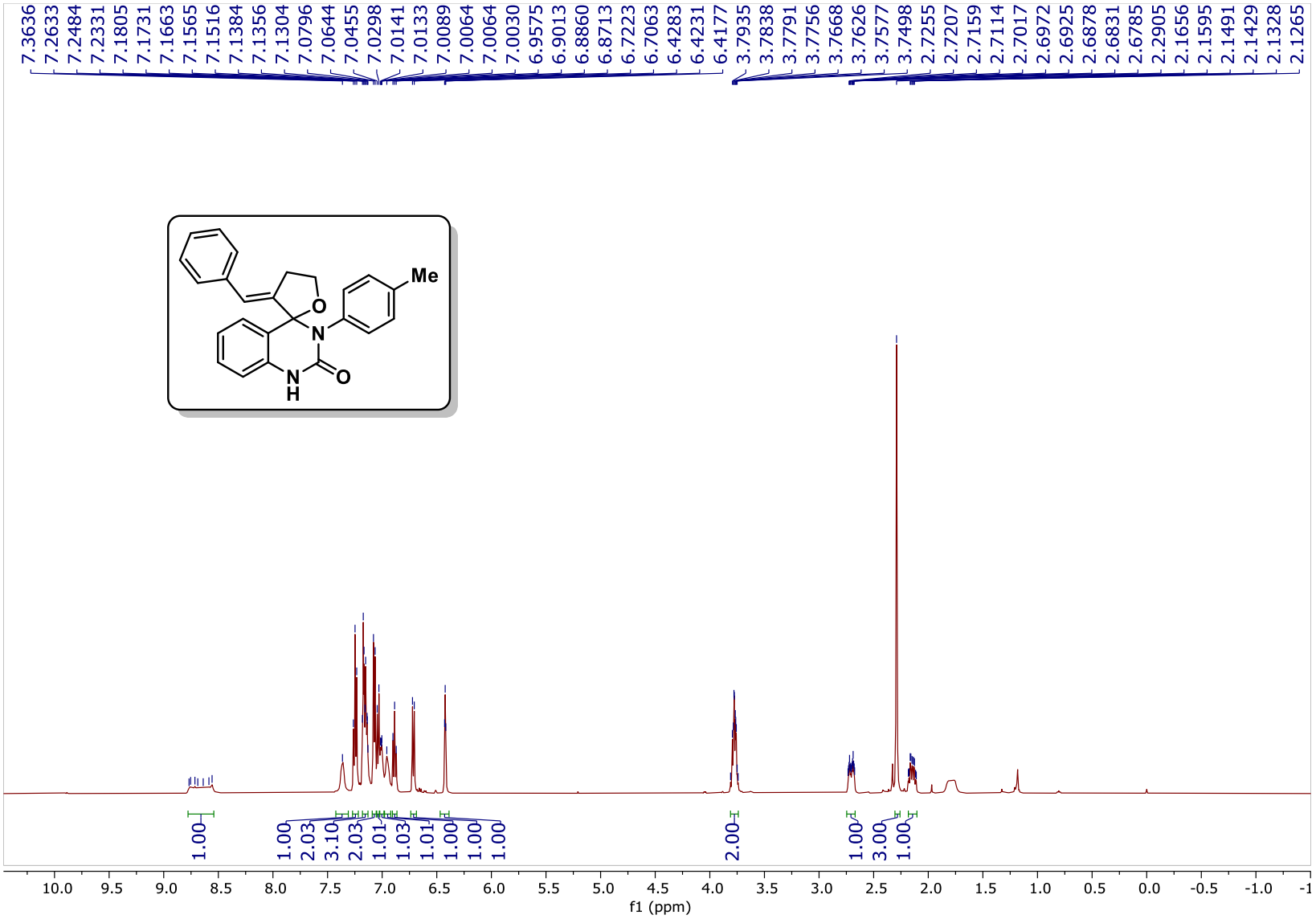
^{1}H spectrum of compound **1l** (400 MHz, $CDCl_3$/DMSO-d_6)



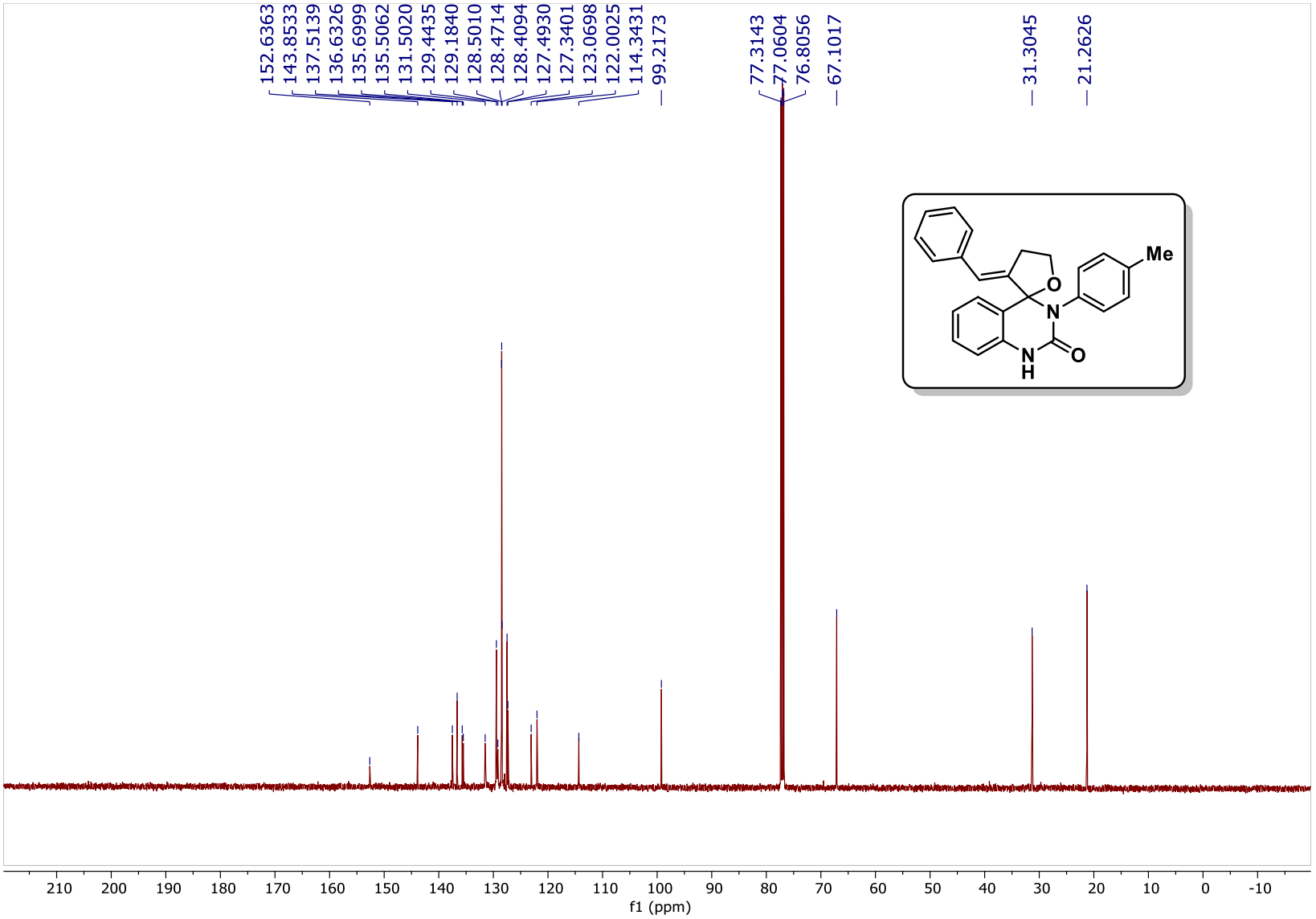
^{13}C spectrum of compound **1l** (125 MHz, $CDCl_3$/DMSO-d_6)



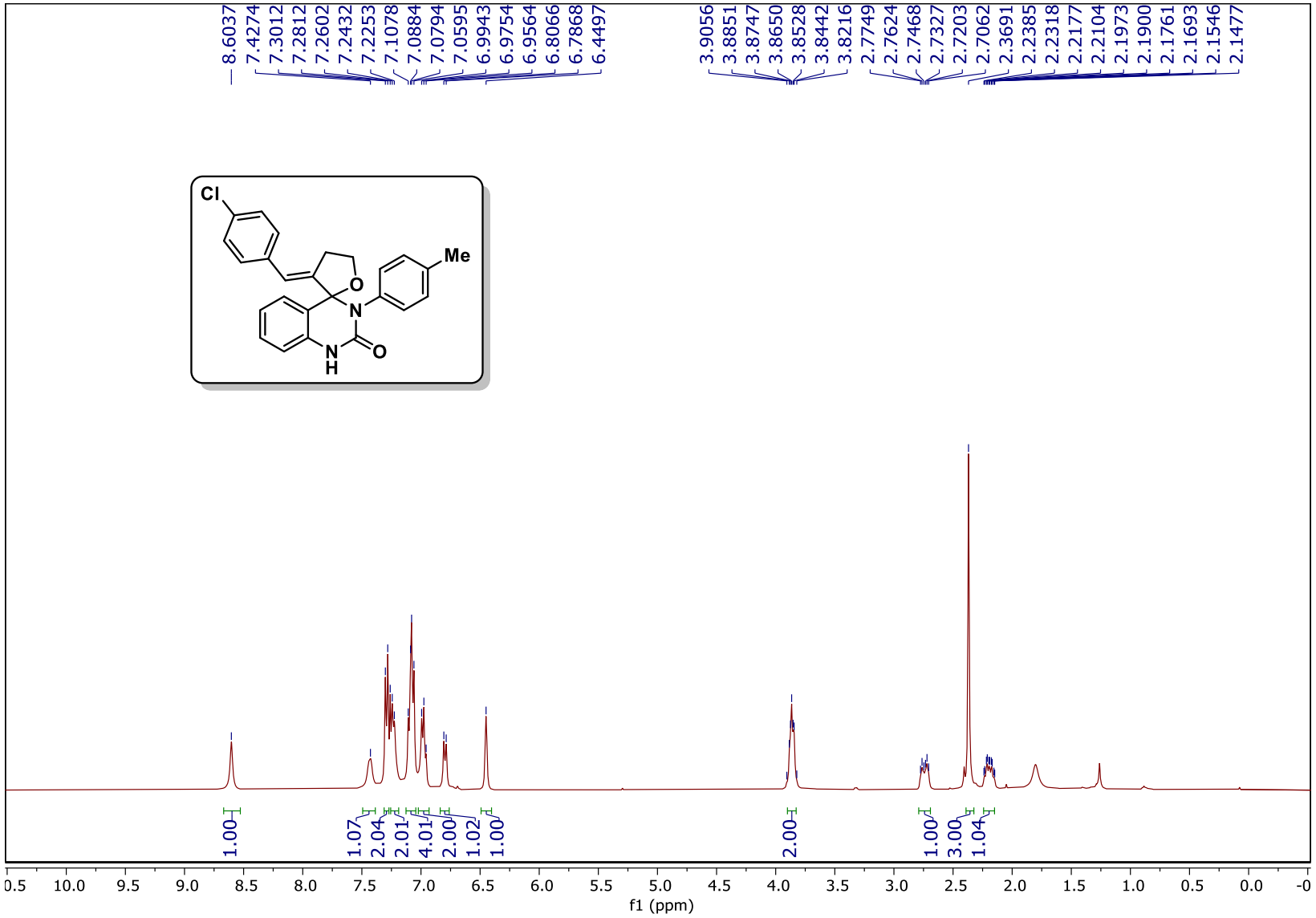
^{1}H spectrum of compound **3aa** (500 MHz, $CDCl_3$)



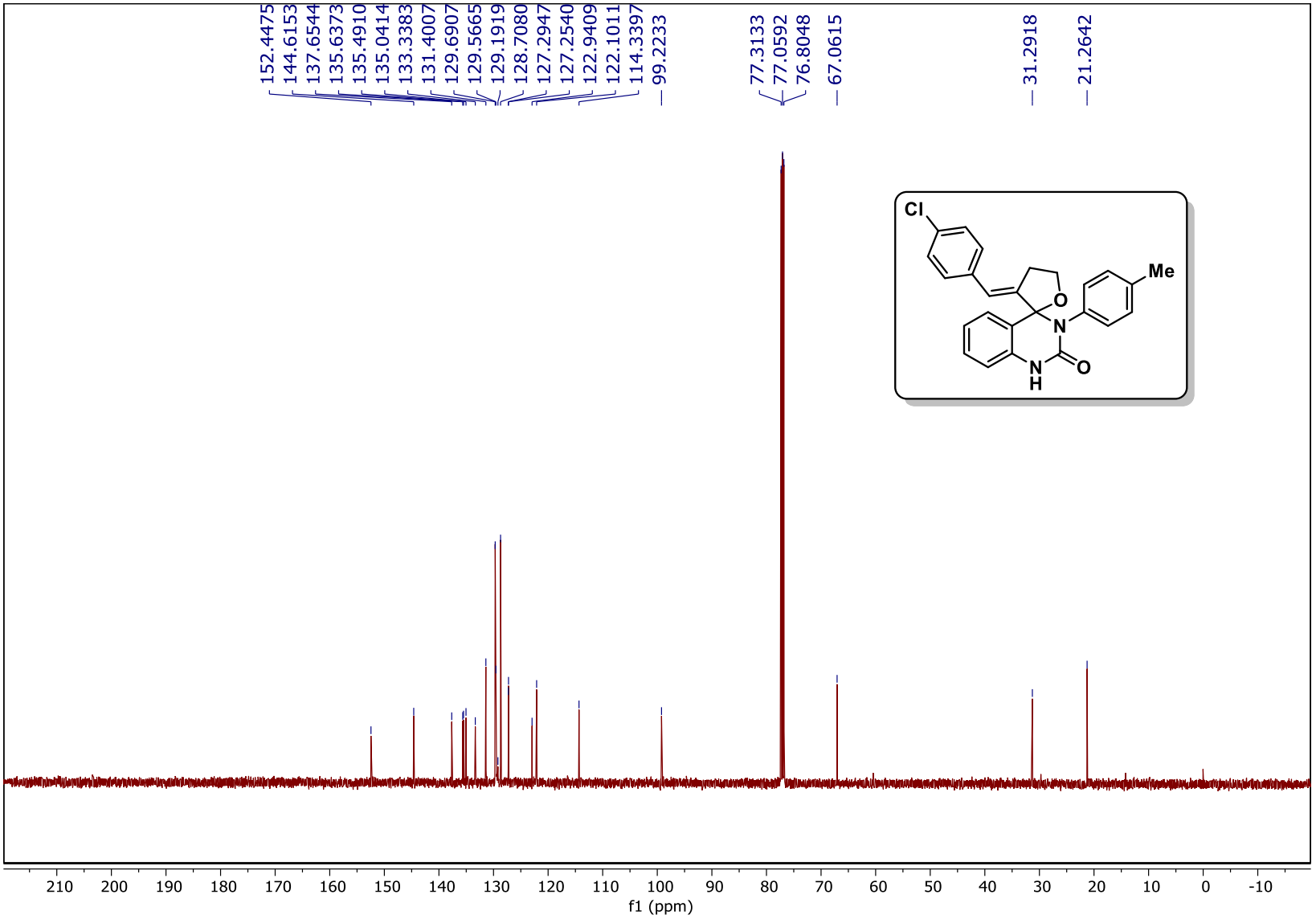
^{13}C spectrum of compound **3aa** (125 MHz, $CDCl_3$)



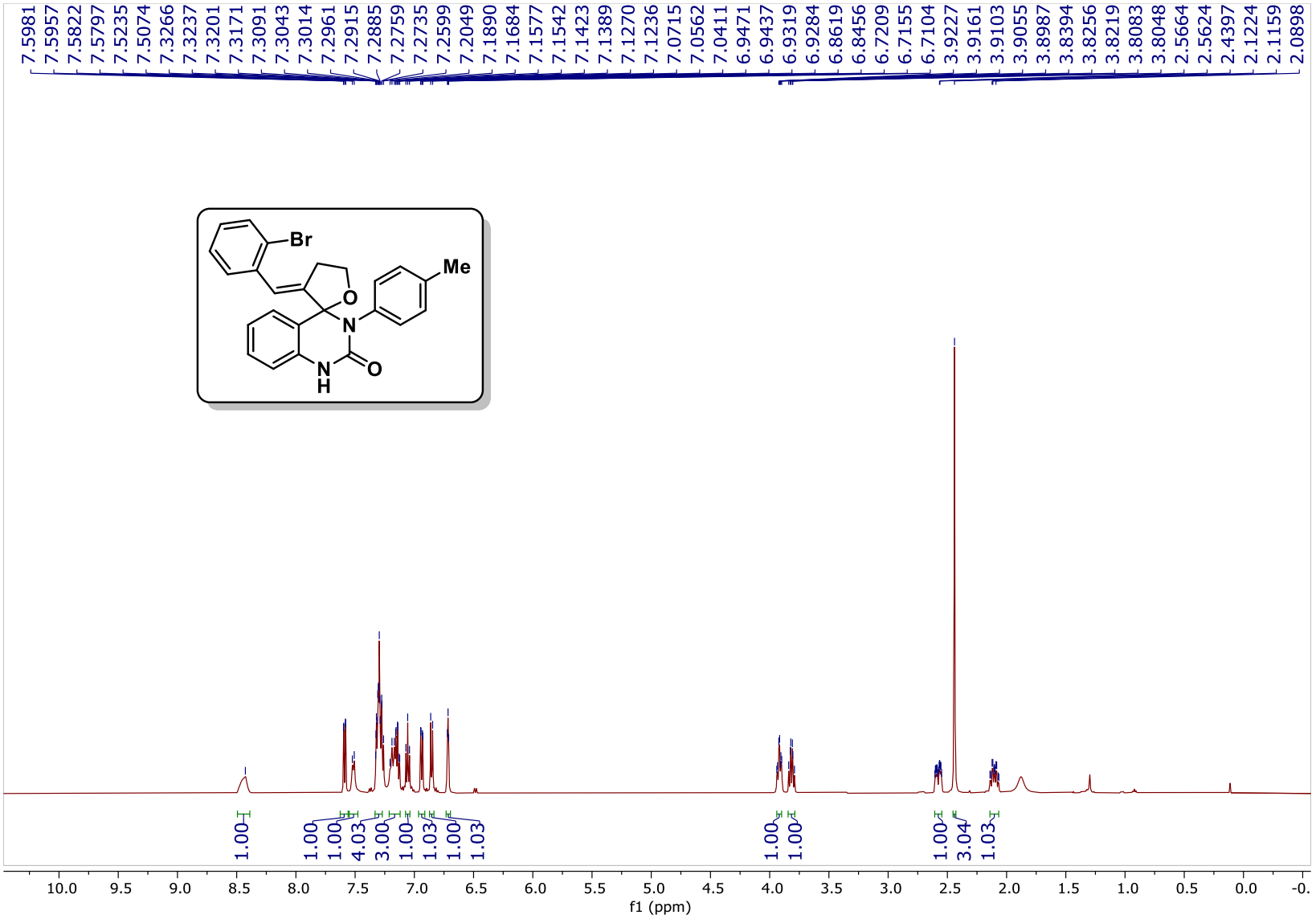
^{1}H spectrum of compound **3ab** (400 MHz, $CDCl_3$)



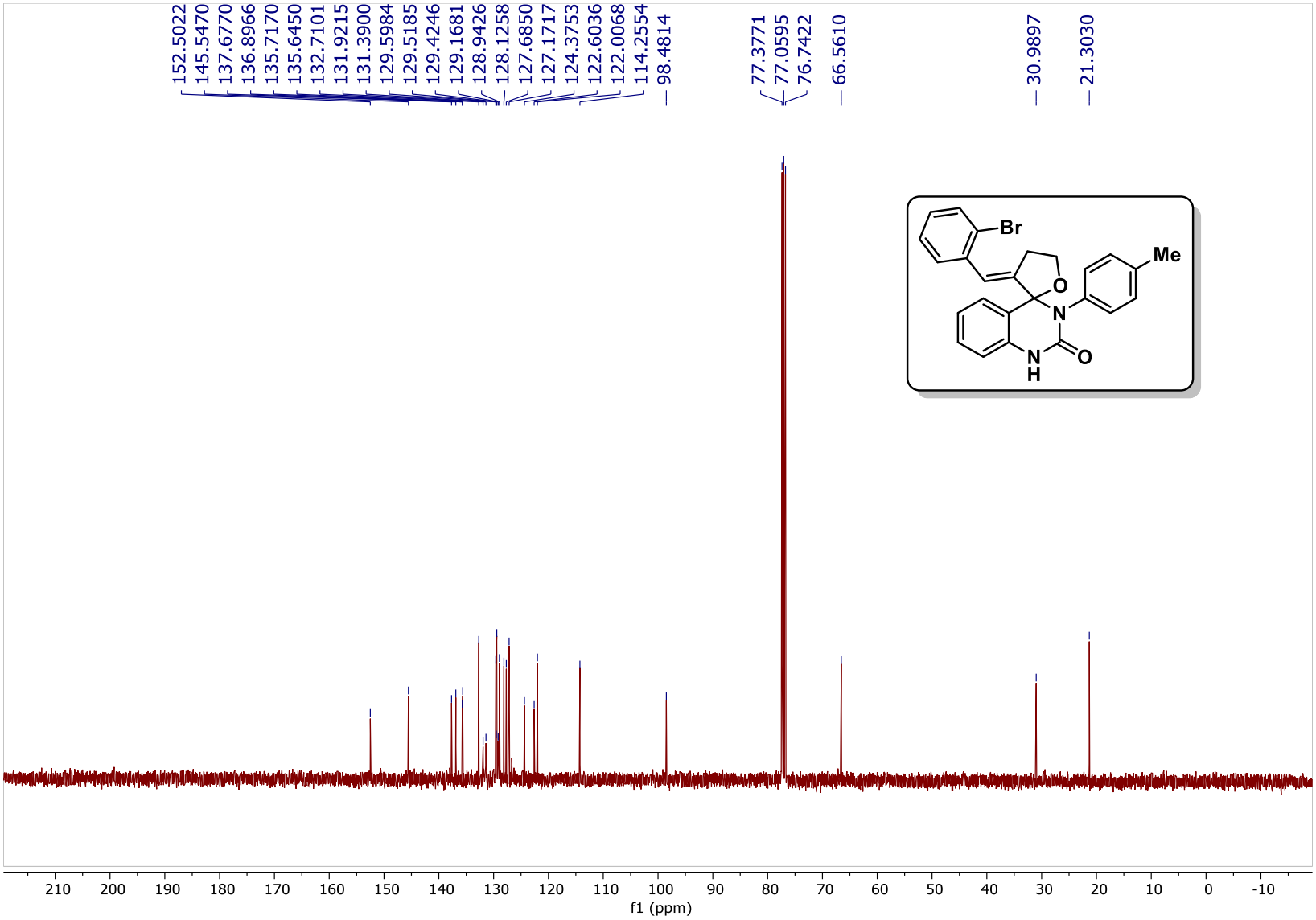
^{13}C spectrum of compound **3ab** (125 MHz, $CDCl_3$)



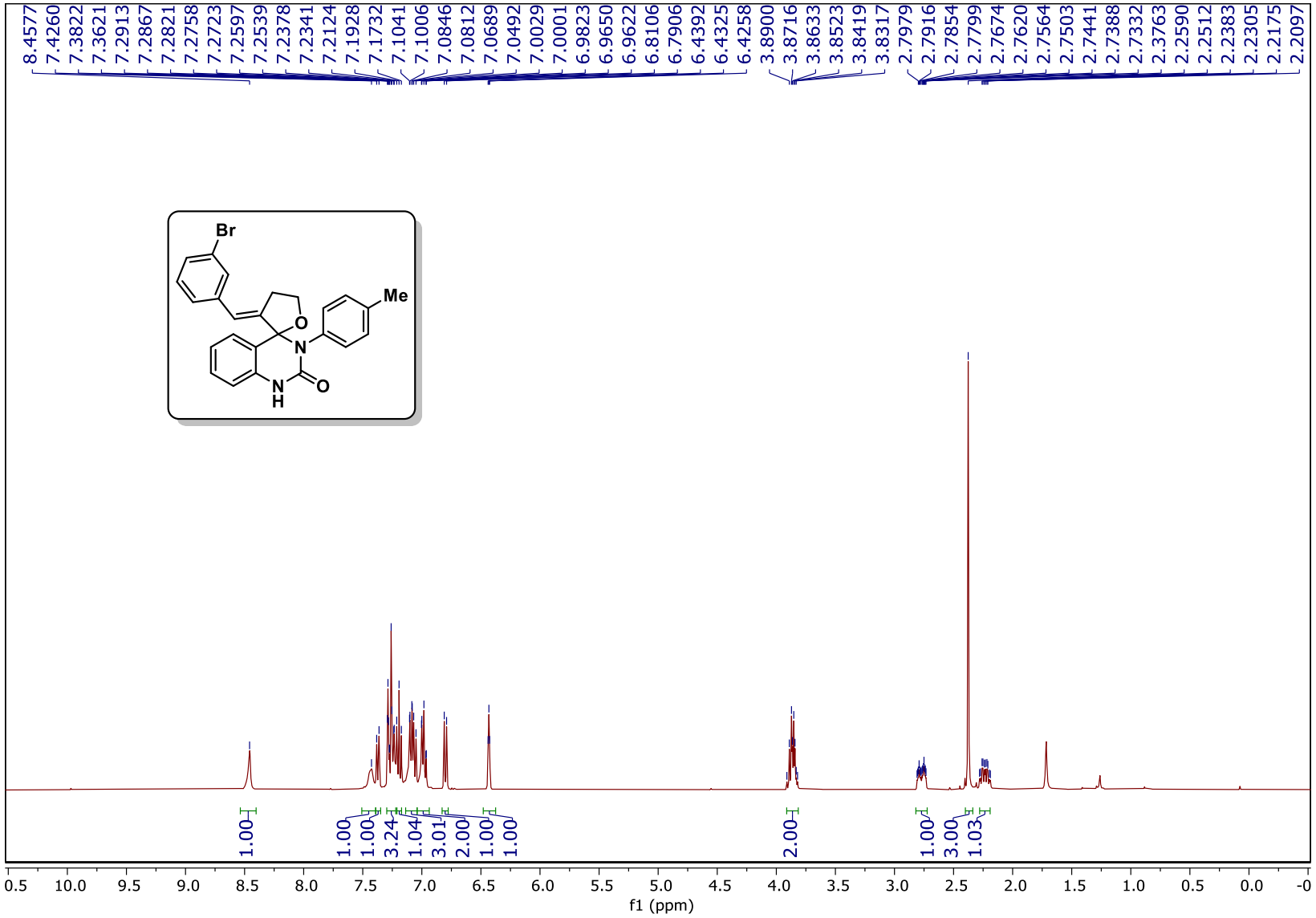
^{1}H spectrum of compound **3ac** (500 MHz, $CDCl_3$)



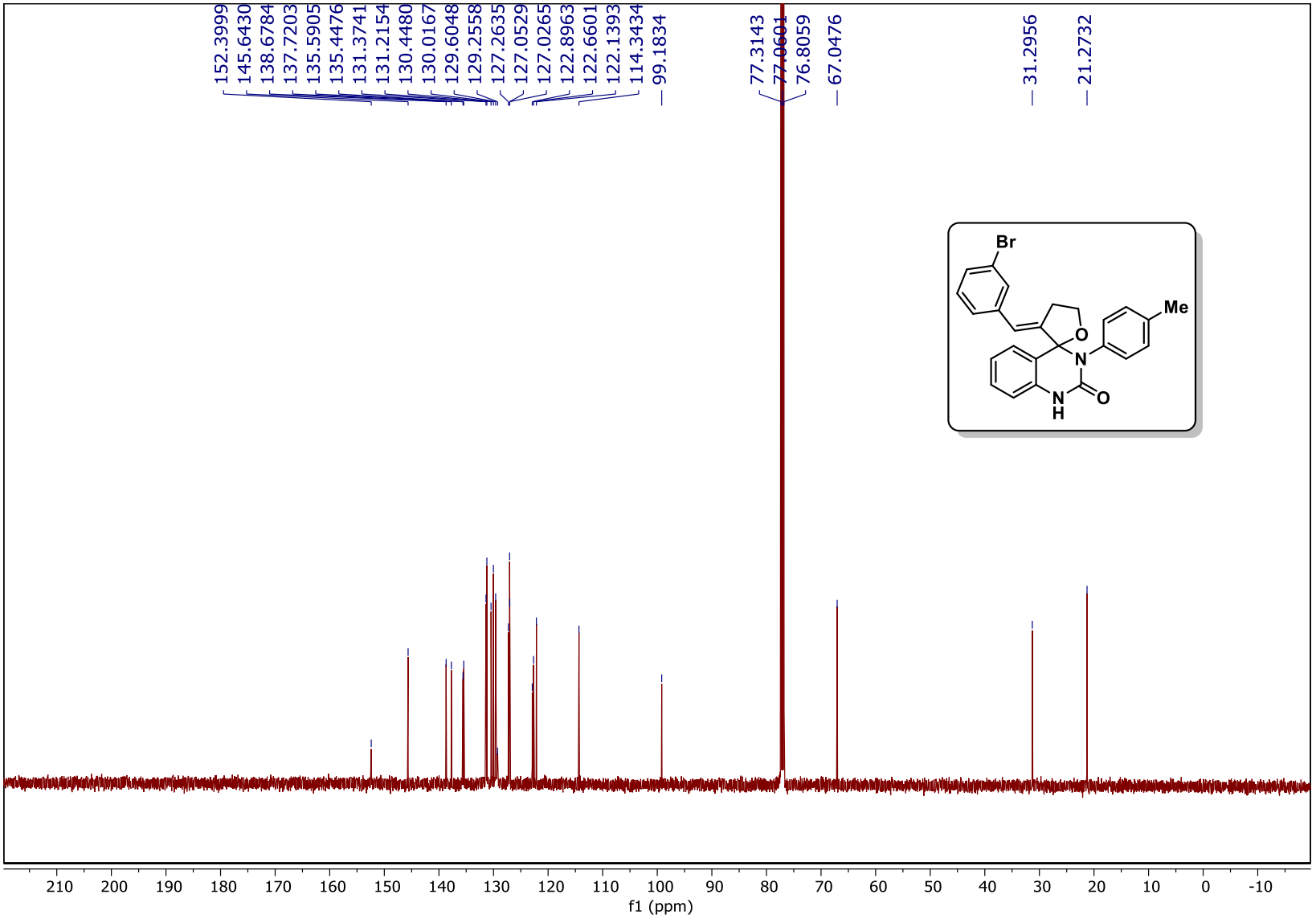
^{13}C spectrum of compound **3ac** (100 MHz, $CDCl_3$)



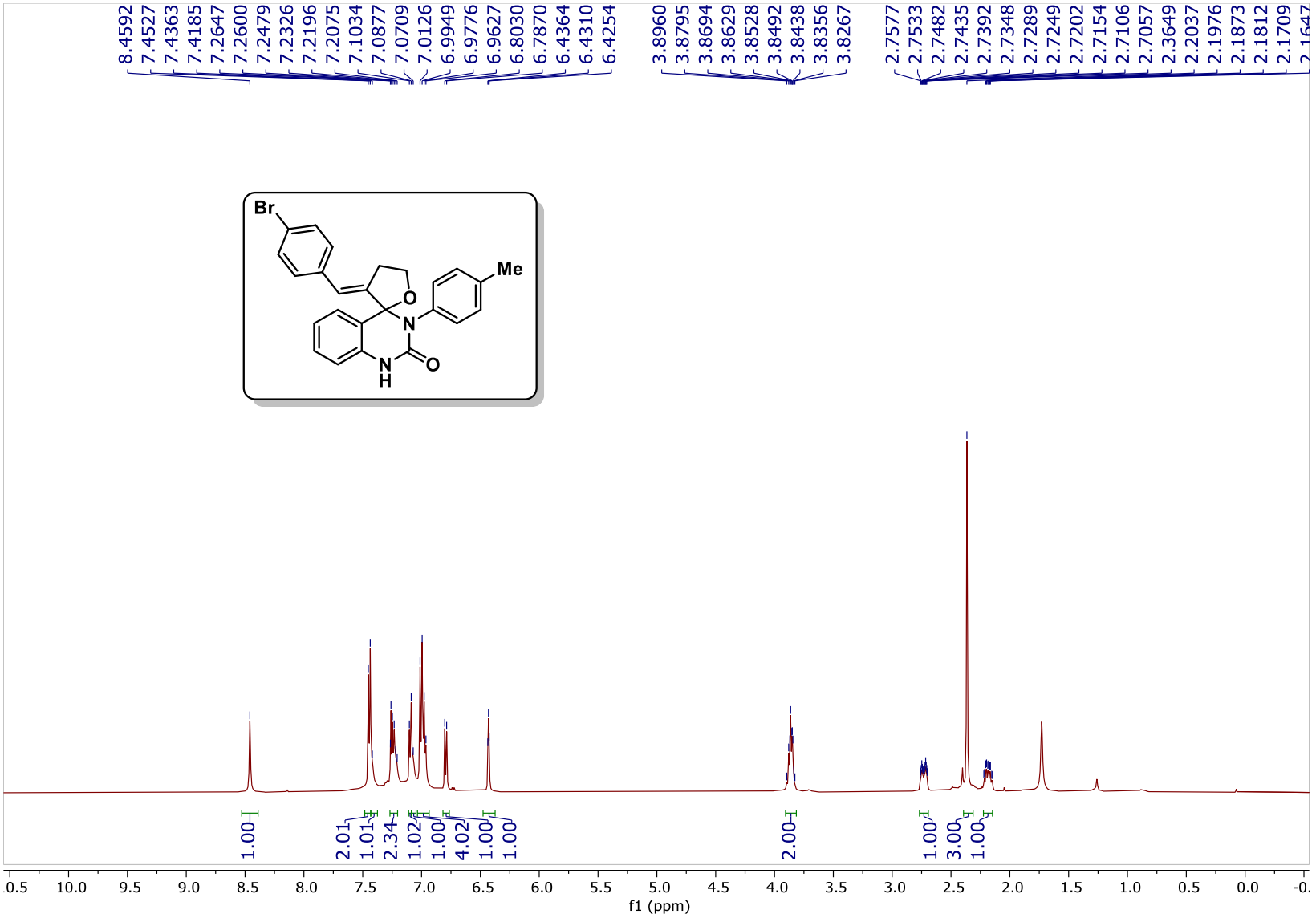
^{1}H spectrum of compound **3ad** (400 MHz, $CDCl_3$)



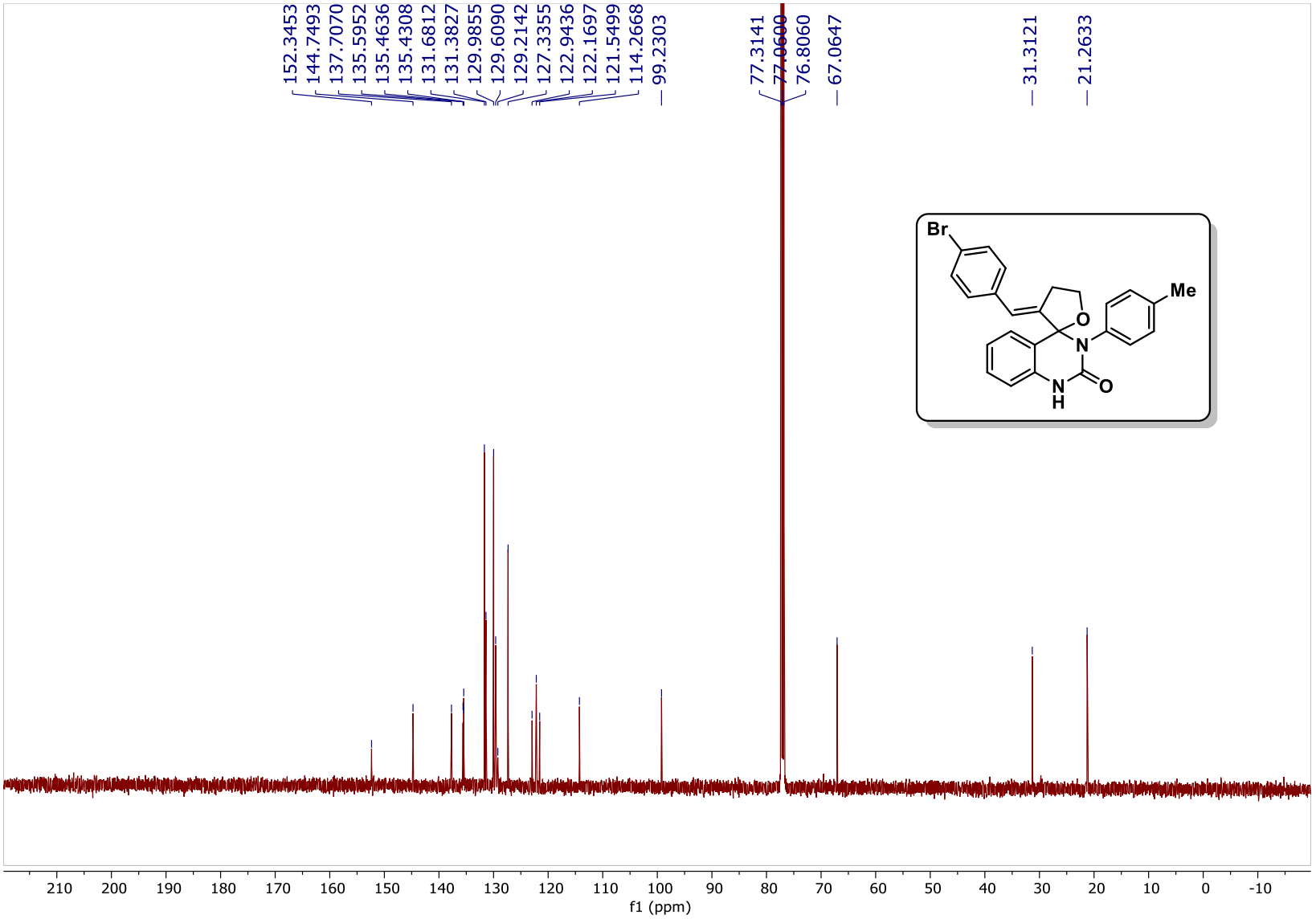
^{13}C spectrum of compound **3ad** (125 MHz, $CDCl_3$)



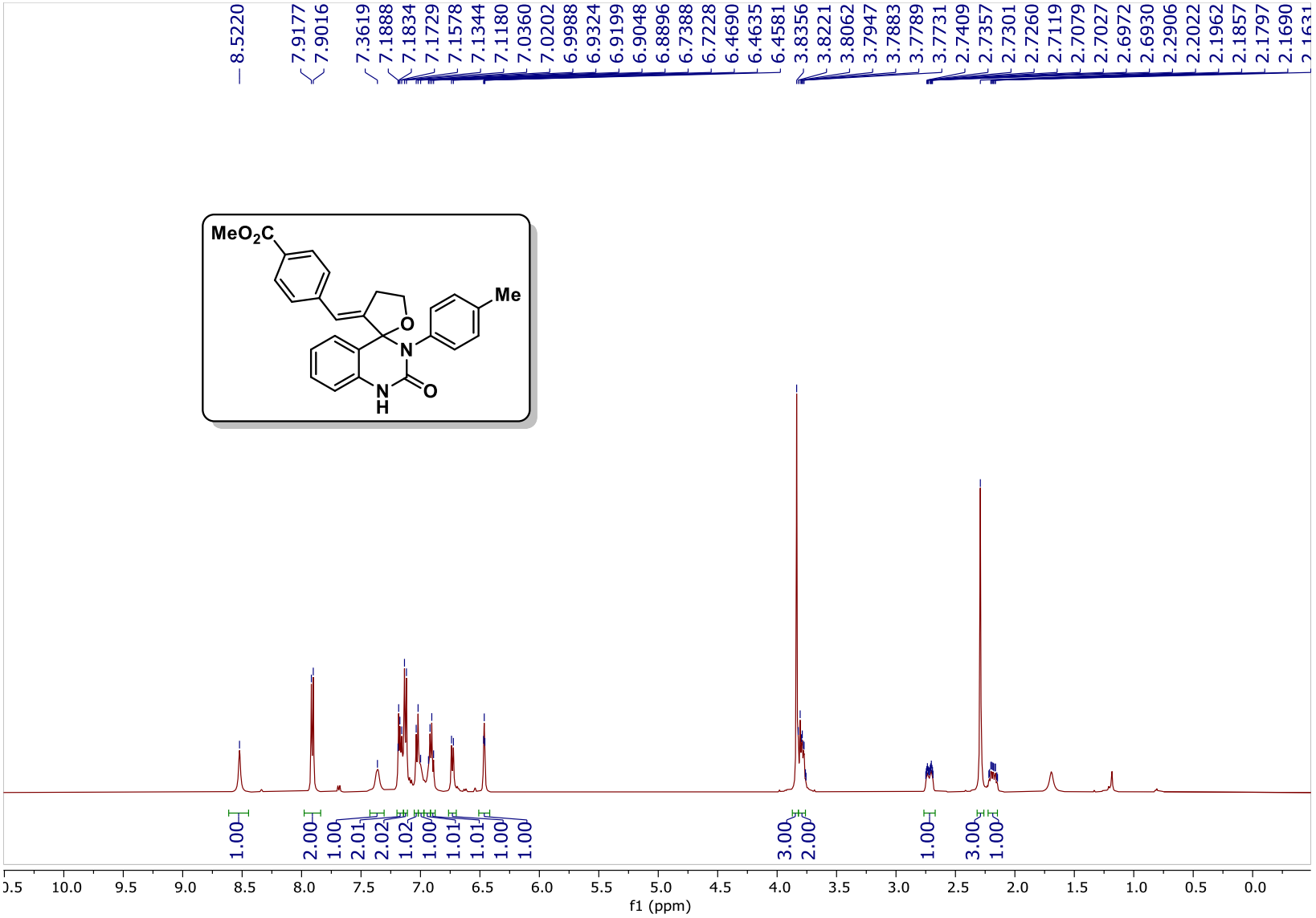
^{1}H spectrum of compound **3ae** (500 MHz, $CDCl_3$)



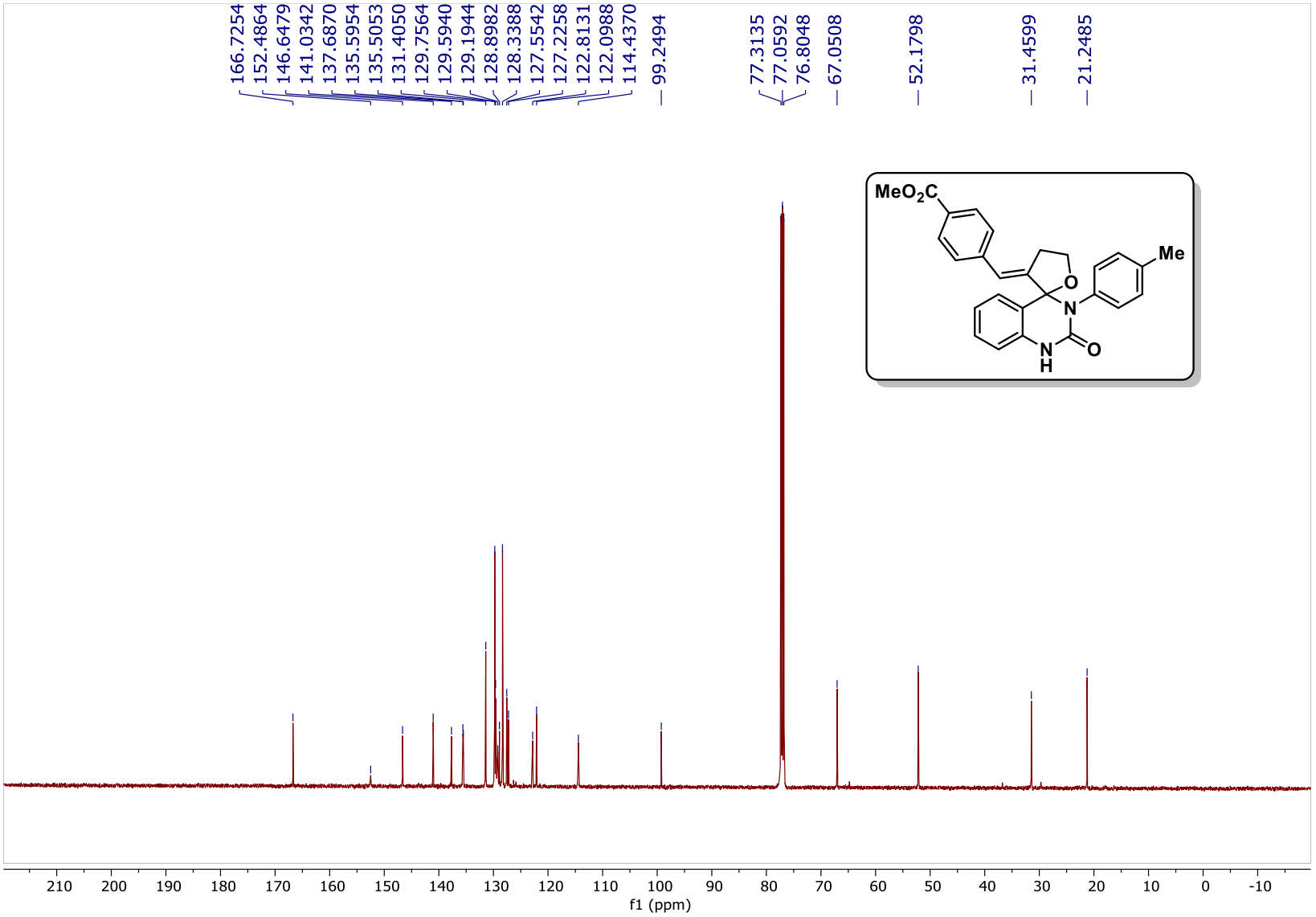
^{13}C spectrum of compound **3ae** (125 MHz, $CDCl_3$)



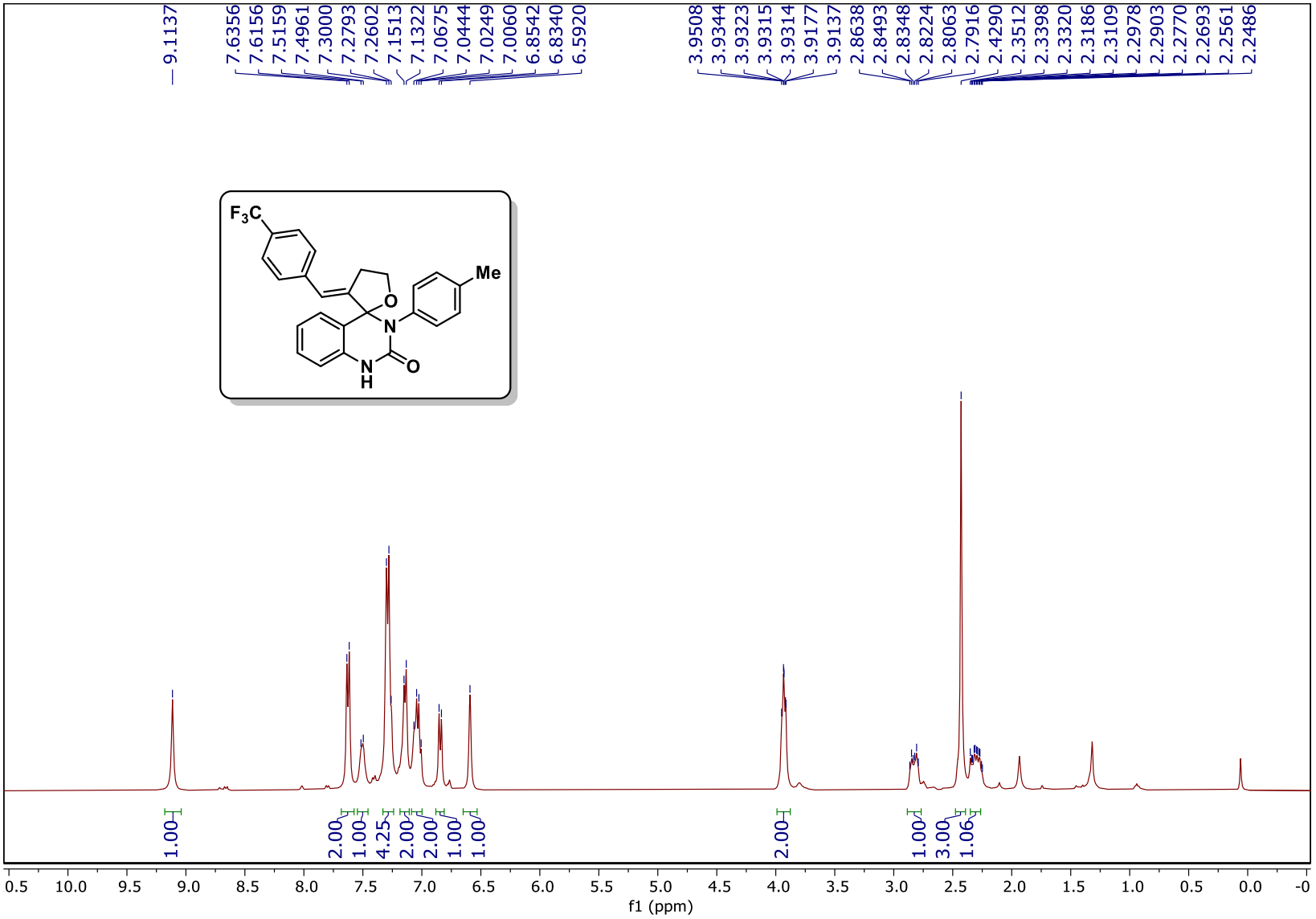
^{1}H spectrum of compound **3af** (500 MHz, $CDCl_3$)



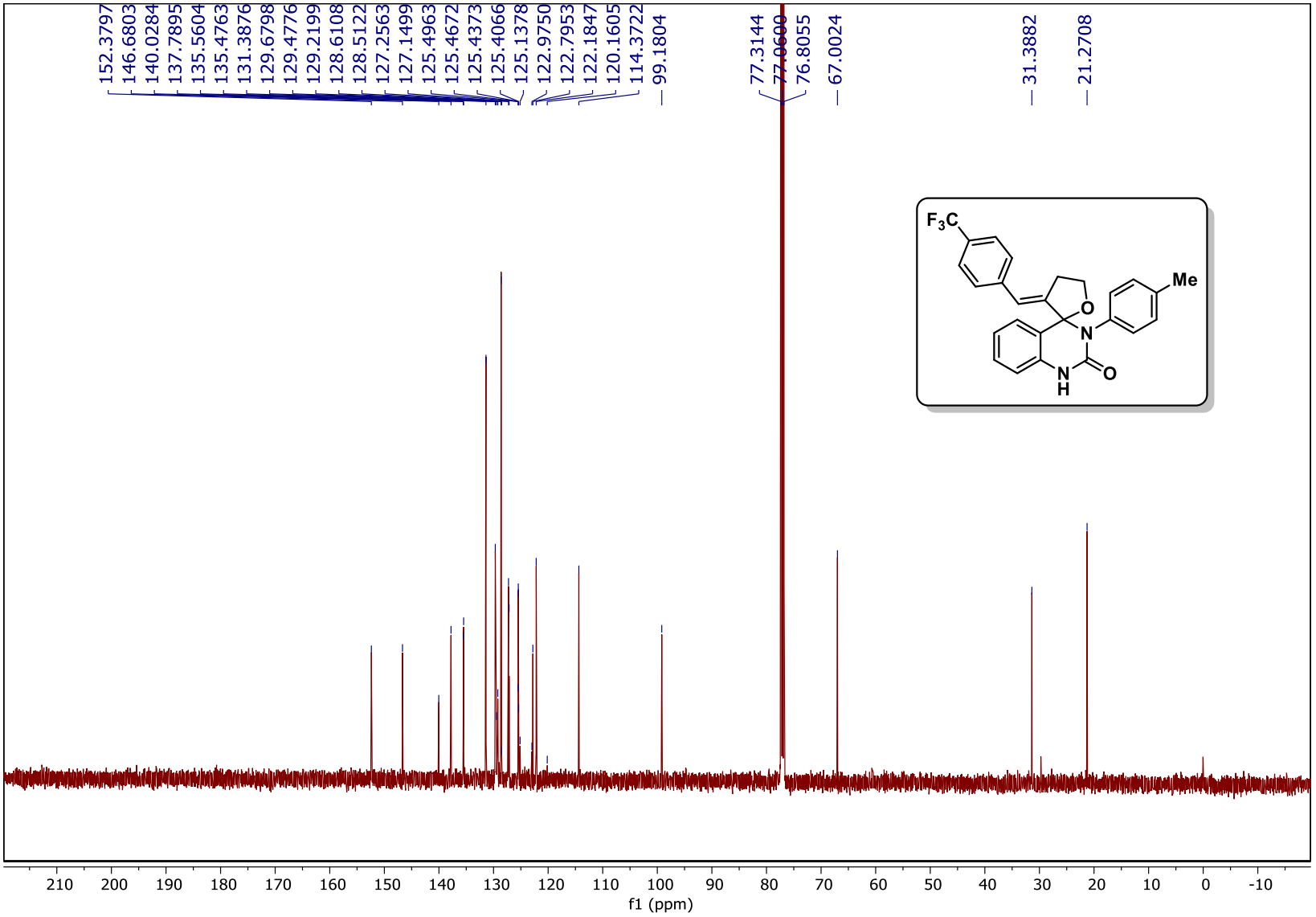
^{13}C spectrum of compound **3af** ($CDCl_3$)



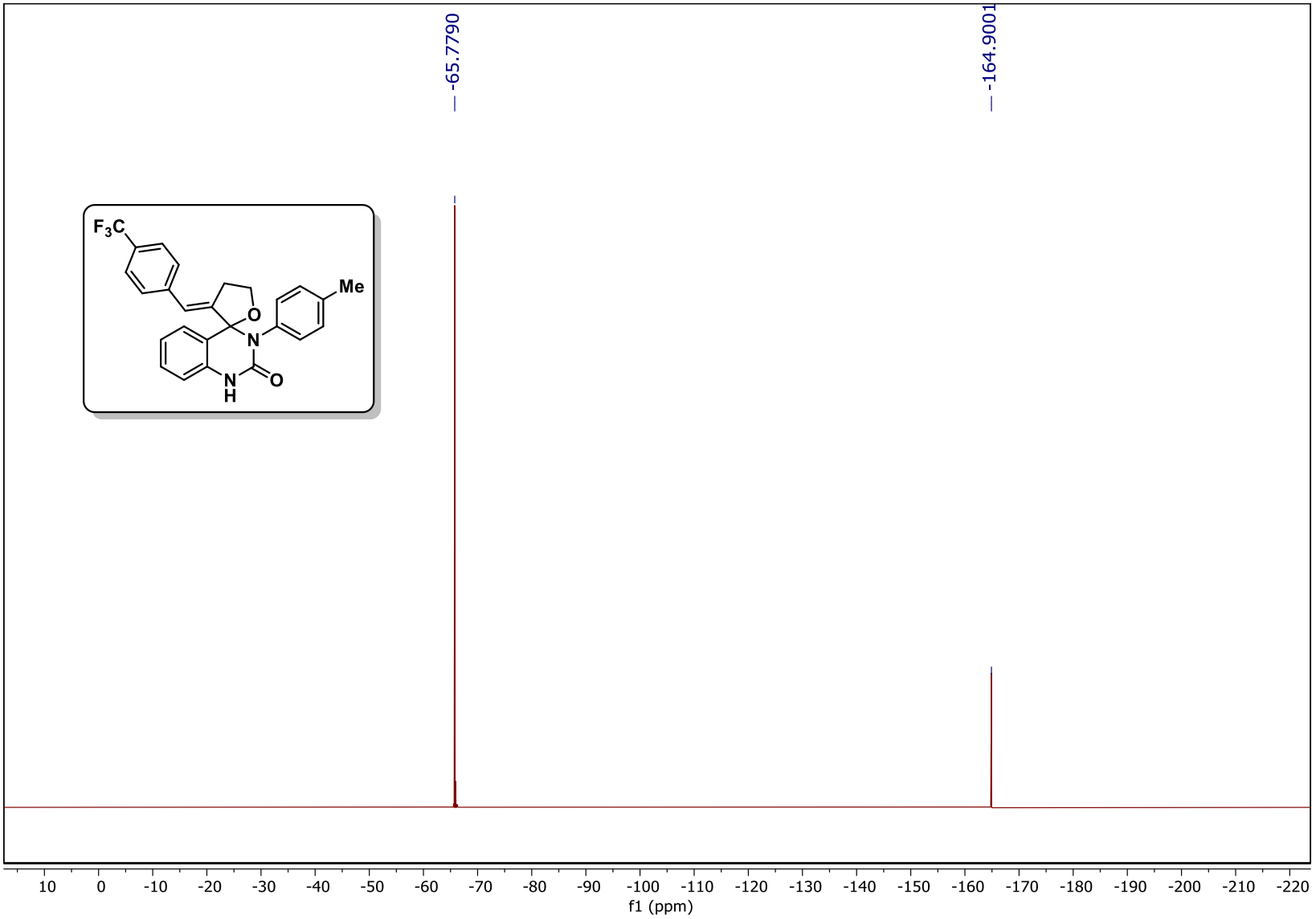
^{1}H spectrum of compound **3ag** (400 MHz, $CDCl_3$)



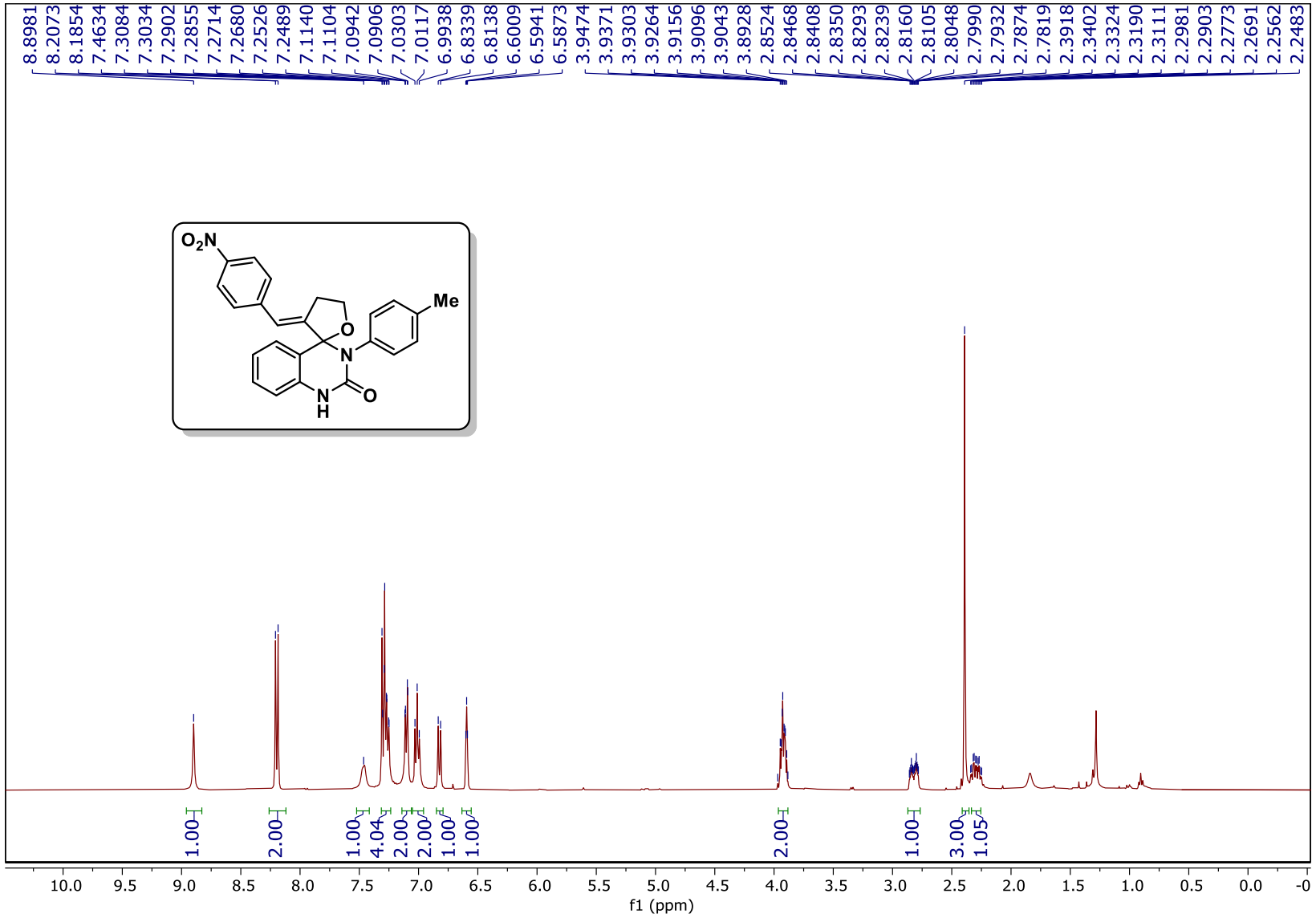
^{13}C spectrum of compound **3ag** (125 MHz, $CDCl_3$)



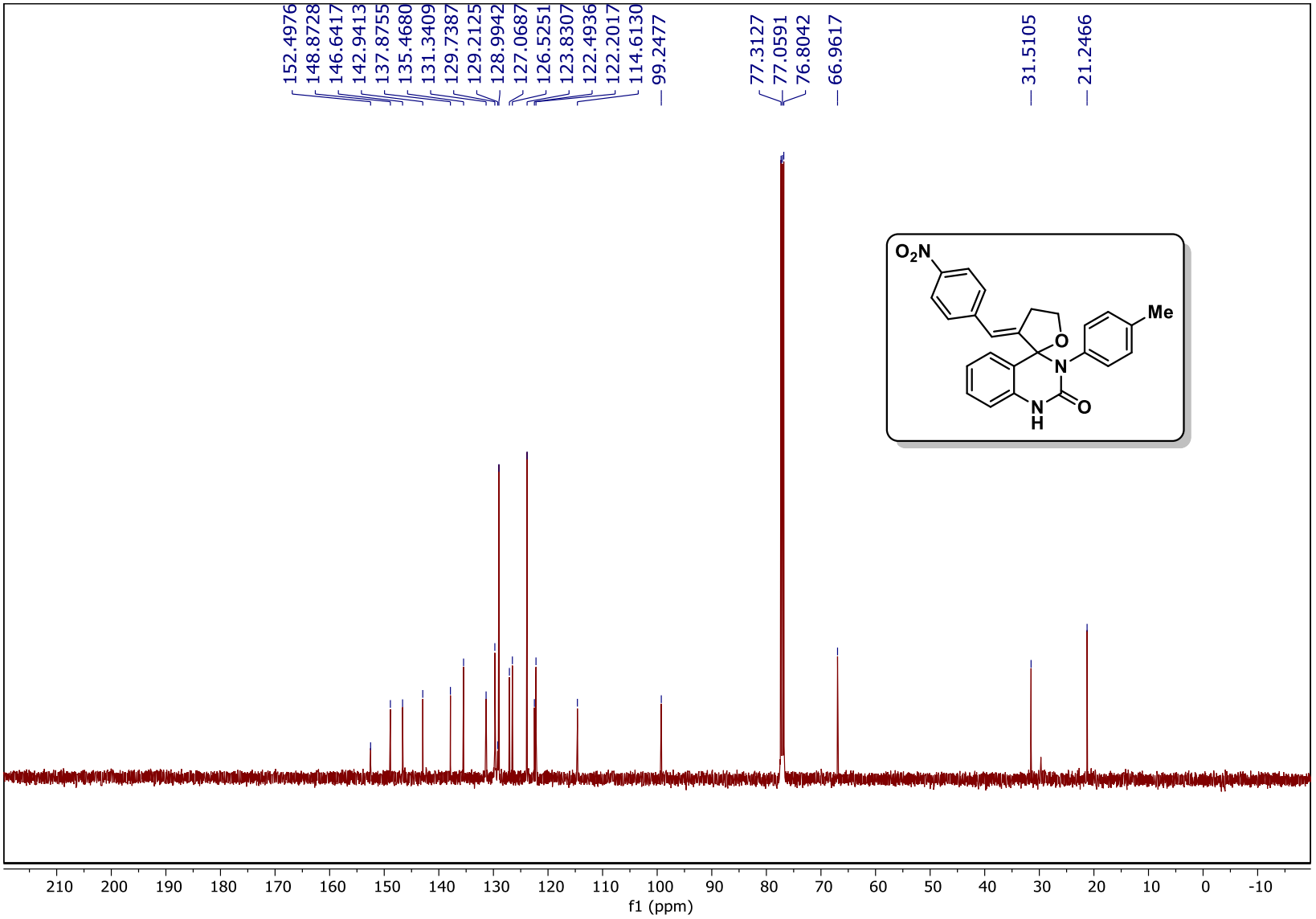
^{19}F spectrum of compound **3ag** (470 MHz, $CDCl_3/C_6F_6$)



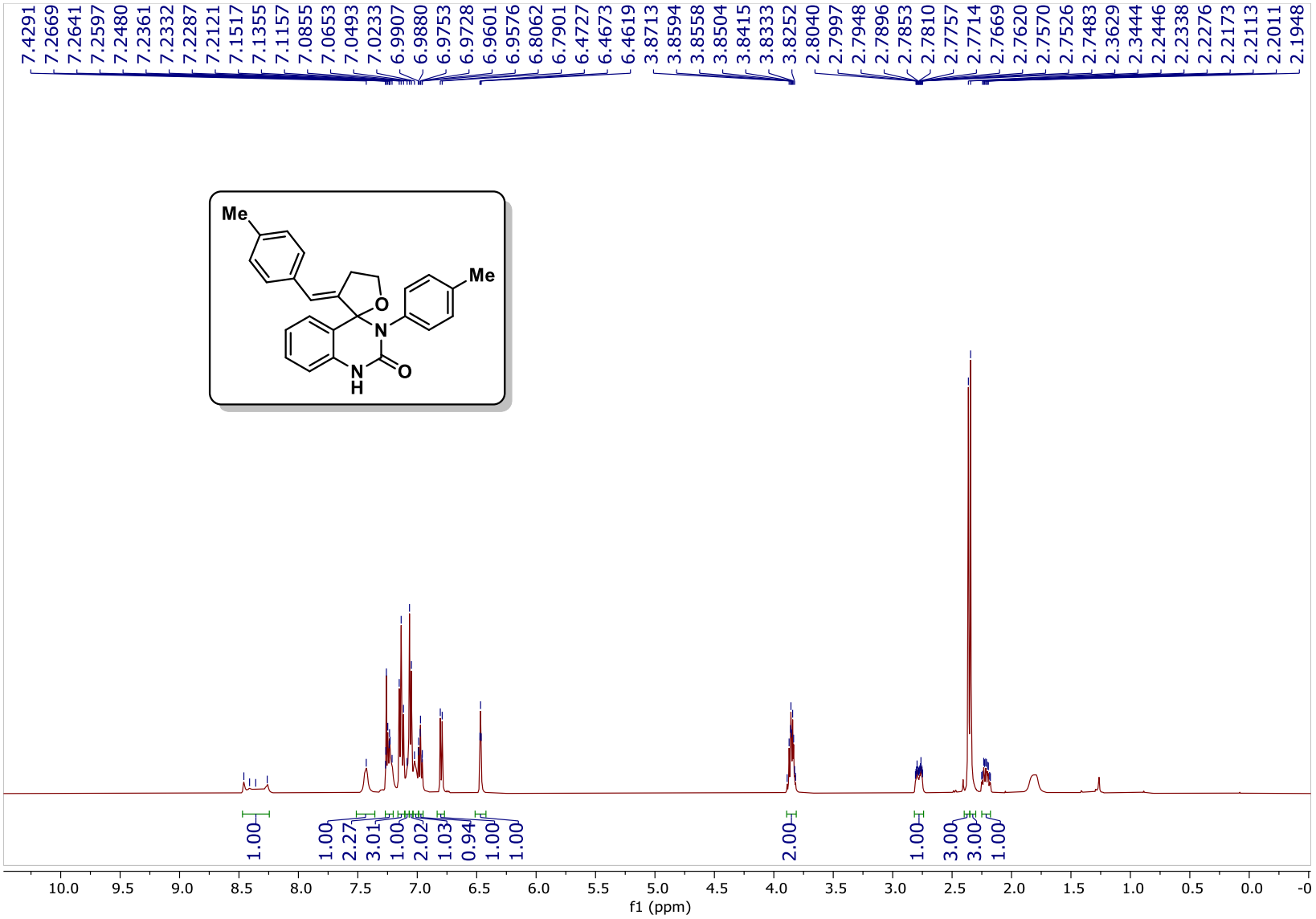
^{1}H spectrum of compound **3ah** (400 MHz, $CDCl_3$)



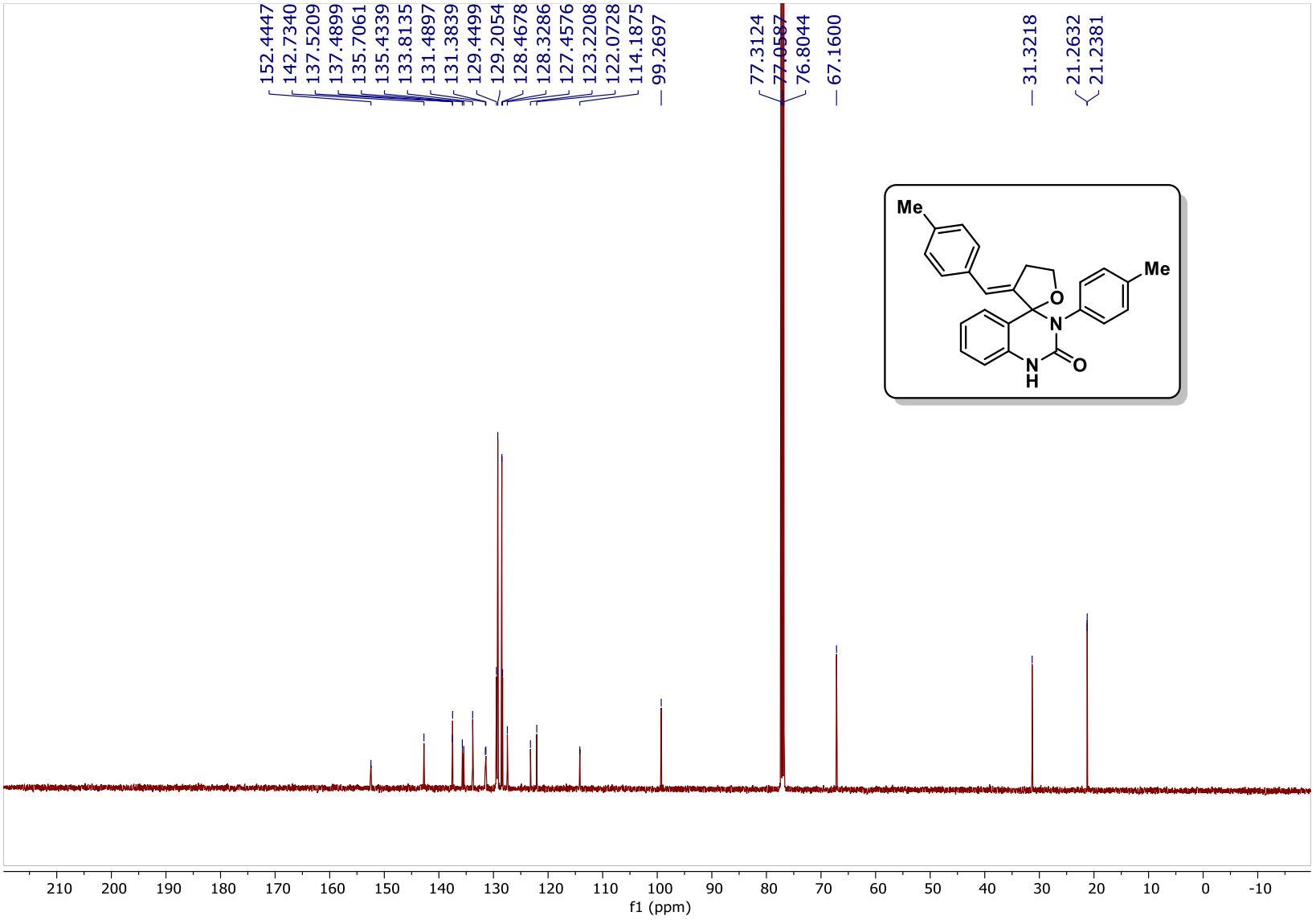
^{13}C spectrum of compound **3ah** (125 MHz, $CDCl_3$)



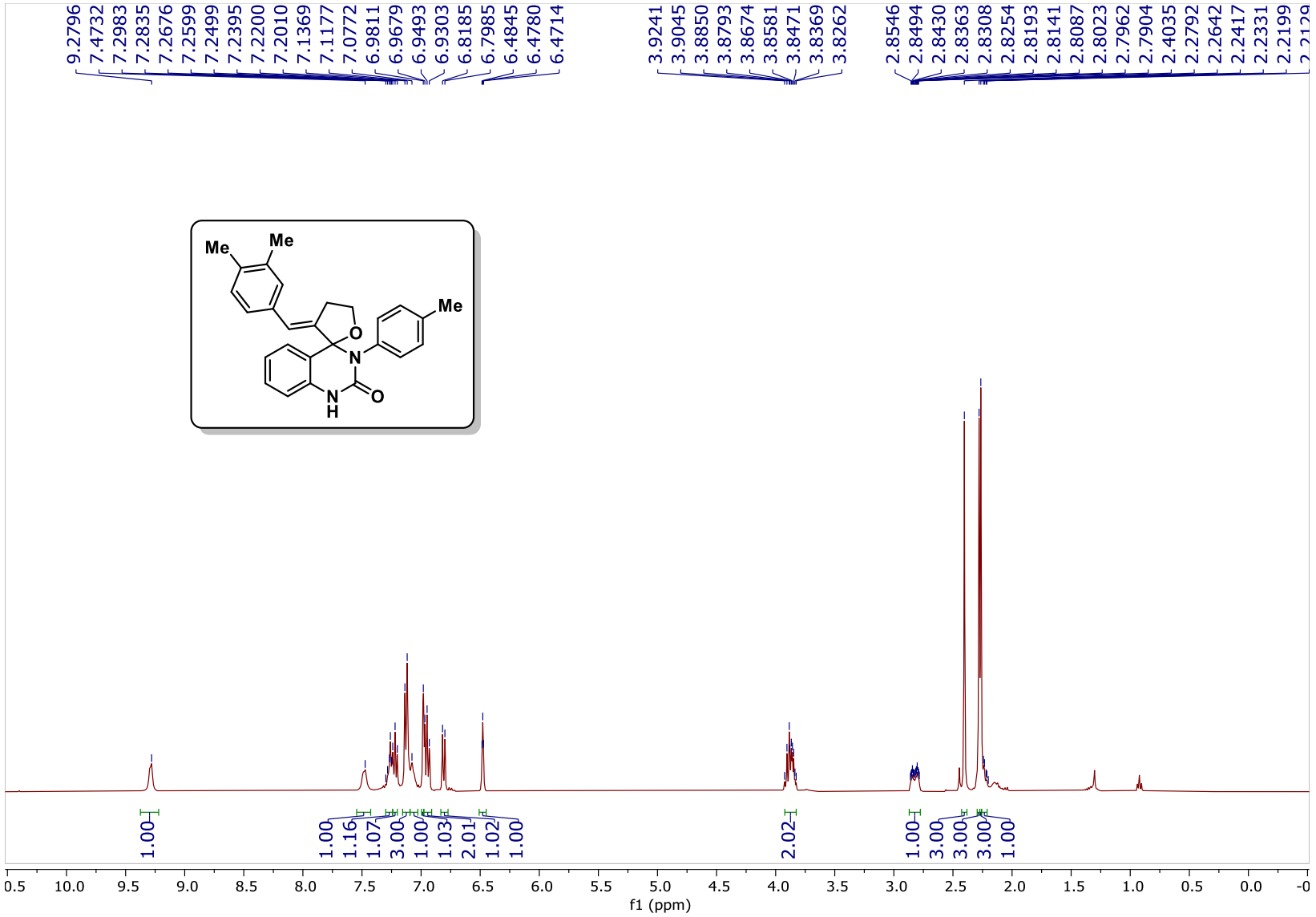
^{1}H spectrum of compound **3ai** (500 MHz, $CDCl_3$)



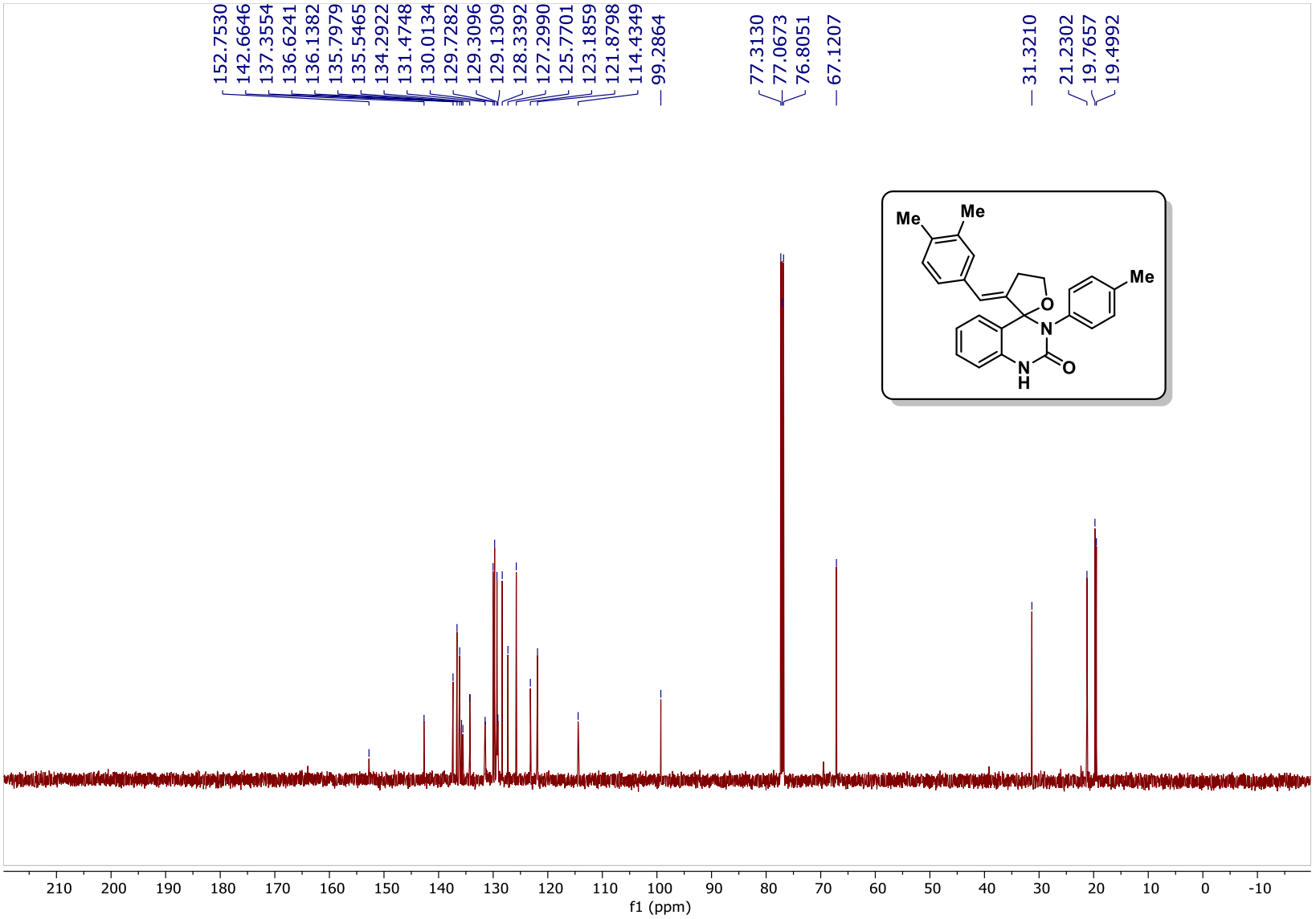
^{13}C spectrum of compound **3ai** (125 MHz, $CDCl_3$)



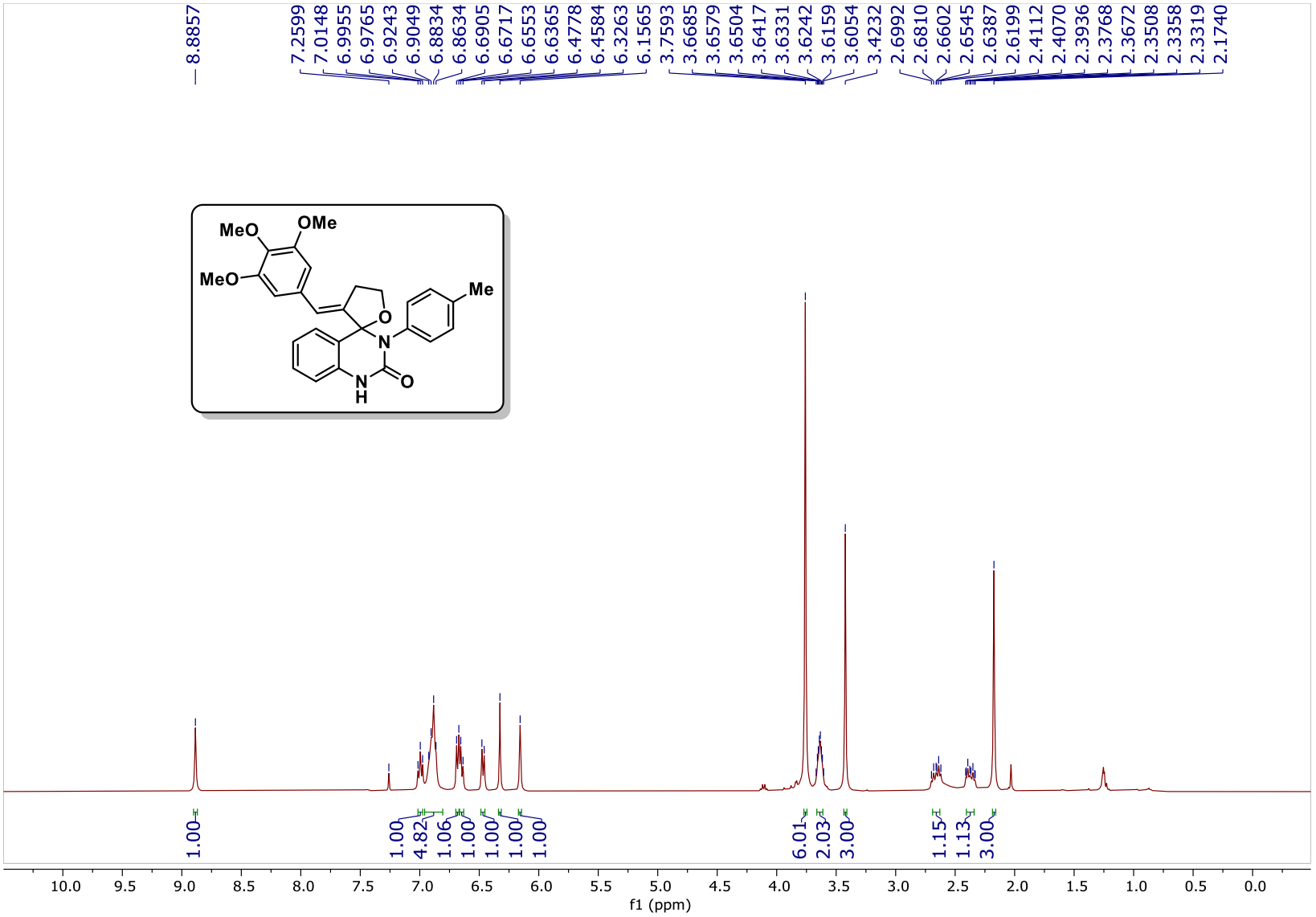
^{1}H spectrum of compound **3aj** (400 MHz, $CDCl_3$)



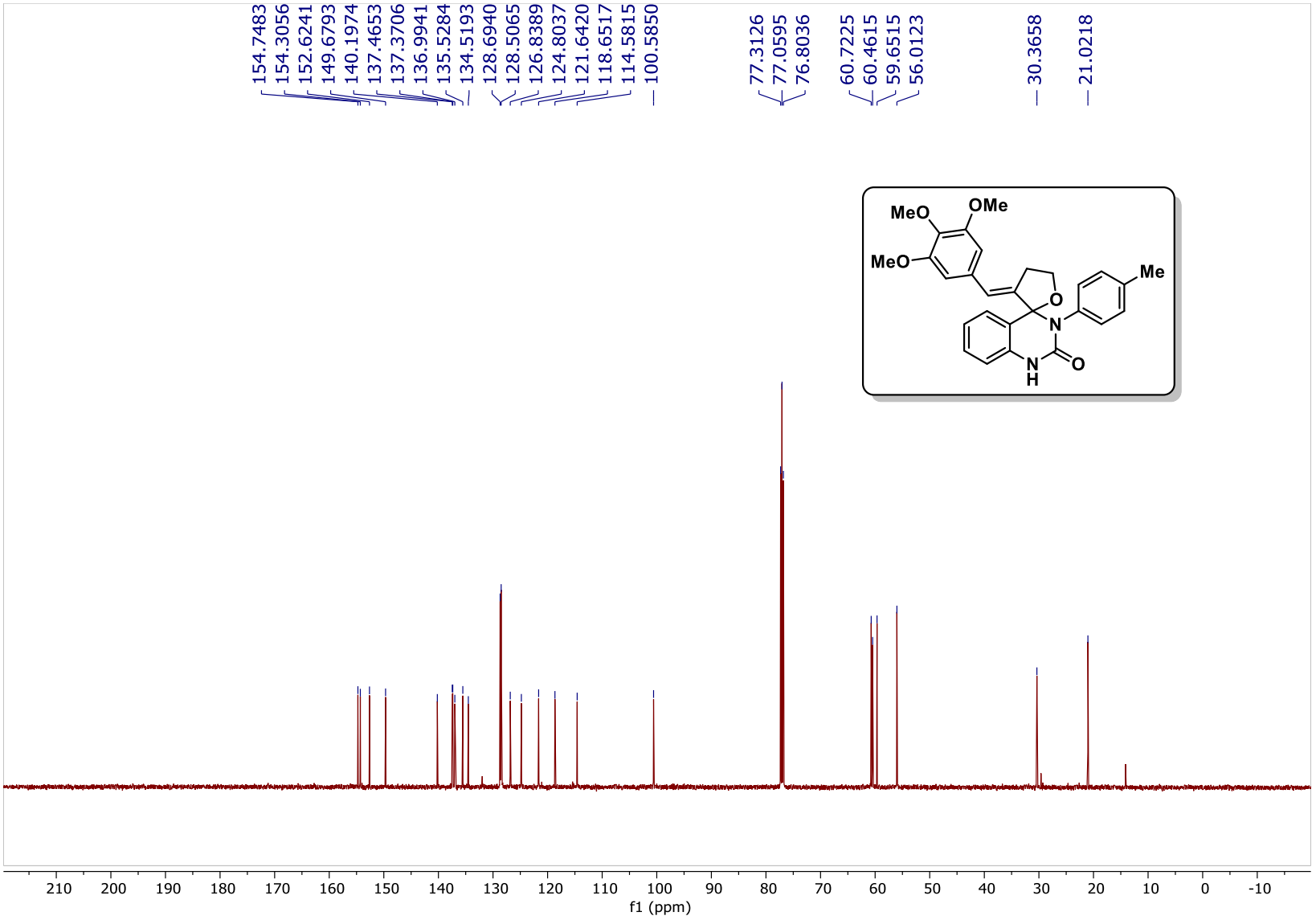
^{13}C spectrum of compound **3aj** (125 MHz, $CDCl_3$)



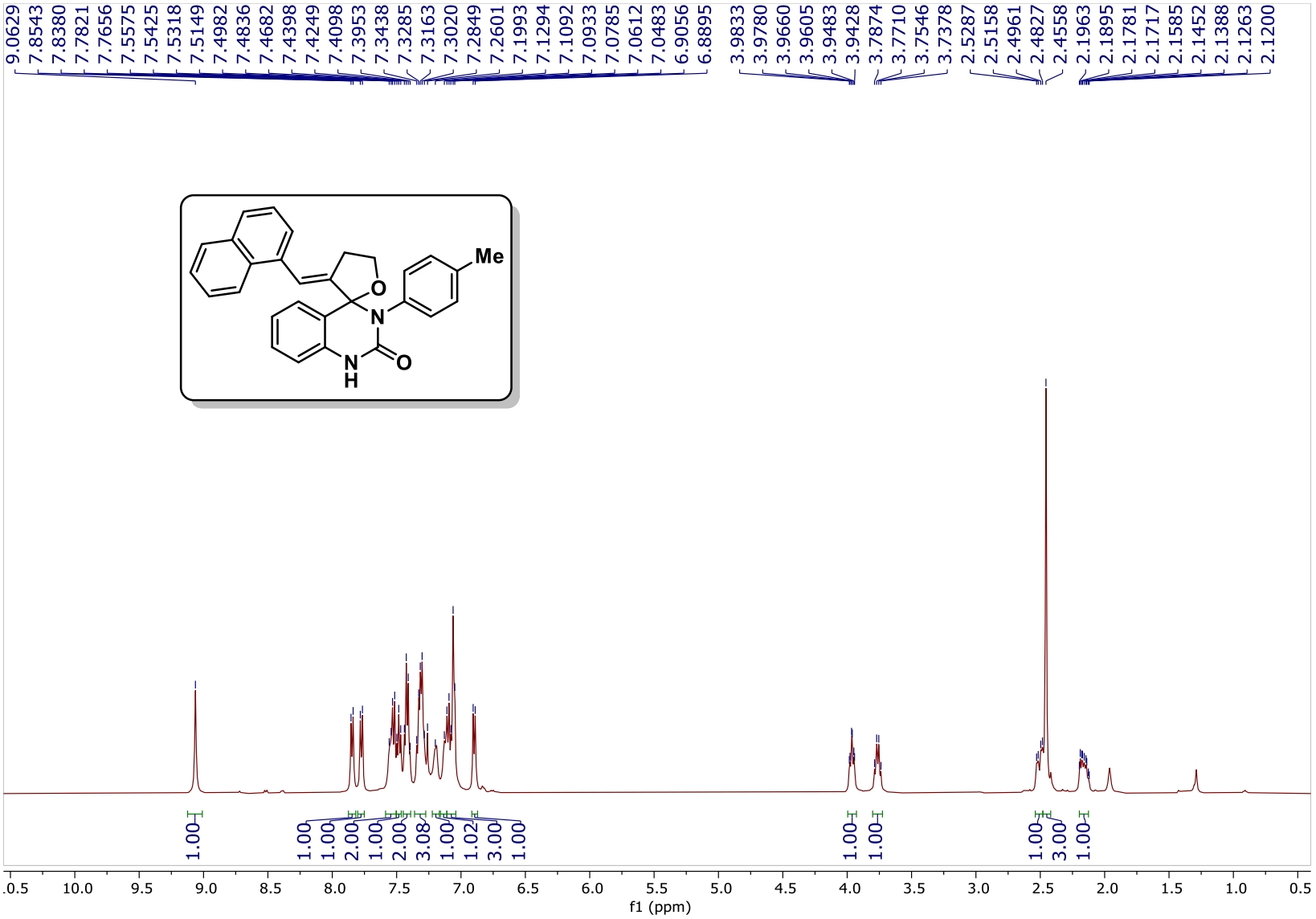
^{1}H spectrum of compound **3ak** (400 MHz, $CDCl_3$)



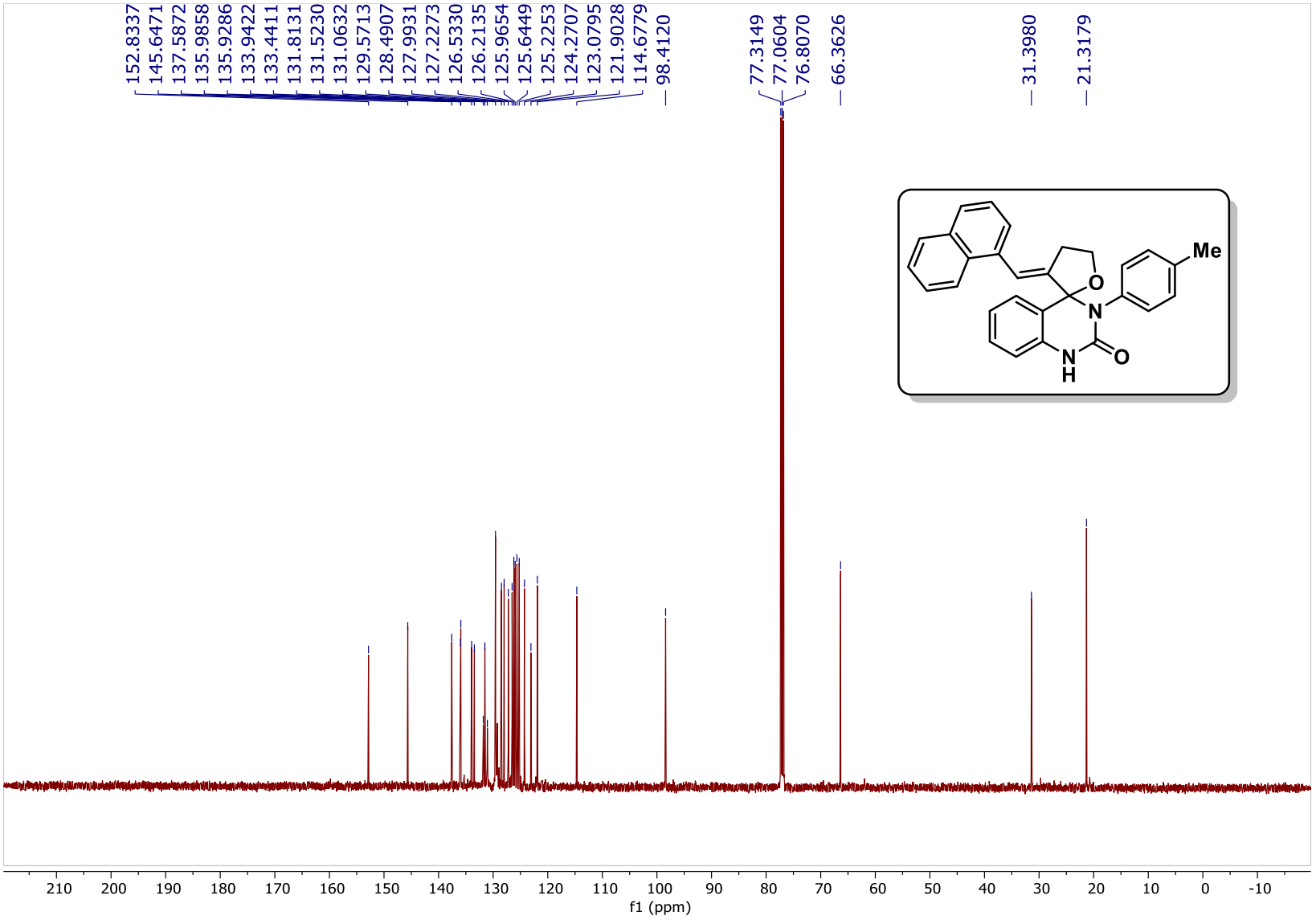
^{13}C spectrum of compound **3ak** (125 MHz, $CDCl_3$)



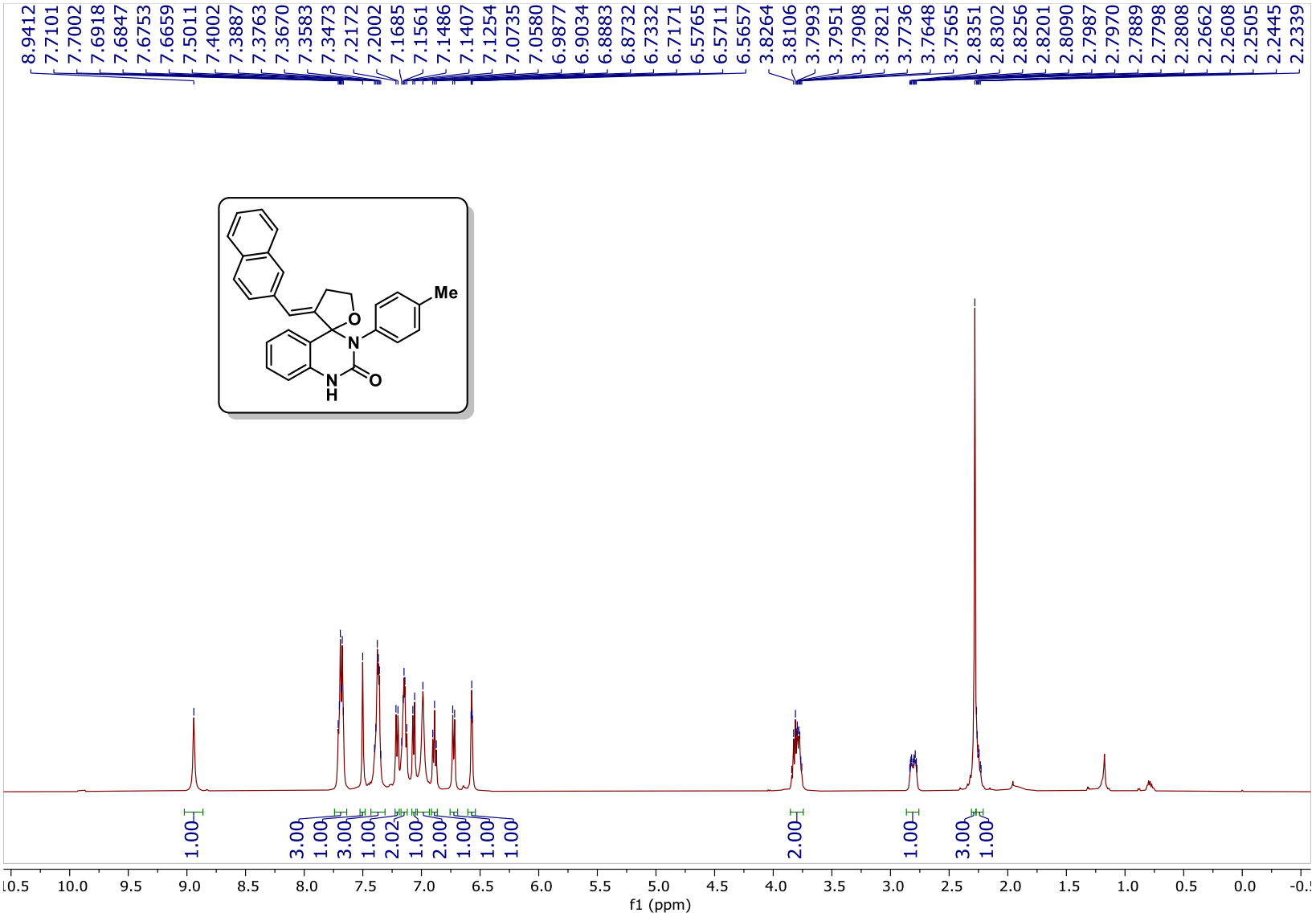
^{1}H spectrum of compound **3al** (500 MHz, $CDCl_3$)



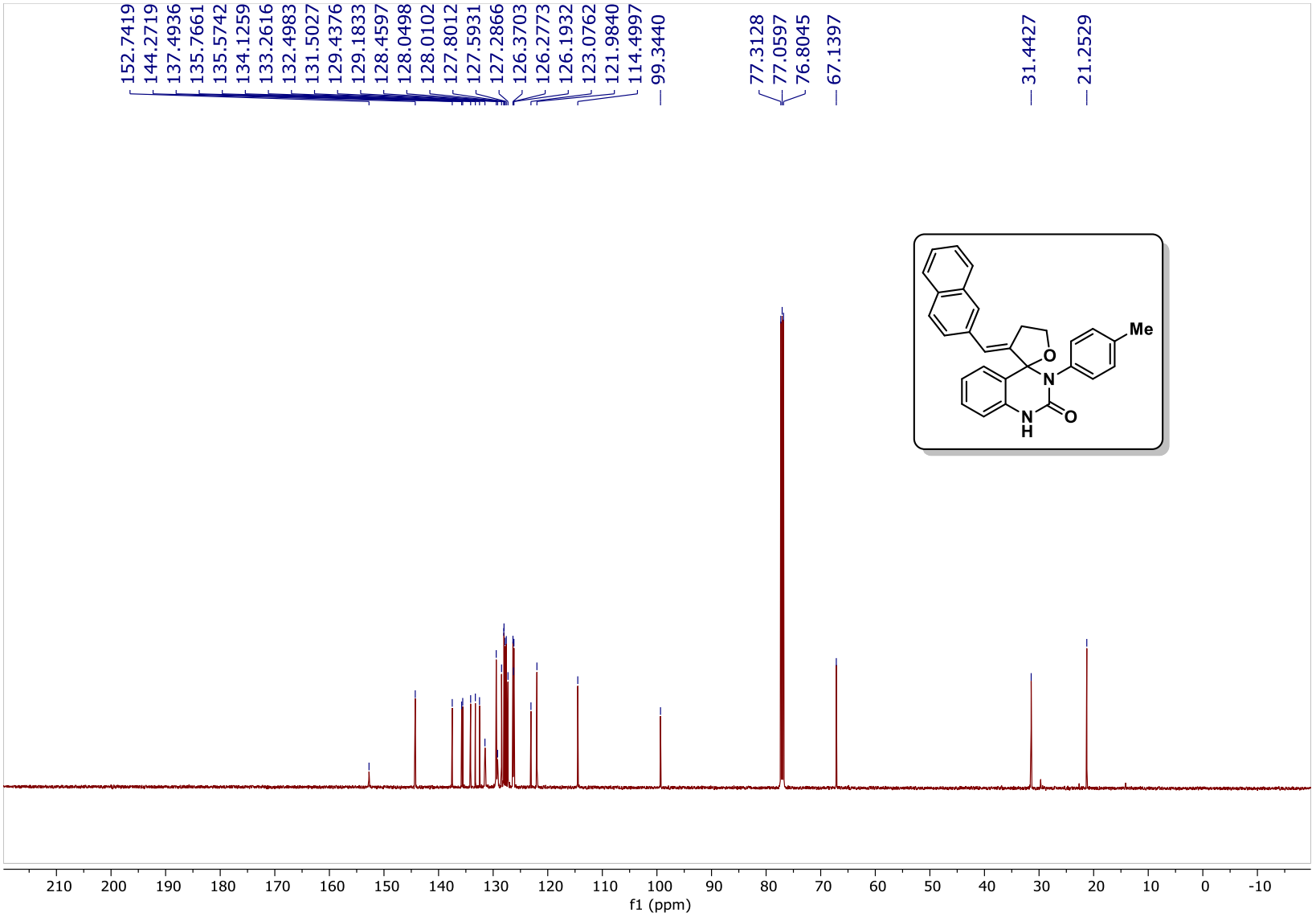
^{13}C spectrum of compound **3al** (125 MHz, $CDCl_3$)



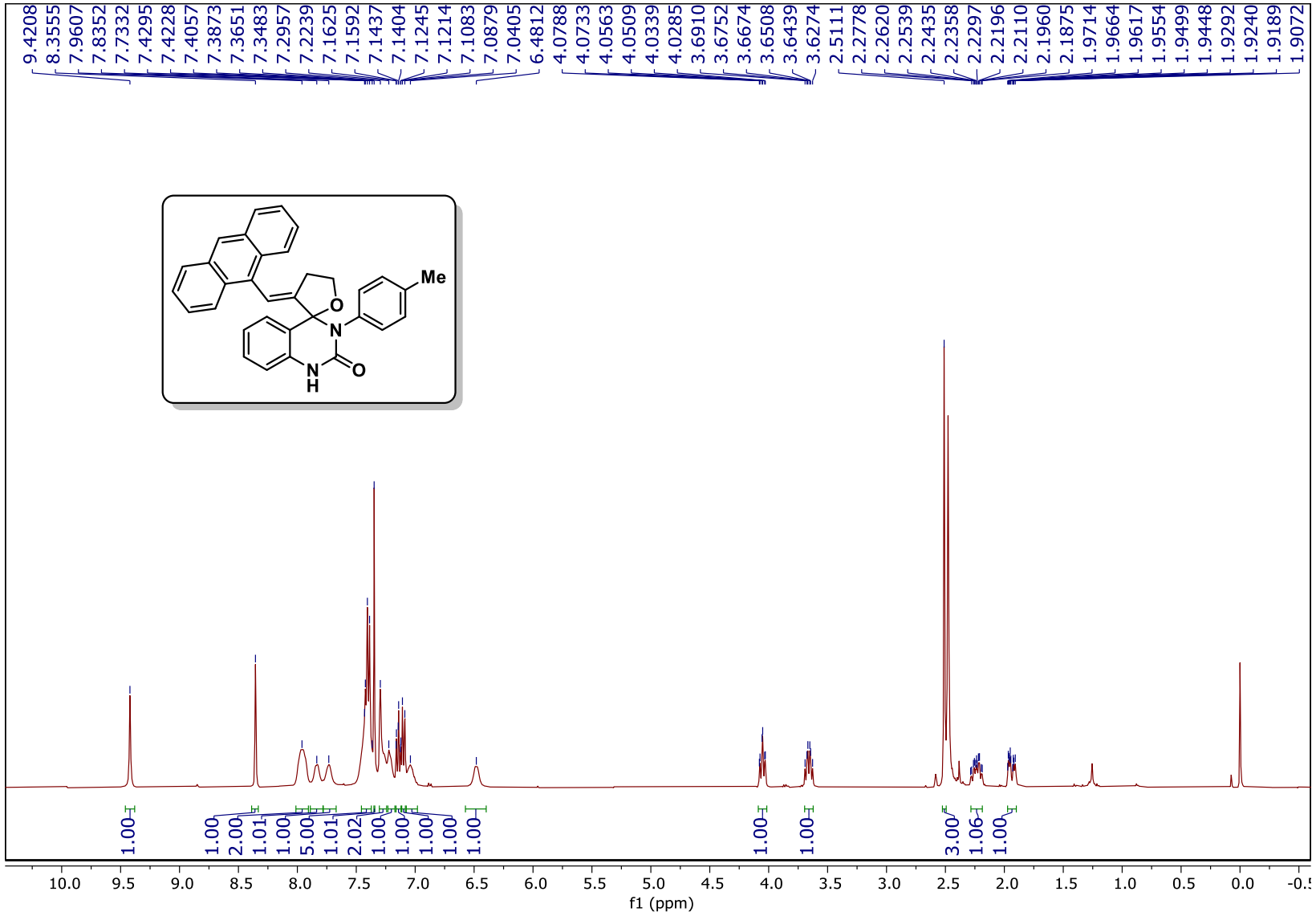
^{1}H spectrum of compound **3am** (500 MHz, $CDCl_3$)



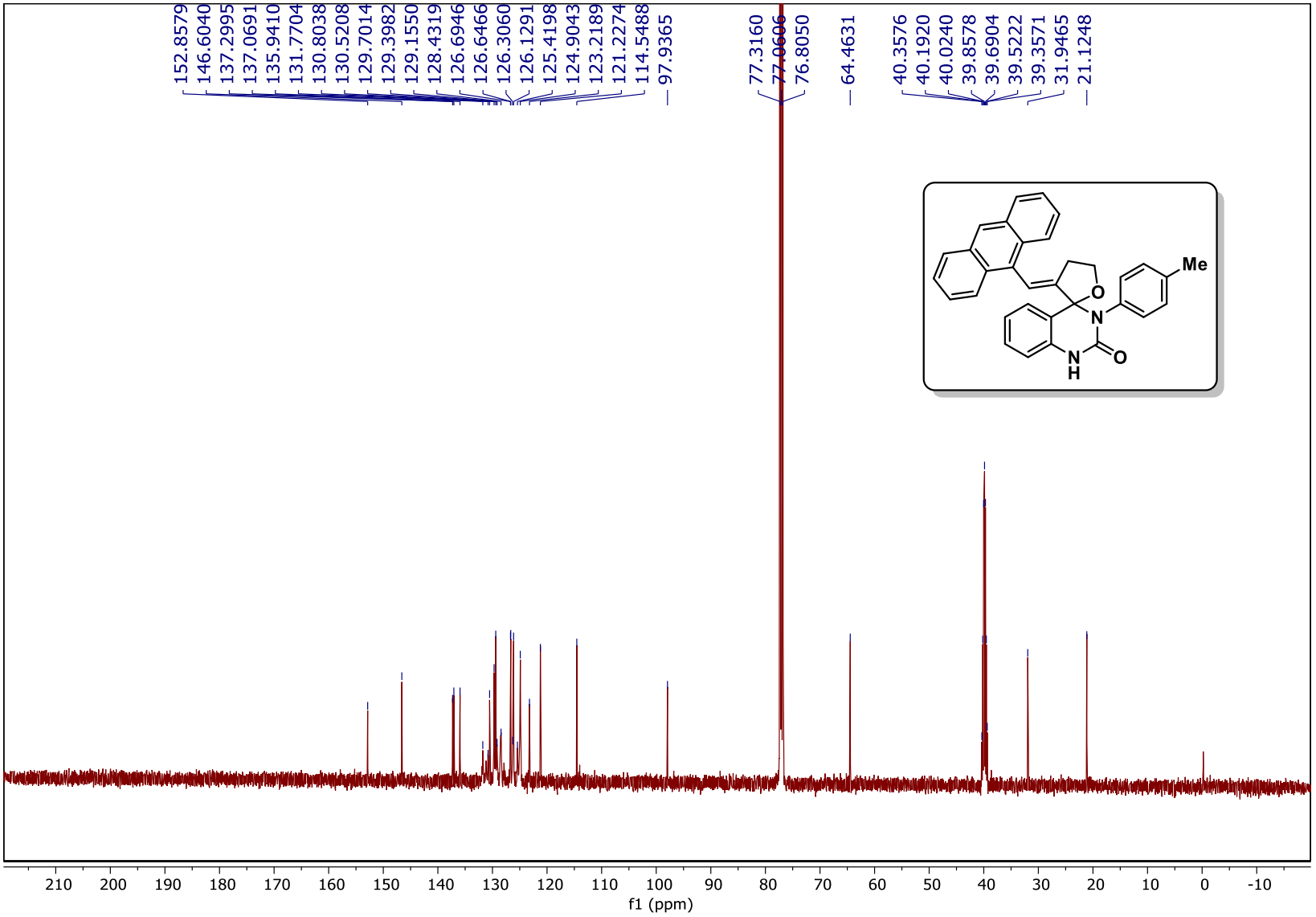
^{13}C spectrum of compound **3am** (125 MHz, $CDCl_3$)



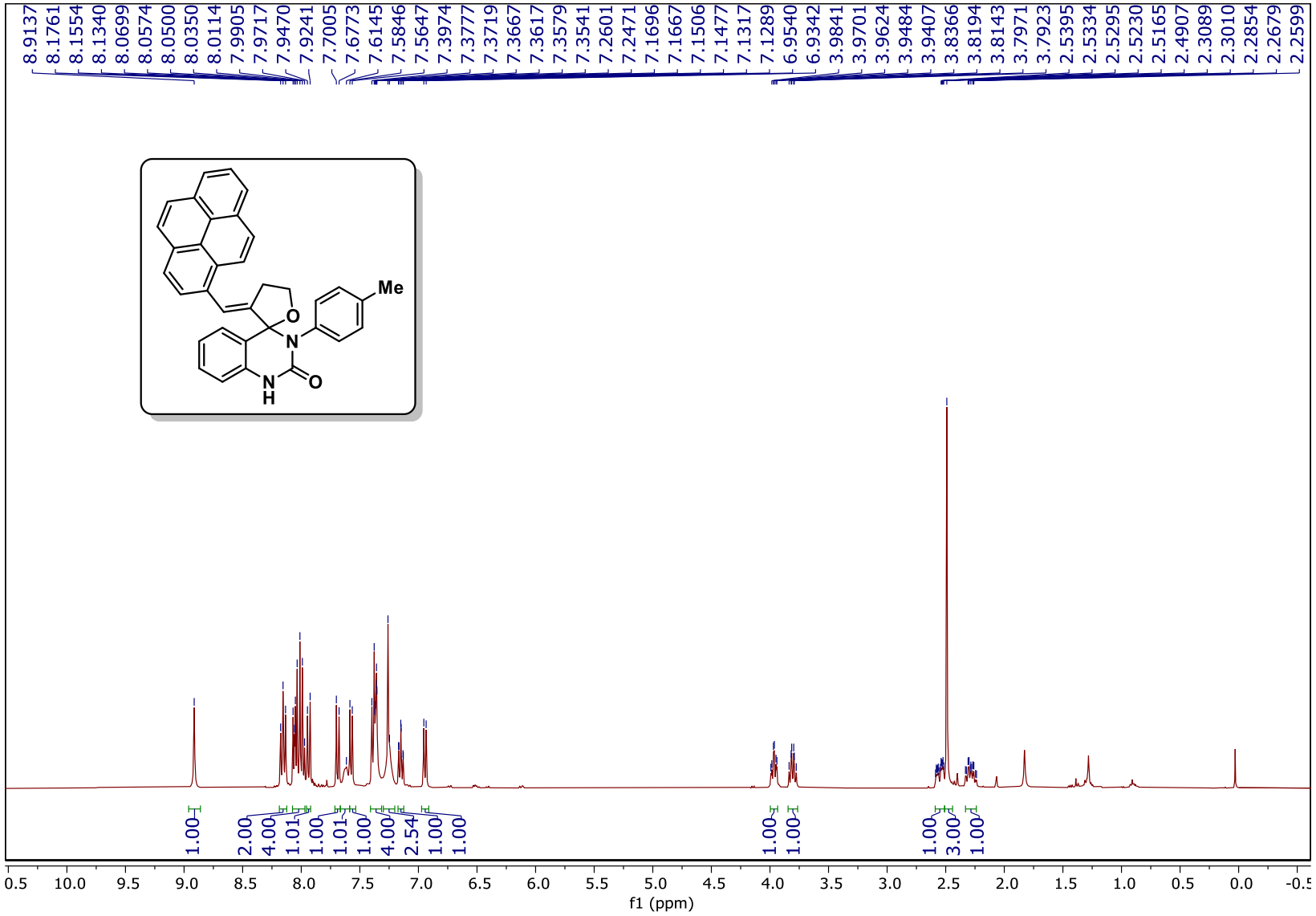
^{1}H spectrum of compound **3an** (400 MHz, $CDCl_3$/DMSO-d_6)



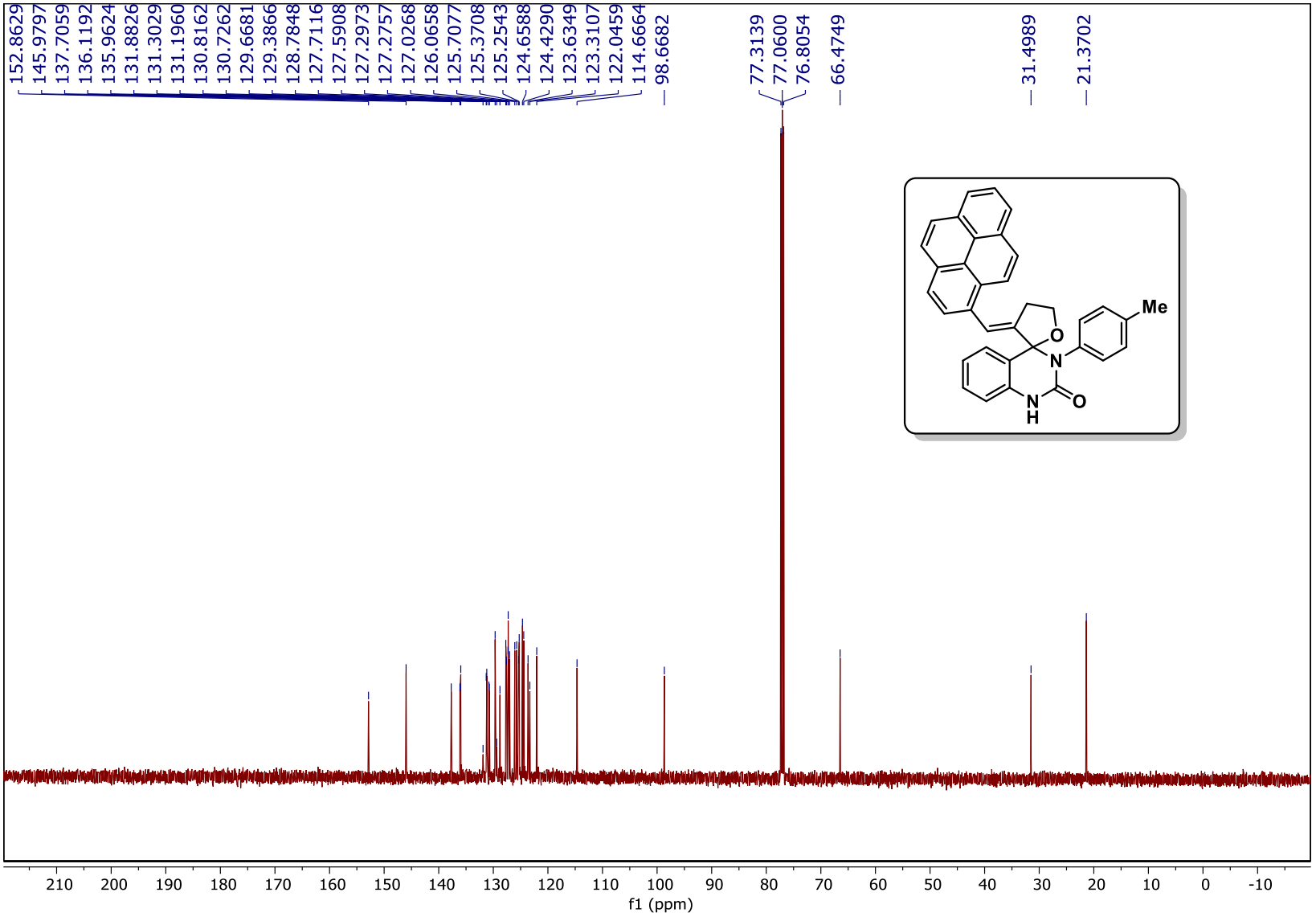
^{13}C spectrum of compound **3an** (125 MHz, $CDCl_3$/DMSO-d_6)



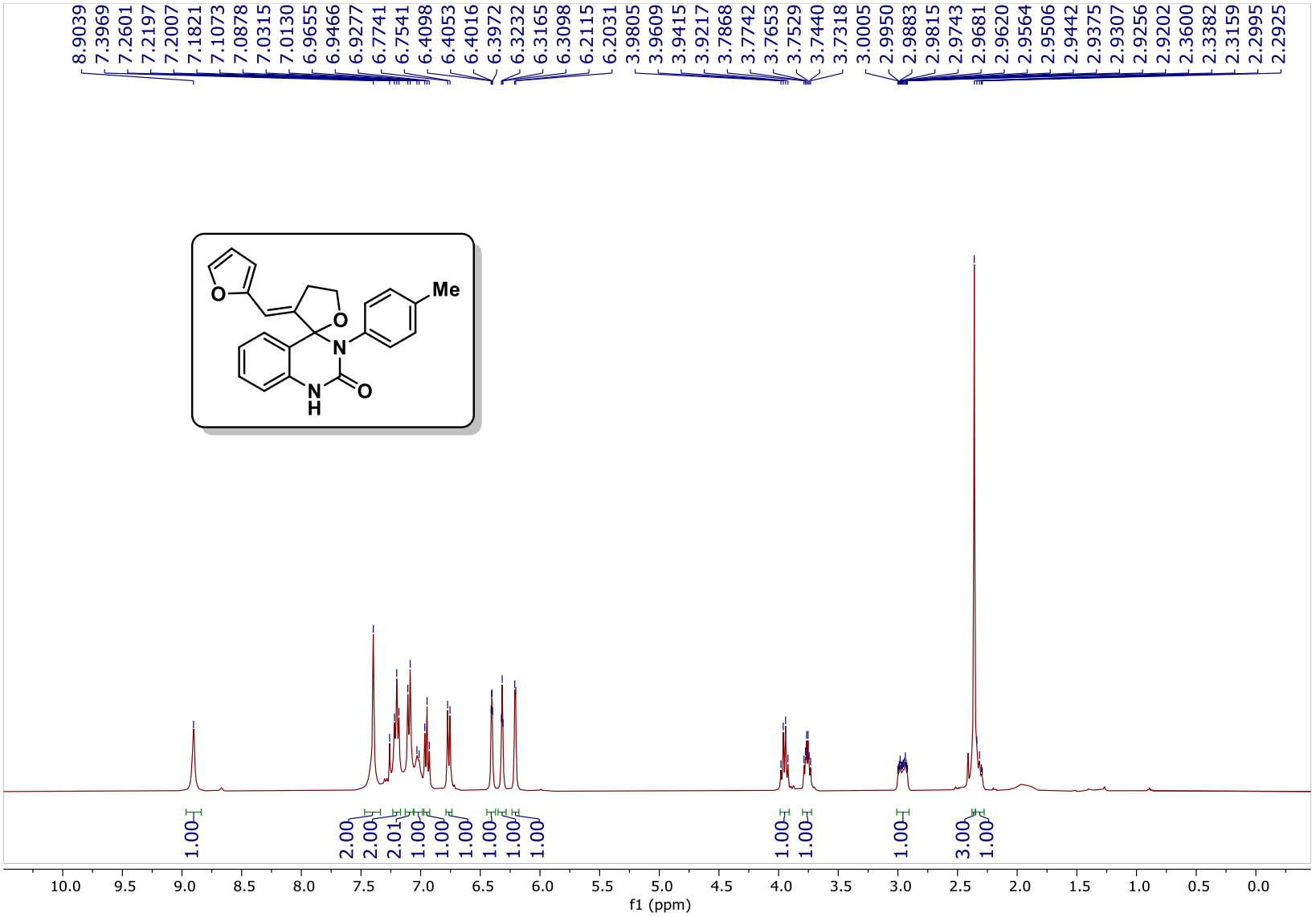
^{1}H spectrum of compound **3ao** (400 MHz, $CDCl_3$)



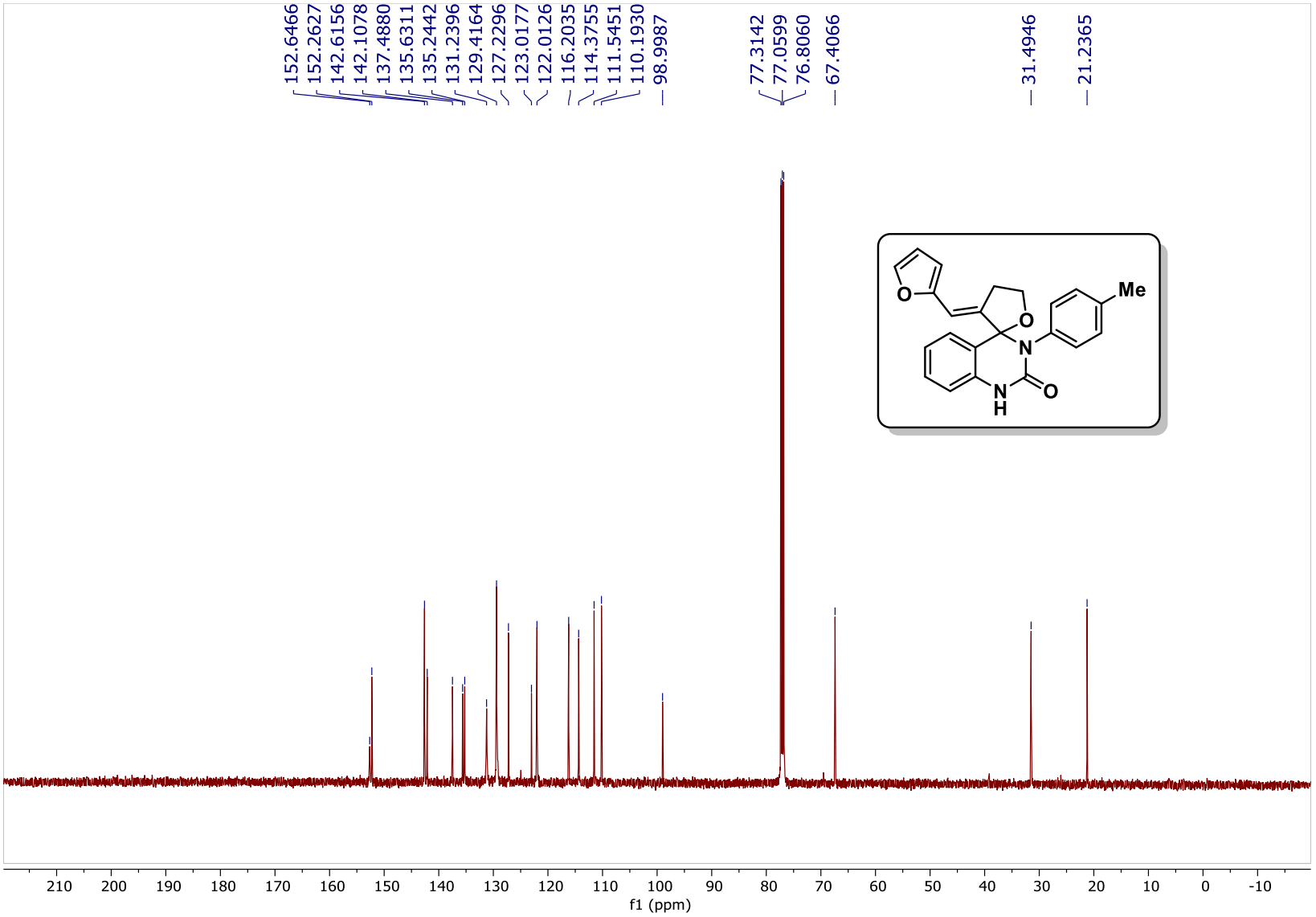
^{13}C spectrum of compound **3ao** (125 MHz, $CDCl_3$)



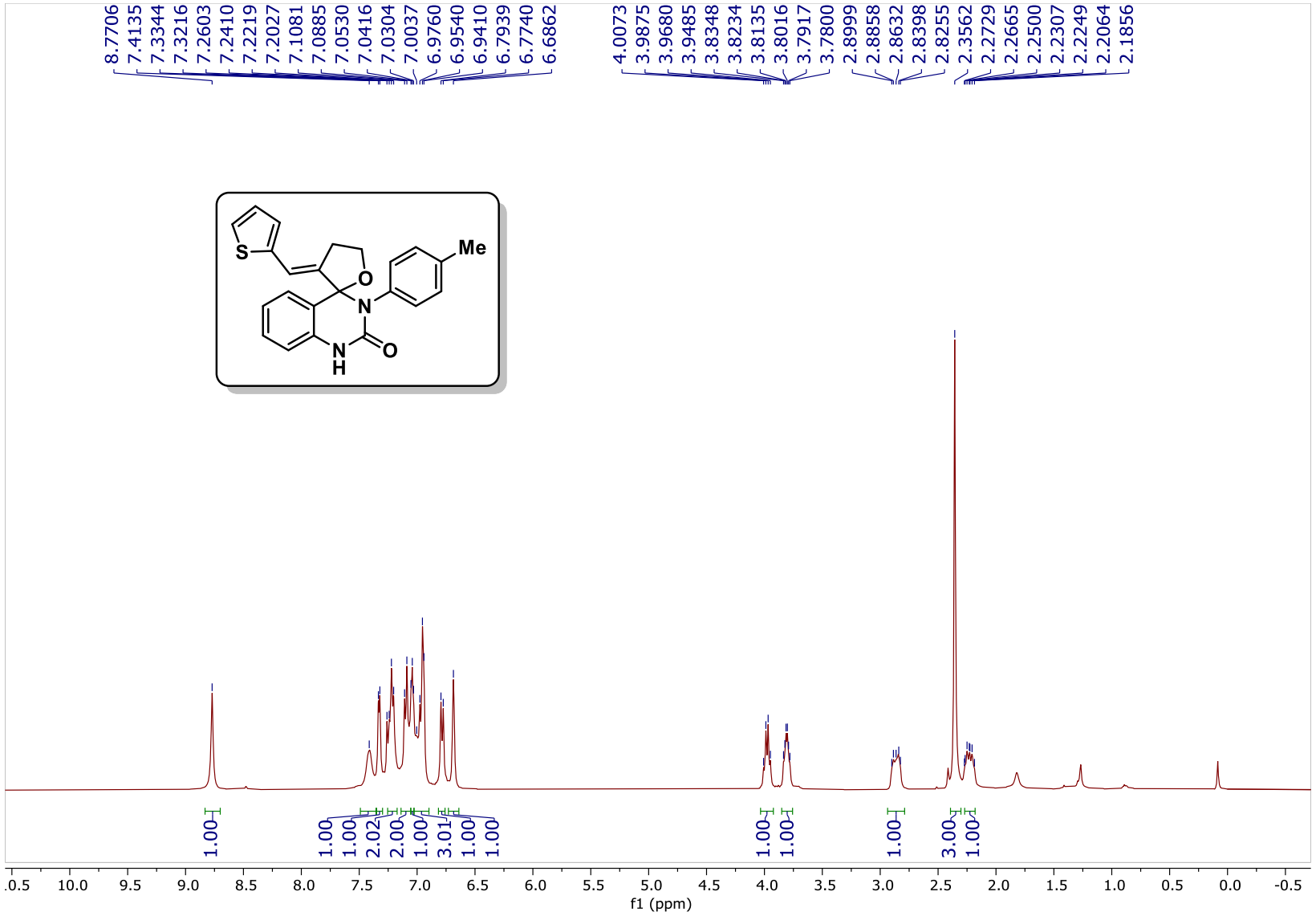
^{1}H spectrum of compound **3ap** (400 MHz, $CDCl_3$)



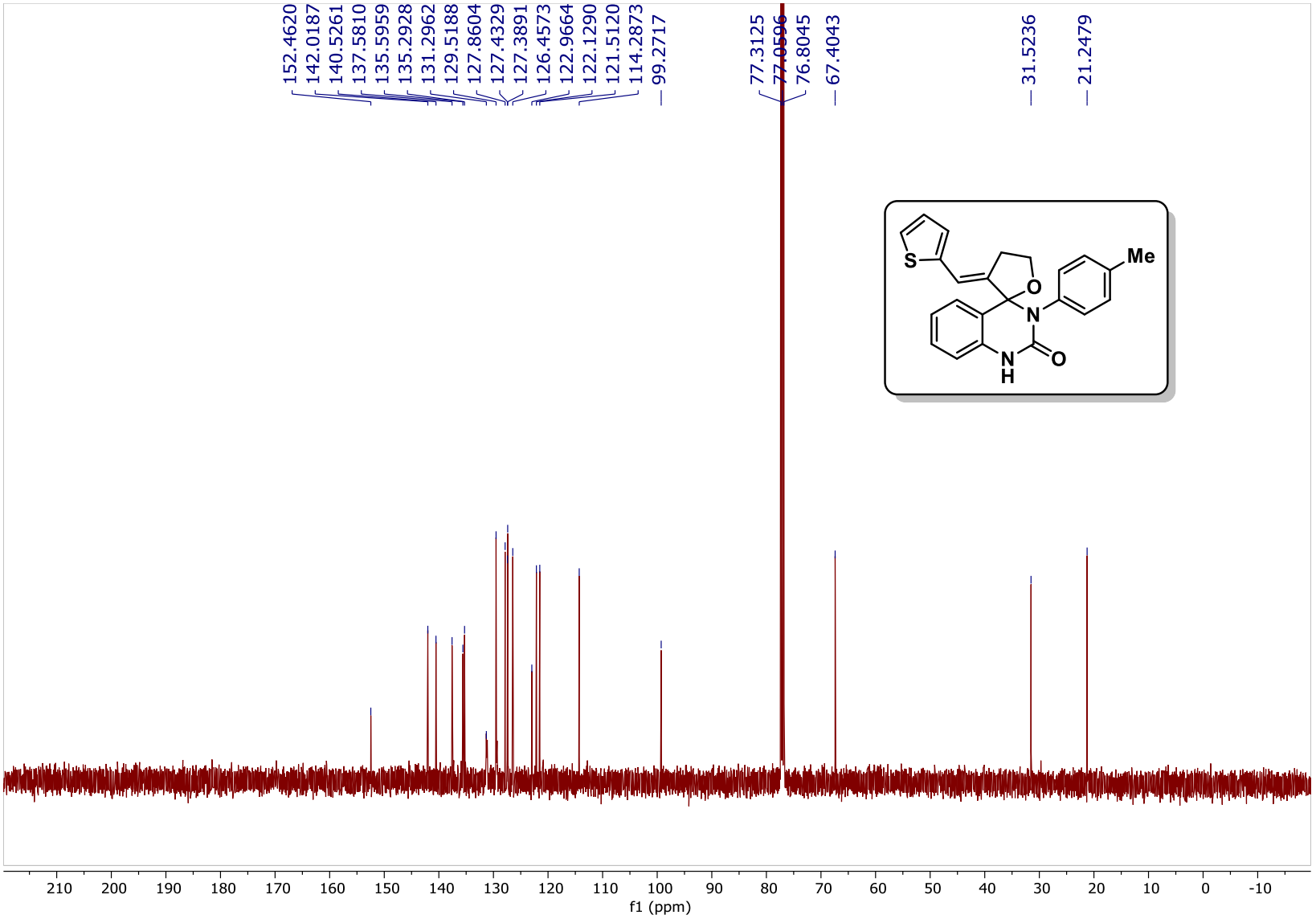
^{13}C spectrum of compound **3ap** (125 MHz, $CDCl_3$)



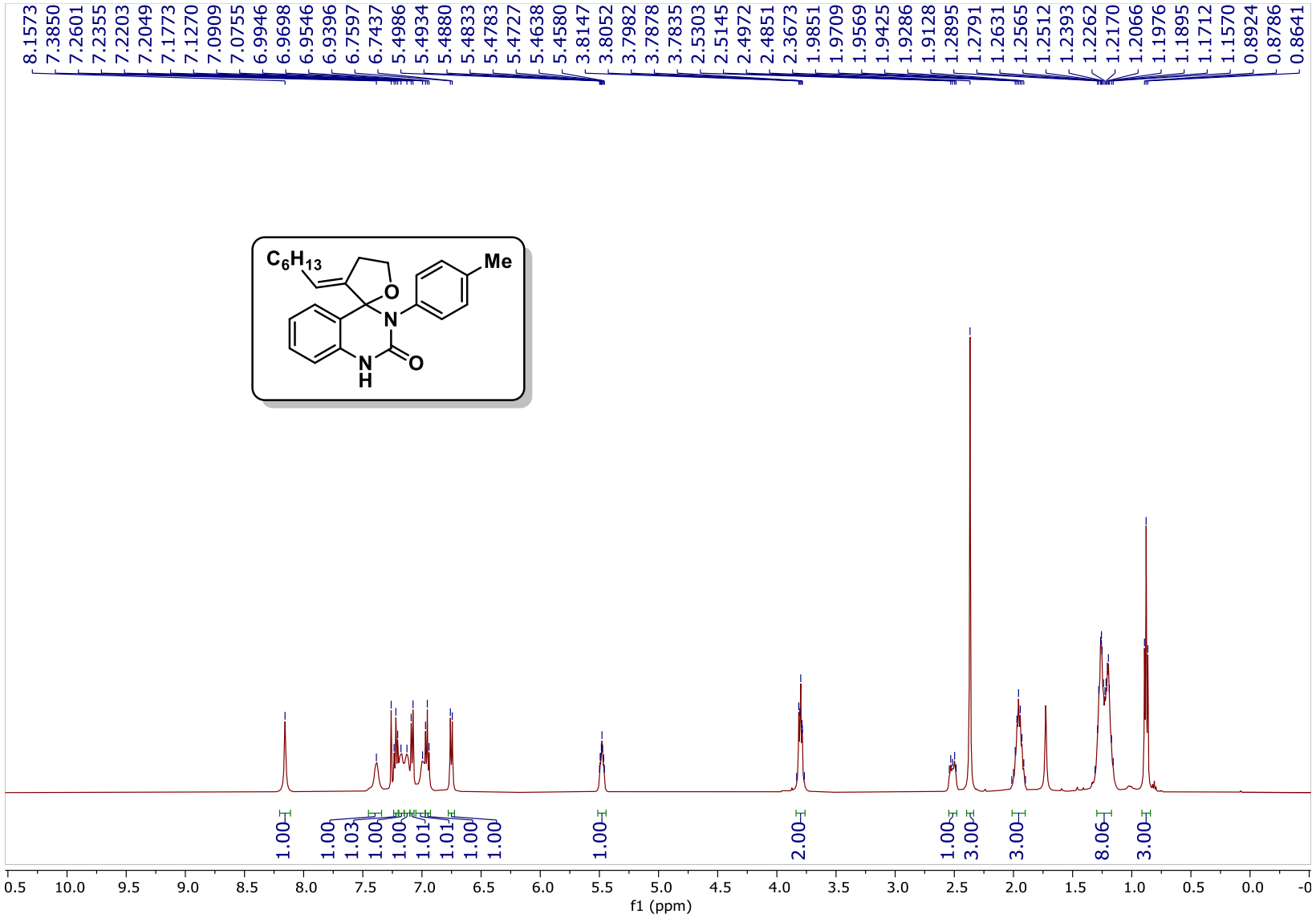
^{1}H spectrum of compound **3aq** (400 MHz, $CDCl_3$)



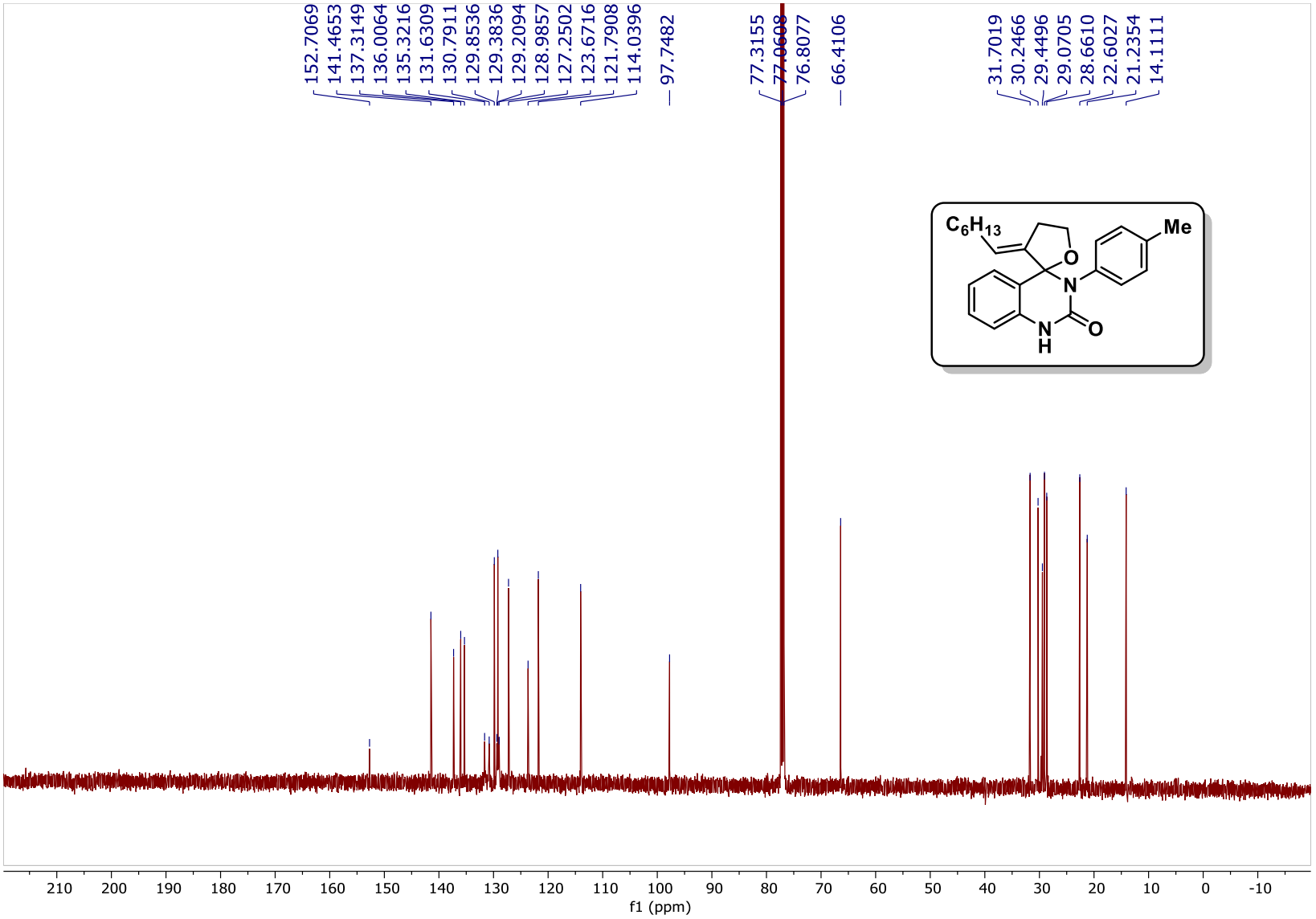
^{13}C spectrum of compound **3aq** ($CDCl_3$)



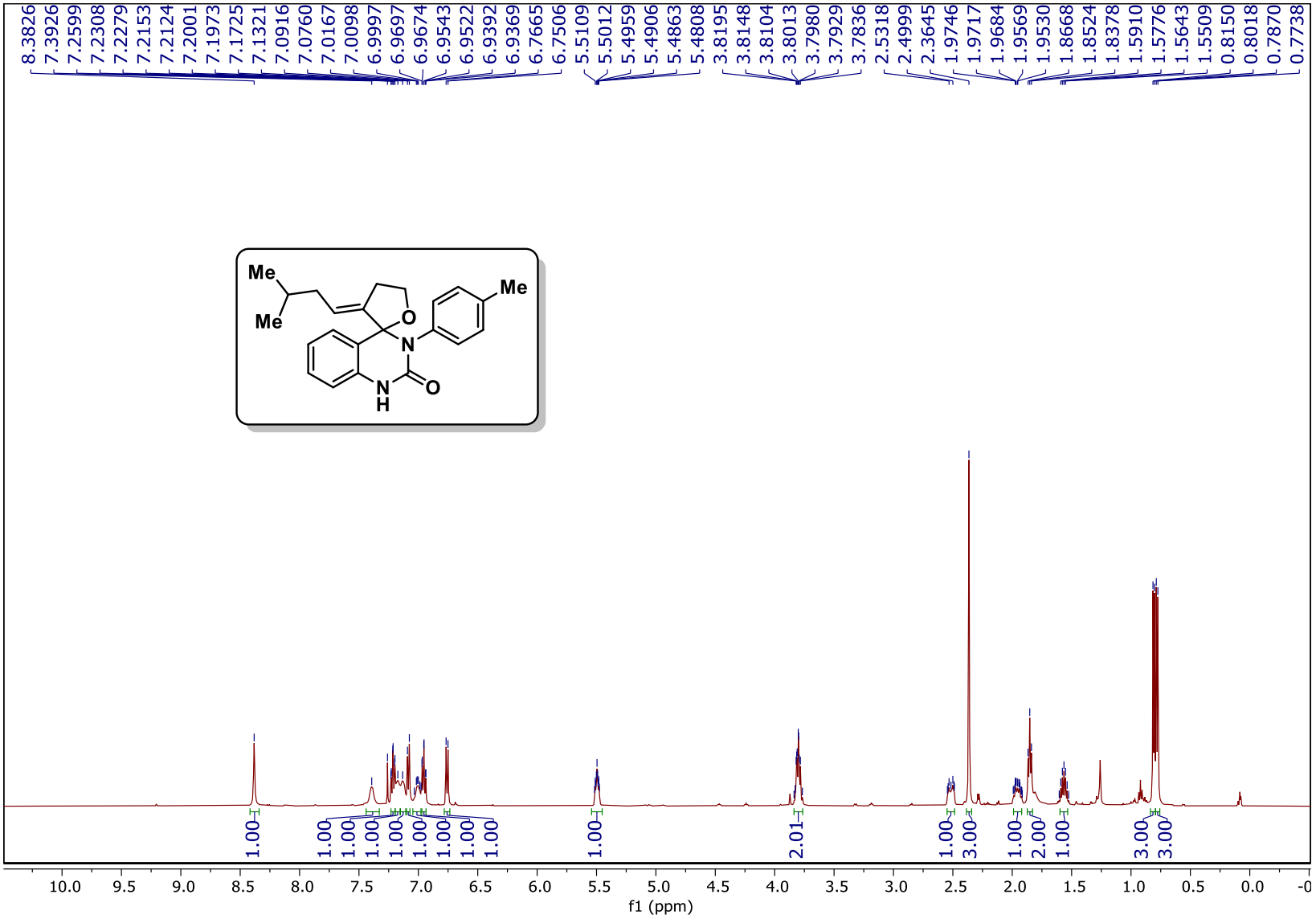
^{1}H spectrum of compound **3ar** (500 MHz, $CDCl_3$)



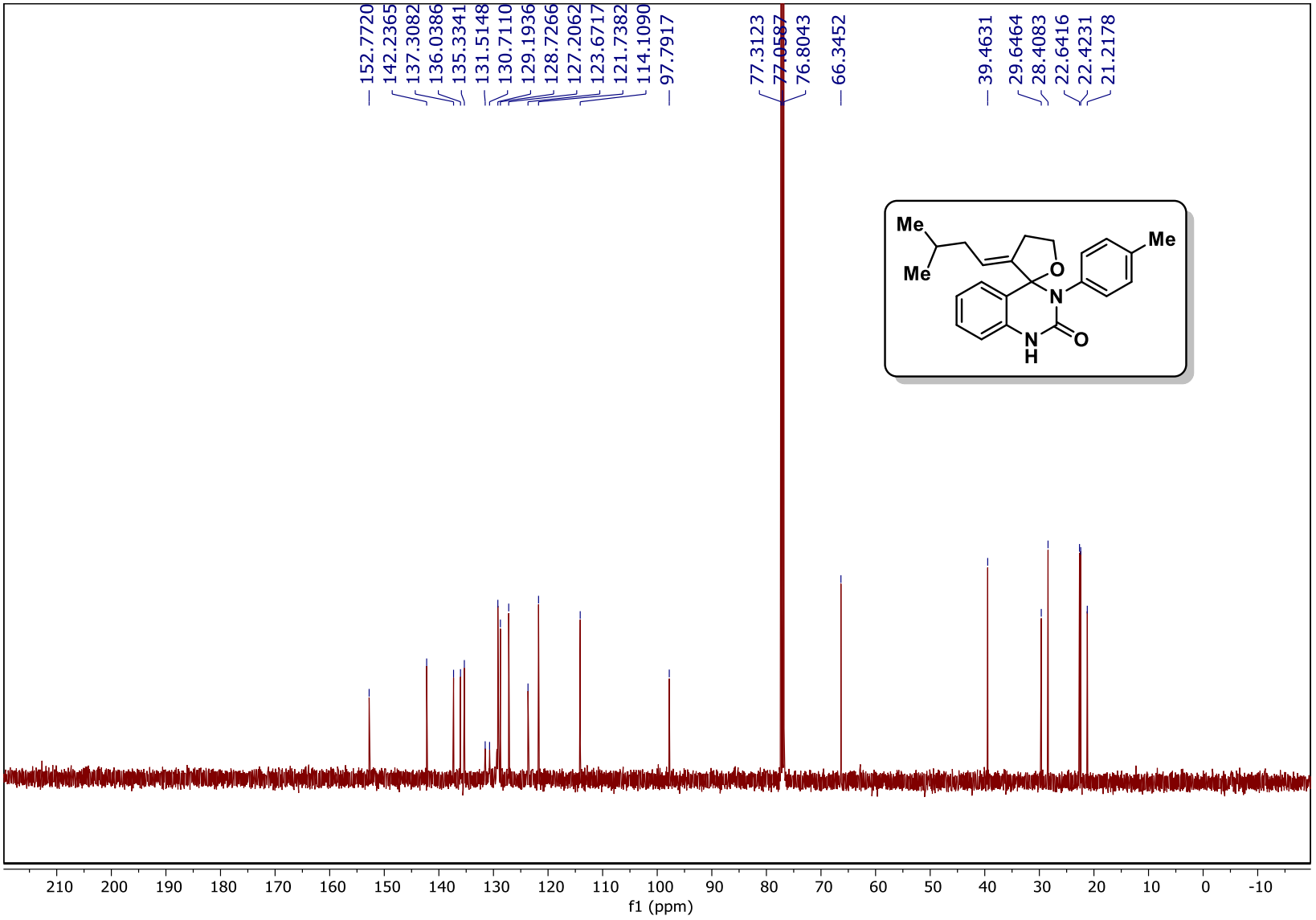
^{13}C spectrum of compound **3ar** ($CDCl_3$)



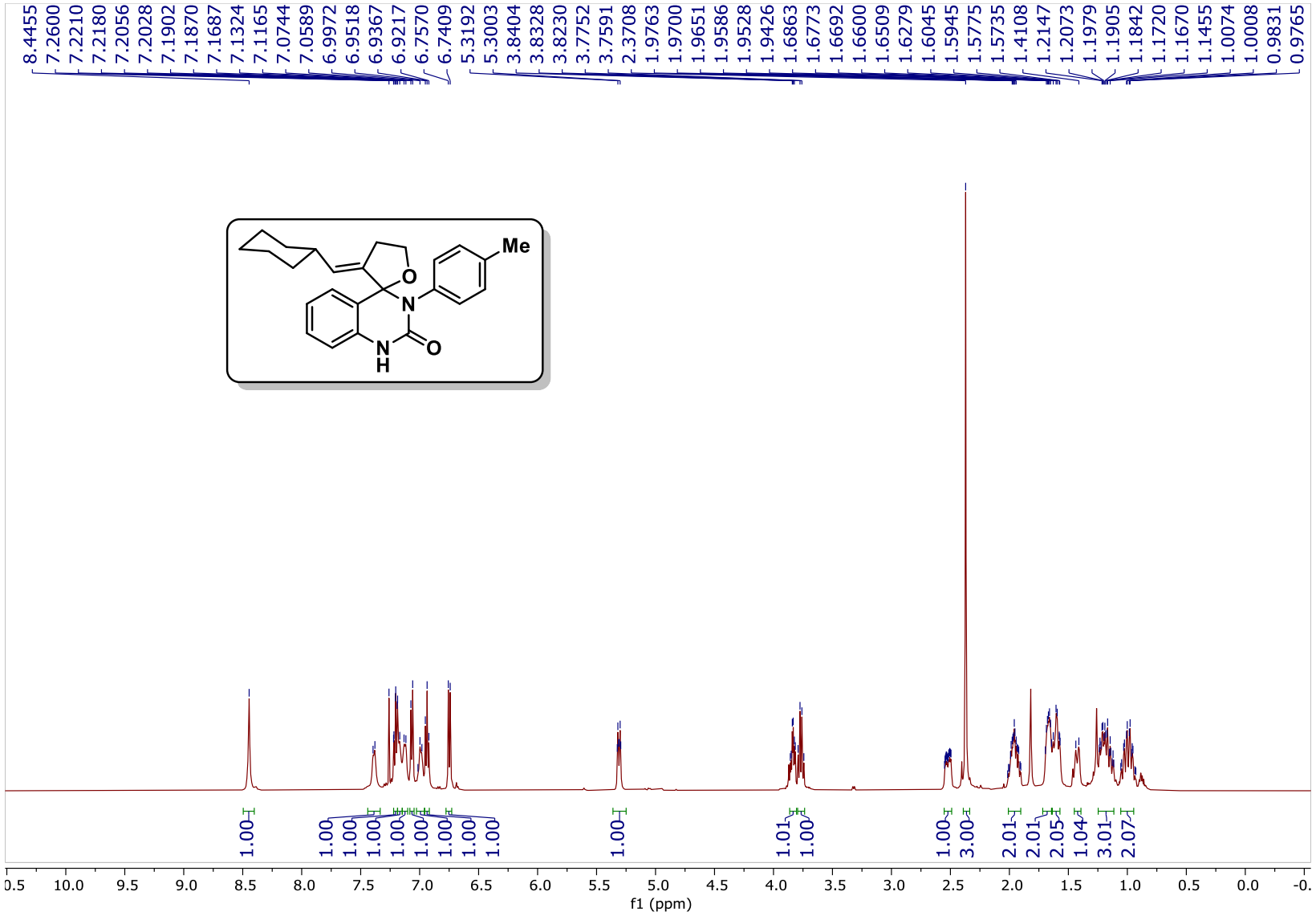
^{1}H spectrum of compound **3as** (500 MHz, $CDCl_3$)



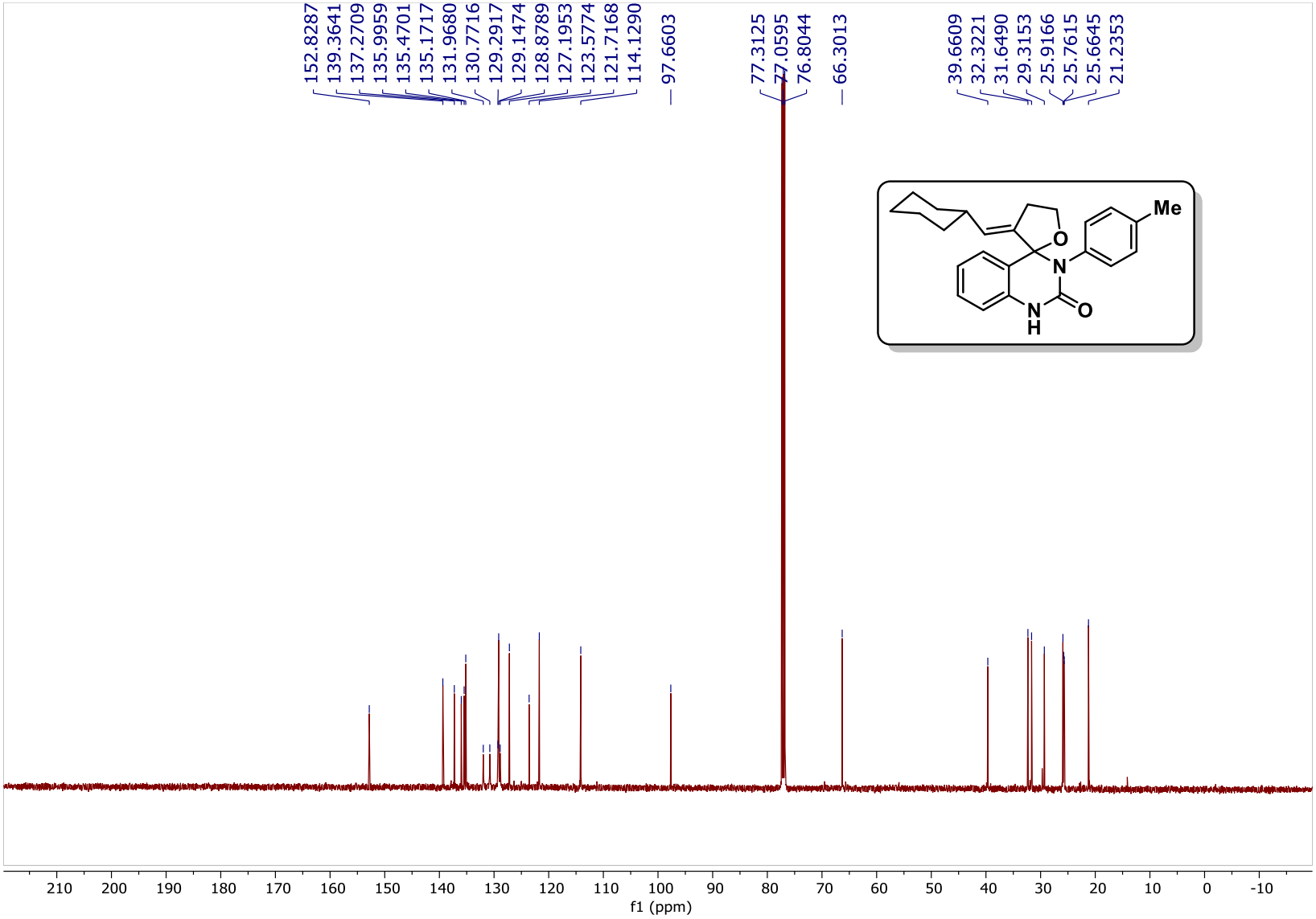
^{13}C spectrum of compound **3as** (125 MHz, $CDCl_3$)



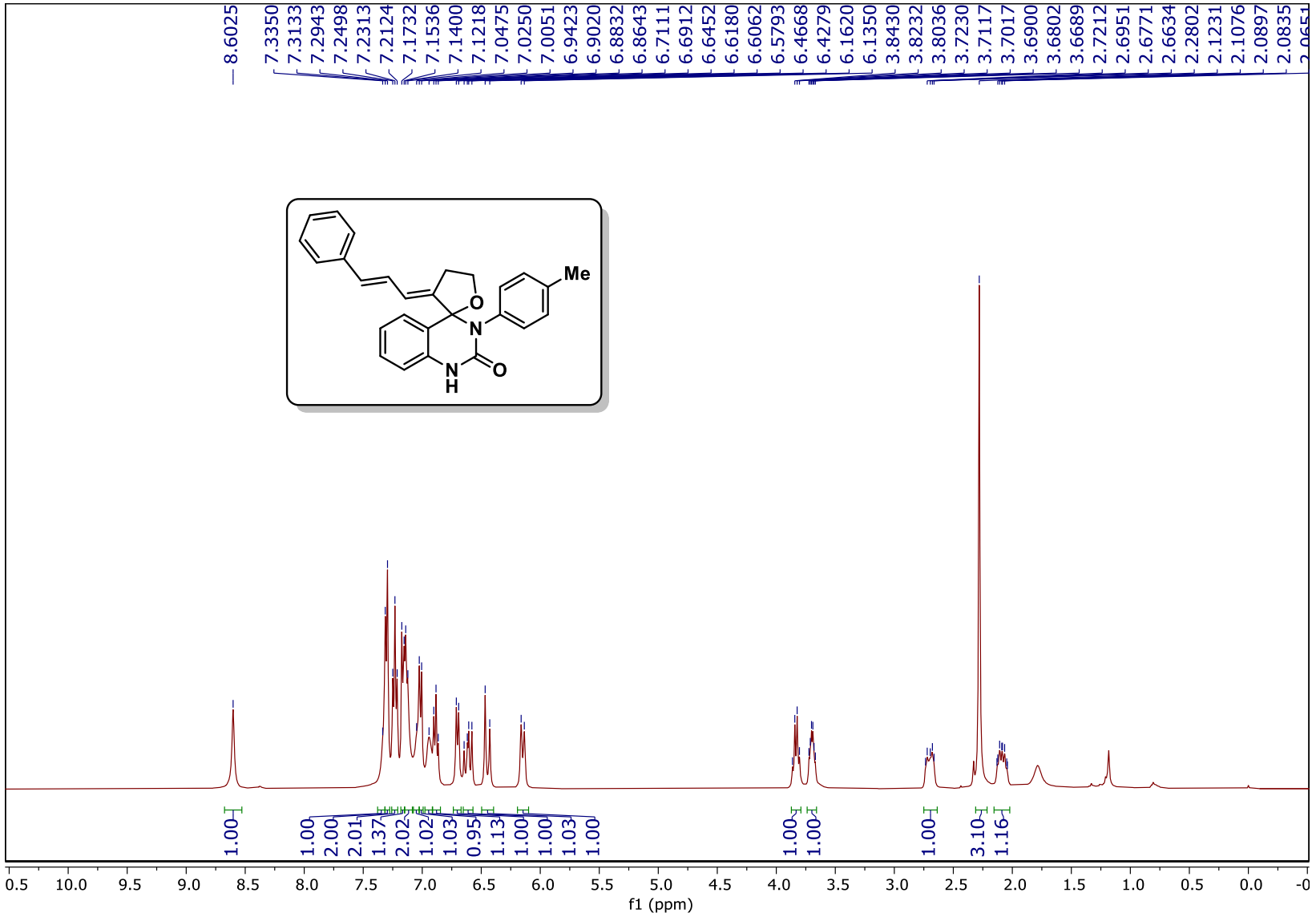
^{1}H spectrum of compound **3at** (500 MHz, $CDCl_3$)



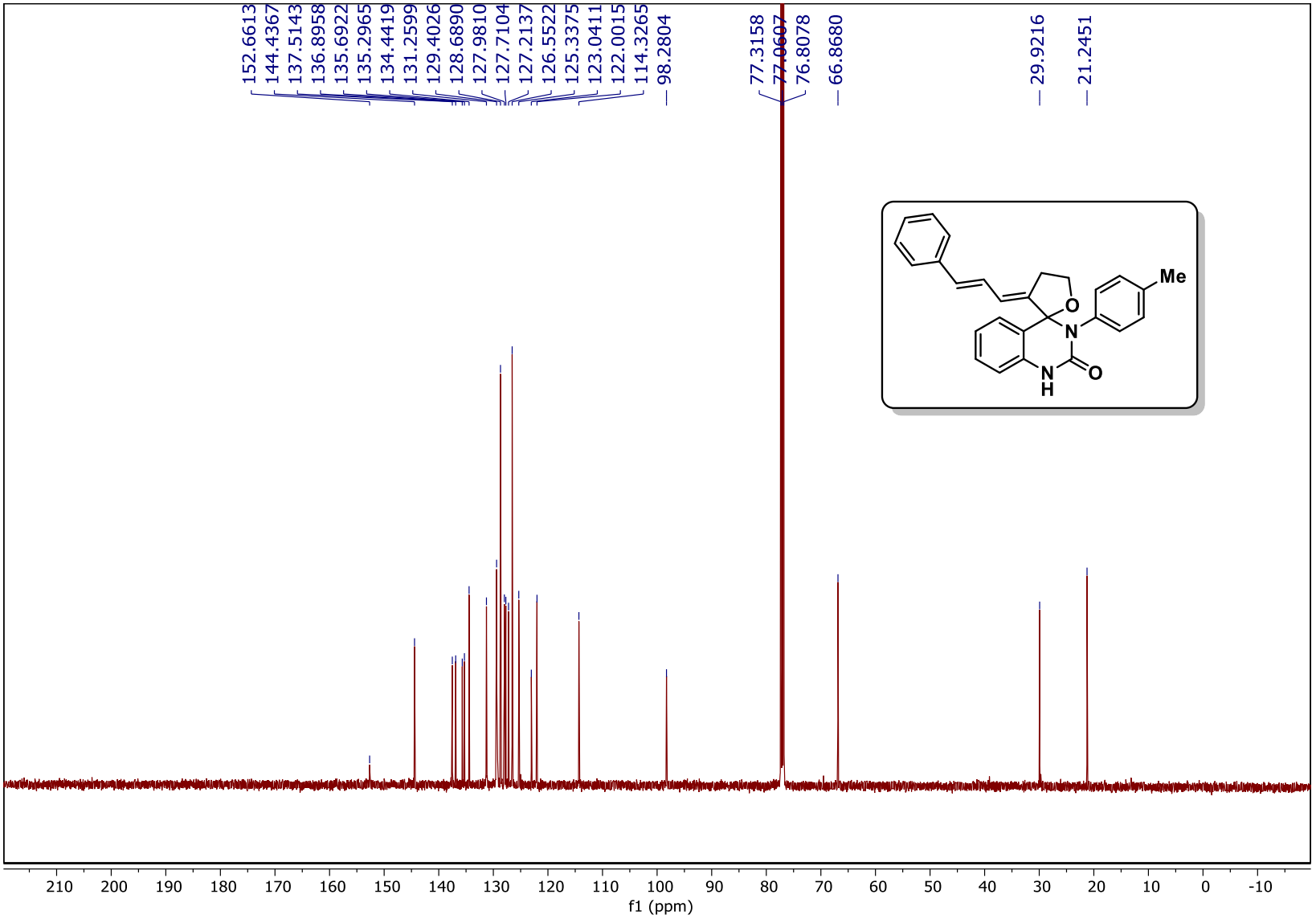
^{13}C spectrum of compound **3at** (125 MHz, $CDCl_3$)



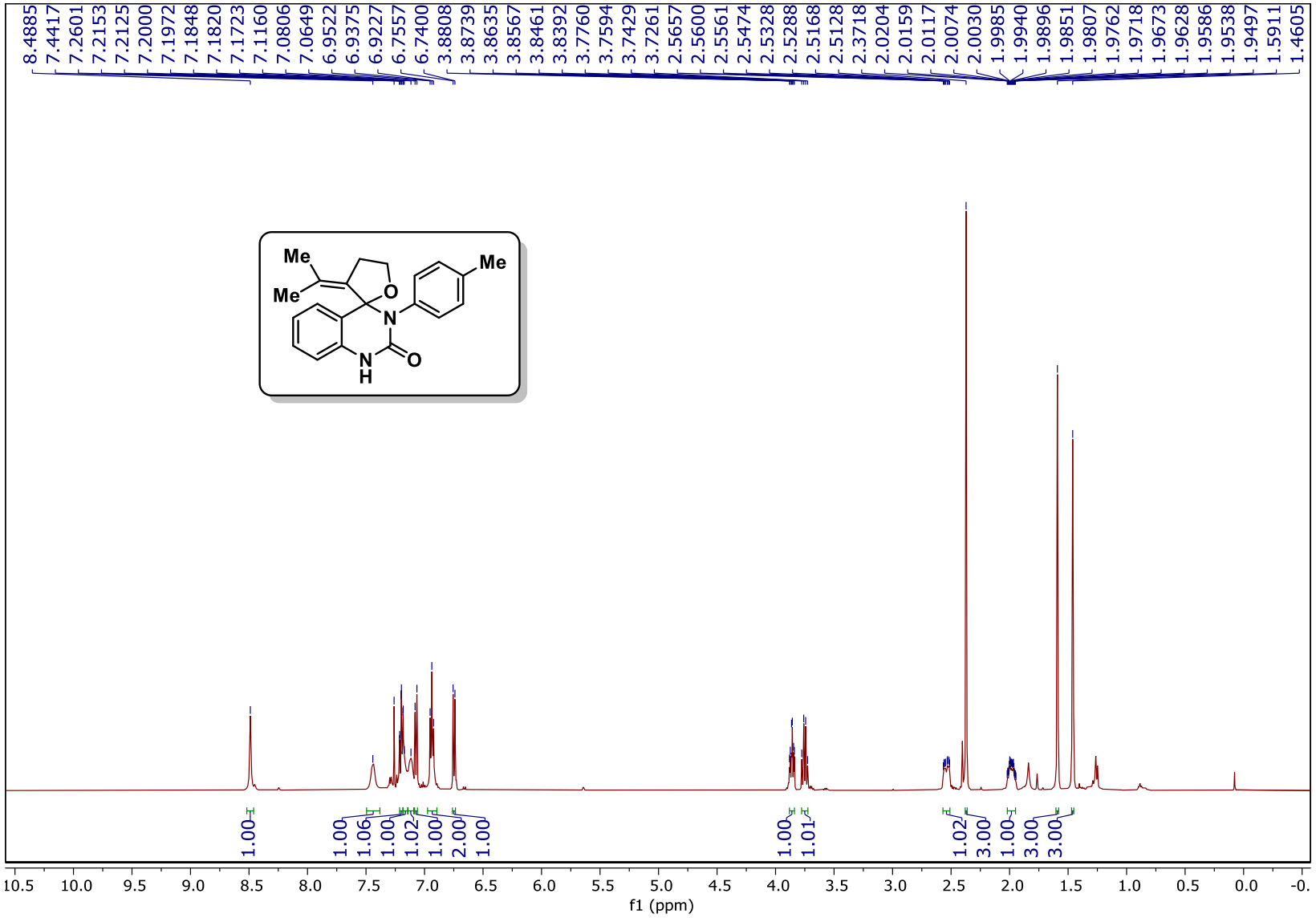
^{1}H spectrum of compound **3au** (400 MHz, $CDCl_3$)



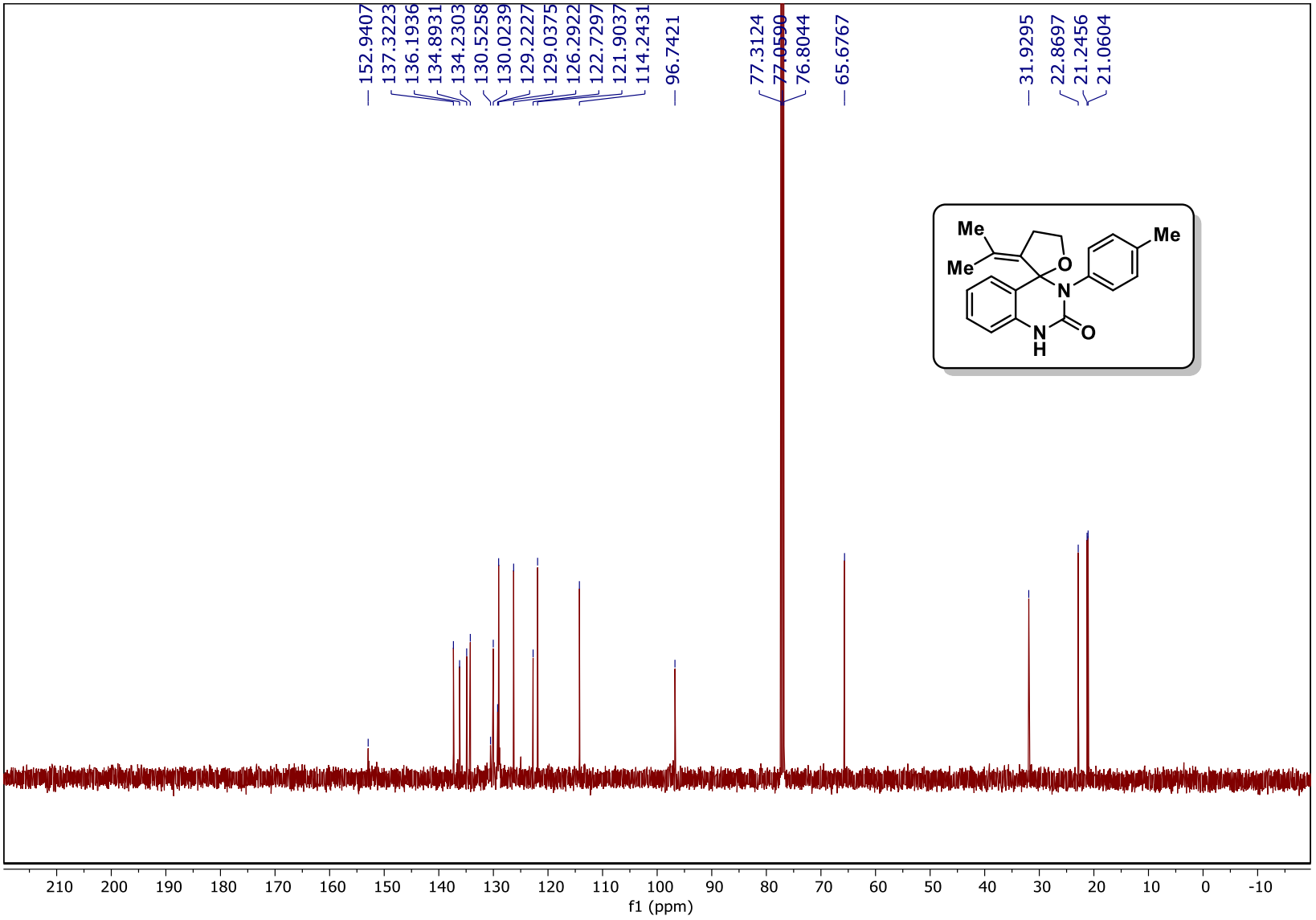
^{13}C spectrum of compound **3au** ($CDCl_3$)



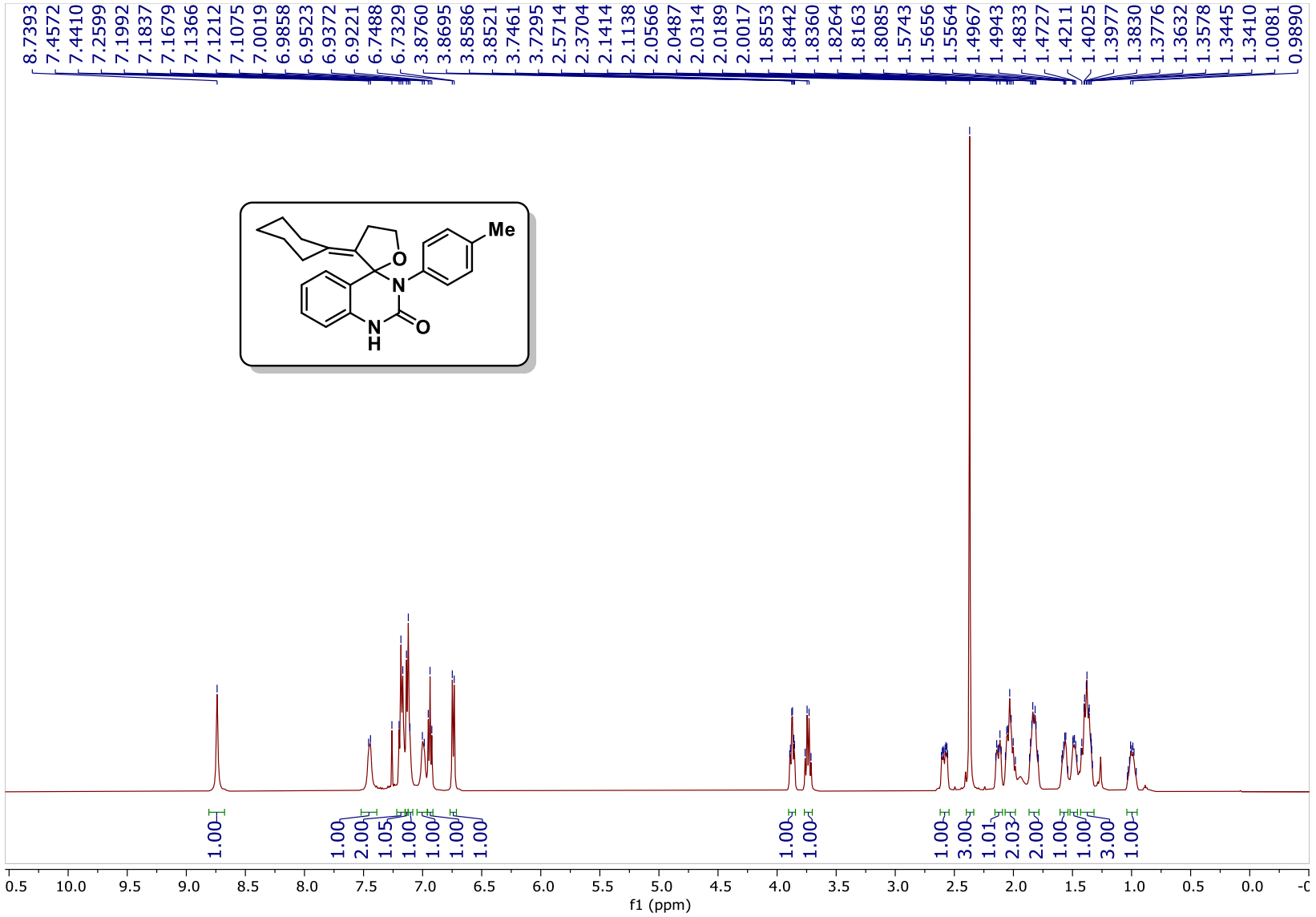
^{1}H spectrum of compound **3av** (500 MHz, $CDCl_3$)



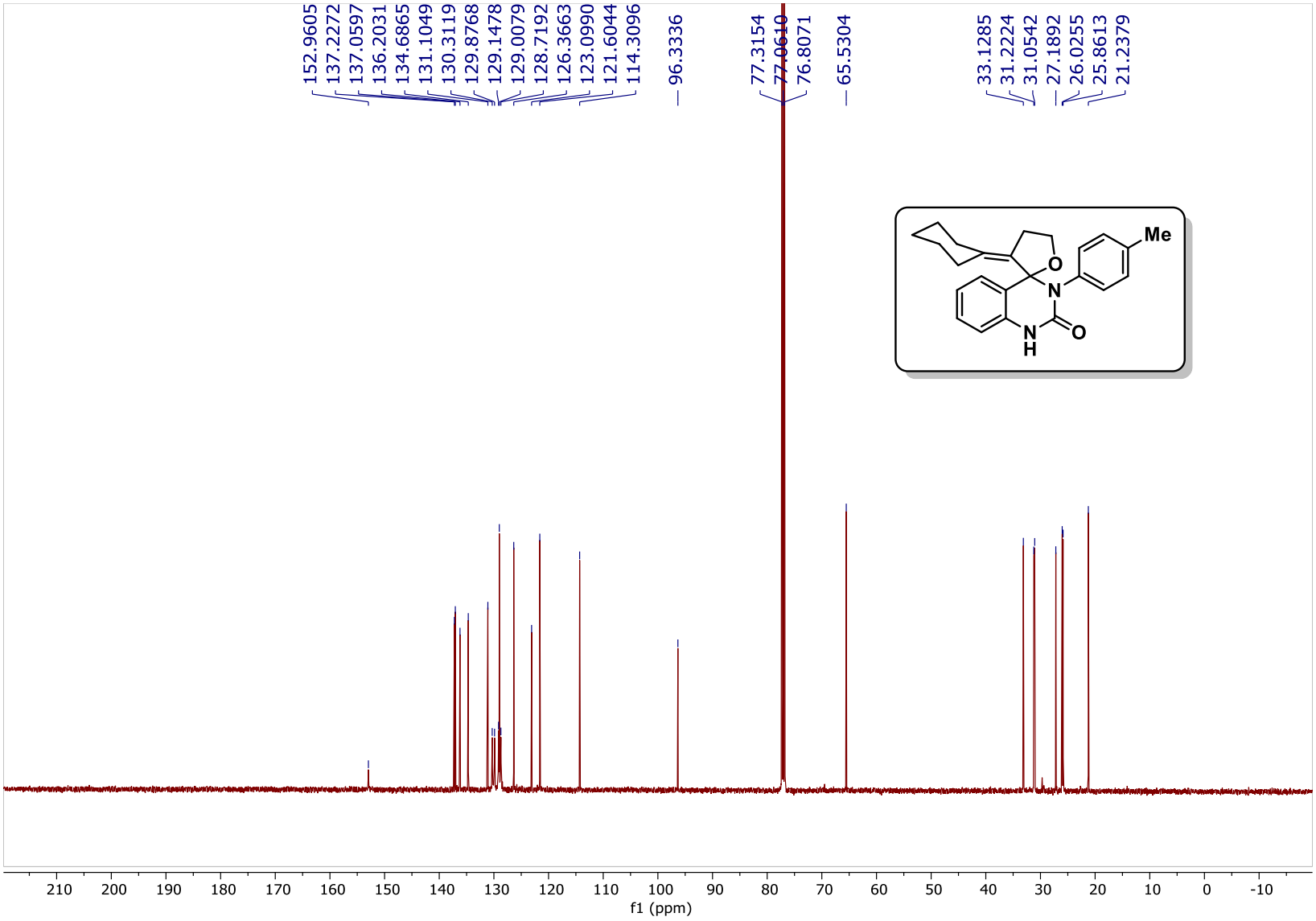
^{13}C spectrum of compound **3av** (125 MHz, $CDCl_3$)



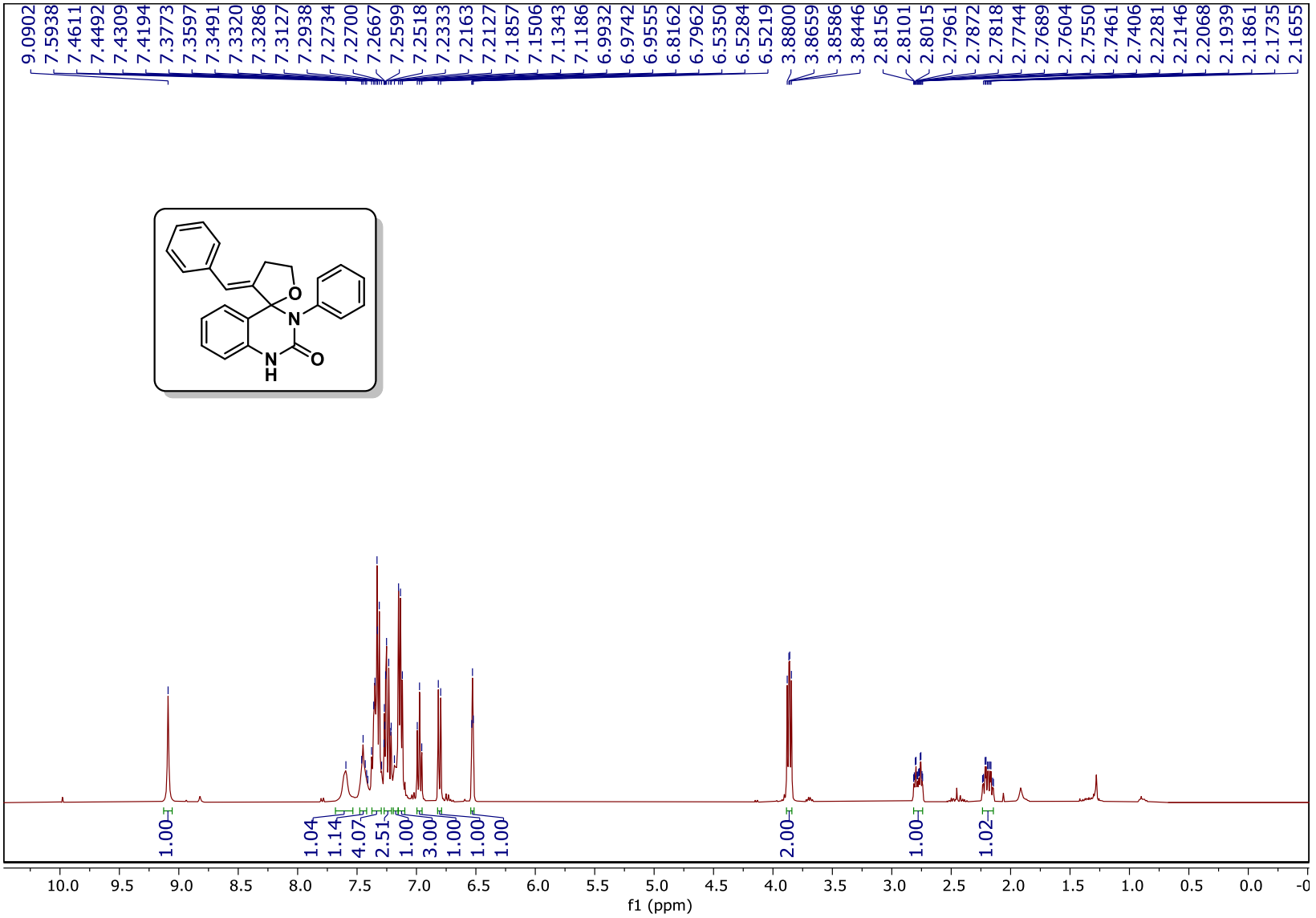
^{1}H spectrum of compound **3aw** (500 MHz, $CDCl_3$)



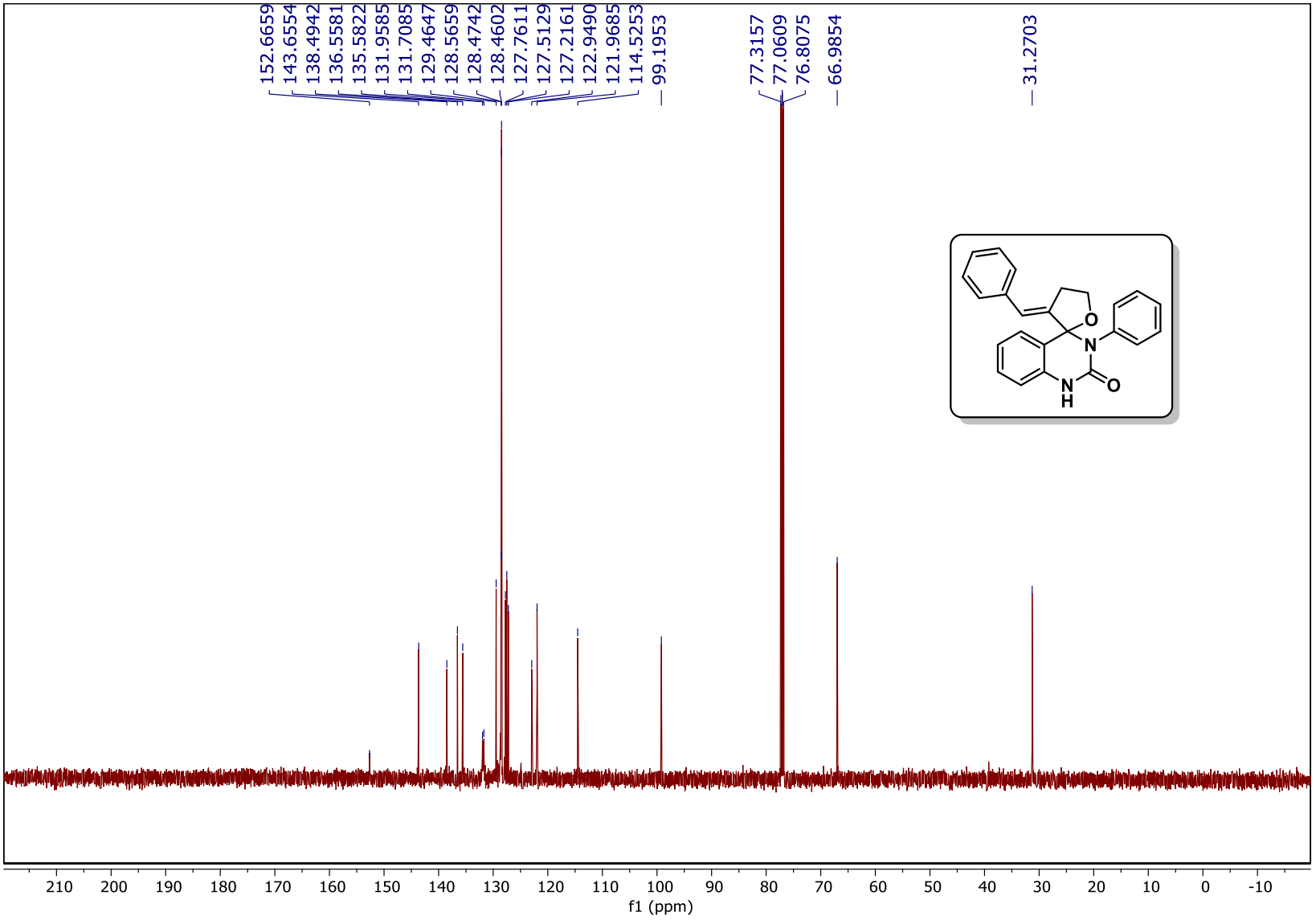
^{13}C spectrum of compound **3aw** (125 MHz, $CDCl_3$)



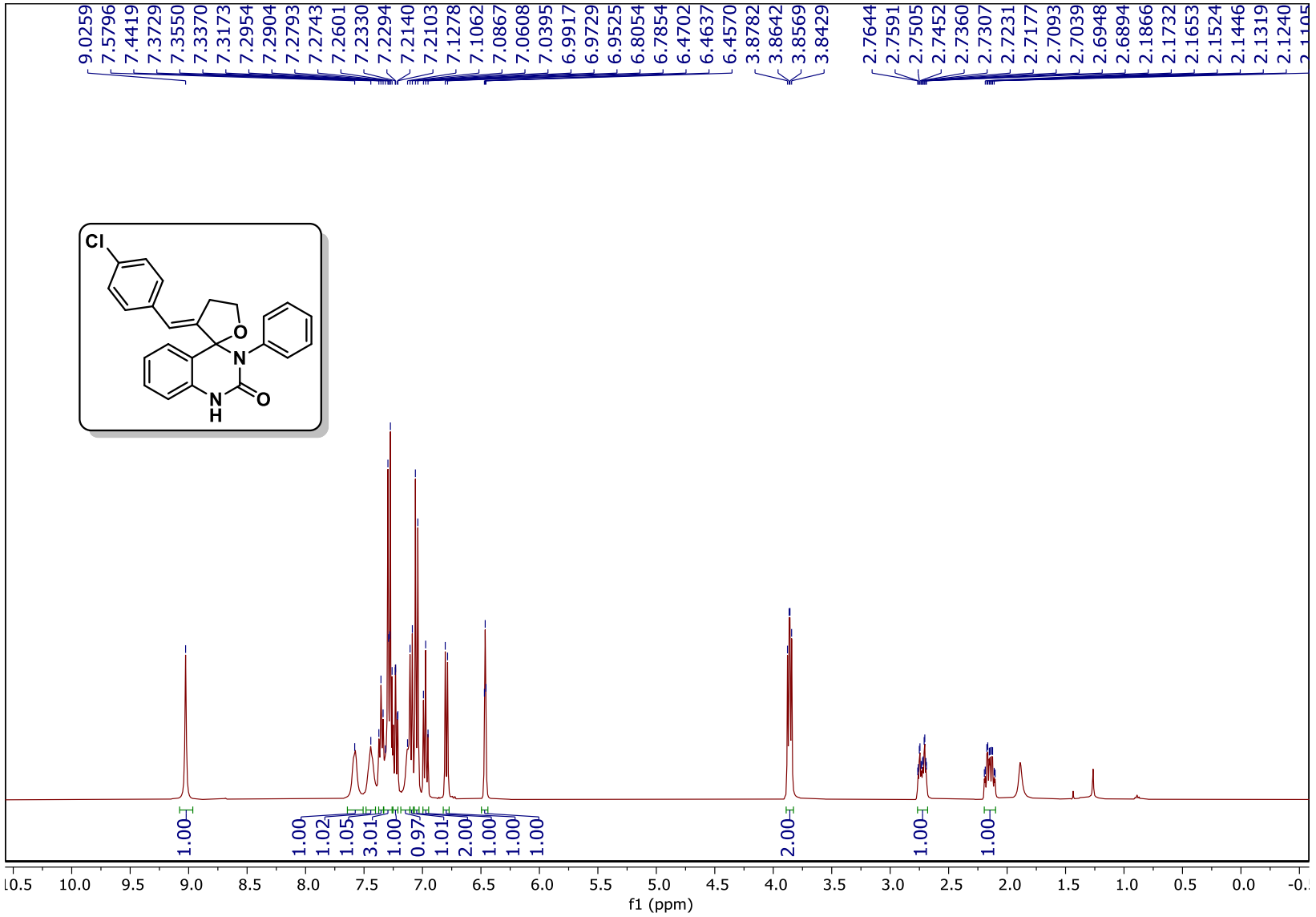
^{1}H spectrum of compound **3ba** (400 MHz, $CDCl_3$)



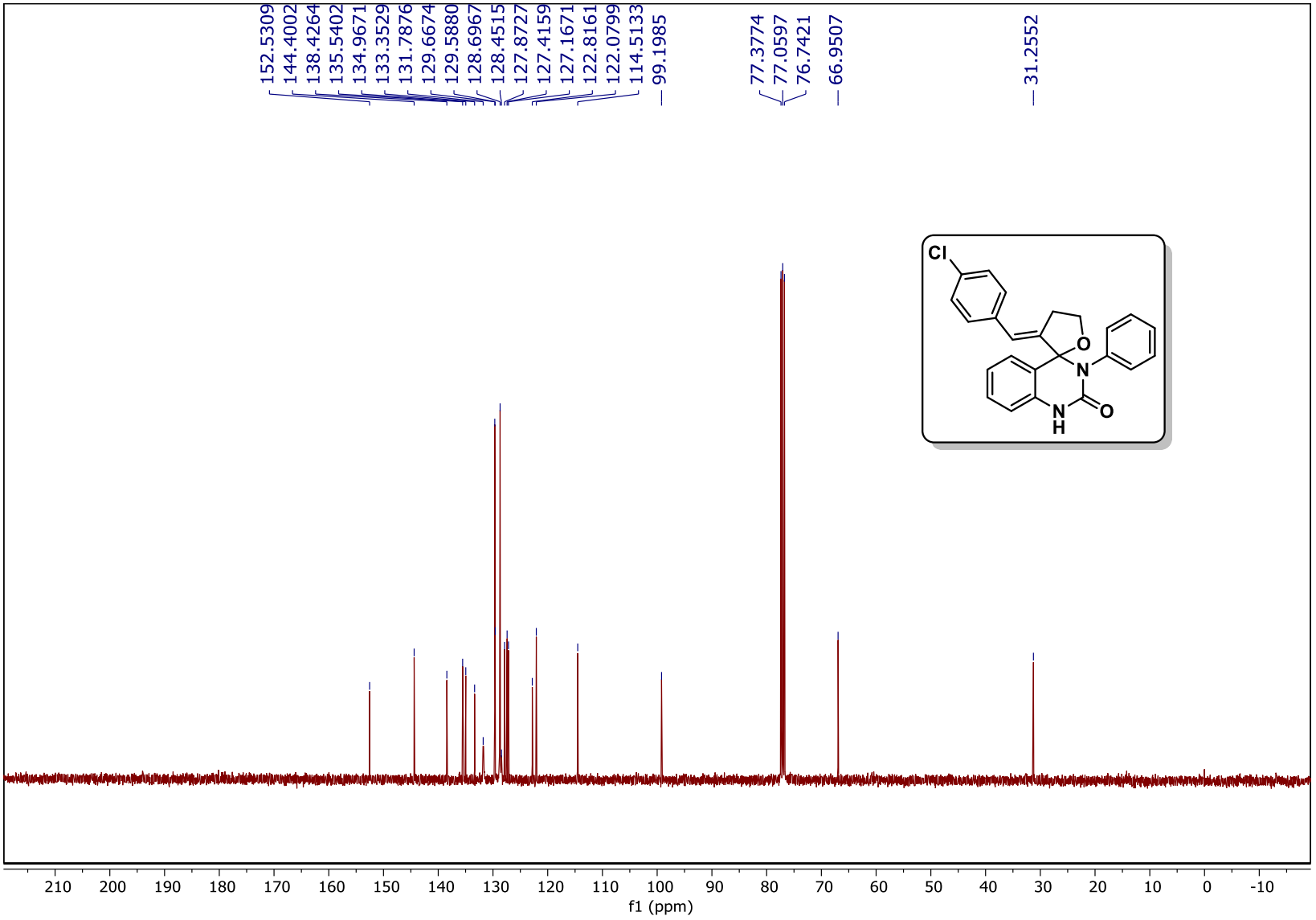
^{13}C spectrum of compound **3ba** ($CDCl_3$)



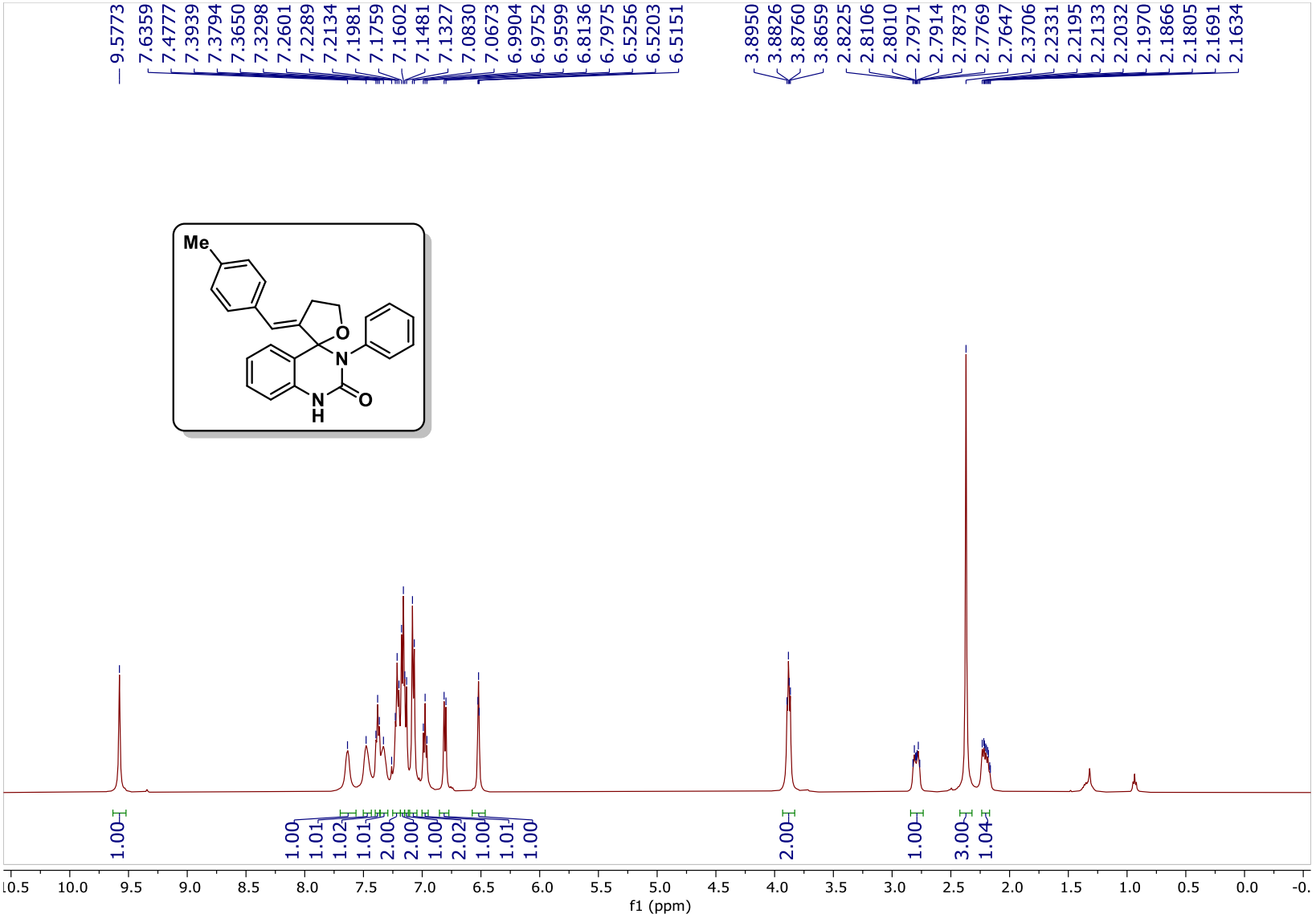
^{1}H spectrum of compound **3bb** (400 MHz, $CDCl_3$)



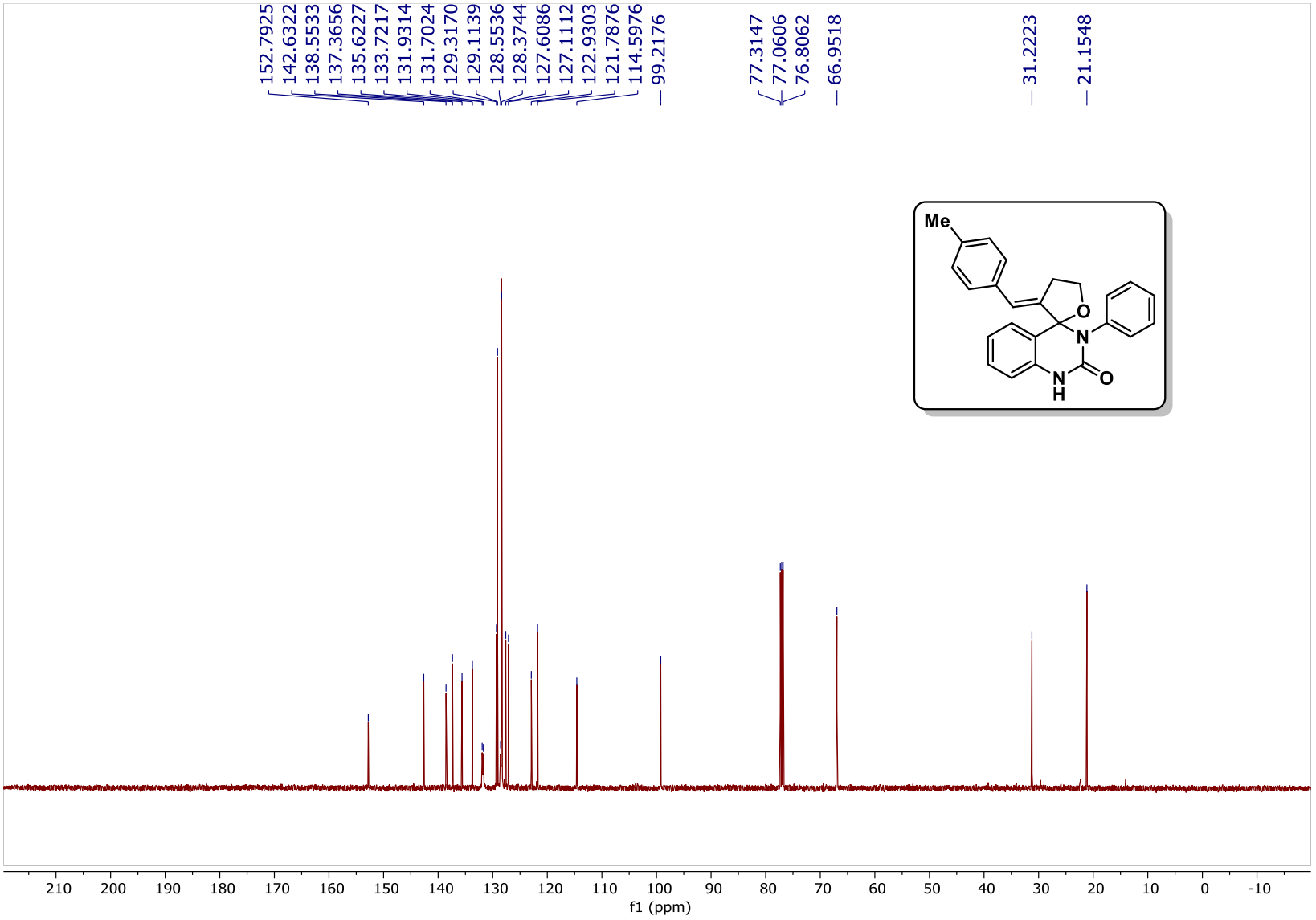
^{13}C spectrum of compound **3bb** (100 MHz, $CDCl_3$)



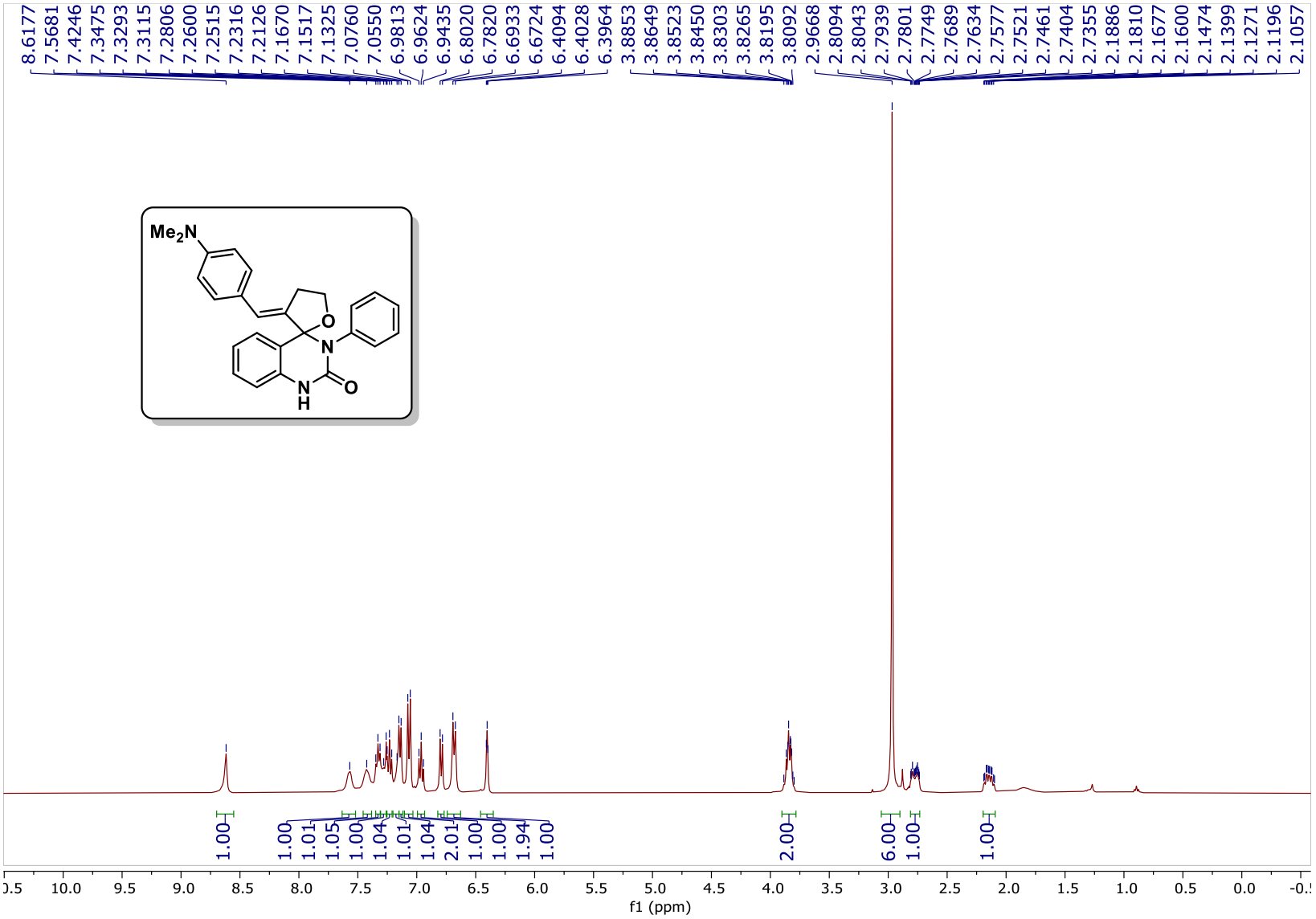
^{1}H spectrum of compound **3bi** (500 MHz, $CDCl_3$)



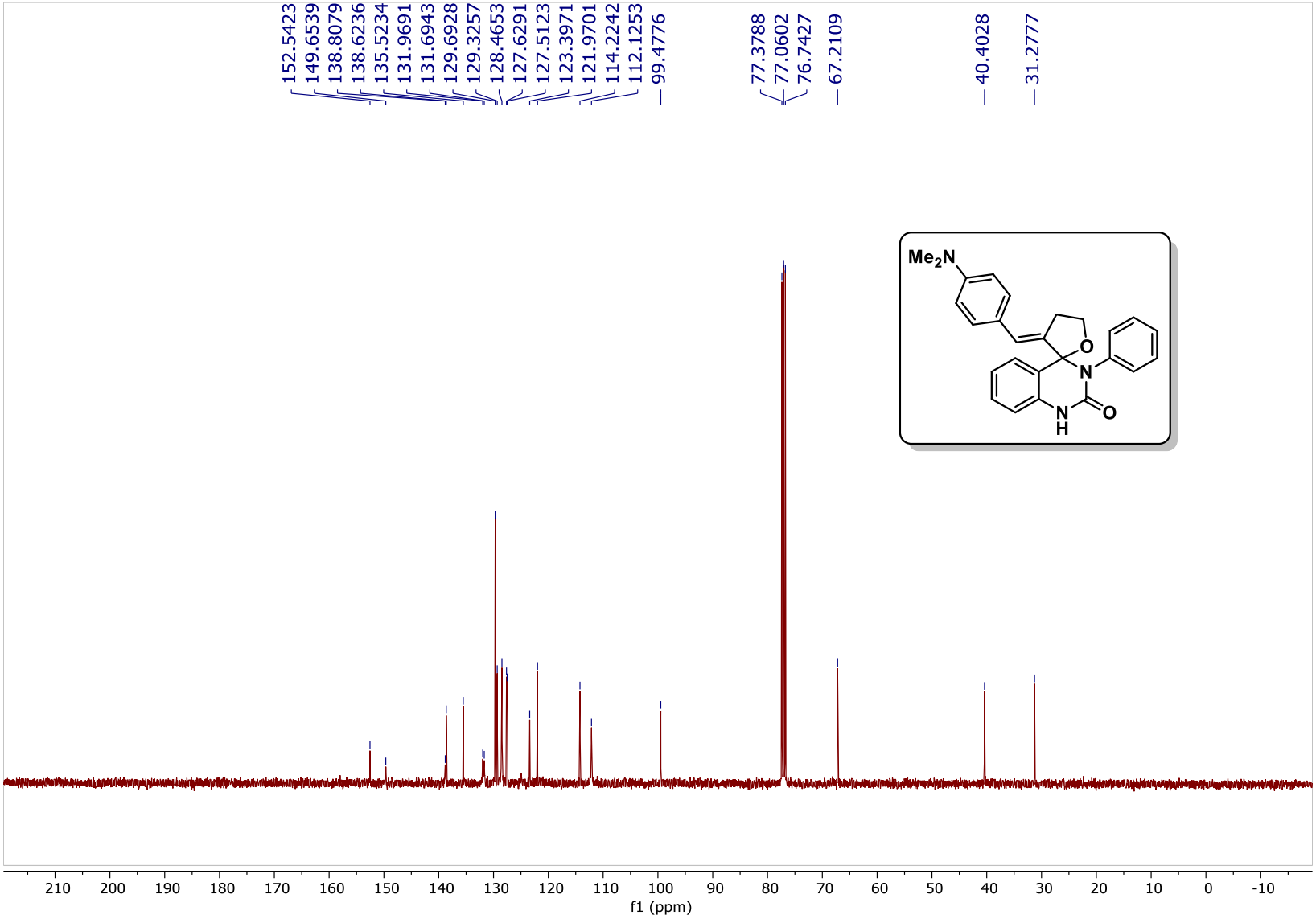
^{13}C spectrum of compound **3bi** (125 MHz, $CDCl_3$)



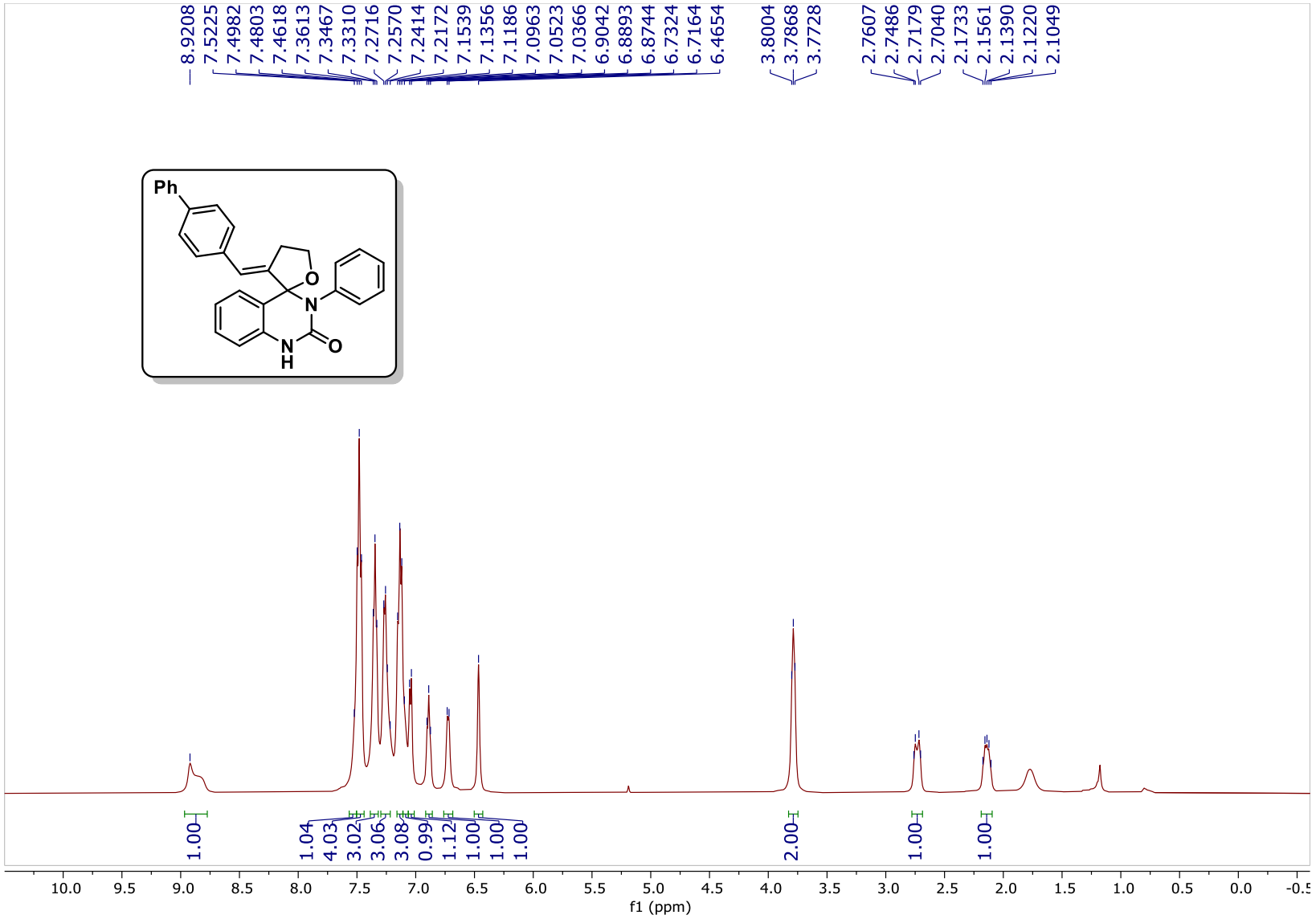
^{1}H spectrum of compound **3bx** (400 MHz, $CDCl_3$)



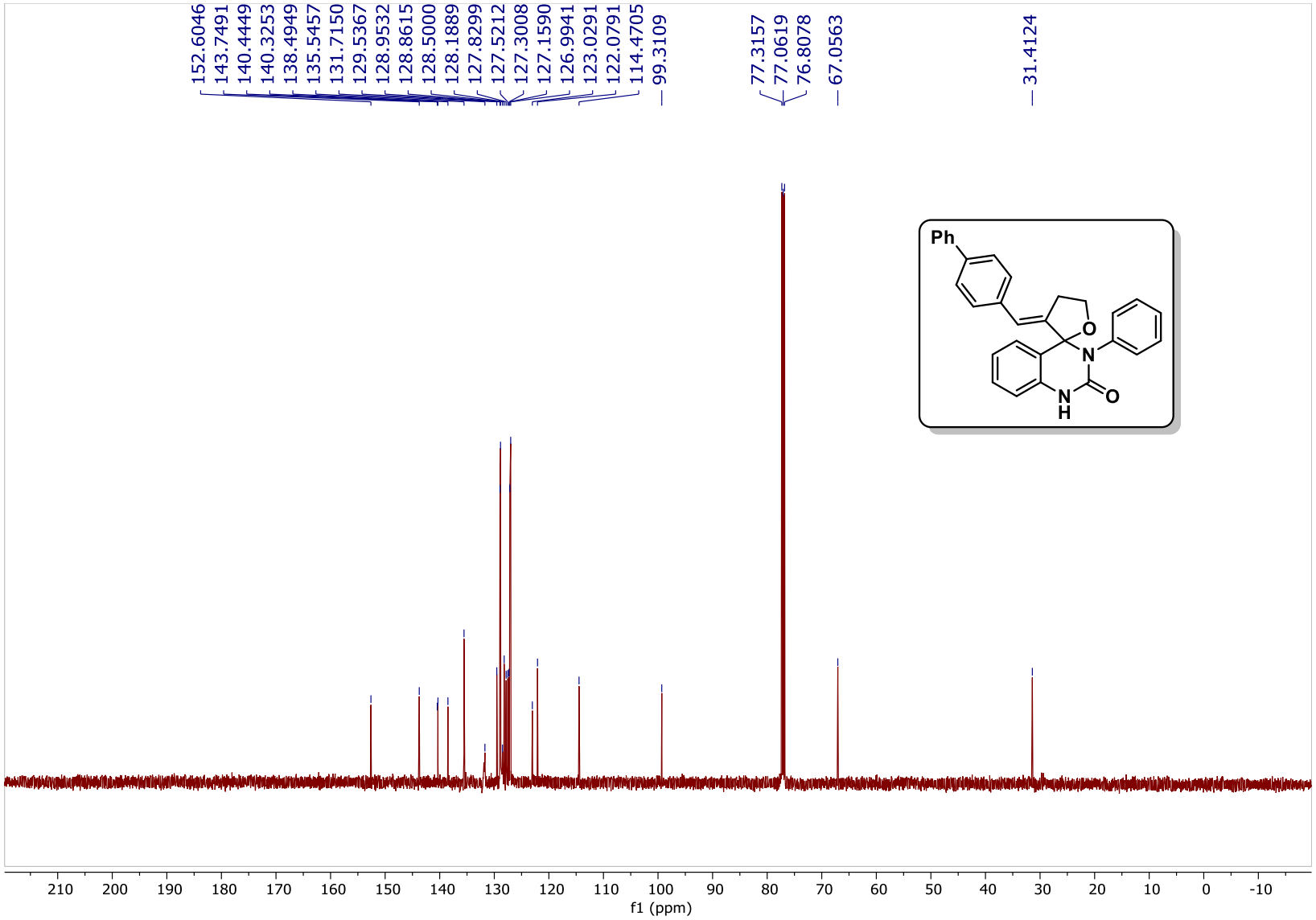
^{13}C spectrum of compound **3bx** (100 MHz, $CDCl_3$)



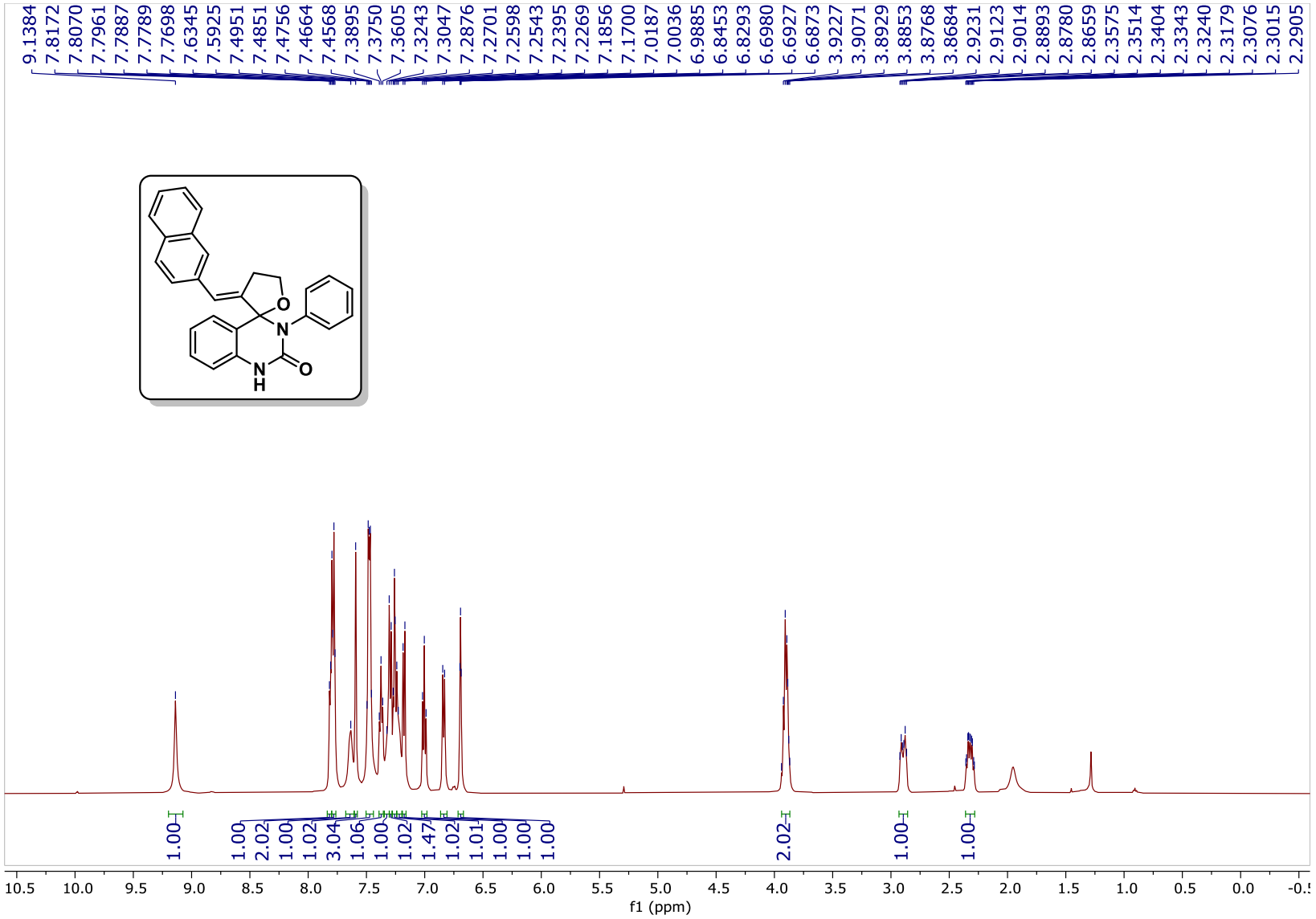
^{1}H spectrum of compound **3by** (500 MHz, $CDCl_3$)



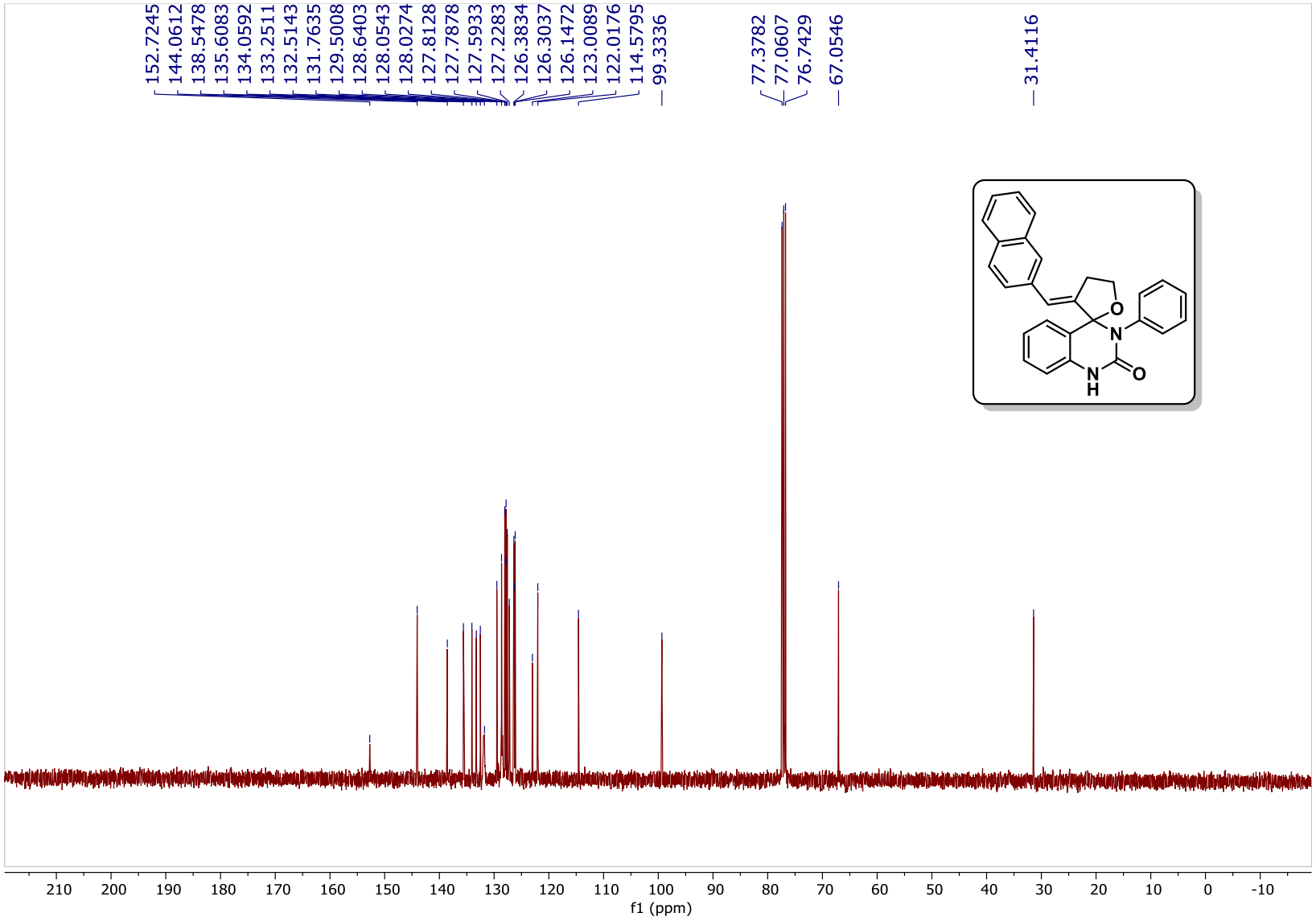
^{13}C spectrum of compound **3by** (125 MHz, $CDCl_3$)



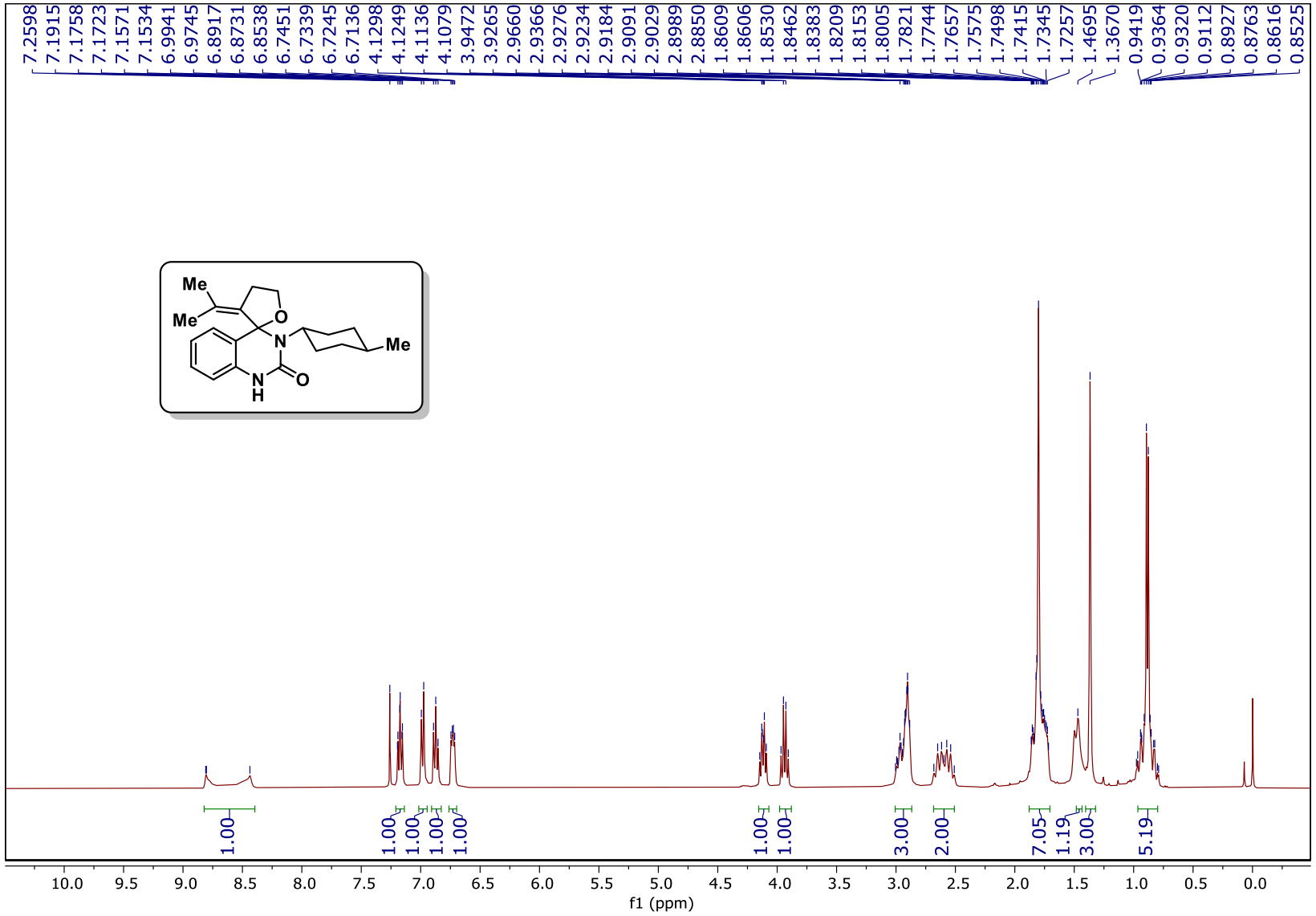
^{1}H spectrum of compound **3bm** (500 MHz, $CDCl_3$)



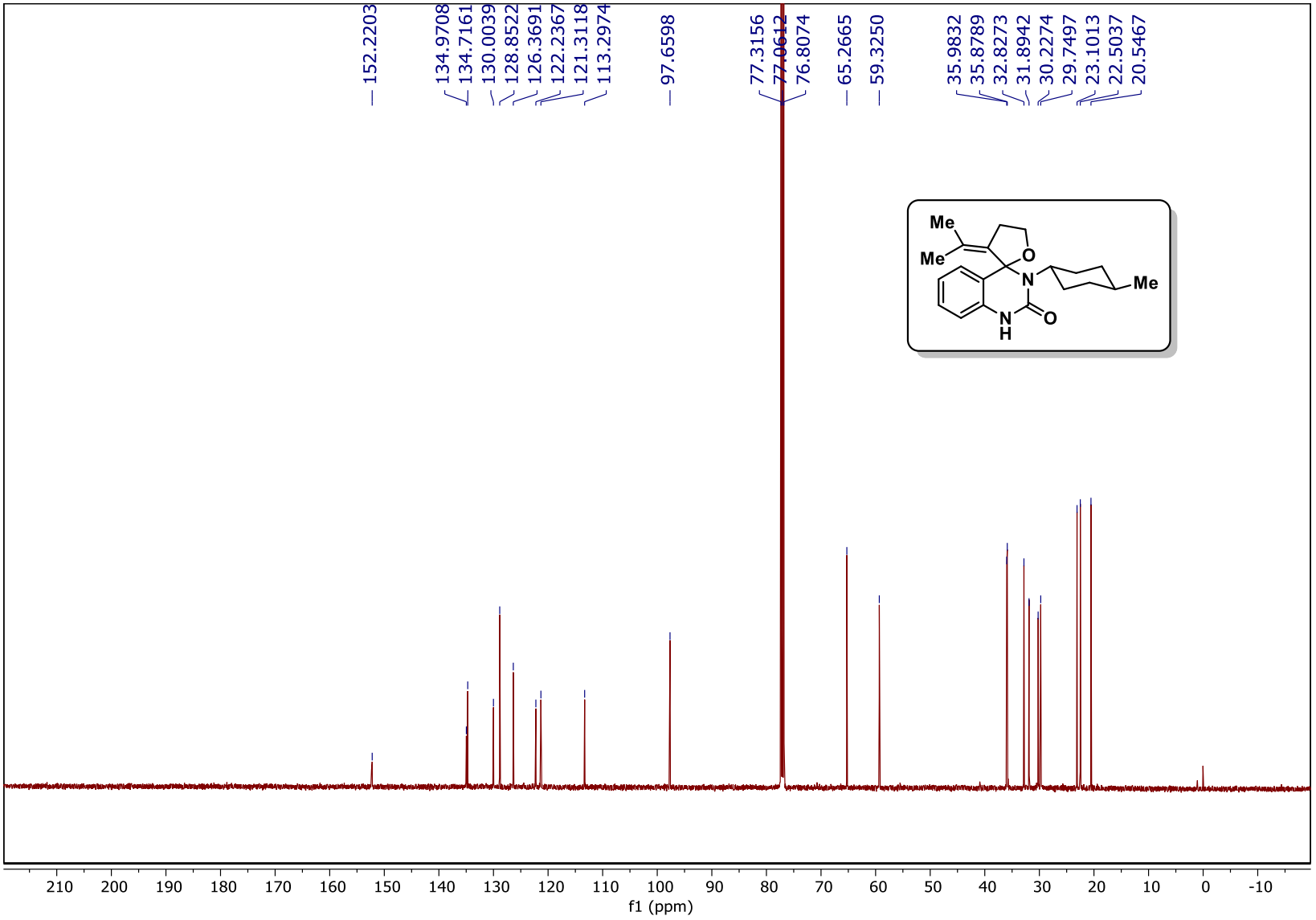
^{13}C spectrum of compound **3bm** (100 MHz, $CDCl_3$)



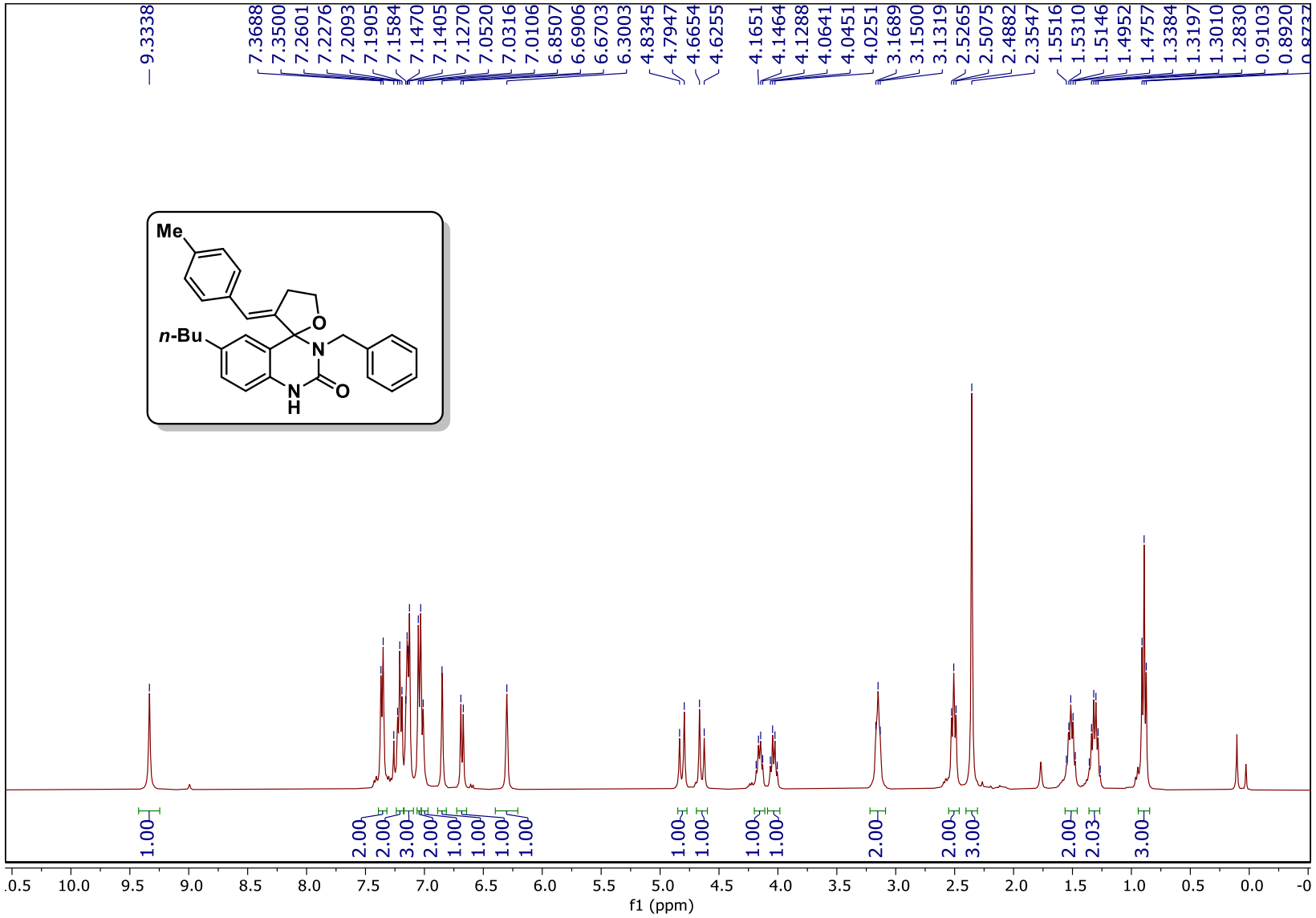
^{1}H spectrum of compound **3cv** (400 MHz, $CDCl_3$)



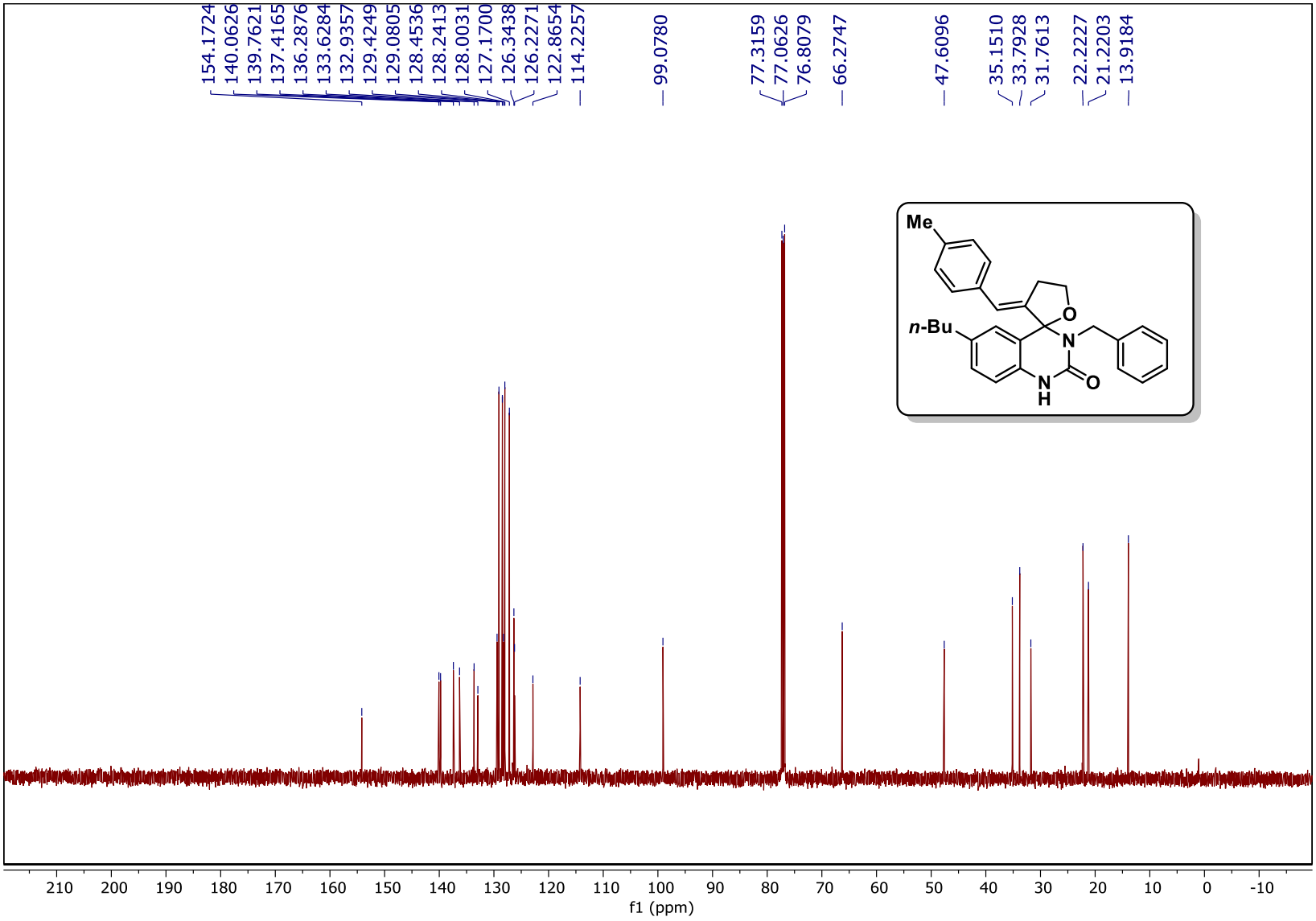
^{13}C spectrum of compound **3cv** (125 MHz, $CDCl_3$)



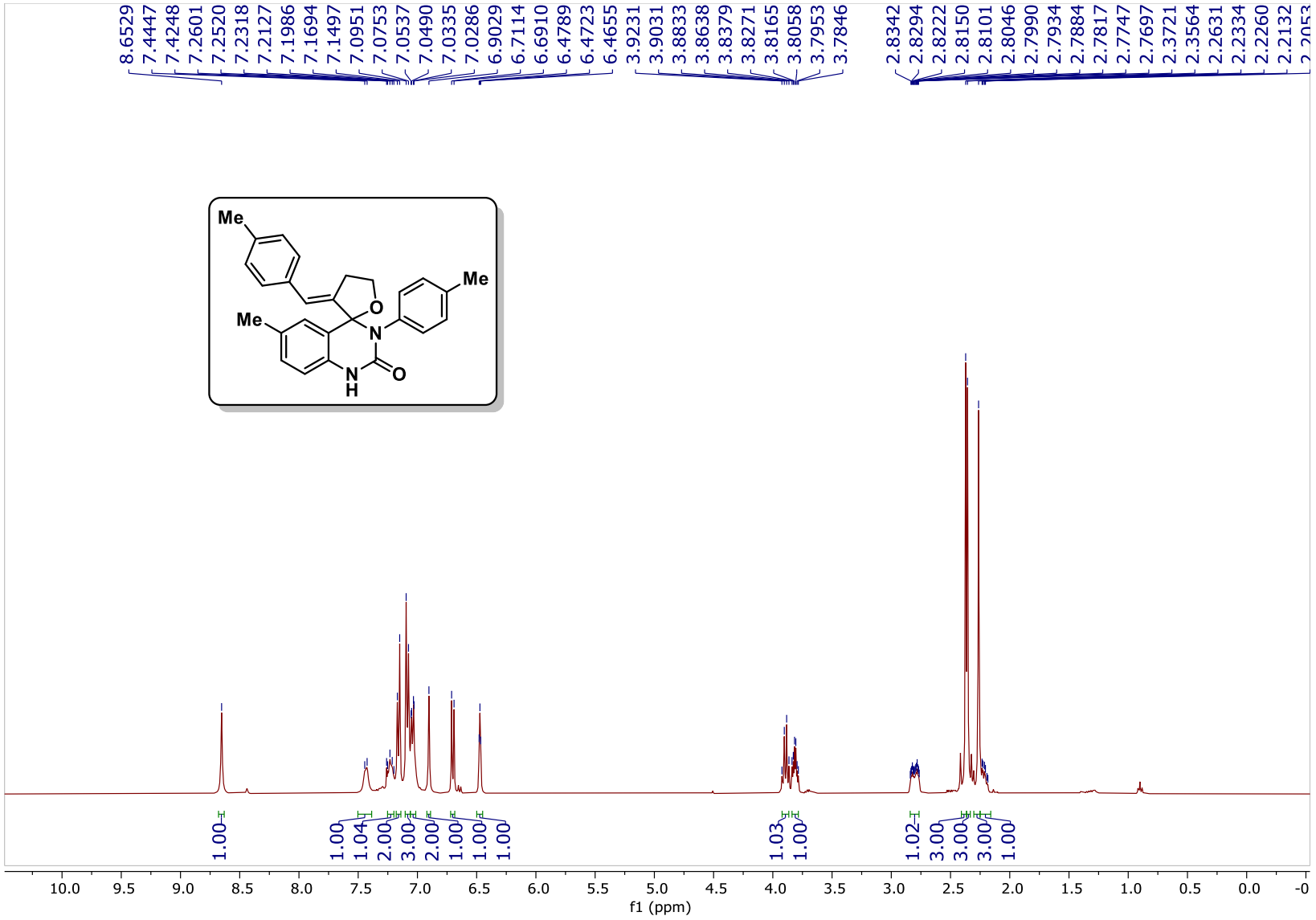
^{1}H spectrum of compound **3di** (400 MHz, $CDCl_3$)



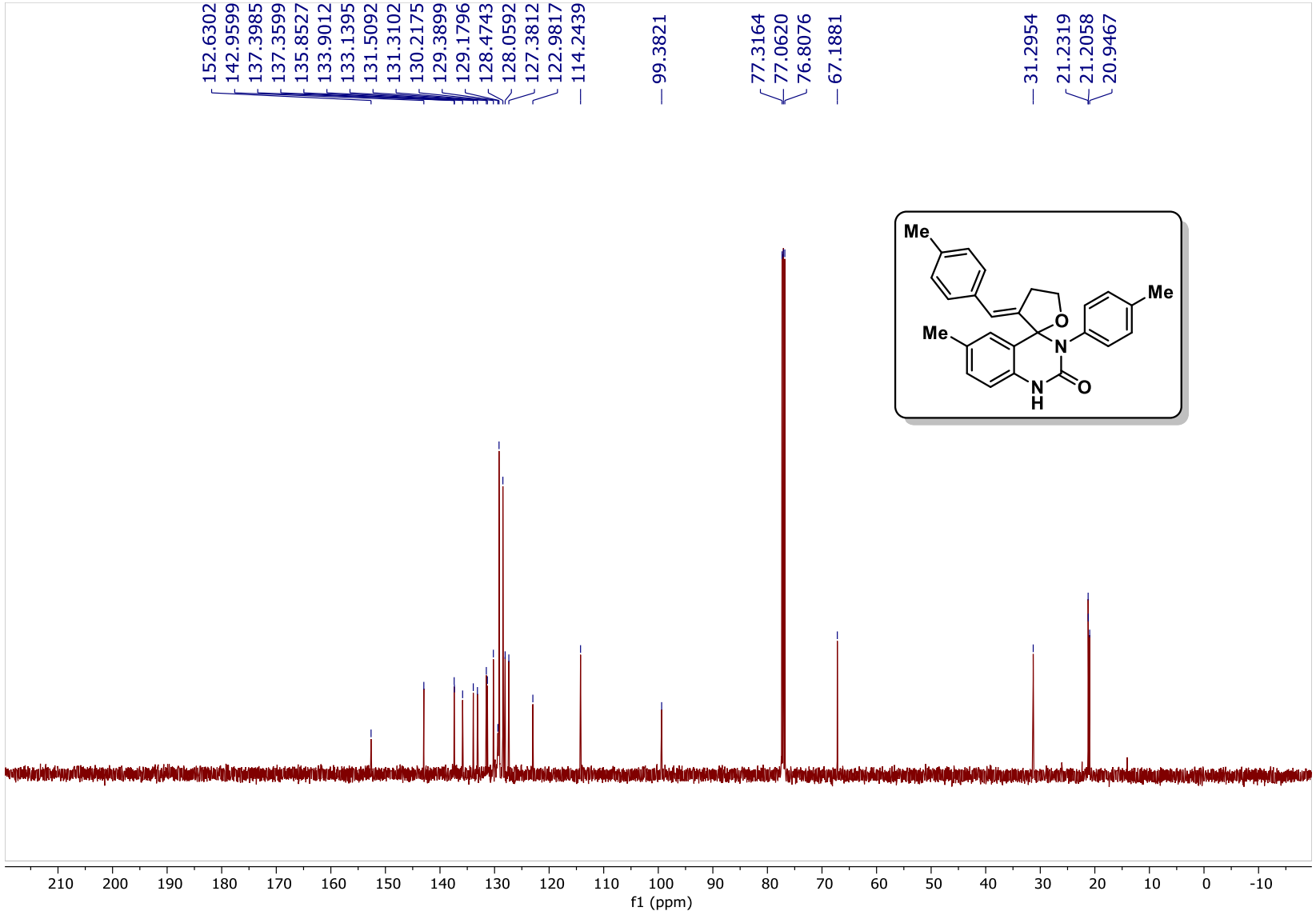
^{13}C spectrum of compound **3di** (125 MHz, $CDCl_3$)



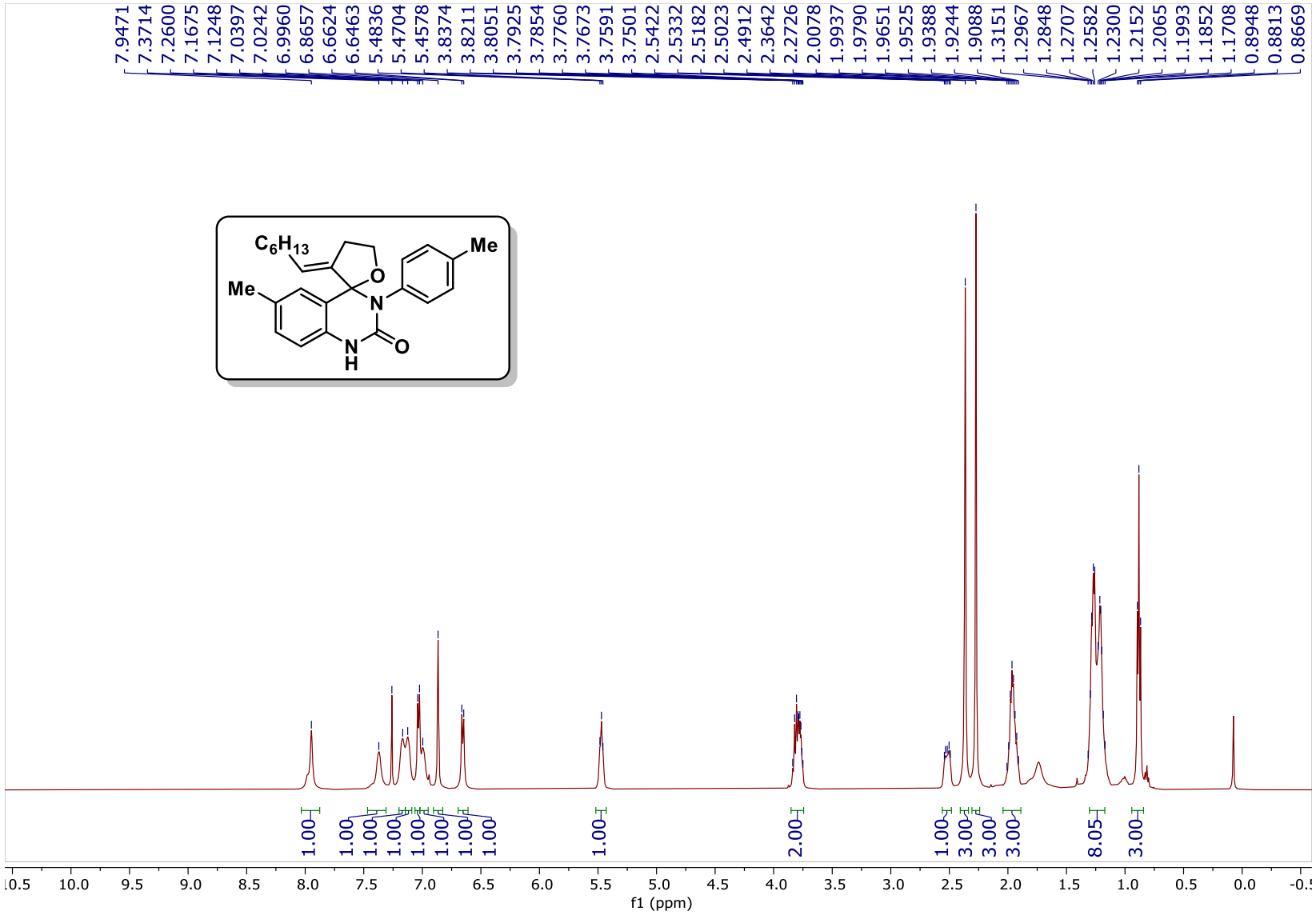
^{1}H spectrum of compound **3ei** (400 MHz, $CDCl_3$)



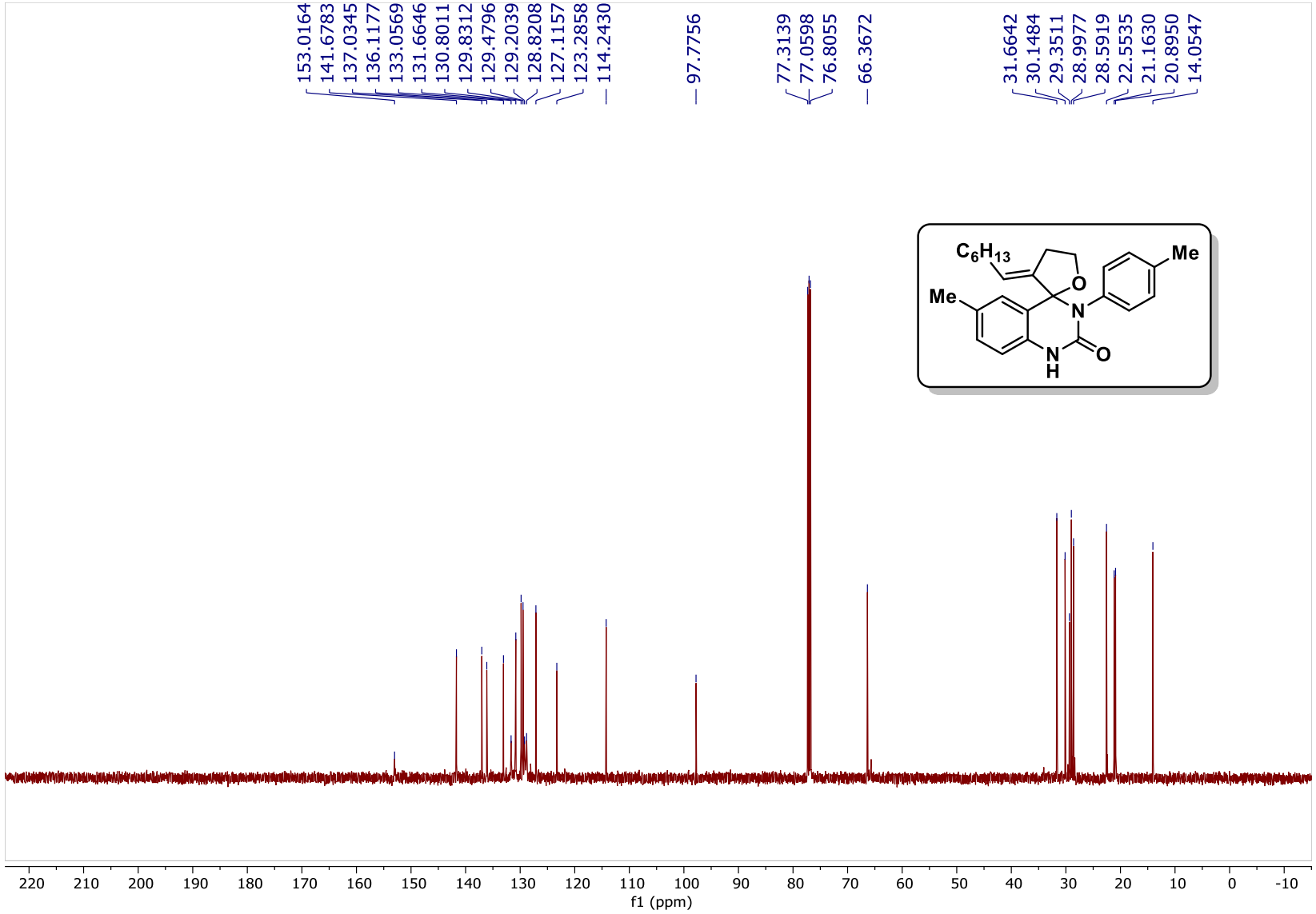
^{13}C spectrum of compound **3ei** (125 MHz, $CDCl_3$)



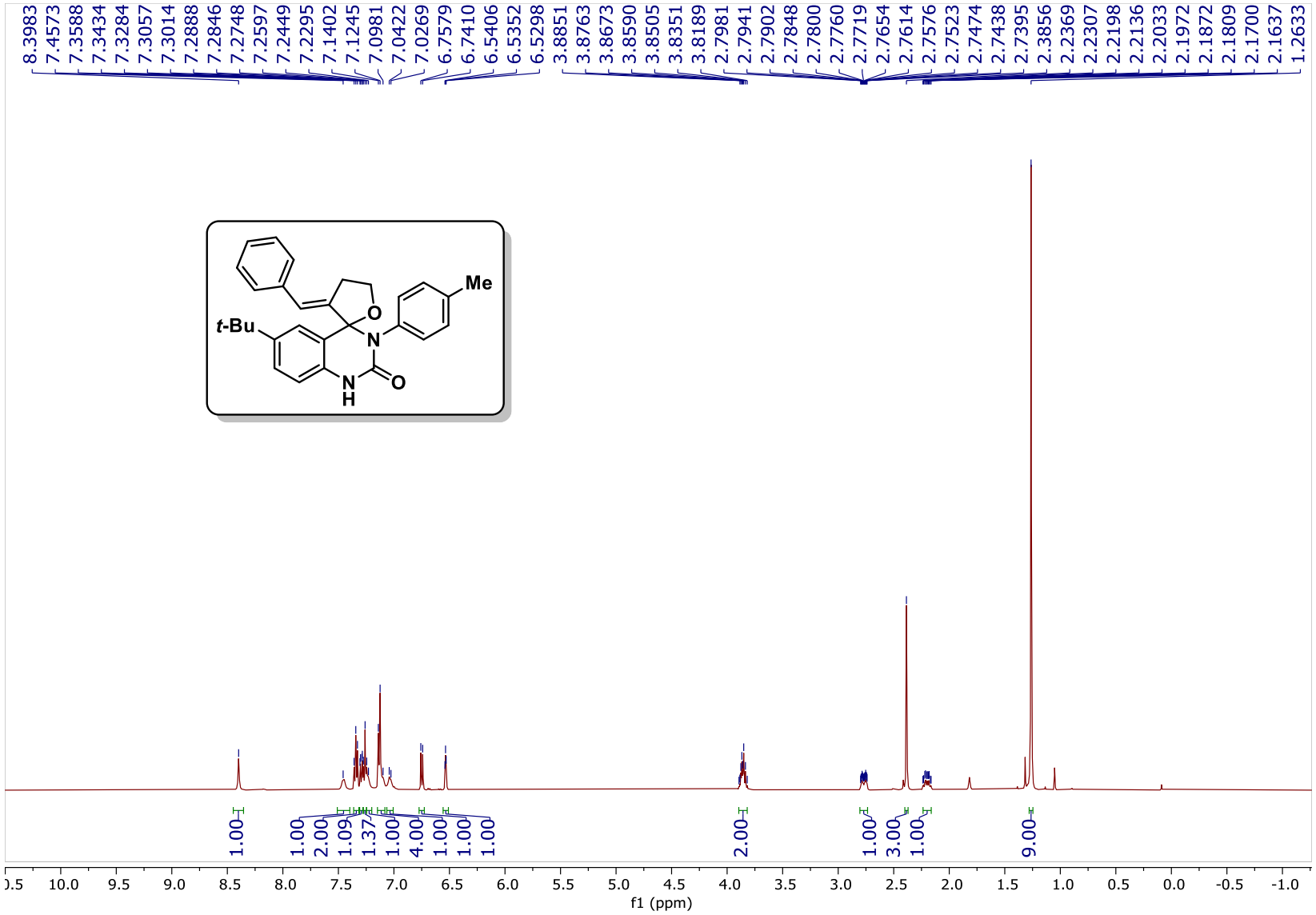
^{1}H spectrum of compound **3er** (500 MHz, $CDCl_3$)



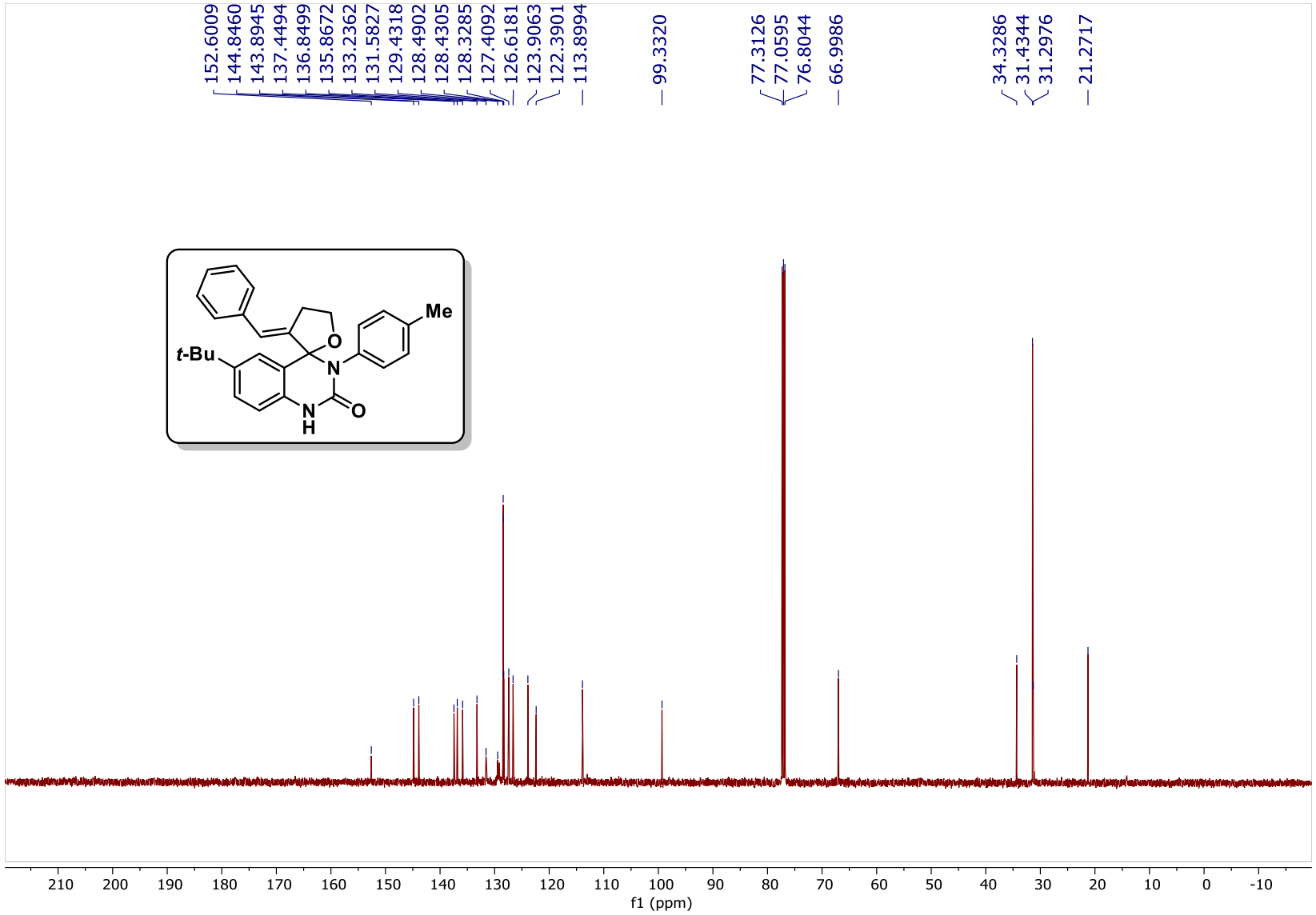
^{13}C spectrum of compound **3er** (125 MHz, $CDCl_3$)



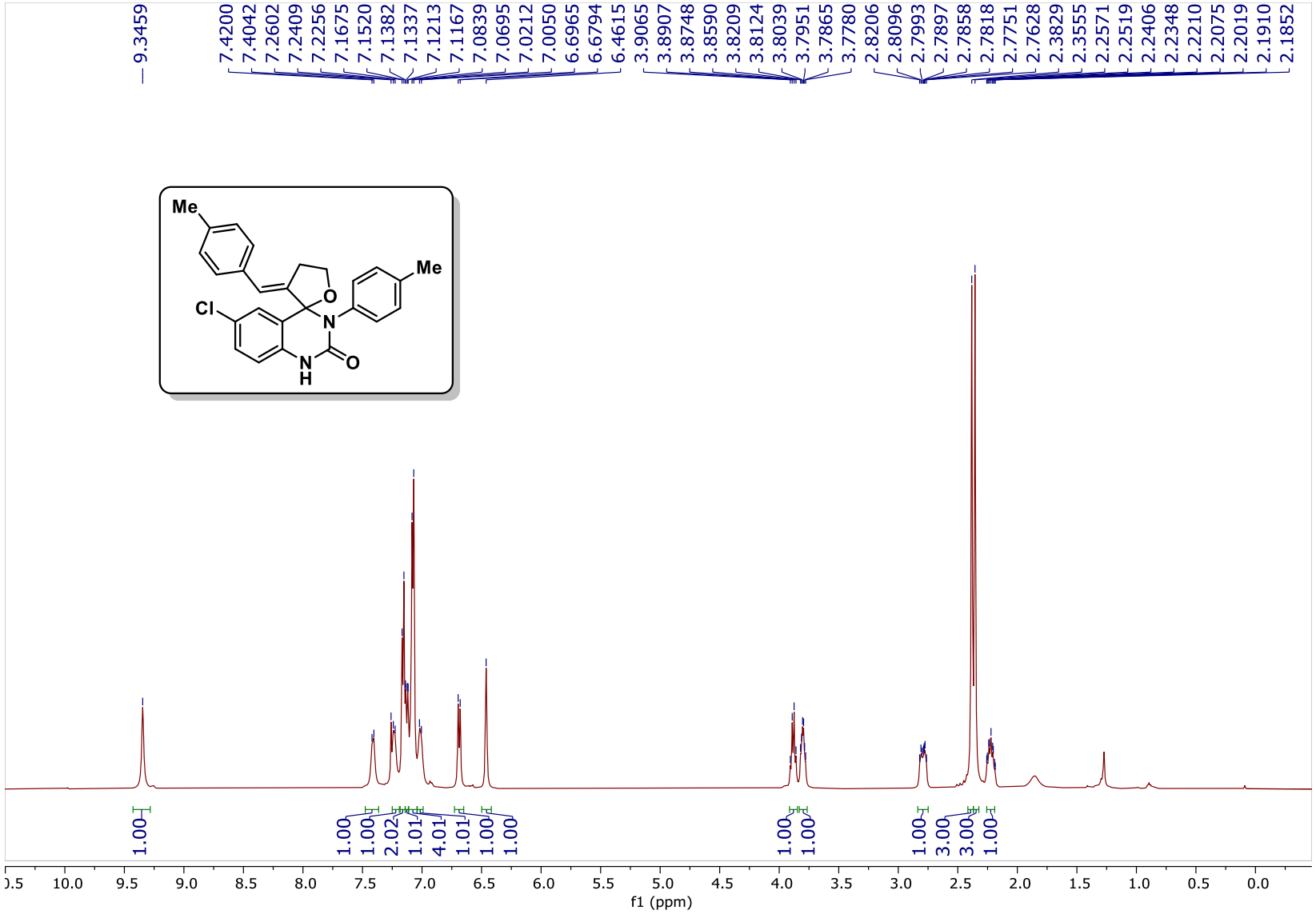
^{1}H spectrum of compound **3fa** (500 MHz, $CDCl_3$)



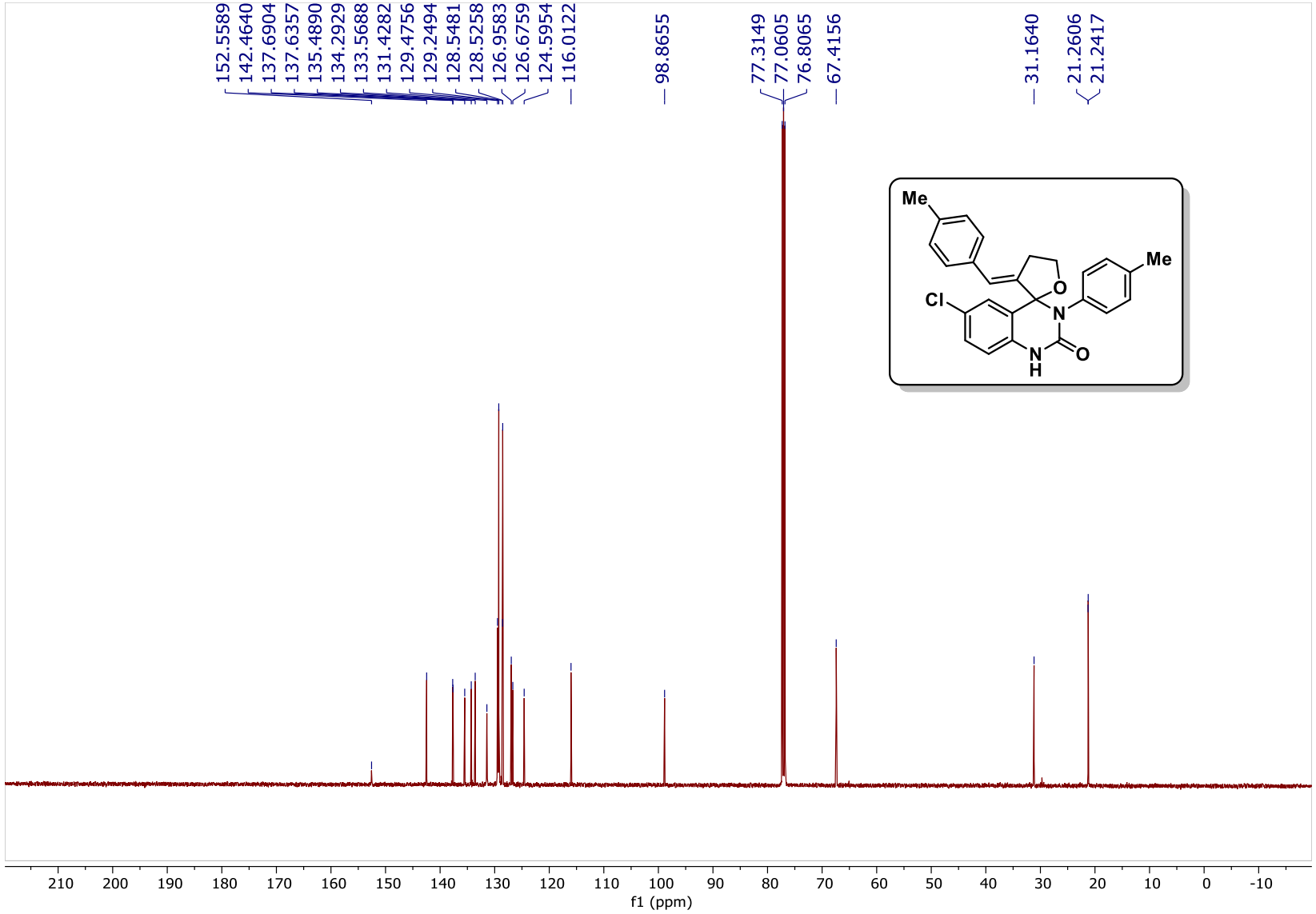
^{13}C spectrum of compound **3fa** (125 MHz, $CDCl_3$)



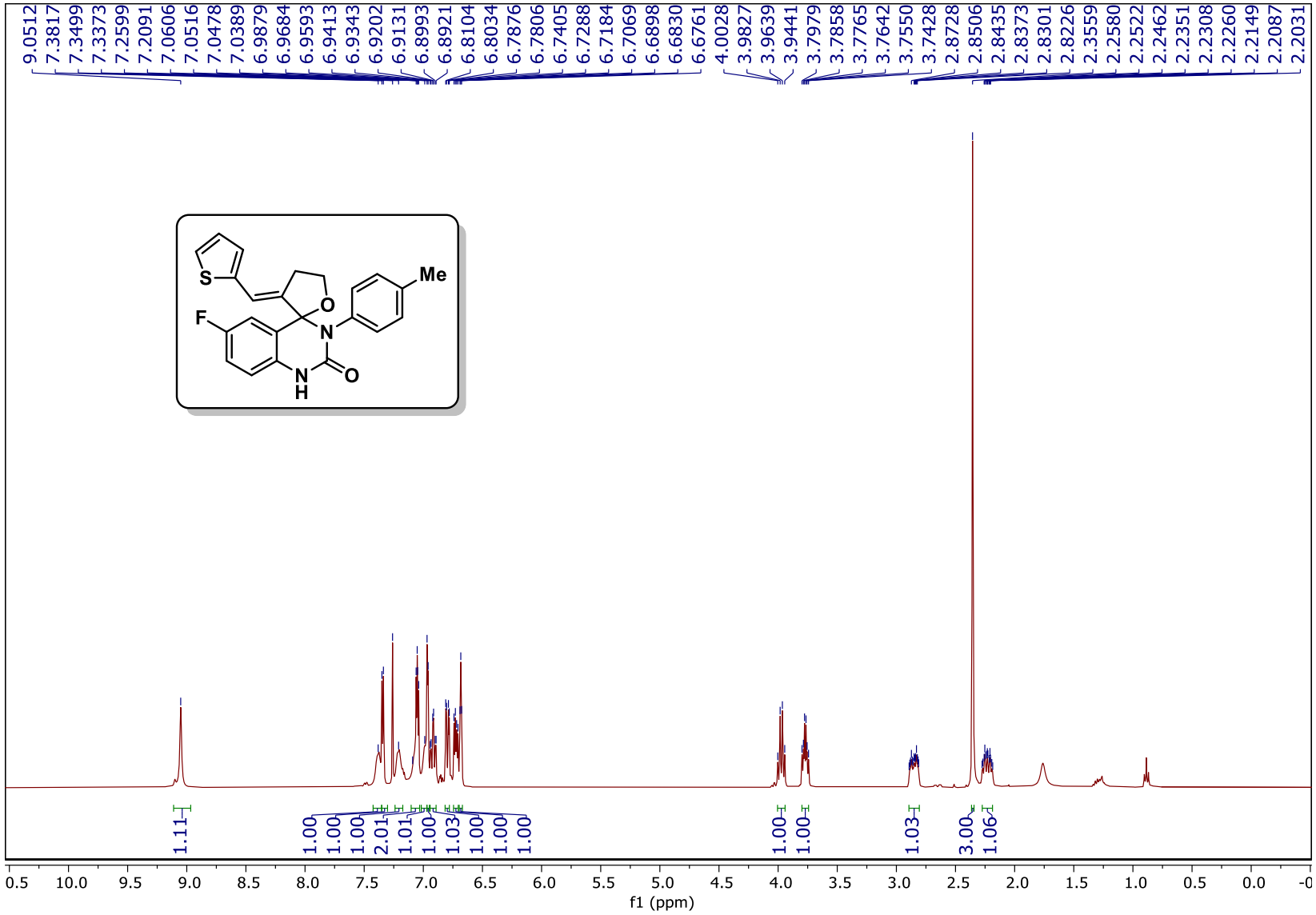
^{1}H spectrum of compound **3gi** (500 MHz, $CDCl_3$)



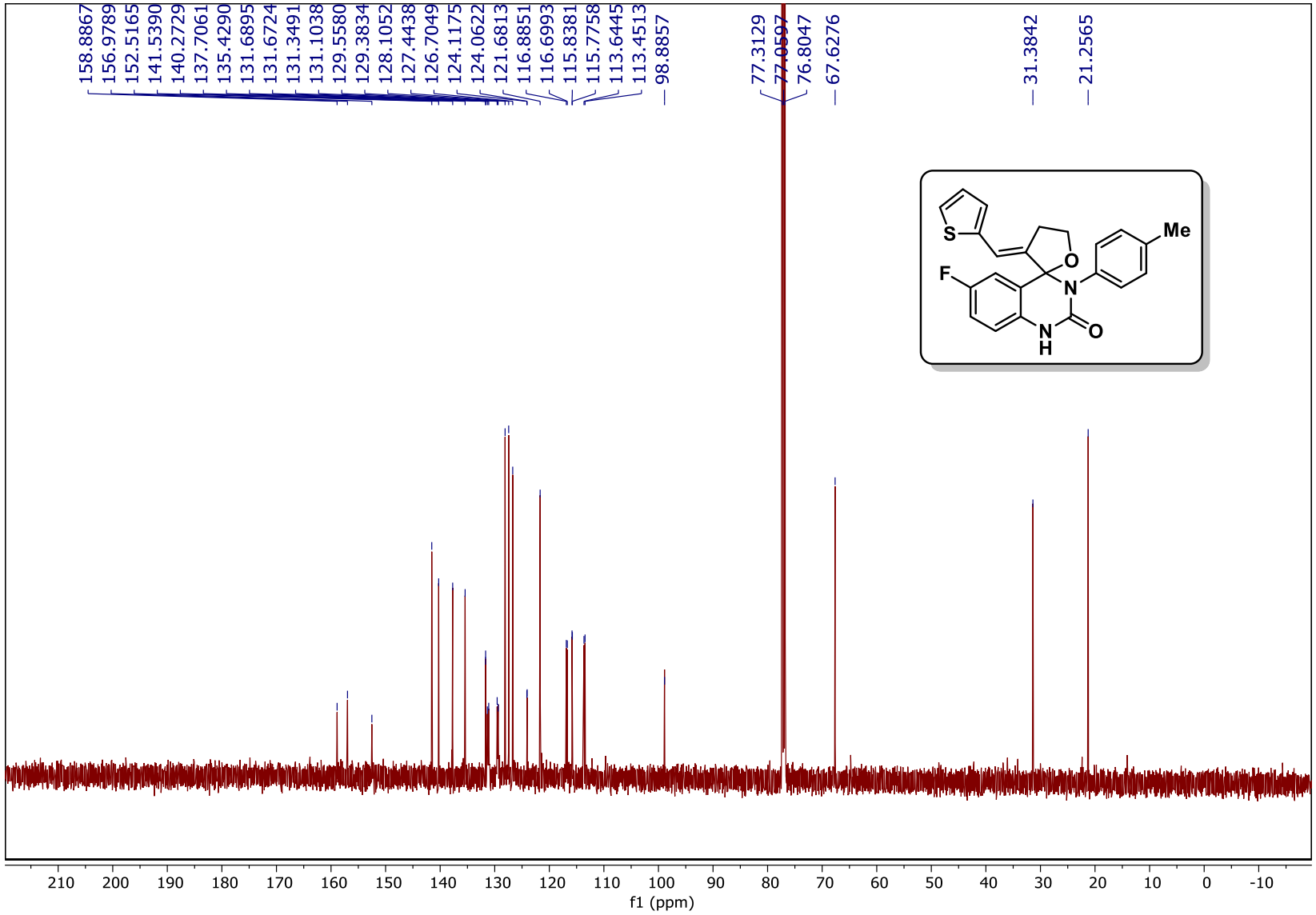
^{13}C spectrum of compound **3gi** (125 MHz, $CDCl_3$)



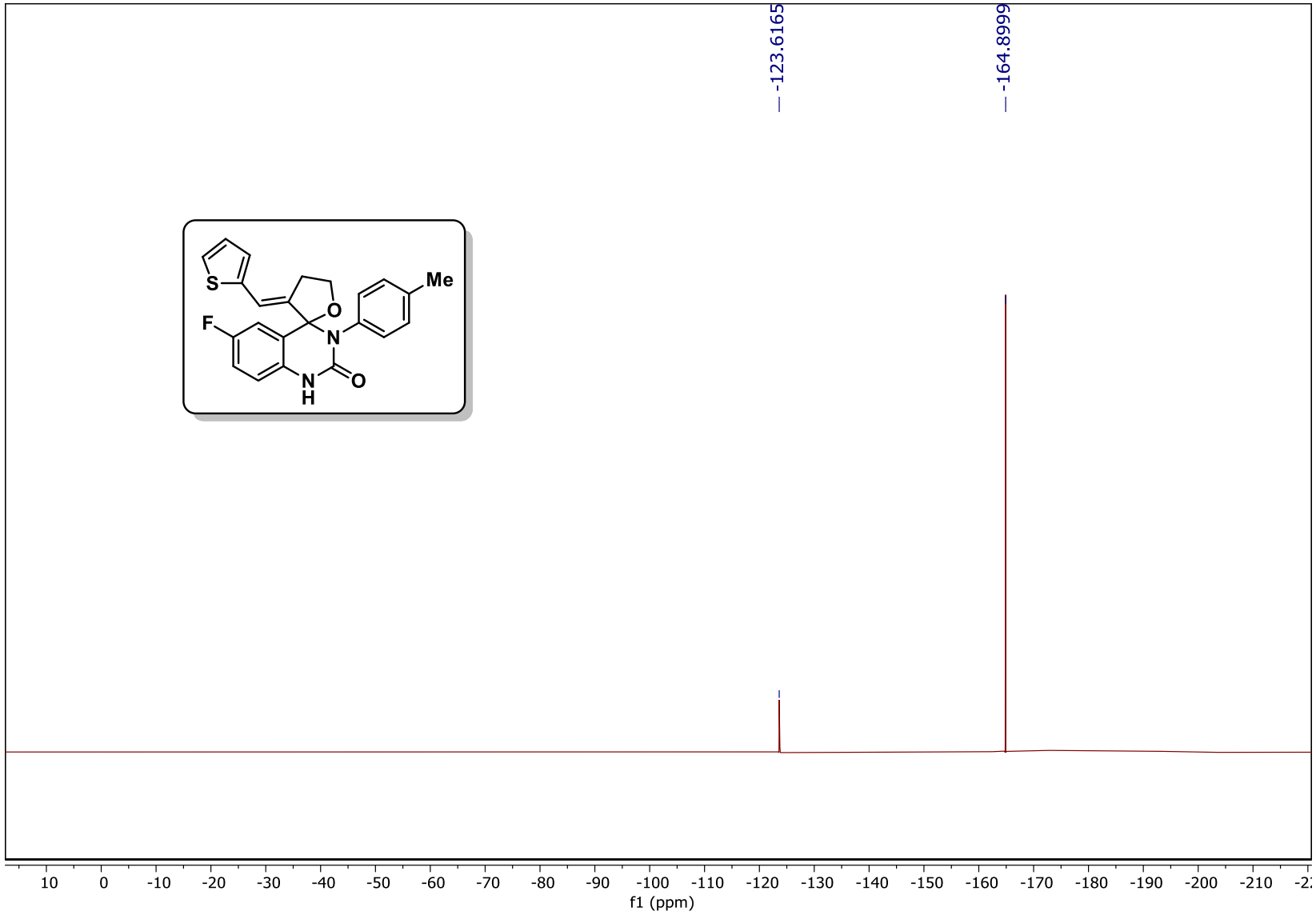
^{1}H spectrum of compound **3hq** (400 MHz, $CDCl_3$)



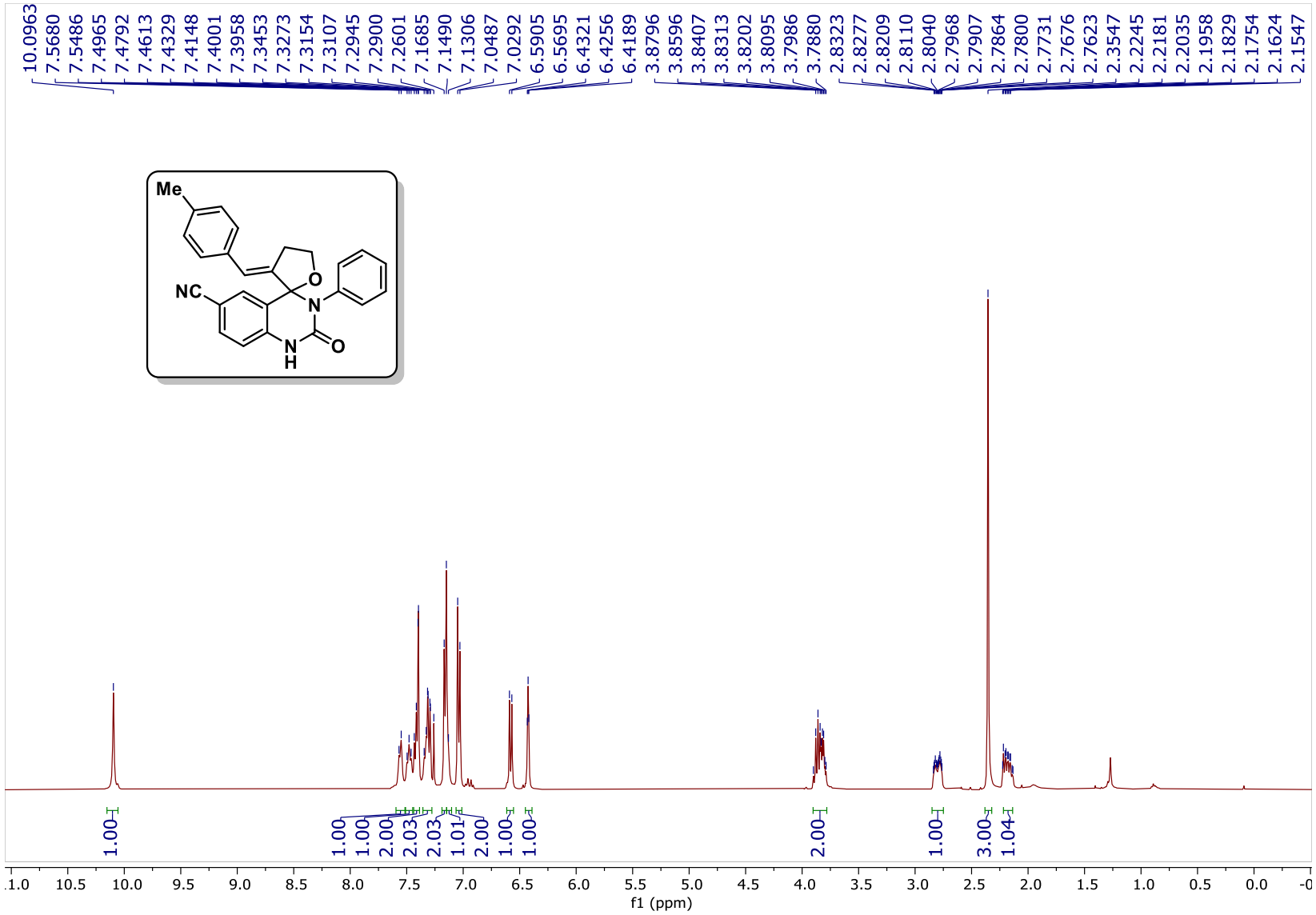
^{13}C spectrum of compound **3hq** (125 MHz, $CDCl_3$)



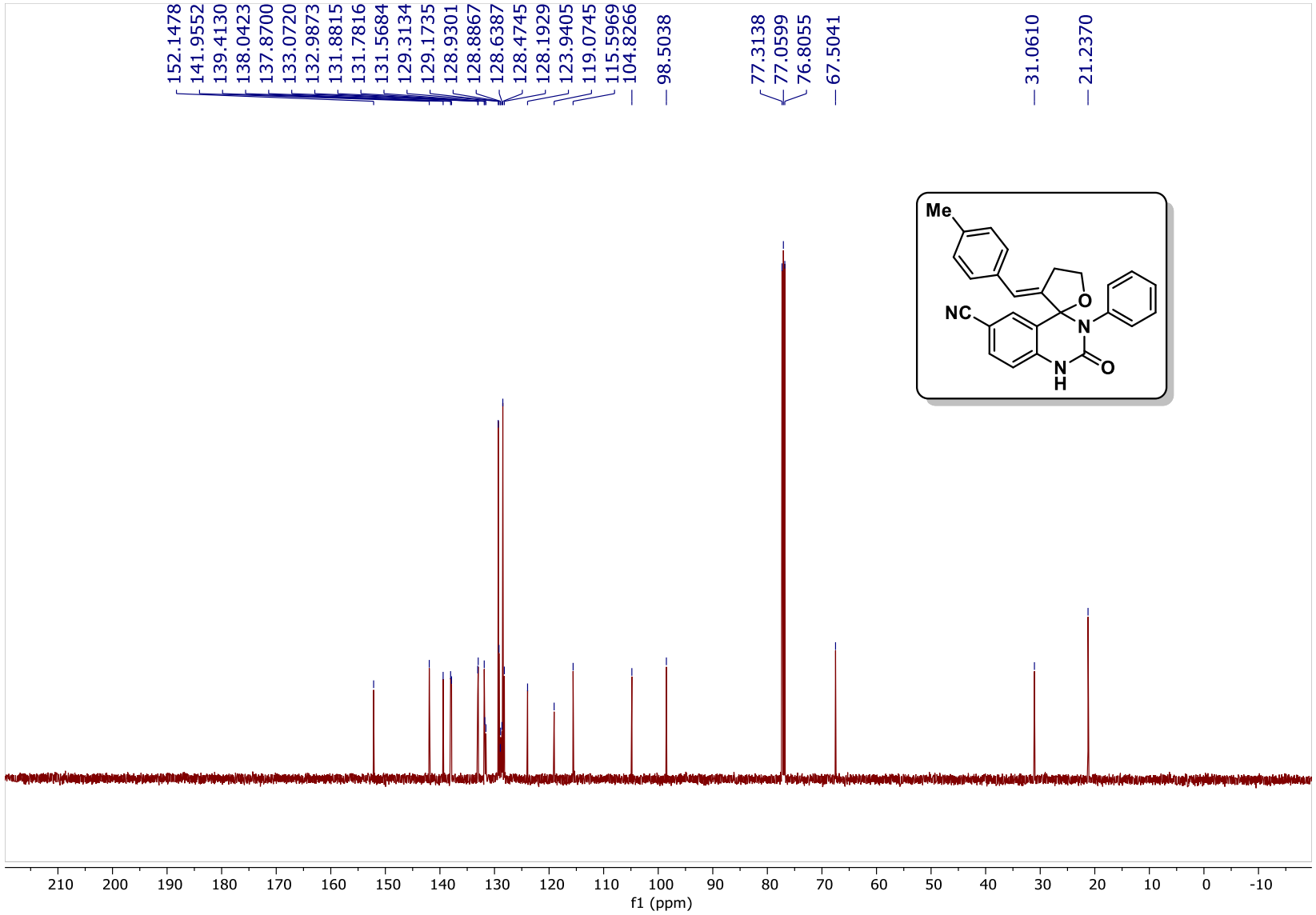
^{19}F spectrum of compound **3hq** (470 MHz, $CDCl_3/C_6F_6$)



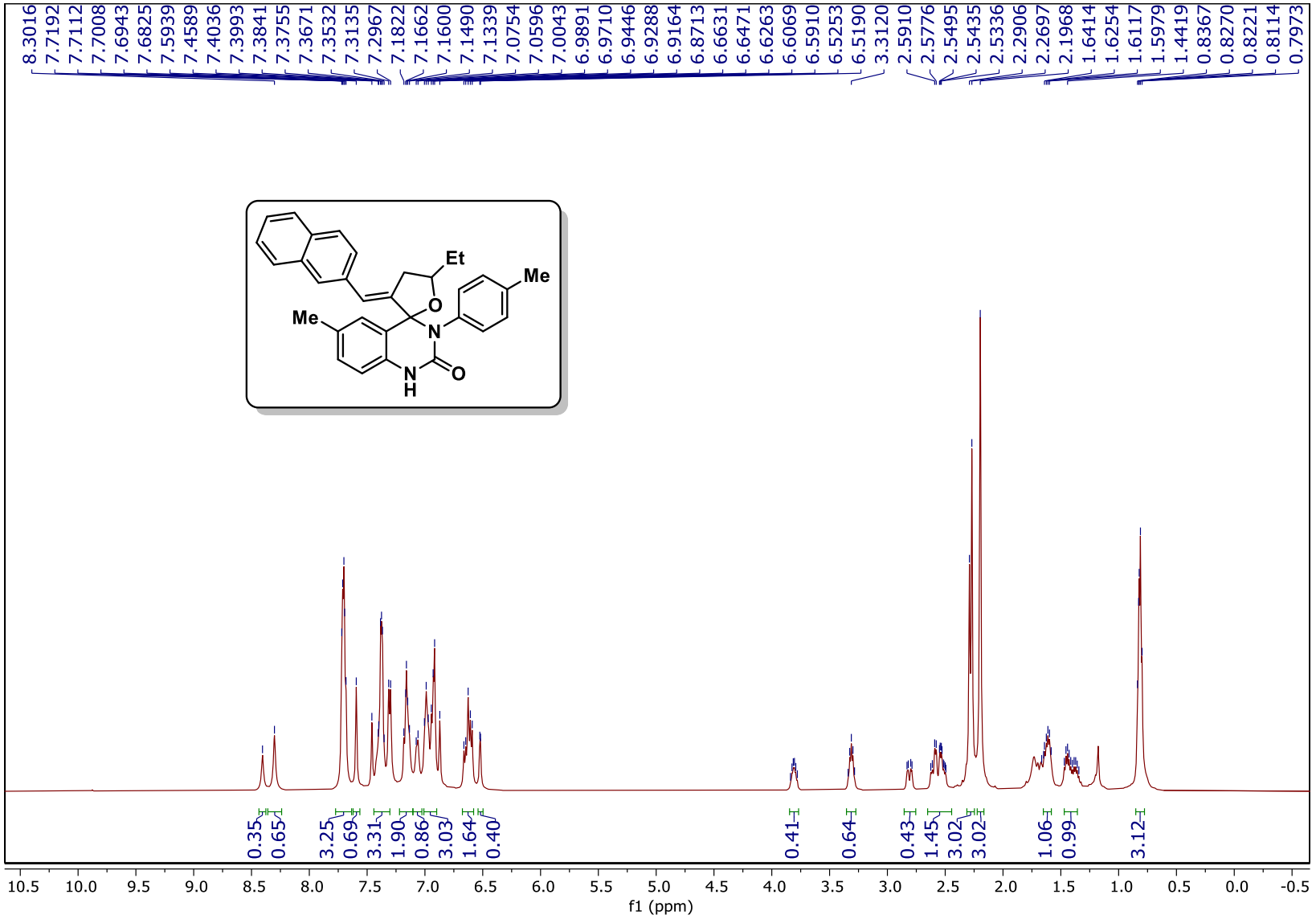
^{1}H spectrum of compound **3ii** (400 MHz, $CDCl_3$)



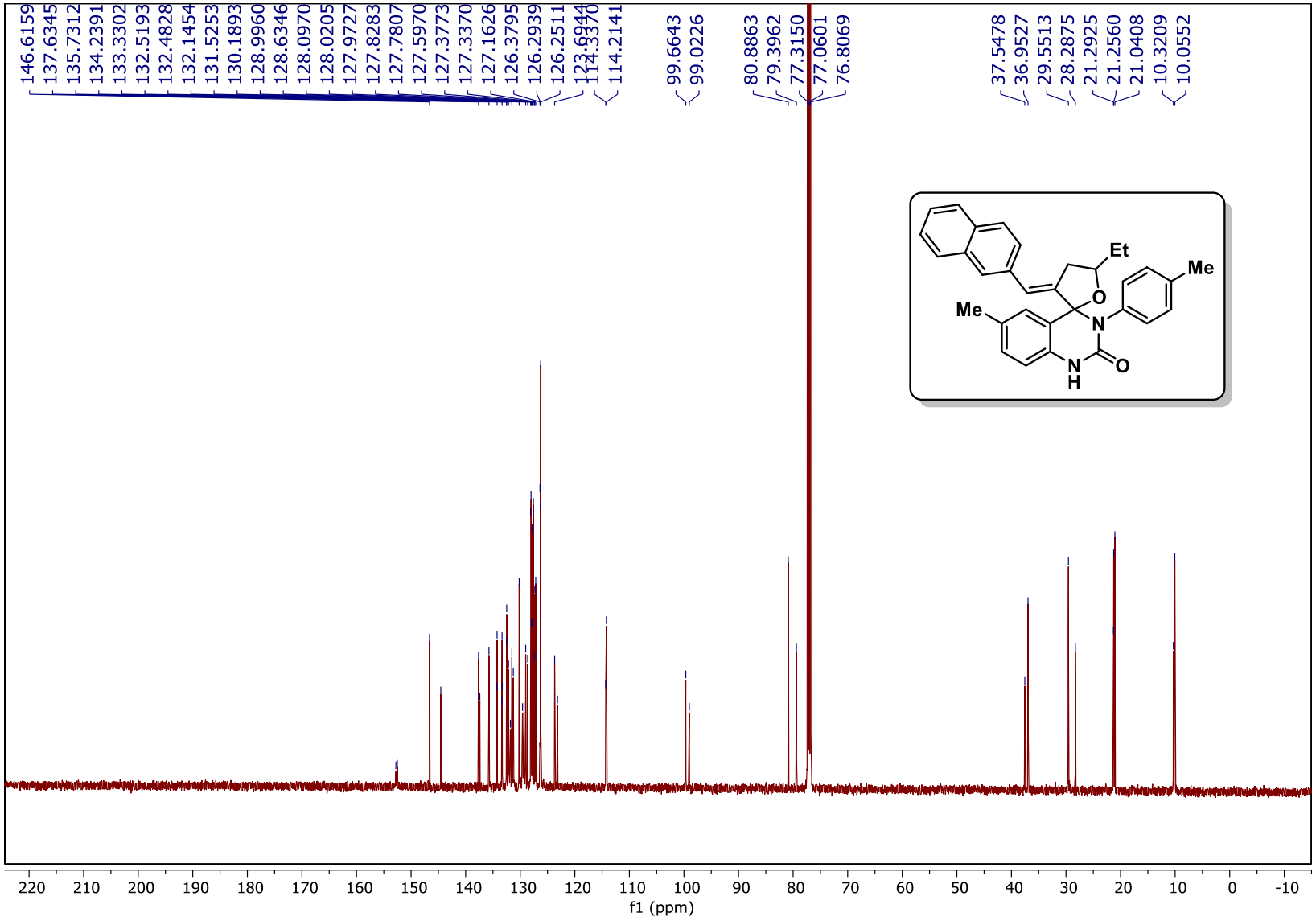
^{13}C spectrum of compound **3ii** (125 MHz, $CDCl_3$)



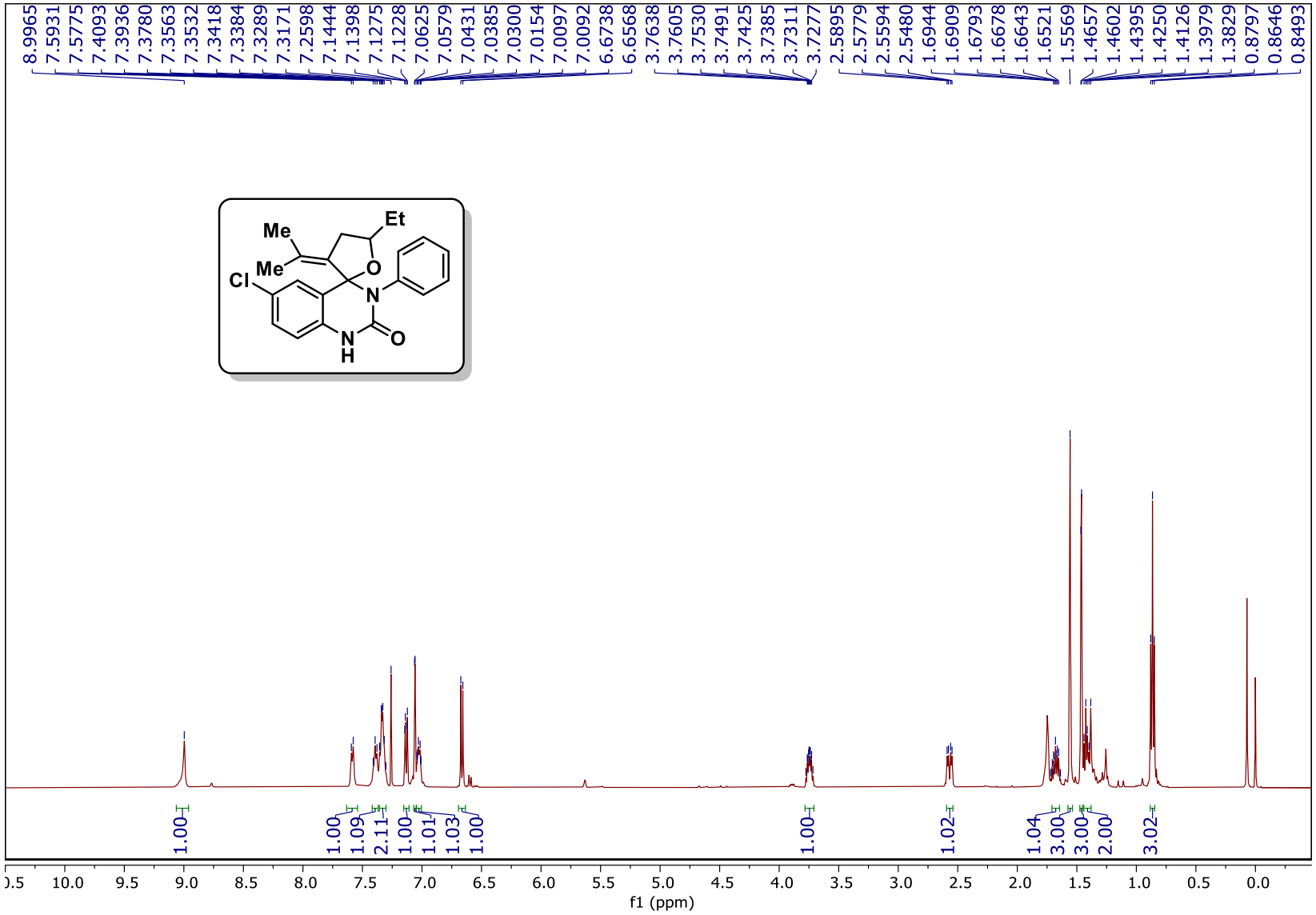
^{1}H spectrum of compound **3jm** (500 MHz, $CDCl_3$)



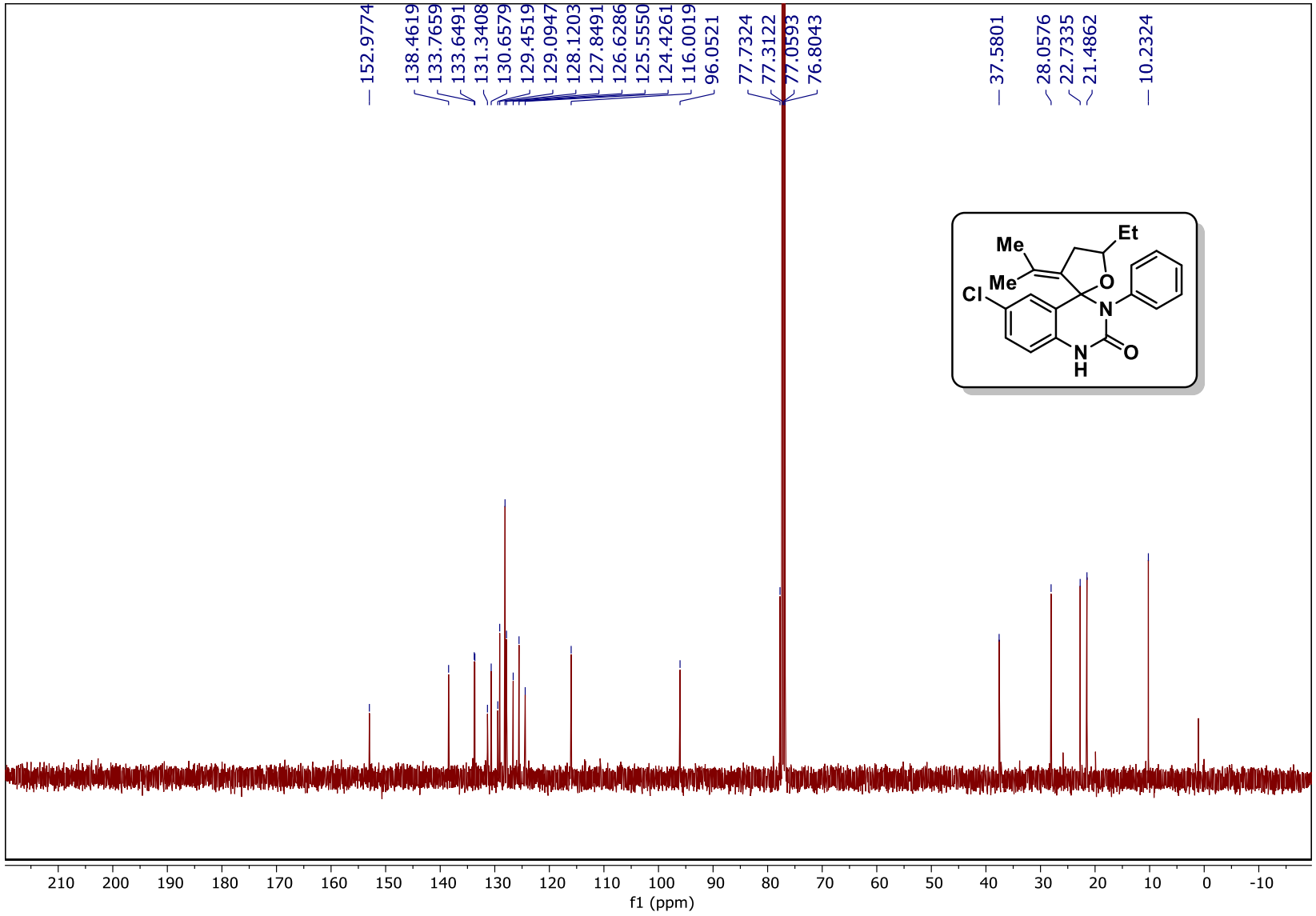
^{13}C spectrum of compound **3jm** (125 MHz, $CDCl_3$)



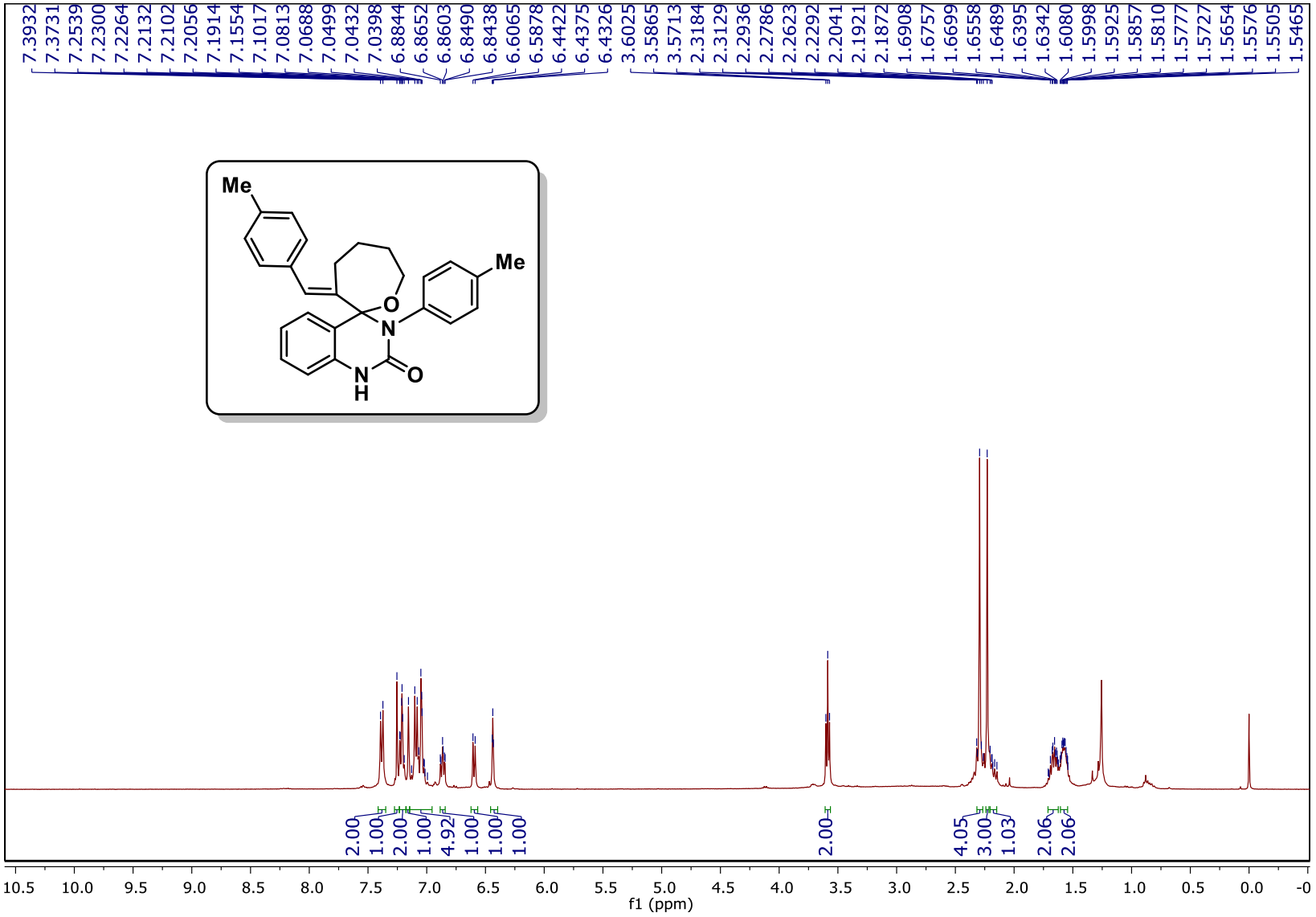
^{1}H spectrum of compound **3kv** (500 MHz, $CDCl_3$)



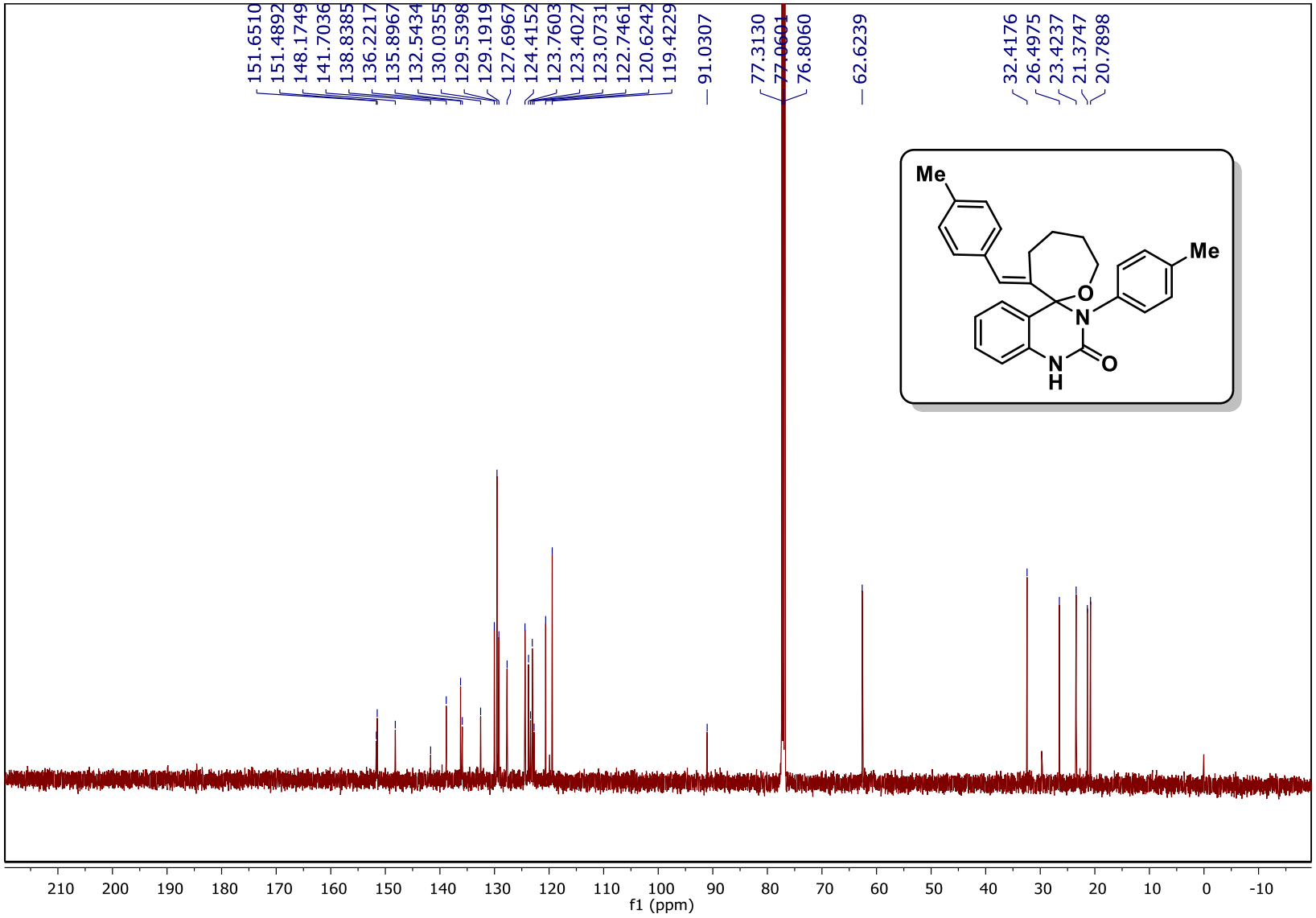
^{13}C spectrum of compound **3kv** (125 MHz, $CDCl_3$)



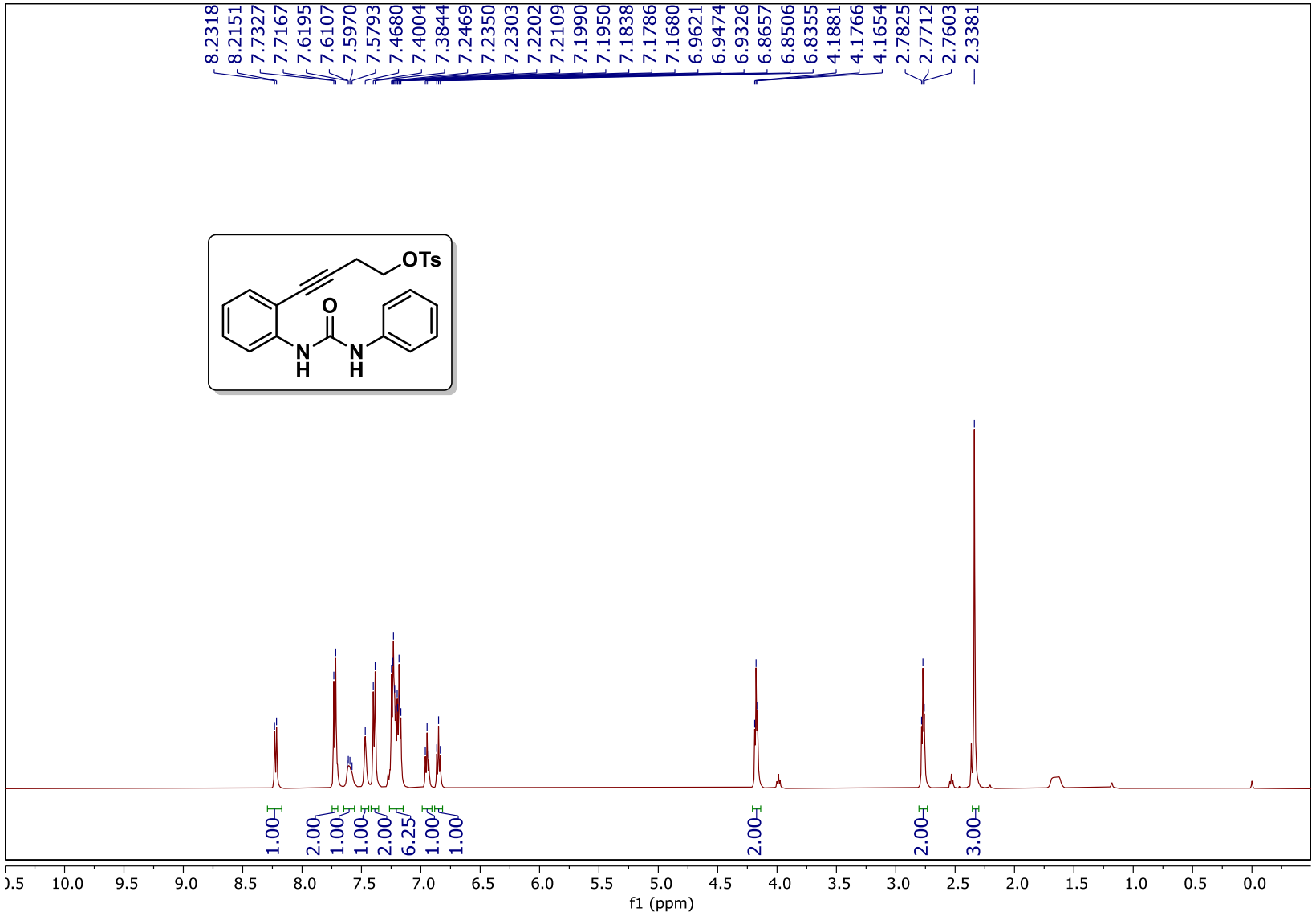
^{1}H spectrum of compound **3li** (400 MHz, $CDCl_3$)



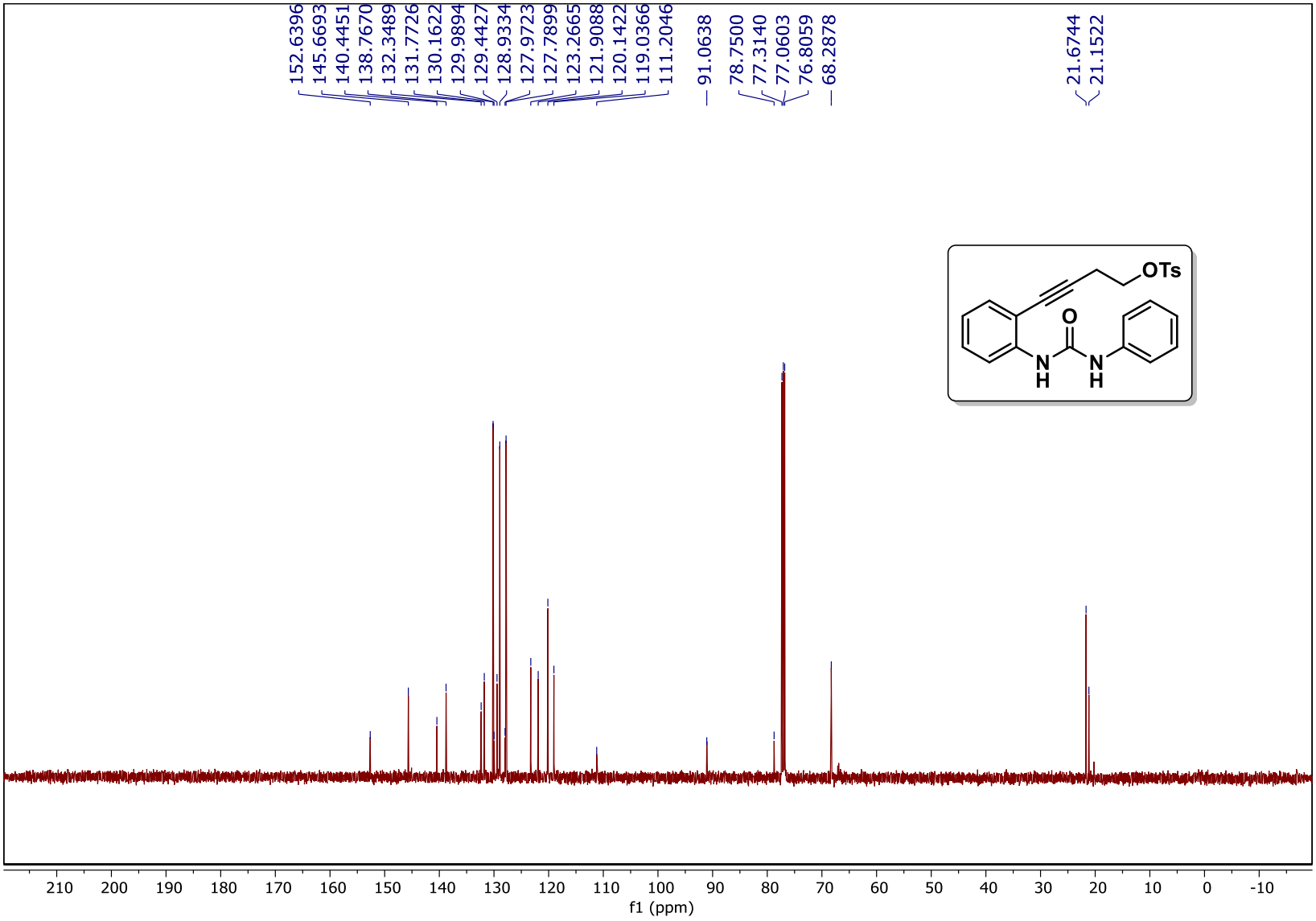
^{13}C spectrum of compound **3li** (125 MHz, $CDCl_3$)



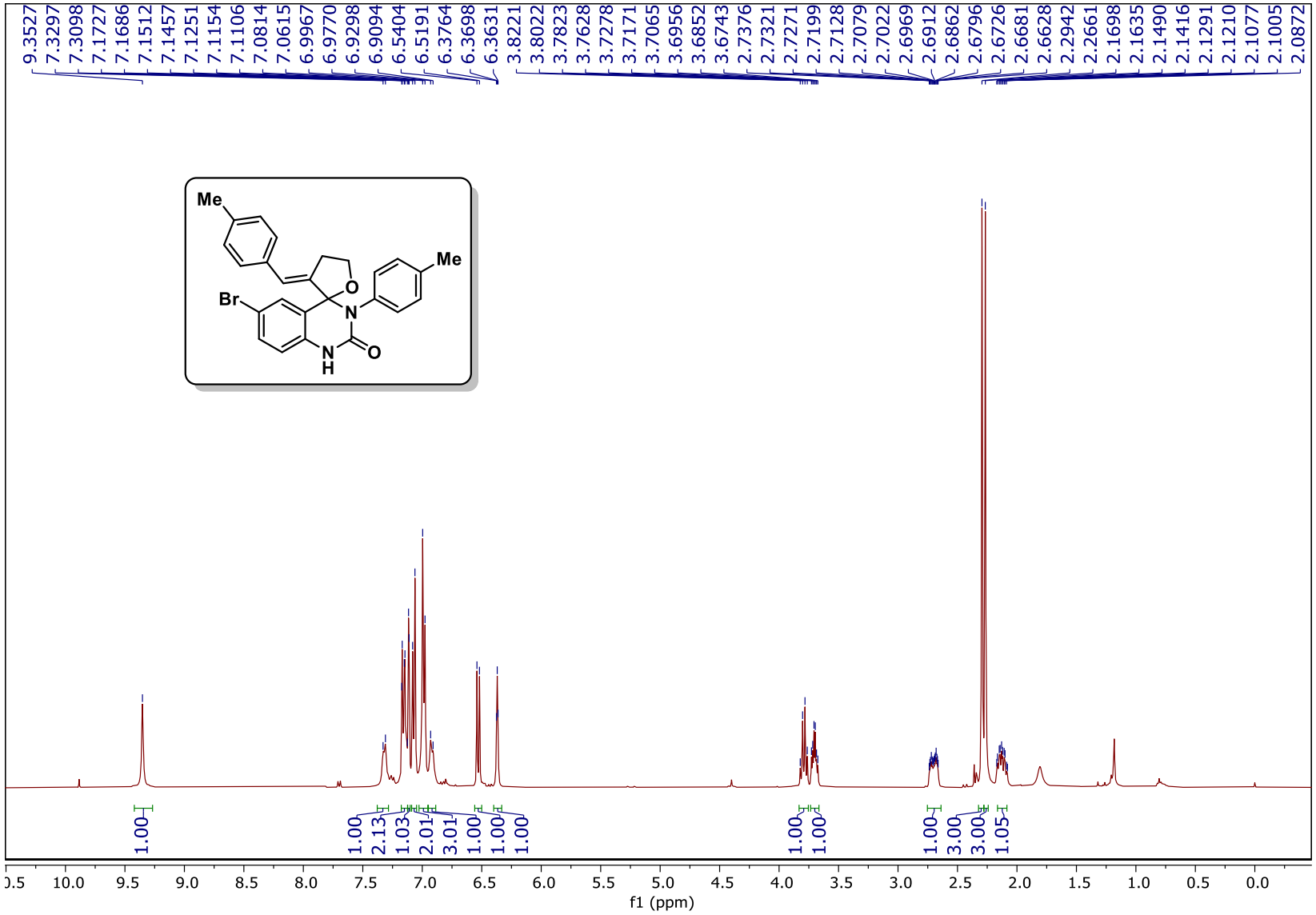
^{1}H spectrum of compound **6** (500 MHz, $CDCl_3$)



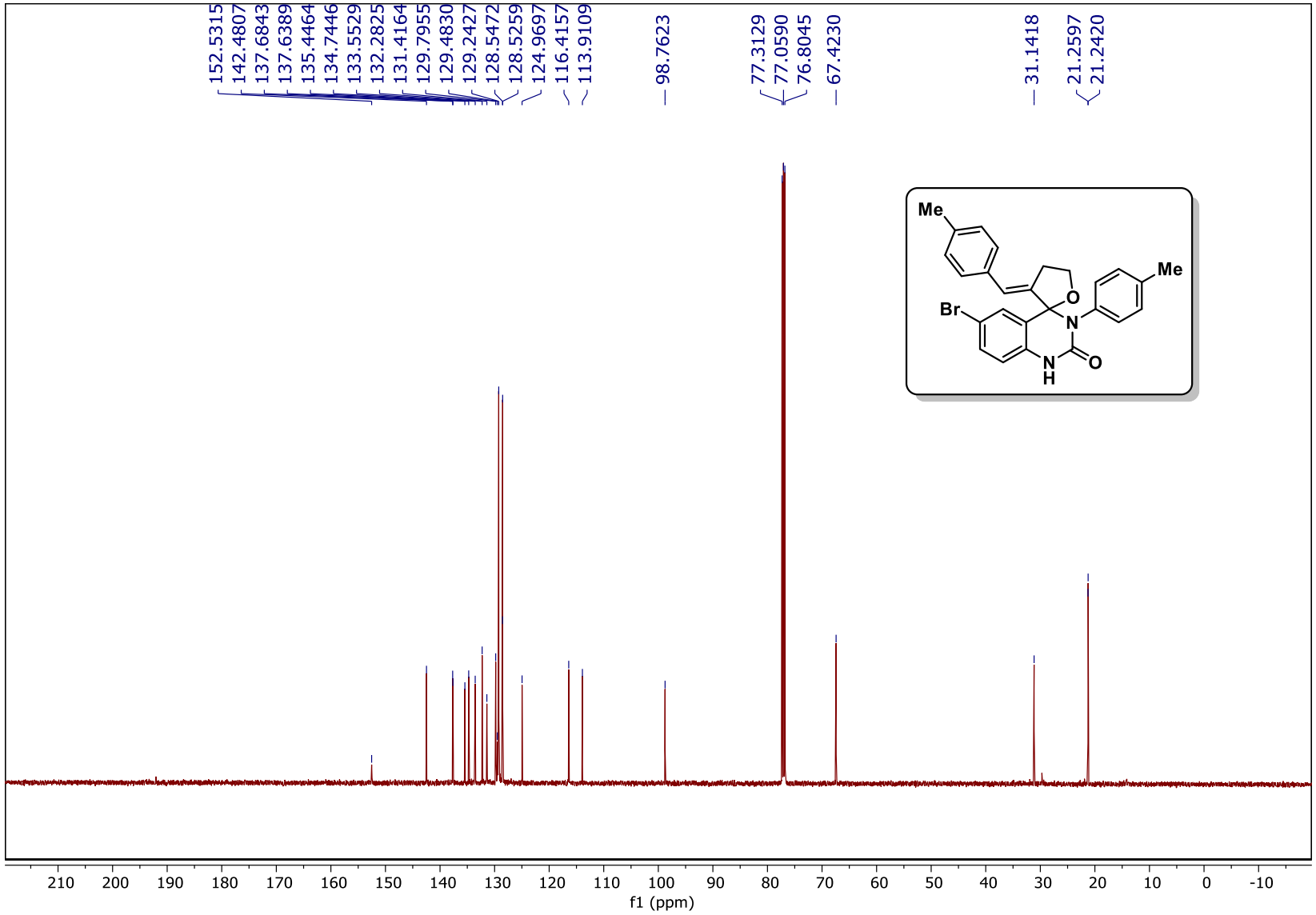
^{13}C spectrum of compound **6** (125 MHz, $CDCl_3$)



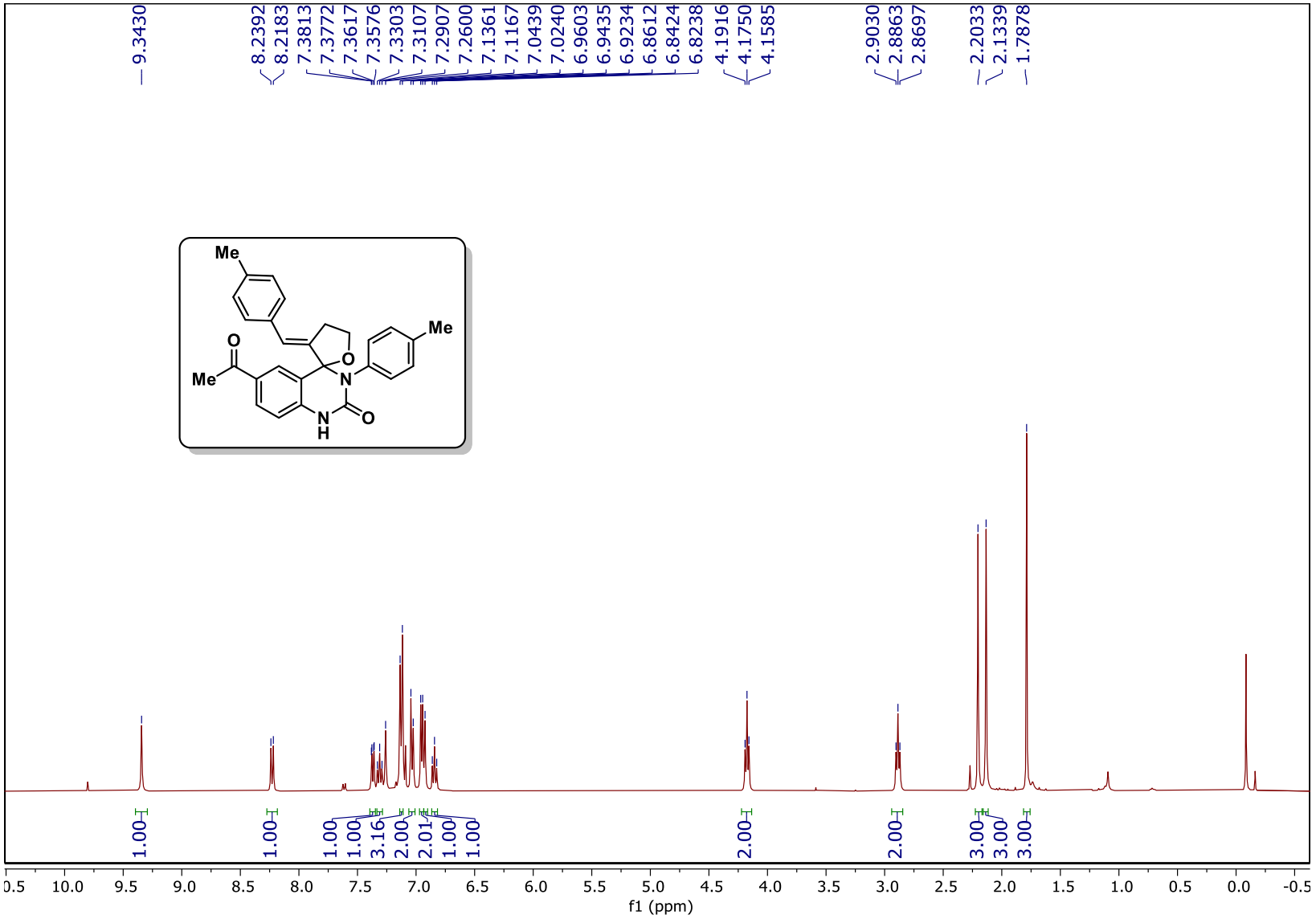
^{1}H spectrum of compound **8** (400 MHz, $CDCl_3$)



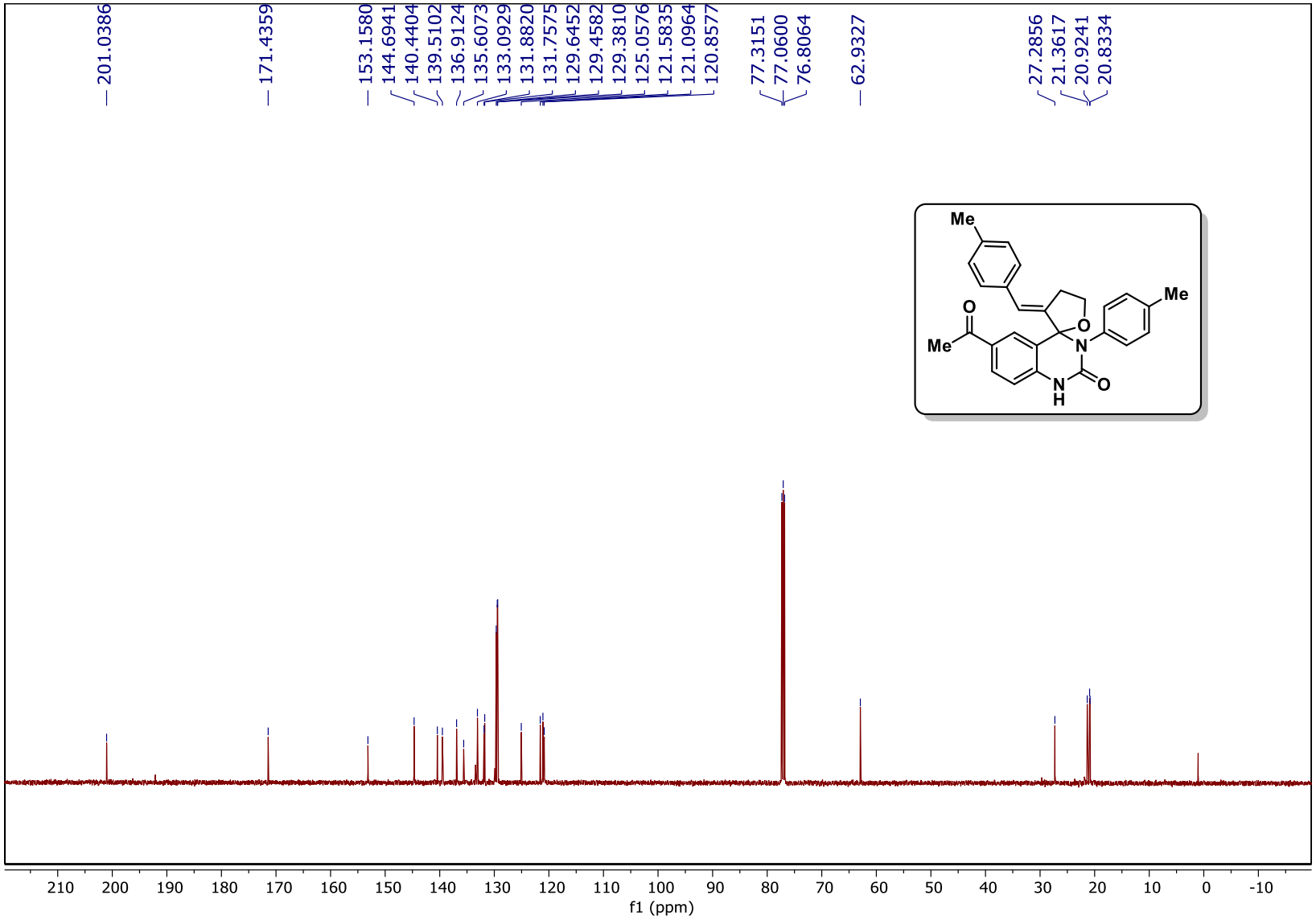
^{13}C spectrum of compound **8** (125 MHz, $CDCl_3$)



^{1}H spectrum of compound **9** (400 MHz, $CDCl_3$)



^{13}C spectrum of compound **9** (125 MHz, $CDCl_3$)



Single crystal X-ray diffraction

Single crystal of compound **3ai** was obtained by slow evaporation of ethyl acetate and methanol solution (9:1). Bruker APEX-II CCD diffractometer was used to collect the intensity data. The instrument is equipped with a fine focus 1.75 kW sealed tube Mo Kα radiation (λ = 0.71073 Å) at 298(2) K. The data acquisition was done with the APEX4 software. APEX4 software was implemented for data integration and reduction. Multi-scan empirical absorption corrections were employed to the data using the program APEX4. Structures were solved by direct methods using SHELXL-2019 and Olex2 1.5 software, and refined with full-matrix least-squares on F2 using SHELXL-2019/1.[4] Structural illustrations have been drawn with ORTEP-3 for Windows.[5] The detailed data collection and structure refinement are summarized in Table S145. CCDC-2279618 contained supplementary crystallographic data for this paper.

References:

4. G. M. Sheldrick, SHELXS-2014, Program for the crystal structure solution; University of Göttingen: Göttingen, Germany, 2014.
5. L. J. Farrugia, XRDIFF: simulation of X-ray diffraction patterns, *J. Appl. Crystallogr.*, 1997, **30**, 565.

Table S145 Crystal parameters of compound **3ai**

Identification code	SB-145
CCDC:	2279618
Empirical formula	$C_{26}H_{24}N_2O_2$
Formula weight	396.47
Temperature	298(2) K
Wavelength	0.71073 Å
Crystal system	Monoclinic
Space group	P 21/n
Unit cell dimensions	a = 12.1441(13) Å α = 90°
	b = 10.4866(11) Å β = 97.791(3)°
	c = 21.897(2) Å γ = 90°
Volume	2762.9(5) $Å^3$
Z	4
Density (calculated)	0.953 mg/m^3
Absorption coefficient	0.061 mm^{-1}
F(000)	840
Crystal size	0.32 × 0.30 × 0.27 mm^3
Theta range for data collection	4.088 to 50°
Index ranges	$-14 \le h \le 14$, $-12 \le k \le 12$, $-26 \le l \le 26$
Reflections collected	62523
Independent reflections	4835 [R_{int} = 0.0358, R_{sigma} = 0.0164]
Completeness to theta = 26.38	99.4%
Refinement method	Full-matrix least-squares on F2
Data / restraints / parameters	4835/0/ 273
Goodness-of-fit on F2	1.061
Final R indices [I>2sigma(I)]	**R_1** = 0.0528
	wR_2 = 0.1520
R indices (all data)	**R_1** = 0.0675
	wR_2 = 0.1771
Extinction coefficient	n/a
Largest diff. peak and hole	0.13 and -0.17 e.Å-3

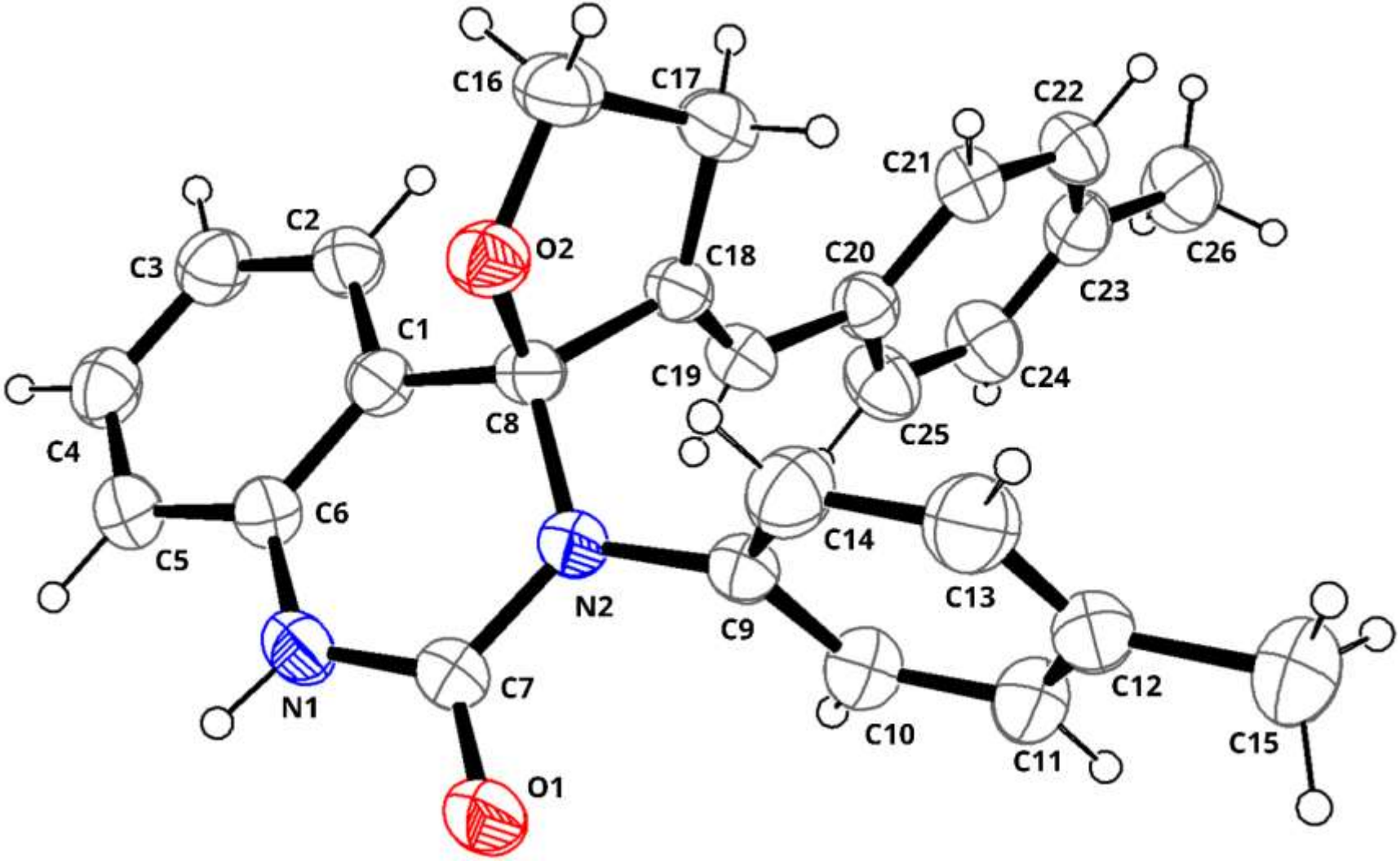


Figure S146 ORTEP diagram of compound **3ai** with thermal ellipsoid of 30% probability.