

Supplementary Materials for**A stereoselective, multicomponent catalytic carbonylative approach to a new class of
 α,β -unsaturated γ -lactam derivatives**

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X-ray Data for Compounds 2a and 2b

X-ray crystallographic data for methyl (Z)-2-(2-oxo-1-phenylpyrrolidin-3-ylidene)acetate 2a¹

| <i>Crystal data</i> | |
|--|---|
| Chemical formula: C ₁₃ H ₁₃ NO ₃ | <i>Mr</i> = 231.24 |
| Triclinic, <i>P</i> -1 | <i>Dx</i> = 1.344 Mg m ⁻³ |
| Hall symbol: -P 1 | Melting point: 375.65 K |
| <i>a</i> = 8.1290 (7) Å | Cell parameters from 124 reflections (θ = 5.5–45.2°) |
| <i>b</i> = 8.2620 (18) Å | μ = 0.10 mm ⁻¹ |
| <i>c</i> = 9.760 (2) Å | <i>T</i> = 293 K |
| α = 98.790 (17)° | Crystal: block, colorless |
| β = 102.096 (13)° | Crystal size: 0.50 × 0.40 × 0.25 mm |
| γ = 112.628 (9)° | <i>F</i> (000) = 244 |
| <i>V</i> = 571.48 (19) Å ³ | <i>Z</i> = 2 |
| <i>Data collection</i> | |
| Bruker-Nonius KappaCCD diffractometer | 2646 independent reflections |
| Radiation source: Mo $K\alpha$ radiation, λ = 0.71073 Å | 1696 reflections with $I > 2\sigma(I)$ |
| φ and ω scans | 13216 measured reflections |
| | R_{int} = 0.061 |
| Absorption correction: multi-scan, SADABS, ² T_{\min} = 0.682, T_{\max} = 0.746 | θ_{\max} = 28.2°, θ_{\min} = 3.8° |
| | H = -10 → 9 |
| | k = -10 → 10 |
| | l = -12 → 12 |
| <i>Refinement</i> | |
| Refinement on F^2 | Hydrogen site location: mixed |
| Least-squares matrix: full | H atoms treated by a mixture of independent and constrained refinement |
| $R[F^2 > 2\sigma(F^2)]$ = 0.059 | $w = 1/[\sigma^2(F_o^2) + (0.1037P)^2 + 0.0154P]$ where $P = (F_o^2 + 2F_c^2)/3$ |
| $wR(F^2)$ = 0.176 | $(\Delta/\sigma)_{\text{max}} < 0.001$ |
| S = 1.07 | $\Delta\rho_{\text{max}} = 0.31 \text{ e \AA}^{-3}$ |
| 2646 reflections | $\Delta\rho_{\text{min}} = -0.34 \text{ e \AA}^{-3}$ |

159 parameters

0 restraints

Programs: SIR2014,³ SHELXL-2014⁴Extinction correction: *SHELXL-2014* (3)

$$Fc^* = kFc[1 + 0.001x Fc^2 \lambda^3 / \sin(2\theta)]^{-1/4}$$

Extinction coefficient: 0.47 (5)

Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (\AA^2)

| | x | y | z | <i>U</i>_{iso}*/<i>U</i>_{eq} |
|------|--------------|-------------|--------------|--|
| O1 | 0.27704 (18) | 0.4468 (2) | 0.66957 (16) | 0.0595 (5) |
| O2 | 0.2922 (2) | 0.5944 (2) | 0.99777 (16) | 0.0646 (5) |
| N2 | 0.0076 (2) | 0.2508 (2) | 0.49816 (16) | 0.0441 (4) |
| O3 | 0.3232 (2) | 0.8039 (2) | 0.87212 (17) | 0.0671 (5) |
| C2 | 0.0786 (3) | 0.1789 (2) | 0.3938 (2) | 0.0434 (5) |
| C3 | -0.0211 (3) | 0.4197 (3) | 0.6966 (2) | 0.0476 (5) |
| C4 | 0.1090 (3) | 0.3782 (3) | 0.6236 (2) | 0.0453 (5) |
| C5 | 0.2315 (3) | 0.6618 (3) | 0.8945 (2) | 0.0501 (5) |
| C6 | -0.1926 (3) | 0.1825 (3) | 0.4833 (2) | 0.0509 (5) |
| H6A | -0.2431 | 0.0572 | 0.4885 | 0.061* |
| H6B | -0.2590 | 0.1887 | 0.3908 | 0.061* |
| C7 | 0.2638 (3) | 0.2600 (3) | 0.3932 (2) | 0.0527 (5) |
| H7 | 0.3481 | 0.3630 | 0.4664 | 0.063* |
| C8 | -0.2131 (3) | 0.3021 (3) | 0.6076 (2) | 0.0564 (6) |
| H8A | -0.2753 | 0.3732 | 0.5716 | 0.068* |
| H8B | -0.2835 | 0.2295 | 0.6627 | 0.068* |
| C9 | -0.0394 (3) | 0.0257 (3) | 0.2862 (2) | 0.0535 (5) |
| H9 | -0.1637 | -0.0331 | 0.2834 | 0.064* |
| C10 | 0.0348 (3) | 0.5451 (3) | 0.8177 (2) | 0.0525 (5) |
| H10A | -0.052 (3) | 0.569 (3) | 0.866 (3) | 0.063* |
| C11 | 0.2079 (4) | 0.0394 (3) | 0.1794 (3) | 0.0622 (6) |
| H11 | 0.2484 | -0.0079 | 0.1076 | 0.075* |
| C12 | 0.3261 (3) | 0.1907 (3) | 0.2856 (3) | 0.0600 (6) |
| H12 | 0.4500 | 0.2493 | 0.2870 | 0.072* |
| C13 | 0.0251 (3) | -0.0430 (3) | 0.1806 (2) | 0.0636 (6) |

| | | | | |
|------|------------|------------|------------|------------|
| H13 | -0.0577 | -0.1479 | 0.1085 | 0.076* |
| C14 | 0.4801 (3) | 0.7026 (4) | 1.0813 (3) | 0.0802 (8) |
| H14A | 0.5131 | 0.6485 | 1.1565 | 0.120* |
| H14B | 0.4952 | 0.8220 | 1.1240 | 0.120* |
| H14C | 0.5597 | 0.7109 | 1.0199 | 0.120* |

Atomic displacement parameters (\AA^2)

| | <i>U</i>11 | <i>U</i>22 | <i>U</i>33 | <i>U</i>12 | <i>U</i>13 | <i>U</i>23 |
|-----|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|
| O1 | 0.0392 (8) | 0.0697 (9) | 0.0522 (9) | 0.0172 (7) | 0.0036 (6) | -0.0019 (7) |
| O2 | 0.0589 (9) | 0.0803 (11) | 0.0519 (9) | 0.0287 (8) | 0.0090 (7) | 0.0232 (8) |
| N2 | 0.0378 (9) | 0.0502 (9) | 0.0413 (9) | 0.0174 (7) | 0.0077 (6) | 0.0131 (7) |
| O3 | 0.0710 (11) | 0.0574 (9) | 0.0600 (10) | 0.0196 (8) | 0.0094 (8) | 0.0146 (8) |
| C2 | 0.0448 (10) | 0.0430 (9) | 0.0408 (10) | 0.0201 (8) | 0.0054 (8) | 0.0134 (8) |
| C3 | 0.0420 (10) | 0.0557 (11) | 0.0471 (11) | 0.0216 (9) | 0.0119 (8) | 0.0194 (9) |
| C4 | 0.0418 (11) | 0.0499 (10) | 0.0417 (10) | 0.0184 (8) | 0.0088 (8) | 0.0137 (8) |
| C5 | 0.0574 (12) | 0.0590 (12) | 0.0381 (10) | 0.0299 (10) | 0.0158 (9) | 0.0090 (9) |
| C6 | 0.0365 (10) | 0.0574 (11) | 0.0523 (11) | 0.0161 (9) | 0.0062 (8) | 0.0173 (9) |
| C7 | 0.0474 (11) | 0.0490 (11) | 0.0547 (12) | 0.0166 (9) | 0.0137 (9) | 0.0077 (9) |
| C8 | 0.0436 (11) | 0.0679 (13) | 0.0566 (12) | 0.0227 (10) | 0.0140 (9) | 0.0177 (10) |
| C9 | 0.0483 (11) | 0.0523 (11) | 0.0493 (11) | 0.0178 (9) | 0.0038 (9) | 0.0093 (9) |
| C10 | 0.0535 (12) | 0.0665 (13) | 0.0446 (11) | 0.0302 (10) | 0.0181 (9) | 0.0173 (10) |
| C11 | 0.0729 (15) | 0.0656 (13) | 0.0529 (13) | 0.0364 (12) | 0.0194 (11) | 0.0097 (11) |
| C12 | 0.0561 (13) | 0.0633 (13) | 0.0632 (14) | 0.0265 (11) | 0.0234 (11) | 0.0136 (11) |
| C13 | 0.0734 (16) | 0.0570 (12) | 0.0488 (12) | 0.0291 (11) | 0.0024 (11) | 0.0005 (10) |
| C14 | 0.0601 (15) | 0.1006 (19) | 0.0646 (15) | 0.0317 (14) | -0.0024 (12) | 0.0160 (14) |

Geometric parameters (\AA , $^\circ$)

| | | | |
|--------|-----------|--------|-----------|
| O1—C4 | 1.207 (2) | C3—C10 | 1.310 (3) |
| O2—C5 | 1.321 (2) | C3—C8 | 1.463 (3) |
| O2—C14 | 1.415 (3) | C3—C4 | 1.504 (3) |
| N2—C4 | 1.343 (2) | C5—C10 | 1.463 (3) |
| N2—C2 | 1.432 (3) | C6—C8 | 1.524 (3) |

| | | | |
|-----------|-------------|-------------|-------------|
| N2—C6 | 1.468 (2) | C7—C12 | 1.396 (3) |
| O3—C5 | 1.207 (3) | C9—C13 | 1.391 (3) |
| C2—C9 | 1.364 (3) | C11—C12 | 1.350 (3) |
| C2—C7 | 1.394 (3) | C11—C13 | 1.379 (3) |
| C5—O2—C14 | 113.97 (19) | N2—C4—C3 | 108.77 (16) |
| C4—N2—C2 | 126.35 (16) | O3—C5—O2 | 124.6 (2) |
| C4—N2—C6 | 110.91 (16) | O3—C5—C10 | 125.13 (18) |
| C2—N2—C6 | 122.65 (15) | O2—C5—C10 | 110.20 (18) |
| C9—C2—C7 | 117.19 (19) | N2—C6—C8 | 107.04 (15) |
| C9—C2—N2 | 119.41 (18) | C2—C7—C12 | 121.78 (19) |
| C7—C2—N2 | 123.38 (17) | C3—C8—C6 | 104.11 (16) |
| C10—C3—C8 | 127.5 (2) | C2—C9—C13 | 120.4 (2) |
| C10—C3—C4 | 123.85 (19) | C3—C10—C5 | 123.1 (2) |
| C8—C3—C4 | 108.60 (18) | C12—C11—C13 | 118.0 (2) |
| O1—C4—N2 | 125.13 (18) | C11—C12—C7 | 120.5 (2) |
| O1—C4—C3 | 126.10 (18) | C11—C13—C9 | 122.1 (2) |

X-ray crystallographic data for methyl (Z)-2-(1-(4-chlorophenyl)-2-oxopyrrolidin-3-ylidene)acetate 2b⁵

| <i>Crystal data</i> | |
|---|--------------------------------------|
| Chemical formula: C ₁₃ H ₁₂ ClNO ₃ | <i>Mr</i> = 265.69 |
| Monoclinic, <i>C</i> 2/ <i>c</i> | <i>Dx</i> = 1.425 Mg m ⁻³ |
| <i>Hall symbol</i> : -C 2yc | Melting point: 368.65 K |
| <i>a</i> = 17.140 (4) Å | Cell parameters from 238 reflections |
| <i>b</i> = 9.005 (3) Å | θ = 2.4–39.5° |
| <i>c</i> = 16.448 (5) Å | μ = 0.31 mm ⁻¹ |
| β = 102.728 (17)° | <i>T</i> = 293 K |
| <i>V</i> = 2476.3 (13) Å ³ | Crystal: block, colorless |
| <i>Z</i> = 8 | Crystal size: 0.34 × 0.24 × 0.18 mm |
| <i>F</i> (000) = 1104 | |
| <i>Data collection</i> | |

| | |
|---|--|
| <u>Bruker-Nonius KappaCCD</u> diffractometer | 2849 independent reflections |
| Radiation source: Mo $K\alpha$ radiation, $\lambda = 0.71073 \text{ \AA}$ | 1988 reflections with $I > 2\sigma(I)$ |
| φ and ω scans | 28230 measured reflections |
| | $R_{\text{int}} = 0.060$ |
| <i>Absorption correction: multi-scan,</i> SADABS, ² $T_{\min} = 0.699$, $T_{\max} = 0.746$ | $\theta_{\max} = 27.5^\circ$, $\theta_{\min} = 3.4^\circ$ $H = -22 \rightarrow 22$ $K = -11 \rightarrow 11$ $L = -21 \rightarrow 21$ |
| Refinement | |
| Refinement on F^2 | Hydrogen site location: mixed |
| Least-squares matrix: full | H atoms treated by a mixture of independent and constrained refinement |
| $R[F^2 > 2\sigma(F^2)] = 0.040$ | $w = 1/[\sigma^2(Fo^2) + (0.1037P)^2 + 0.0154P]$ where $P = (Fo^2 + 2Fc^2)/3$ |
| $wR(F^2) = 0.099$ | $(\Delta/\sigma)_{\text{max}} < 0.001$ |
| $S = 1.02$ | $\Delta\rho_{\max} = 0.18 \text{ e \AA}^{-3}$ |
| 2849 reflections | $\Delta\rho_{\min} = -0.19 \text{ e \AA}^{-3}$ |
| 168 parameters | Extinction correction: SHEXL-2014 (3) |
| 0 restraints | $Fc^* = kFc[1 + 0.001xFc^2\lambda^3/\sin(2\theta)]^{-1/4}$ |
| Programs: SIR2014, ³ HELXL-2014 ⁴ | Extinction coefficient: 0.0058 (5) |

Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (\AA^2)

| | x | y | z | $U_{\text{iso}}^*/U_{\text{eq}}$ |
|-----|--------------|--------------|---------------|--|
| Cl1 | 0.36140 (4) | 0.28077 (7) | 0.19067 (4) | 0.0744 (2) |
| O1 | 0.60917 (7) | 0.30353 (13) | -0.07714 (8) | 0.0519 (3) |
| N1 | 0.62275 (8) | 0.12000 (14) | 0.02375 (8) | 0.0428 (3) |
| O2 | 0.62083 (8) | 0.21774 (14) | -0.25023 (9) | 0.0617 (4) |
| C2 | 0.56157 (10) | 0.16225 (18) | 0.06441 (10) | 0.0417 (4) |
| C3 | 0.63977 (10) | 0.18922 (18) | -0.04420 (11) | 0.0424 (4) |
| C4 | 0.53006 (11) | 0.05738 (19) | 0.11046 (10) | 0.0474 (4) |
| H4 | 0.5504 | -0.0387 | 0.1152 | 0.057* |
| C5 | 0.53137 (12) | 0.30670 (19) | 0.05982 (11) | 0.0507 (4) |

| | | | | |
|------|--------------|---------------|---------------|------------|
| H5 | 0.5524 | 0.3788 | 0.0303 | 0.061* |
| C6 | 0.43917 (11) | 0.2359 (2) | 0.14260 (11) | 0.0502 (4) |
| C15 | 0.66552 (11) | -0.02241 (18) | 0.04229 (11) | 0.0504 (4) |
| H15A | 0.6295 | -0.1055 | 0.0257 | 0.061* |
| H15B | 0.6894 | -0.0308 | 0.1013 | 0.061* |
| C7 | 0.46901 (11) | 0.0939 (2) | 0.14924 (10) | 0.0505 (4) |
| H7 | 0.4482 | 0.0229 | 0.1796 | 0.061* |
| O3 | 0.72945 (9) | 0.35962 (17) | -0.20874 (9) | 0.0698 (4) |
| C9 | 0.72980 (11) | -0.0186 (2) | -0.00858 (13) | 0.0586 (5) |
| H9A | 0.7815 | 0.0064 | 0.0263 | 0.070* |
| H9B | 0.7338 | -0.1137 | -0.0350 | 0.070* |
| C10 | 0.69283 (11) | 0.2483 (2) | -0.20222 (11) | 0.0505 (4) |
| C11 | 0.47046 (12) | 0.3431 (2) | 0.09881 (11) | 0.0538 (5) |
| H11 | 0.4505 | 0.4395 | 0.0956 | 0.065* |
| C12 | 0.70200 (10) | 0.10022 (19) | -0.07254 (12) | 0.0477 (4) |
| C13 | 0.72371 (11) | 0.1250 (2) | -0.14379 (14) | 0.0562 (5) |
| H13A | 0.7597 (12) | 0.068 (2) | -0.1611 (13) | 0.067* |
| C14 | 0.58596 (15) | 0.3348 (3) | -0.30646 (15) | 0.0796 (7) |
| H14A | 0.5325 | 0.3075 | -0.3340 | 0.119* |
| H14B | 0.6176 | 0.3499 | -0.3472 | 0.119* |
| H14C | 0.5846 | 0.4248 | -0.2756 | 0.119* |

Atomic displacement parameters (\AA^2)

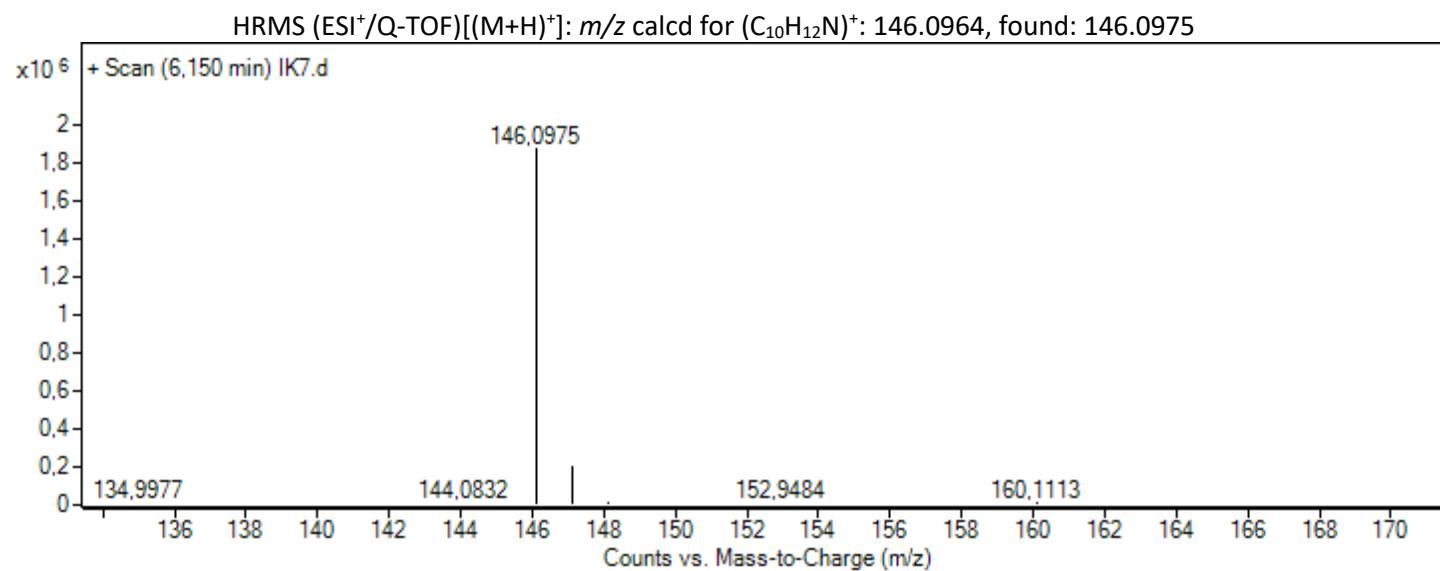
| | <i>U</i>11 | <i>U</i>22 | <i>U</i>33 | <i>U</i>12 | <i>U</i>13 | <i>U</i>23 |
|-----|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|
| Cl1 | 0.0765 (4) | 0.0859 (4) | 0.0669 (3) | -0.0009 (3) | 0.0288 (3) | -0.0111 (3) |
| O1 | 0.0582 (8) | 0.0420 (7) | 0.0568 (7) | 0.0093 (6) | 0.0155 (6) | 0.0114 (6) |
| N1 | 0.0487 (8) | 0.0317 (7) | 0.0437 (8) | 0.0010 (6) | 0.0008 (6) | 0.0027 (6) |
| O2 | 0.0621 (8) | 0.0496 (8) | 0.0666 (9) | -0.0049 (6) | -0.0005 (7) | -0.0040 (7) |
| C2 | 0.0502 (10) | 0.0352 (8) | 0.0349 (8) | -0.0034 (7) | -0.0007 (7) | -0.0011 (7) |
| C3 | 0.0421 (9) | 0.0355 (9) | 0.0458 (9) | -0.0025 (7) | 0.0017 (7) | -0.0008 (7) |
| C4 | 0.0604 (11) | 0.0362 (9) | 0.0407 (9) | -0.0025 (8) | 0.0004 (8) | 0.0025 (7) |

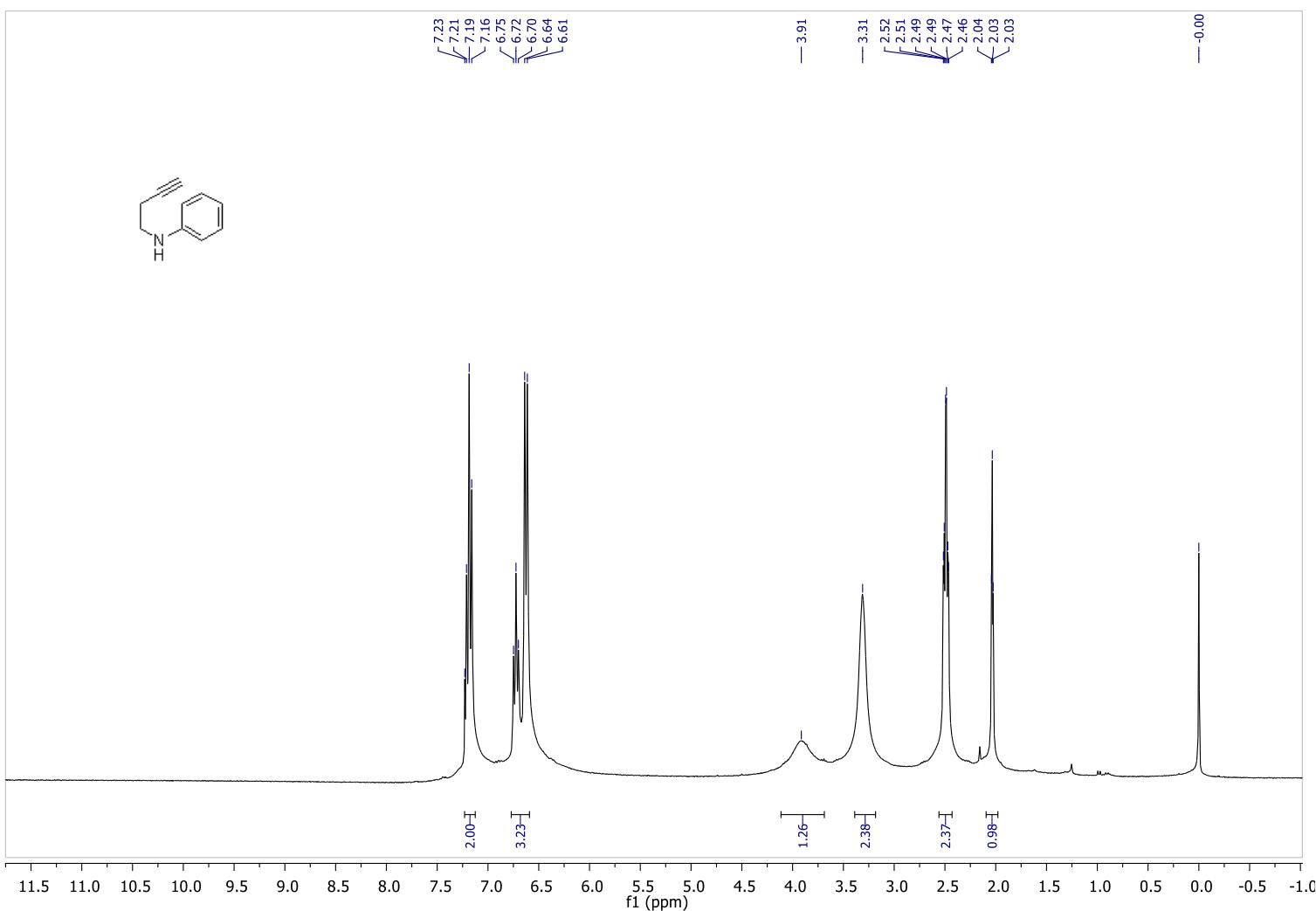
| | | | | | | |
|-----|-------------|-------------|-------------|-------------|--------------|--------------|
| C5 | 0.0691 (12) | 0.0351 (9) | 0.0486 (10) | -0.0014 (8) | 0.0142 (9) | 0.0030 (8) |
| C6 | 0.0557 (11) | 0.0552 (11) | 0.0380 (9) | -0.0055 (8) | 0.0063 (8) | -0.0075 (8) |
| C15 | 0.0585 (11) | 0.0365 (9) | 0.0487 (10) | 0.0057 (8) | -0.0046 (8) | 0.0028 (7) |
| C7 | 0.0624 (11) | 0.0477 (10) | 0.0382 (9) | -0.0109 (9) | 0.0042 (8) | 0.0024 (8) |
| O3 | 0.0629 (9) | 0.0728 (10) | 0.0755 (10) | -0.0179 (8) | 0.0192 (7) | 0.0032 (8) |
| C9 | 0.0495 (11) | 0.0468 (10) | 0.0731 (13) | 0.0094 (8) | -0.0002 (9) | 0.0041 (9) |
| C10 | 0.0483 (10) | 0.0545 (11) | 0.0525 (10) | -0.0003 (8) | 0.0191 (8) | -0.0081 (8) |
| C11 | 0.0695 (12) | 0.0419 (10) | 0.0501 (10) | 0.0051 (9) | 0.0133 (9) | -0.0017 (8) |
| C12 | 0.0391 (9) | 0.0418 (9) | 0.0593 (11) | 0.0013 (7) | 0.0043 (8) | -0.0030 (8) |
| C13 | 0.0453 (10) | 0.0532 (11) | 0.0705 (13) | 0.0068 (8) | 0.0139 (9) | -0.0073 (10) |
| C14 | 0.0871 (16) | 0.0613 (13) | 0.0763 (15) | 0.0013 (12) | -0.0121 (13) | 0.0007 (12) |

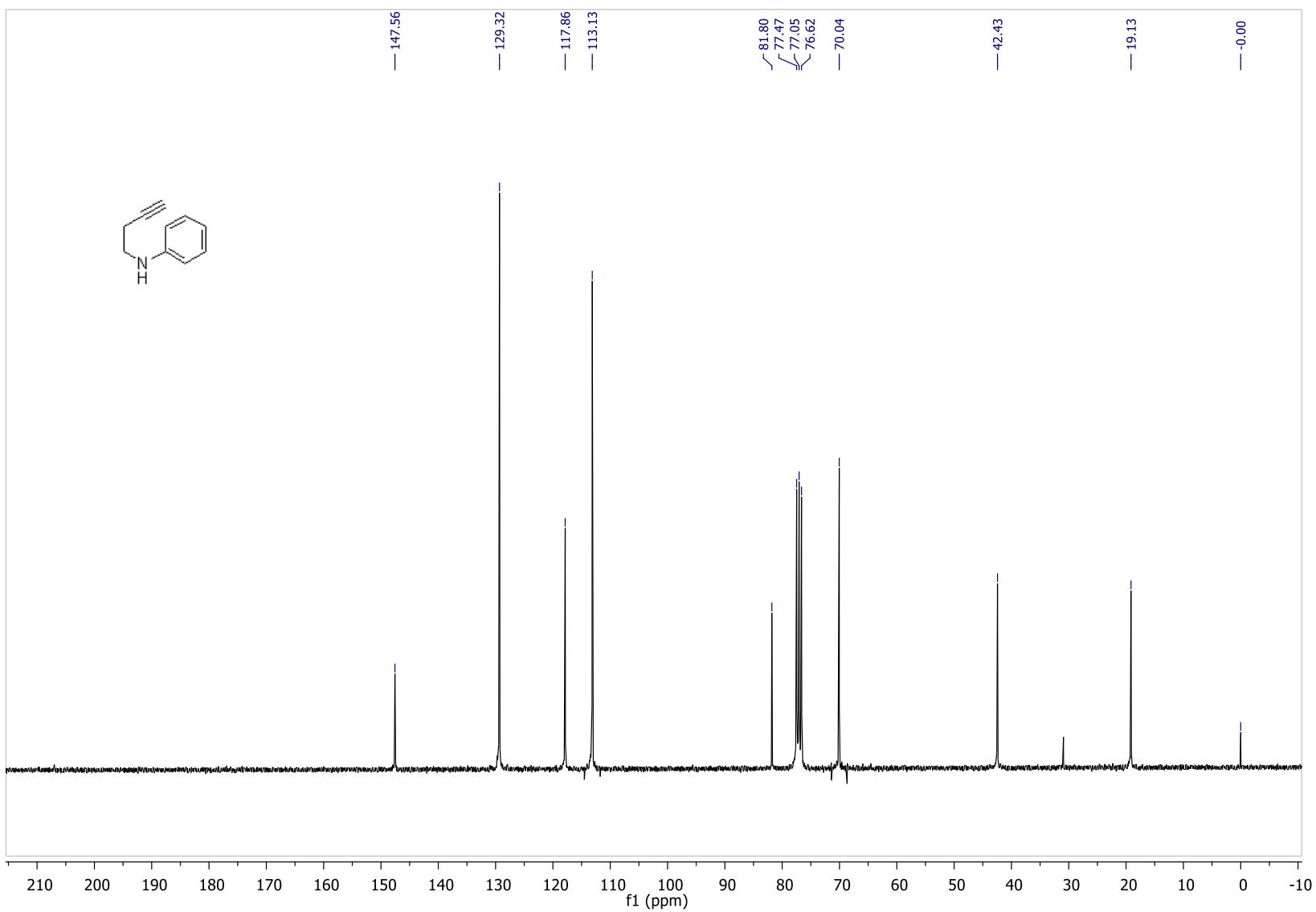
Geometric parameters (Å, °)

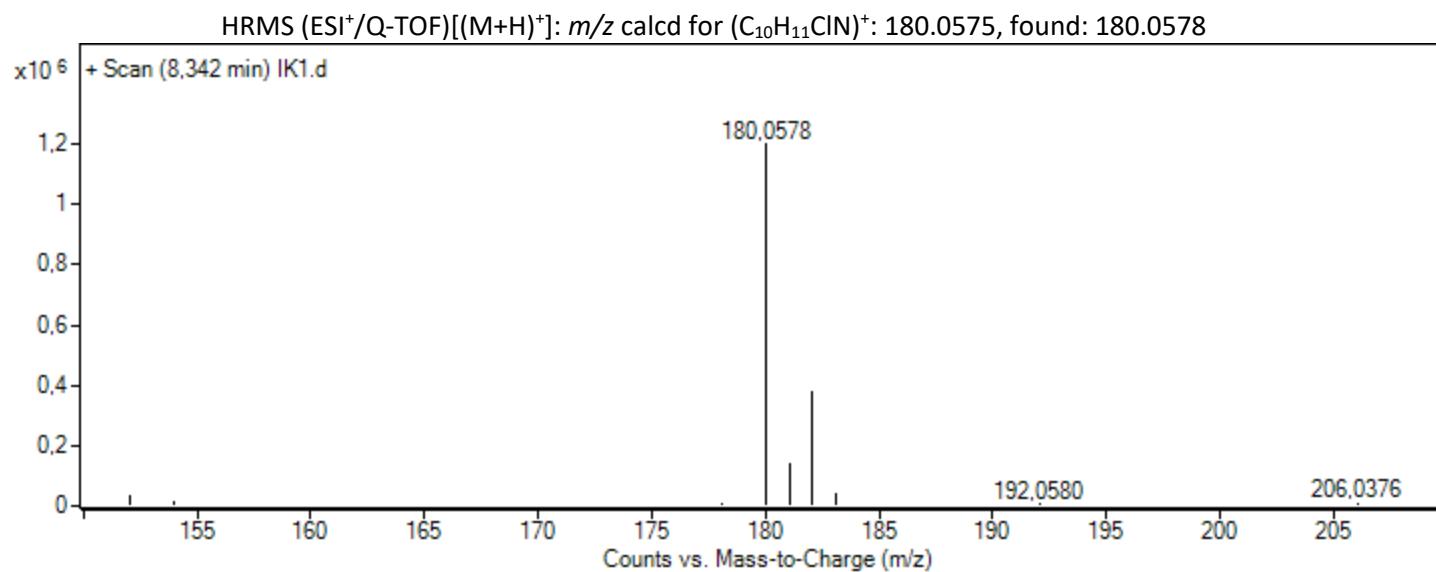
| | | | |
|-----------|-------------|------------|-------------|
| Cl1—C6 | 1.741 (2) | C4—C7 | 1.380 (3) |
| O1—C3 | 1.2259 (19) | C5—C11 | 1.380 (3) |
| N1—C3 | 1.366 (2) | C6—C7 | 1.373 (3) |
| N1—C2 | 1.414 (2) | C6—C11 | 1.382 (3) |
| N1—C15 | 1.475 (2) | C15—C9 | 1.524 (3) |
| O2—C10 | 1.339 (2) | O3—C10 | 1.200 (2) |
| O2—C14 | 1.442 (2) | C9—C12 | 1.502 (3) |
| C2—C4 | 1.392 (2) | C10—C13 | 1.488 (3) |
| C2—C5 | 1.396 (2) | C12—C13 | 1.324 (3) |
| C3—C12 | 1.489 (2) | | |
| C3—N1—C2 | 125.33 (14) | C7—C6—Cl1 | 119.30 (15) |
| C3—N1—C15 | 112.32 (14) | C11—C6—Cl1 | 119.92 (15) |
| C2—N1—C15 | 121.50 (14) | N1—C15—C9 | 104.67 (14) |

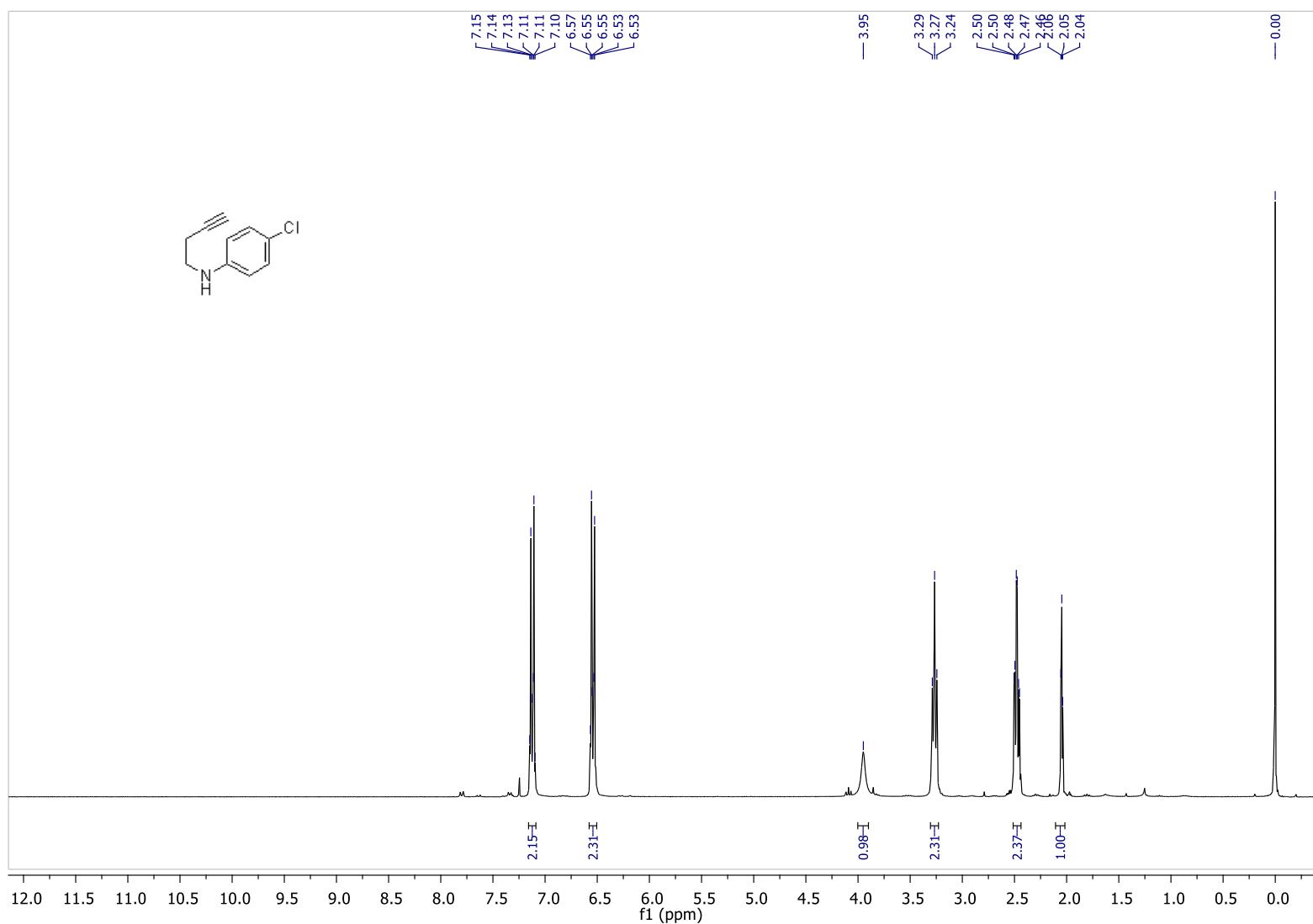
| | | | |
|------------|-------------|-------------|-------------|
| C10—O2—C14 | 115.20 (16) | C6—C7—C4 | 119.51 (17) |
| C4—C2—C5 | 118.55 (17) | C12—C9—C15 | 104.16 (14) |
| C4—C2—N1 | 119.46 (15) | O3—C10—O2 | 123.47 (18) |
| C5—C2—N1 | 121.99 (15) | O3—C10—C13 | 124.23 (18) |
| O1—C3—N1 | 126.65 (16) | O2—C10—C13 | 112.20 (16) |
| O1—C3—C12 | 125.45 (16) | C5—C11—C6 | 119.76 (18) |
| N1—C3—C12 | 107.90 (14) | C13—C12—C3 | 123.14 (17) |
| C7—C4—C2 | 120.96 (16) | C13—C12—C9 | 129.21 (18) |
| C11—C5—C2 | 120.40 (17) | C3—C12—C9 | 107.62 (16) |
| C7—C6—C11 | 120.79 (18) | C12—C13—C10 | 124.63 (17) |

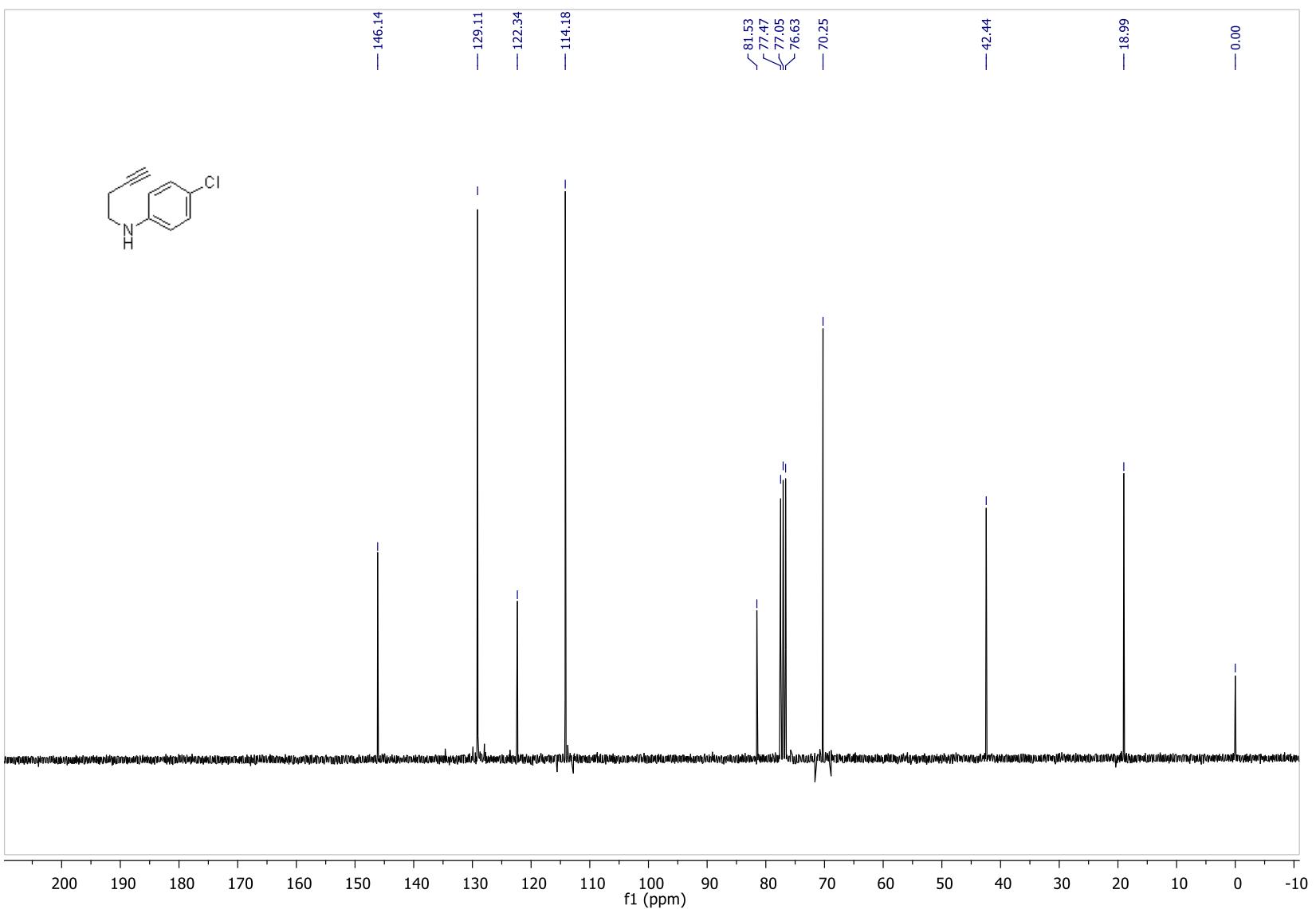
Copies of HRMS, ^1H NMR, and ^{13}C NMR Spectra***N*-(but-3-yn-1-yl)aniline **1a****

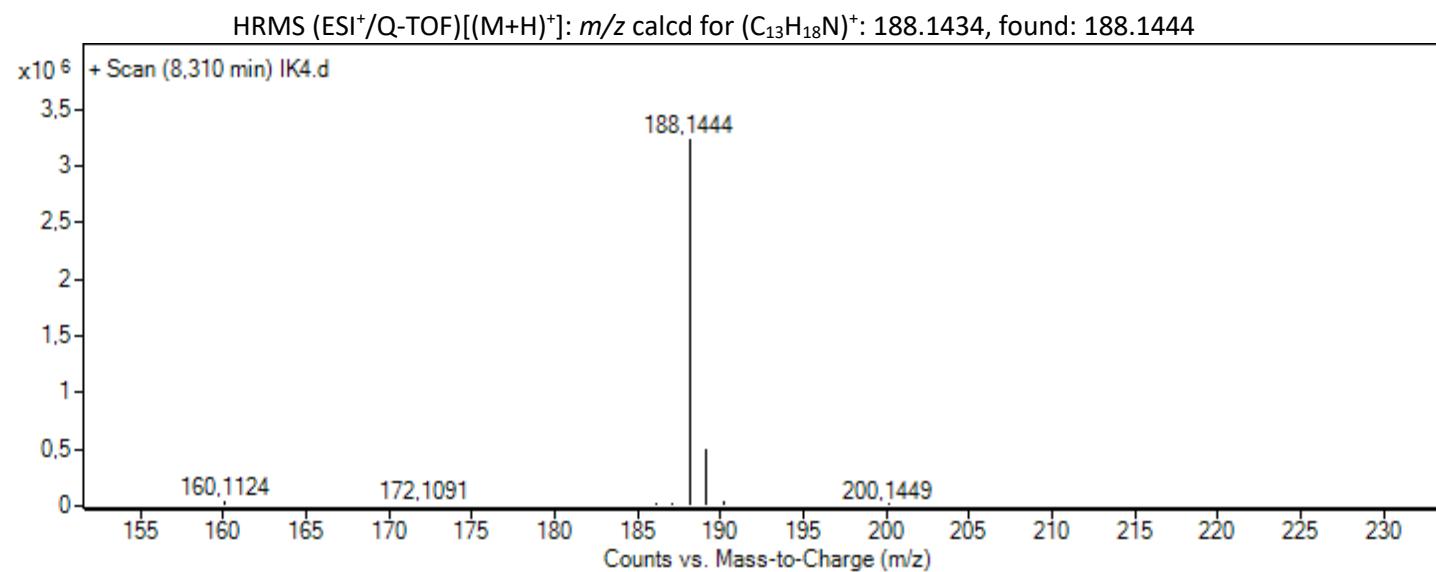
^1H NMR (CDCl_3)

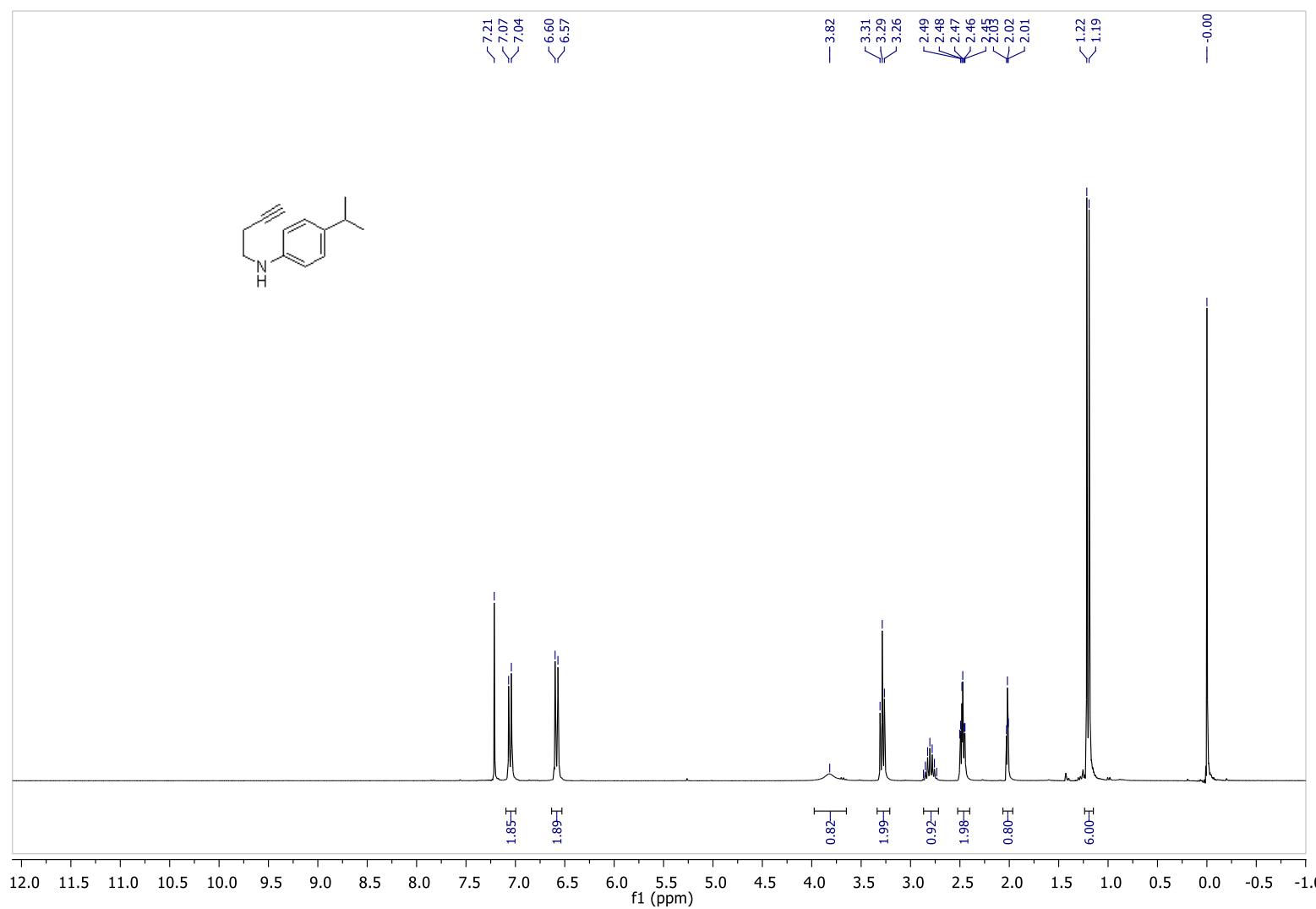
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

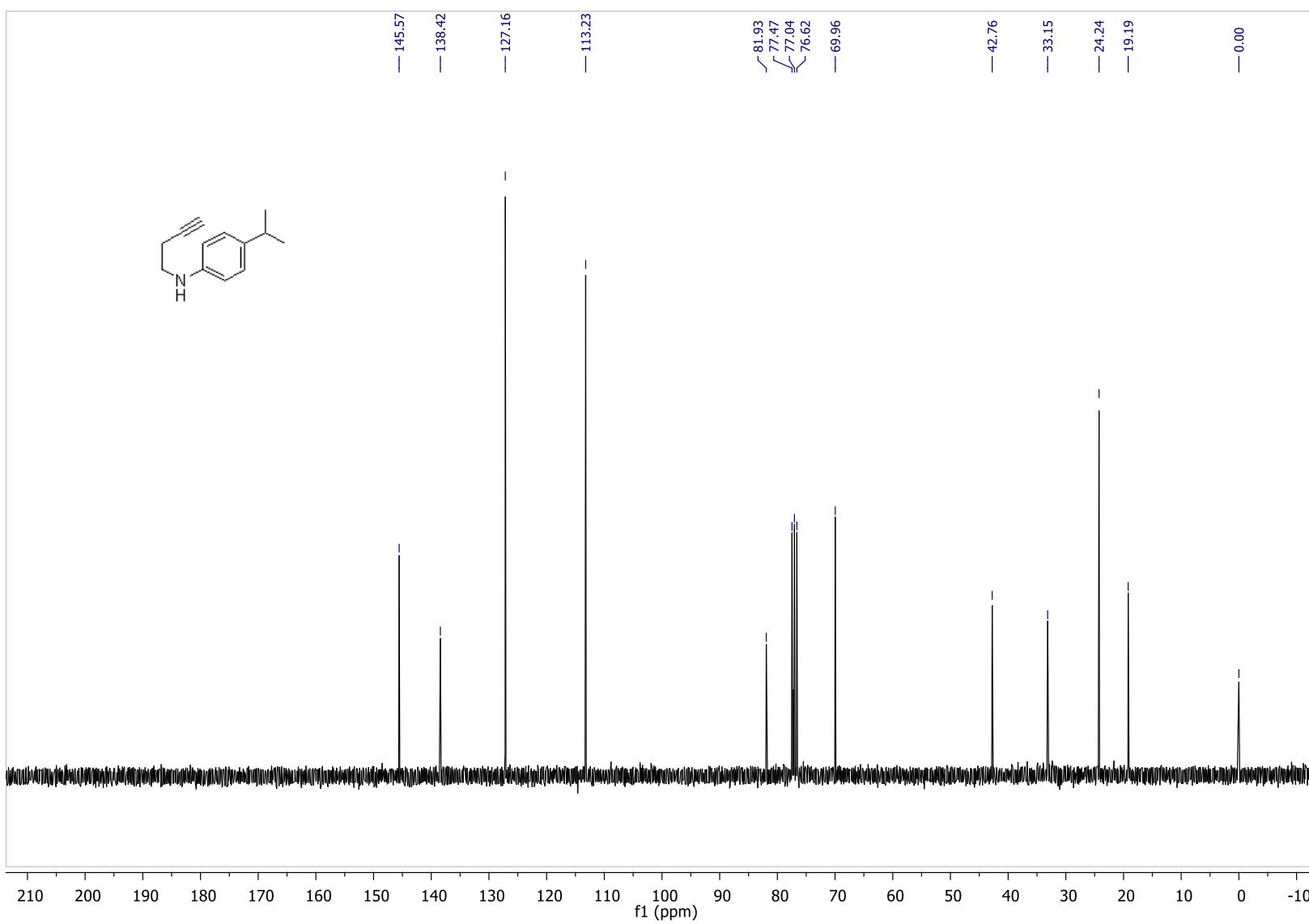
N-(but-3-yn-1-yl)-4-chloroaniline **1b**

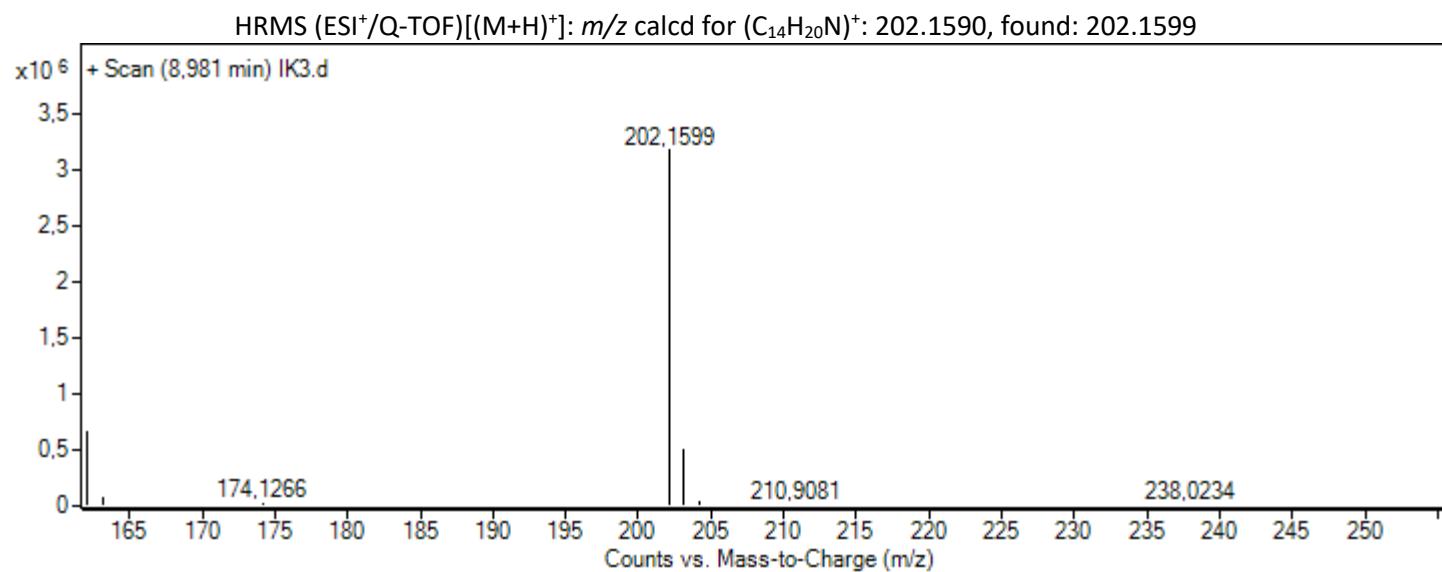
^1H NMR (CDCl_3)

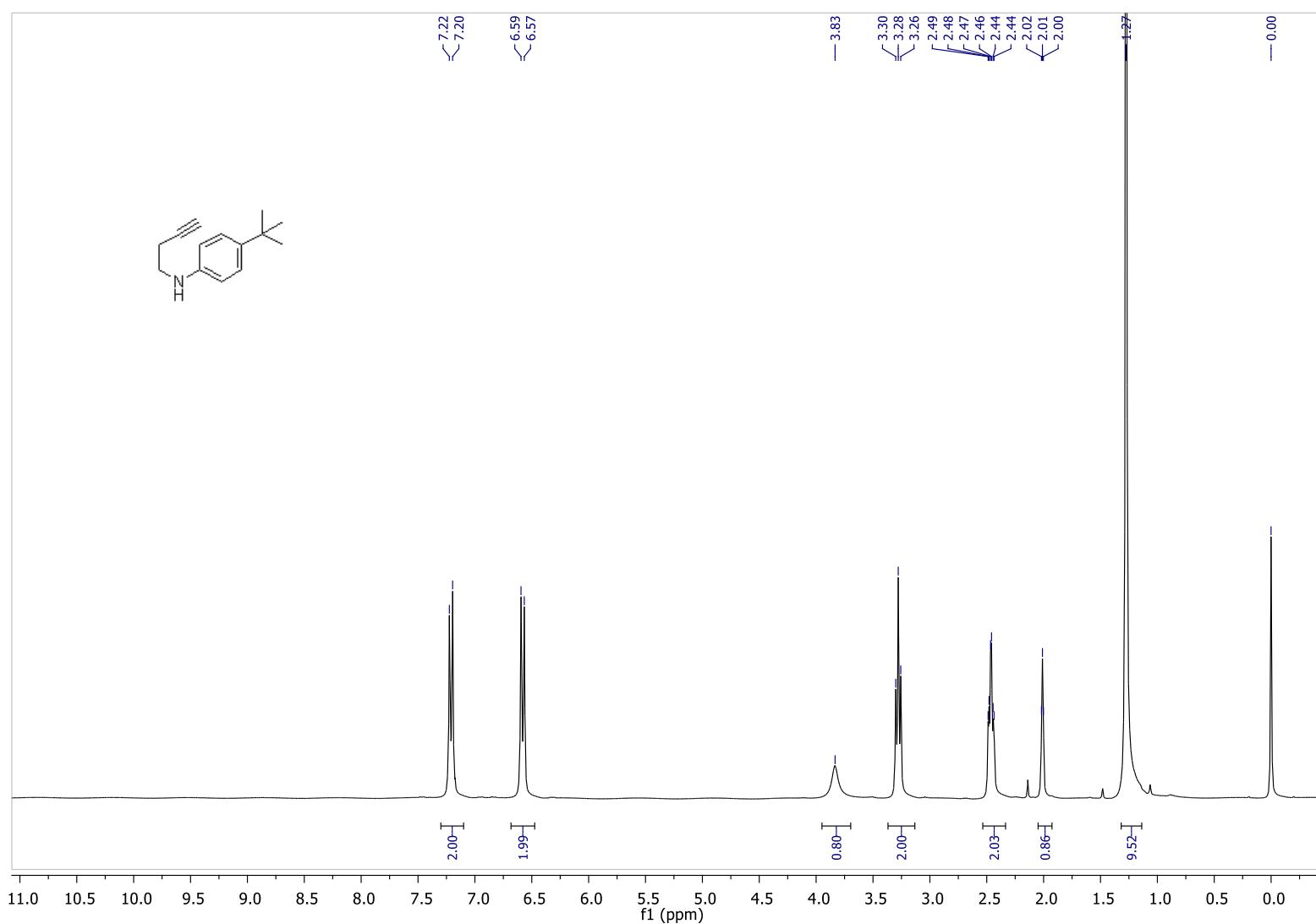
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

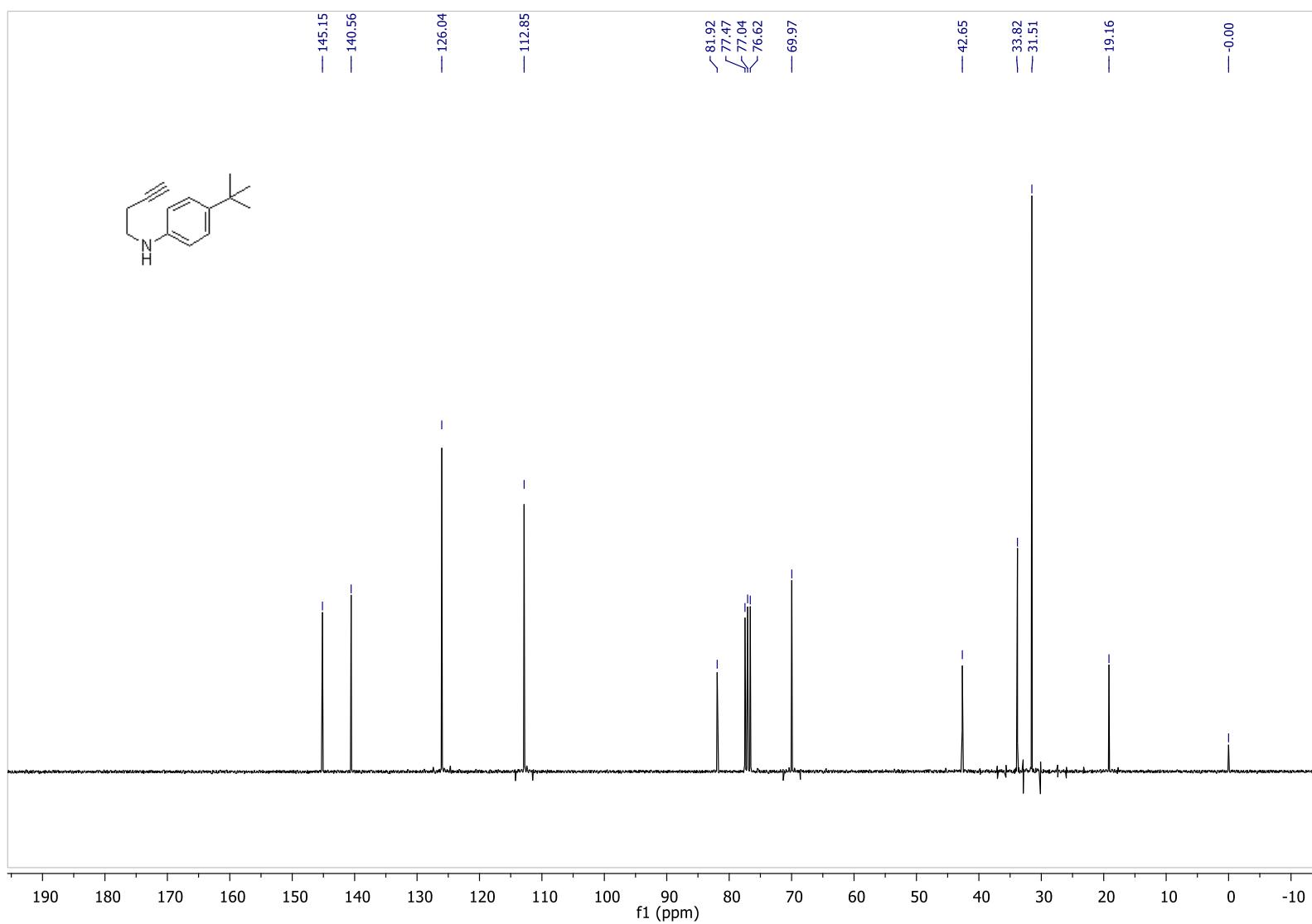
N-(but-3-yn-1-yl)-4-isopropylaniline **1c**

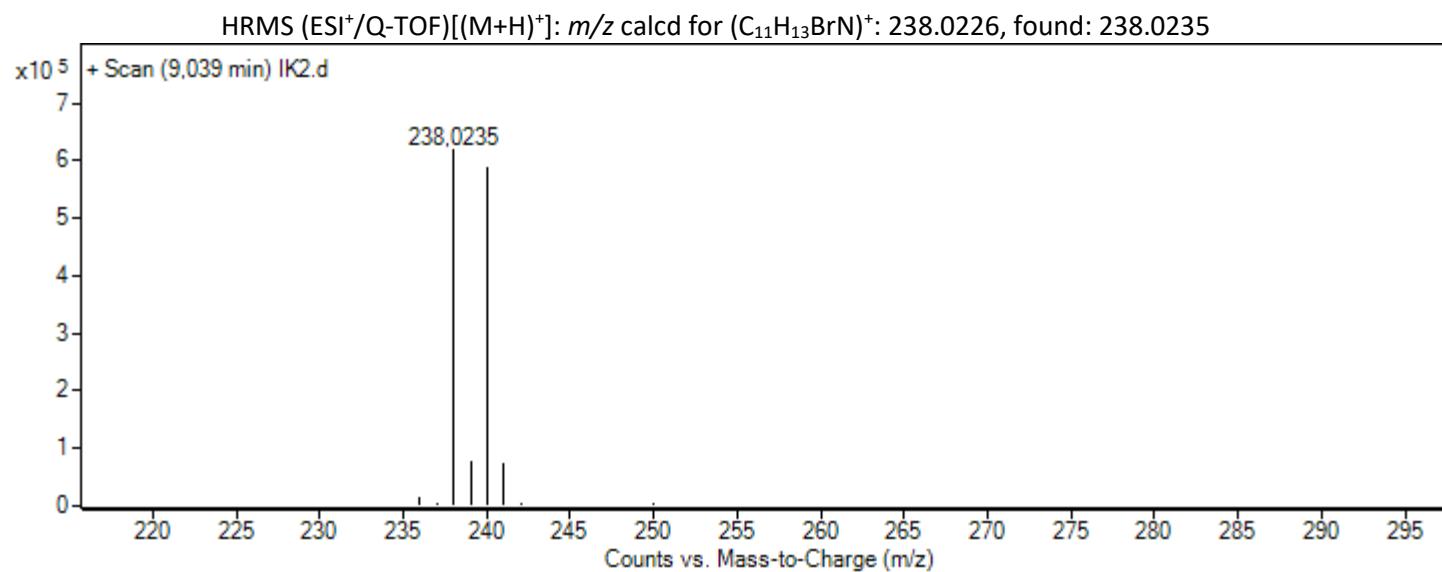
^1H NMR (CDCl_3)

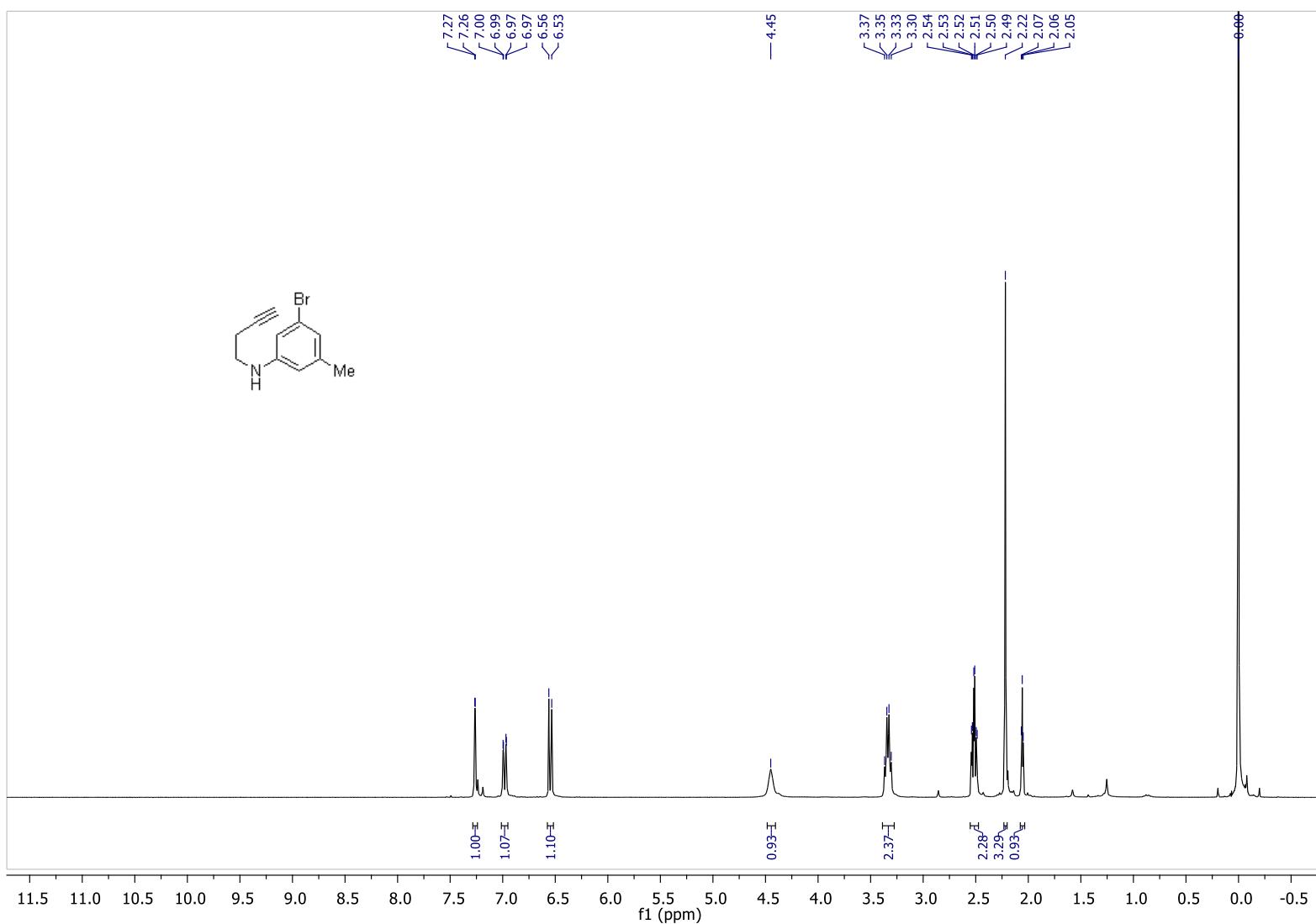
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

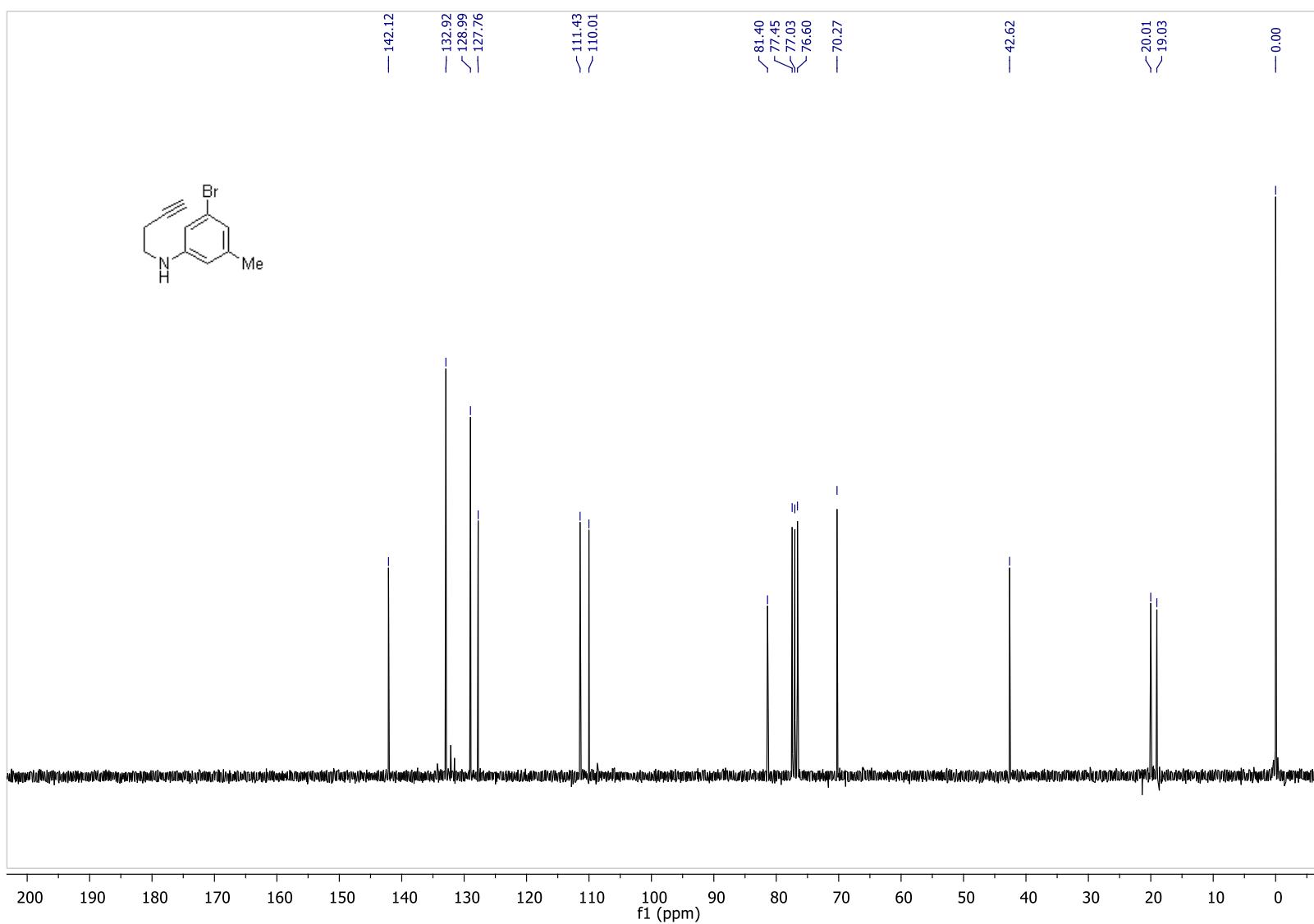
N-(but-3-yn-1-yl)-4-(tert-butyl)aniline **1d**

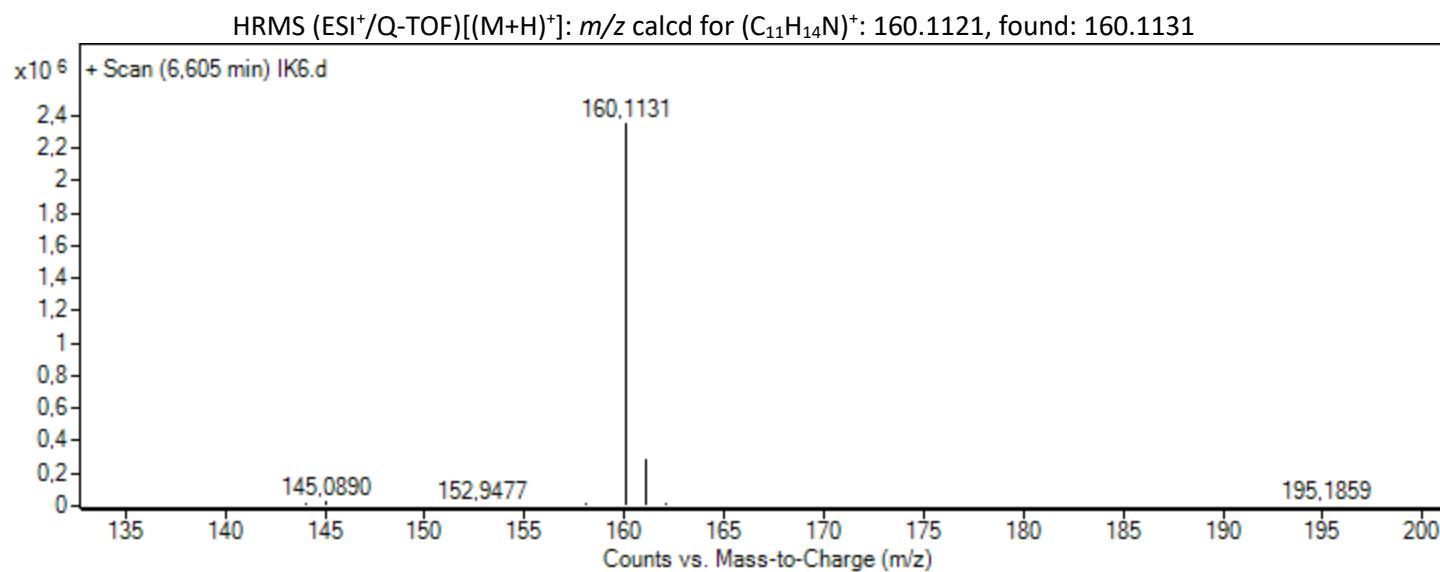
^1H NMR (CDCl_3)

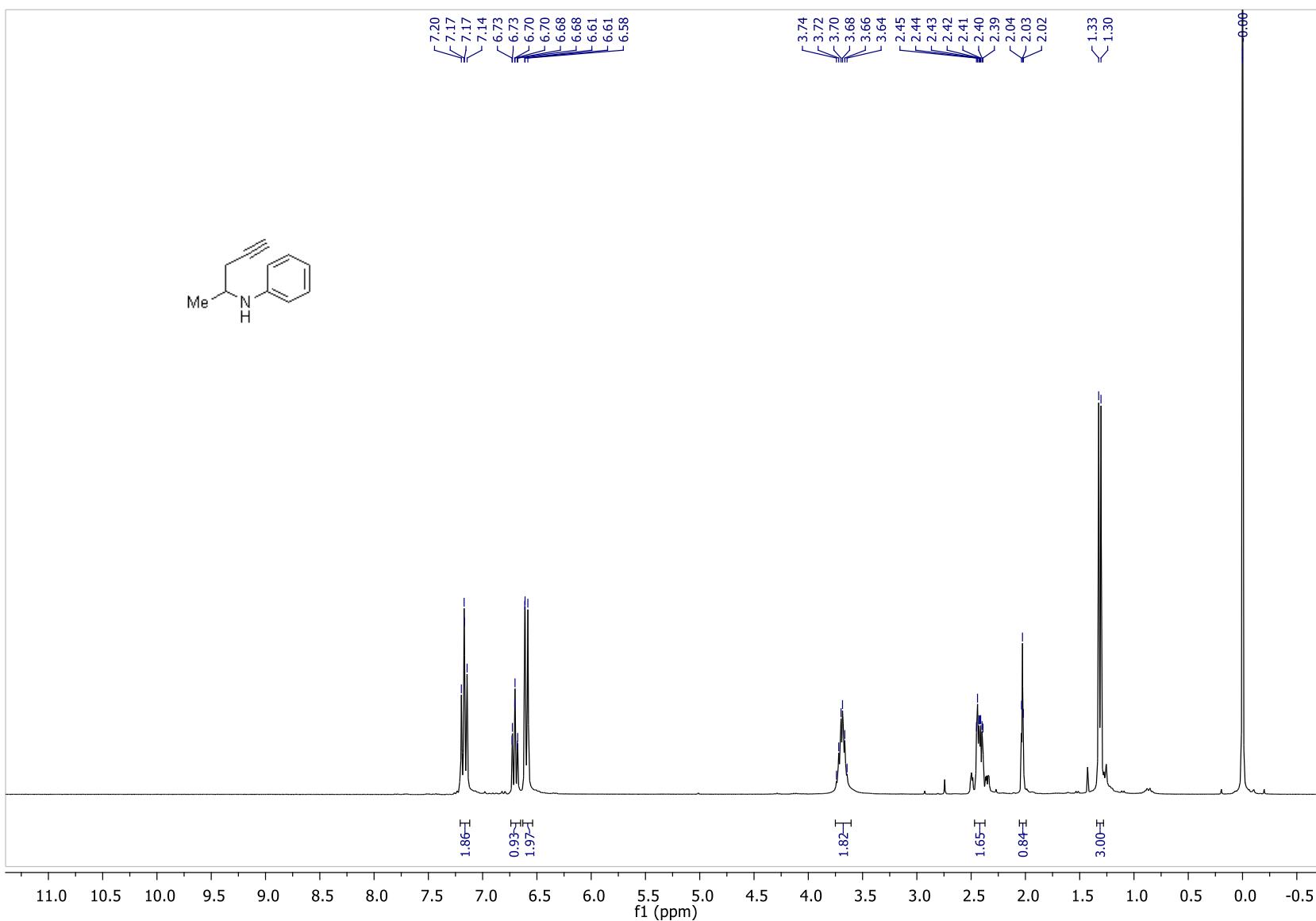
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

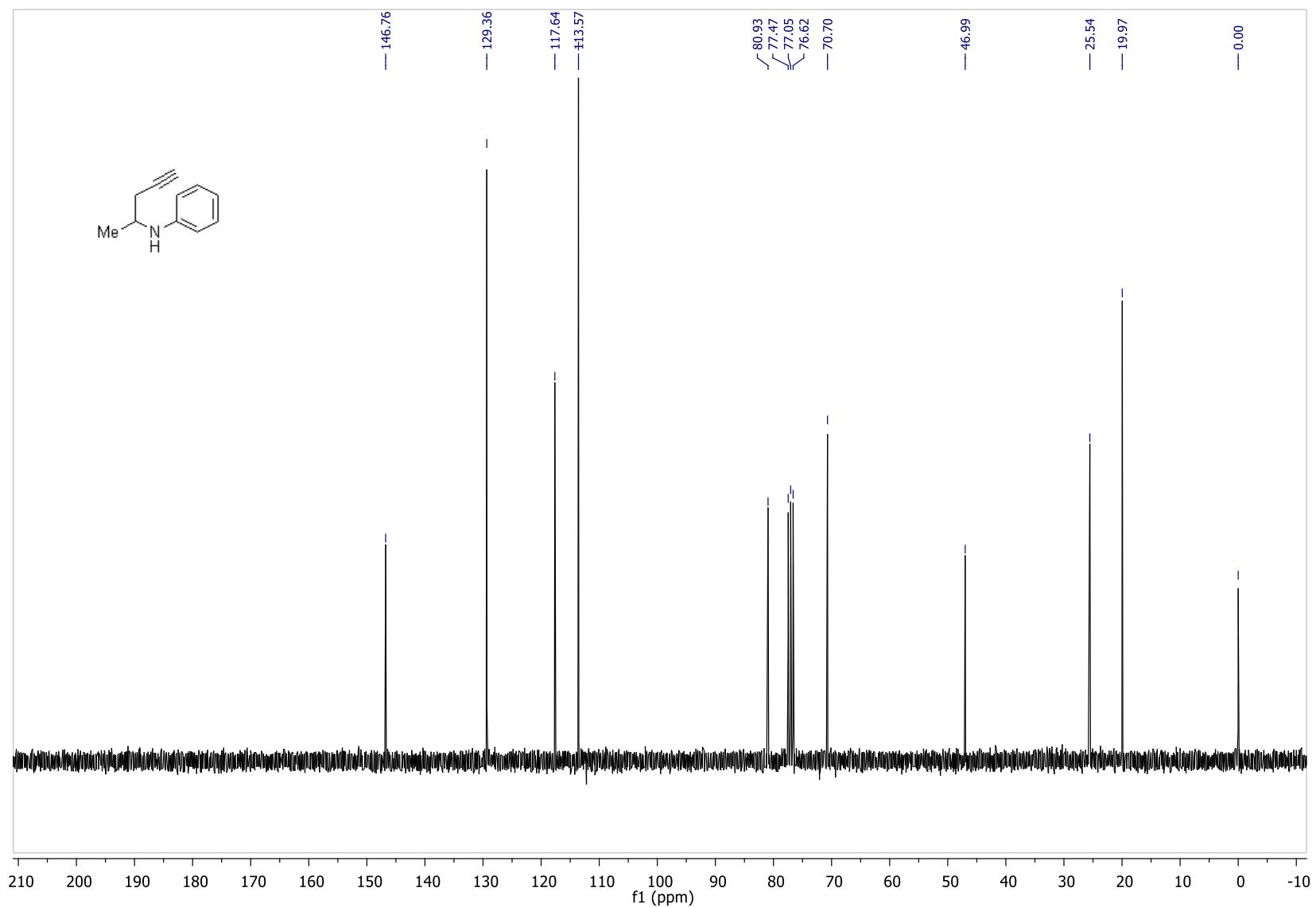
3-bromo-N-(but-3-yn-1-yl)-5-methylaniline **1e**

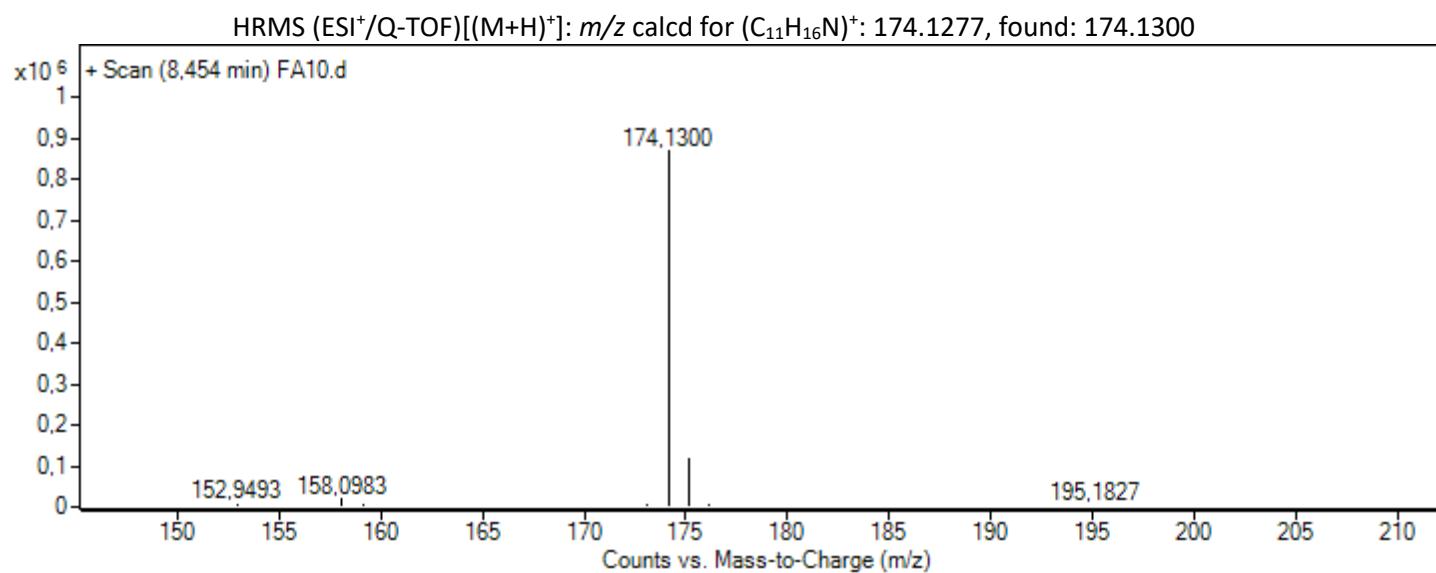
^1H NMR (CDCl_3)

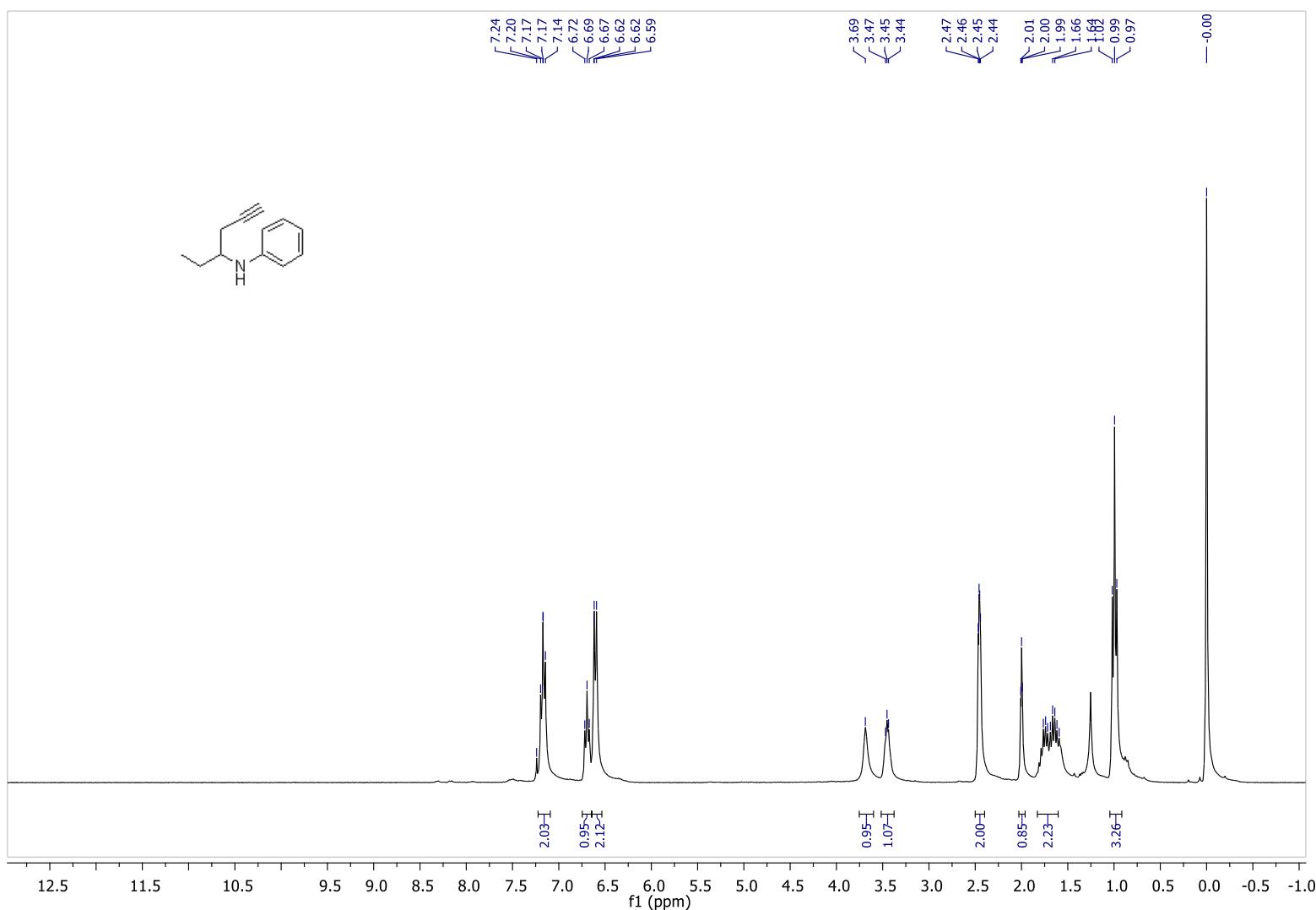
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

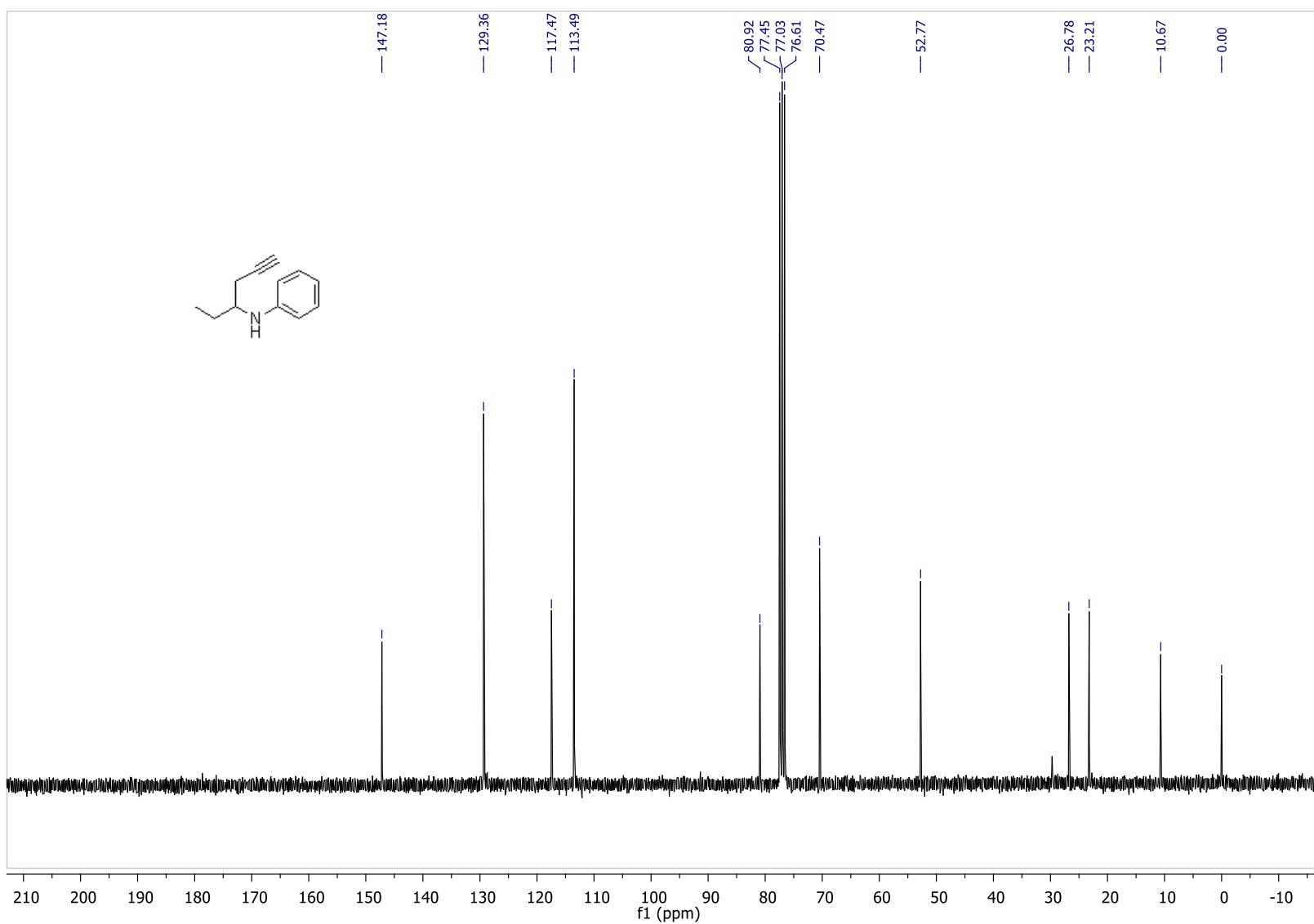
N-(pent-4-yn-2-yl)aniline **1f**

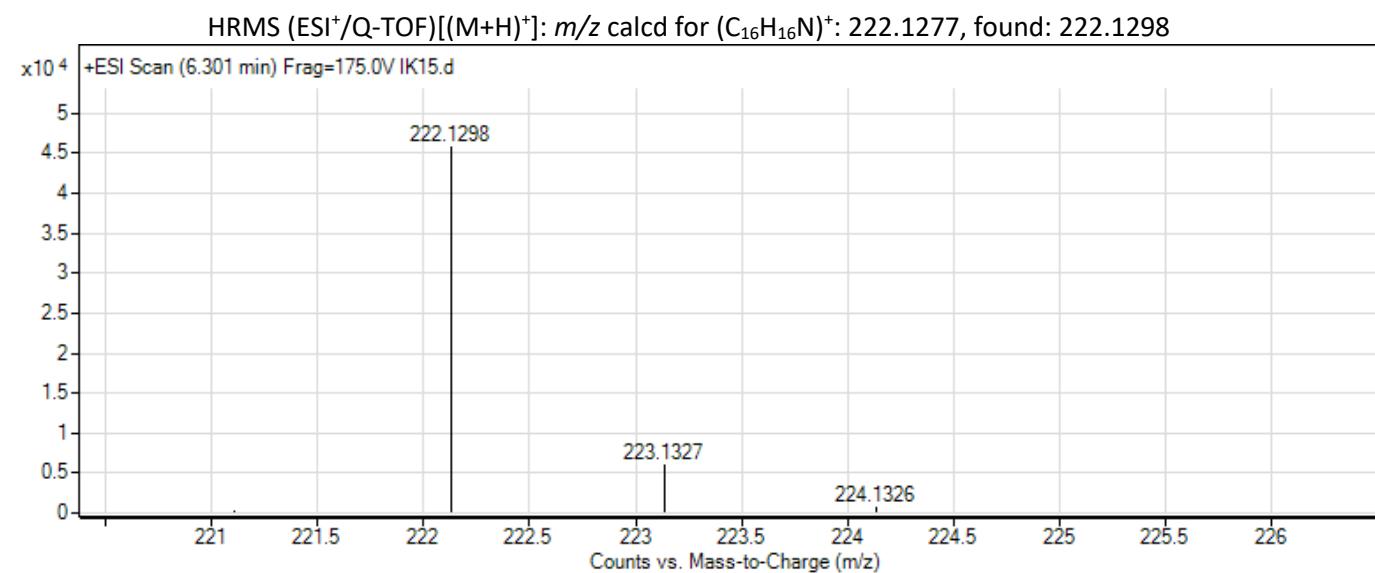
^1H NMR (CDCl_3)

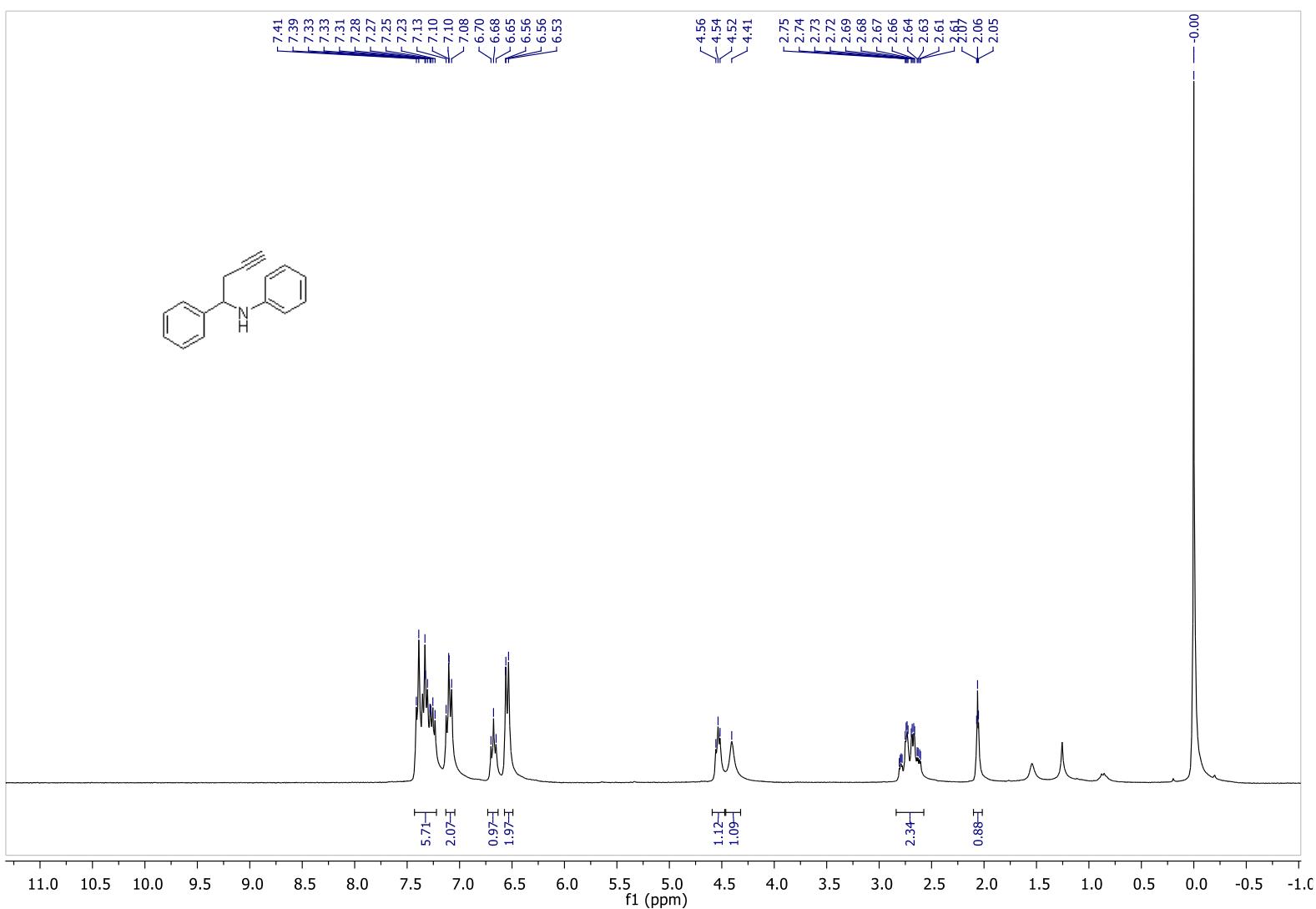
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

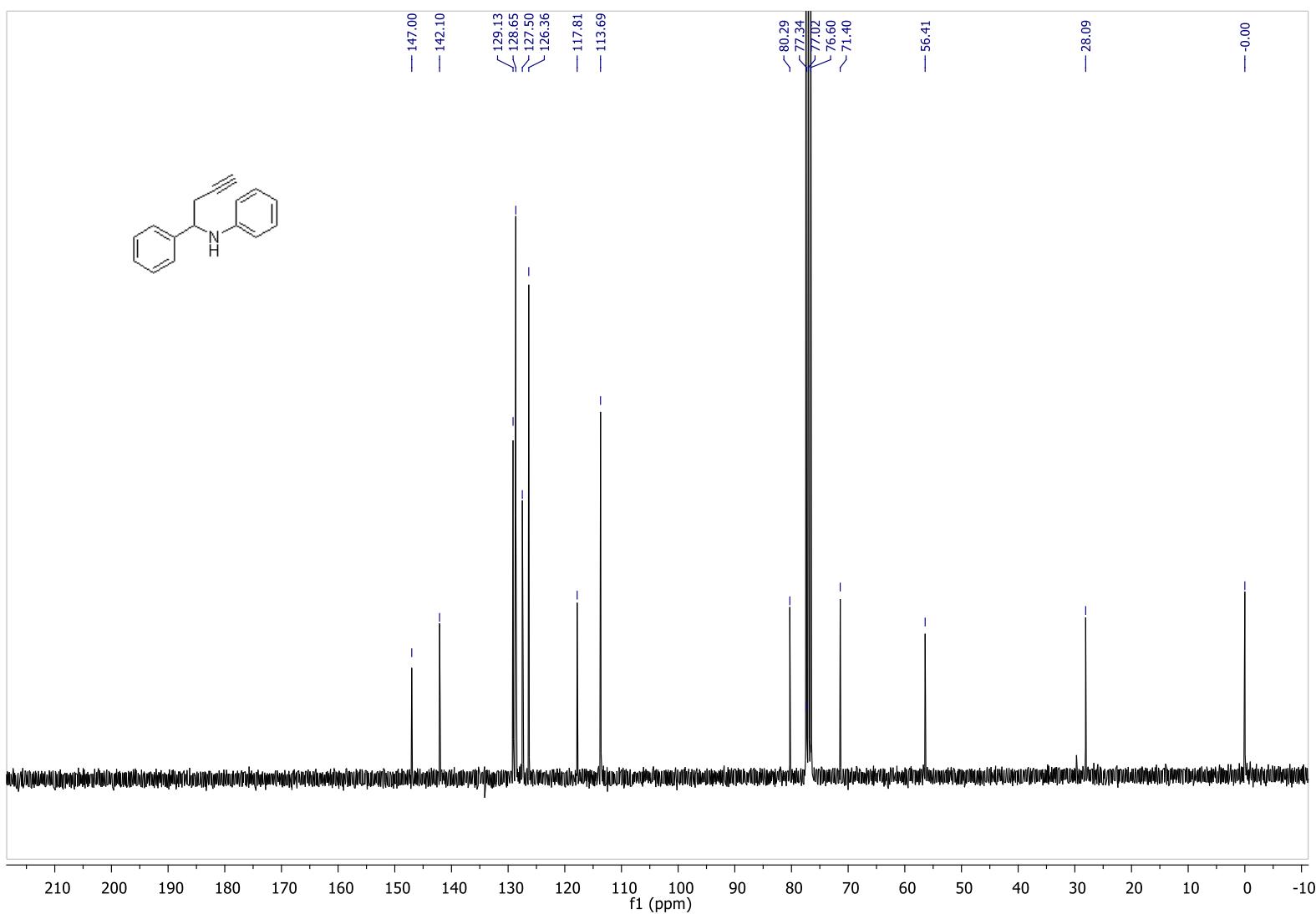
N-(hex-5-yn-3-yl)aniline **1g**

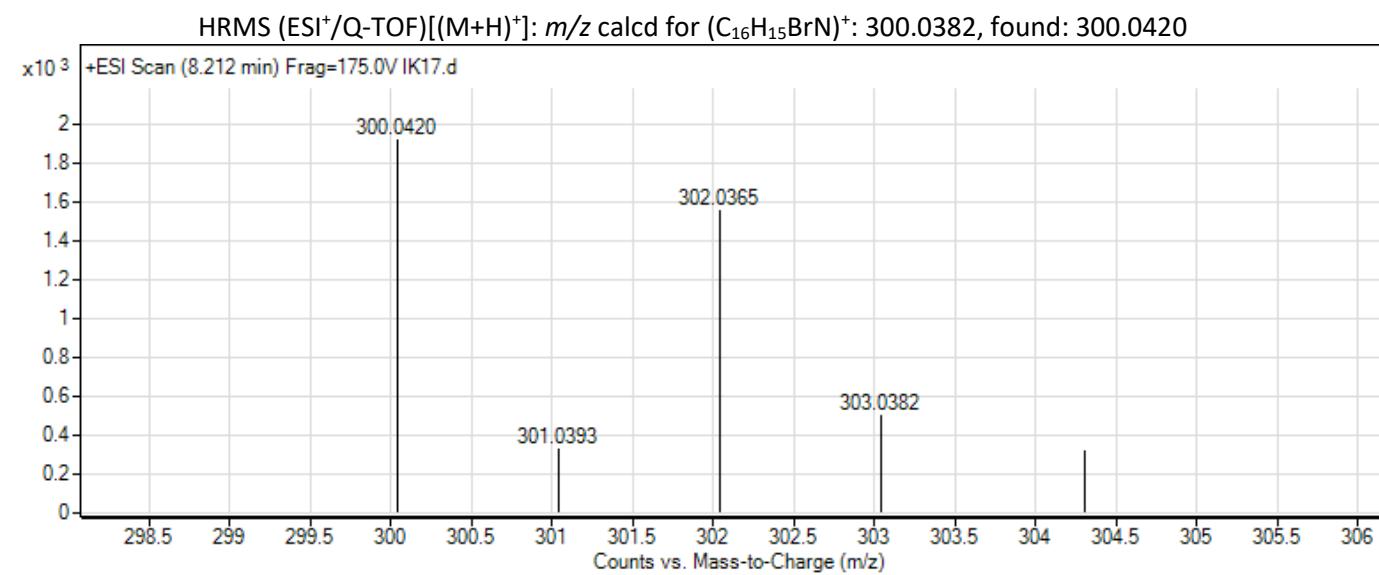
^1H NMR (CDCl_3)

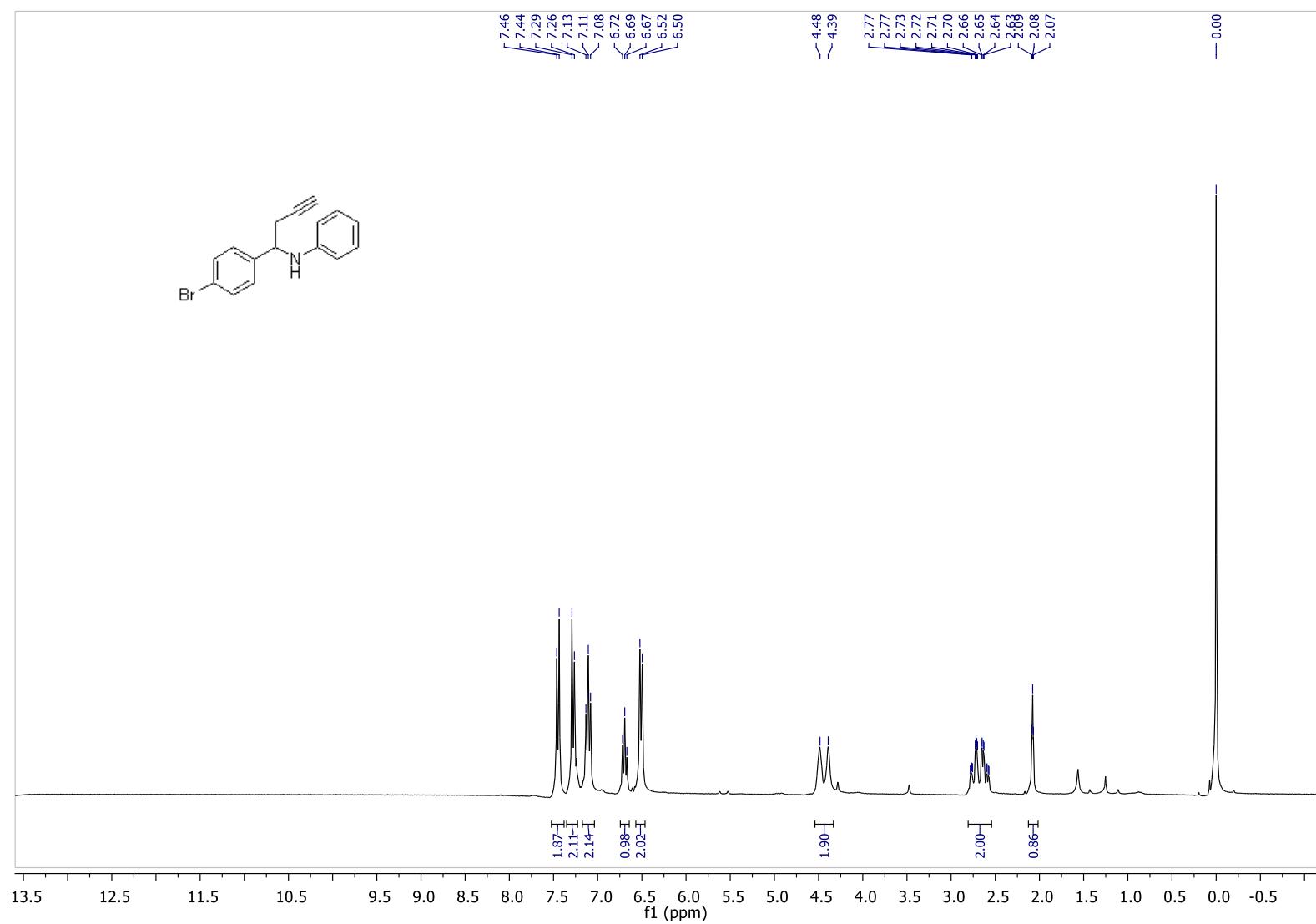
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

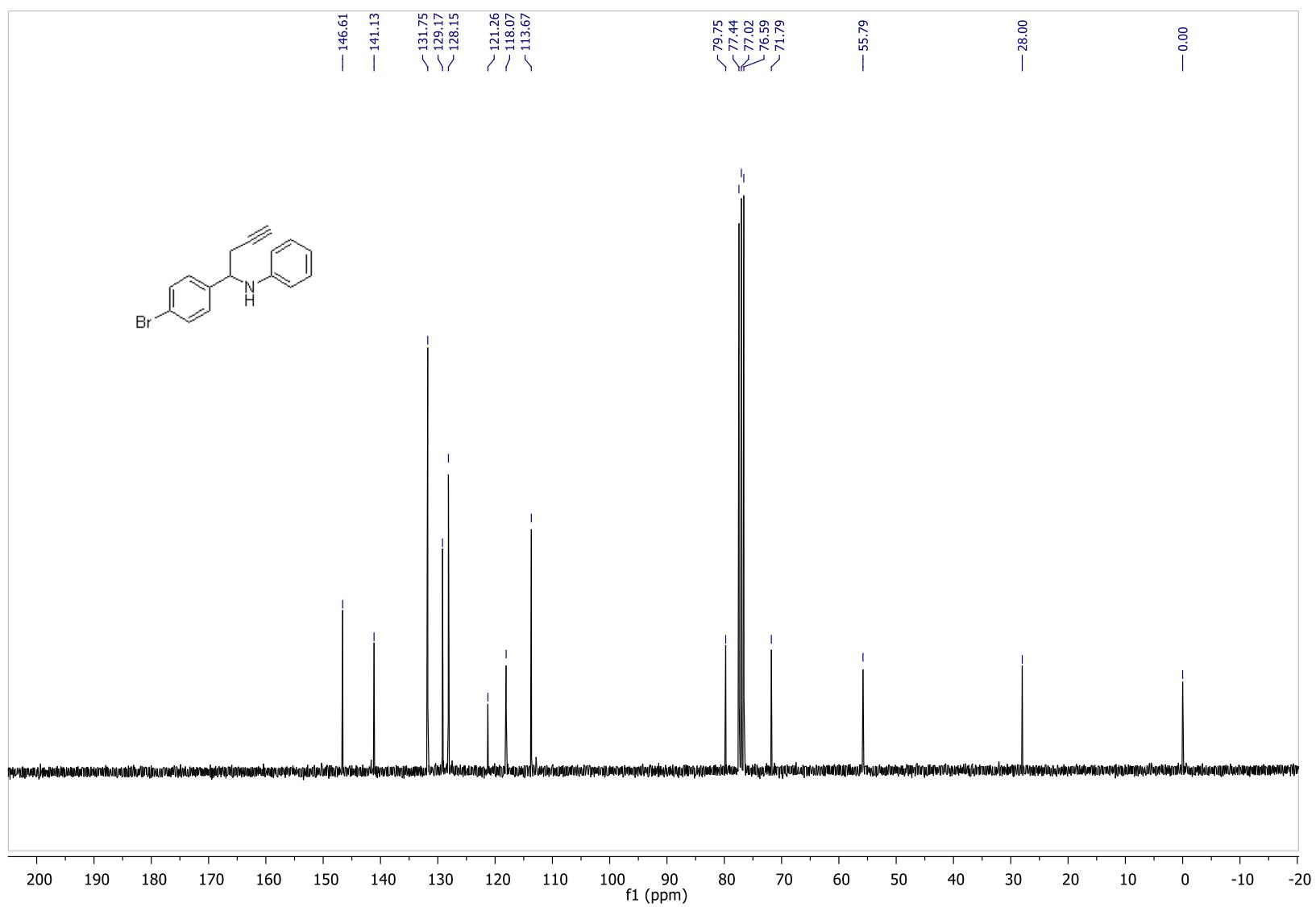
N-(1-phenylbut-3-yn-1-yl)aniline **1h**

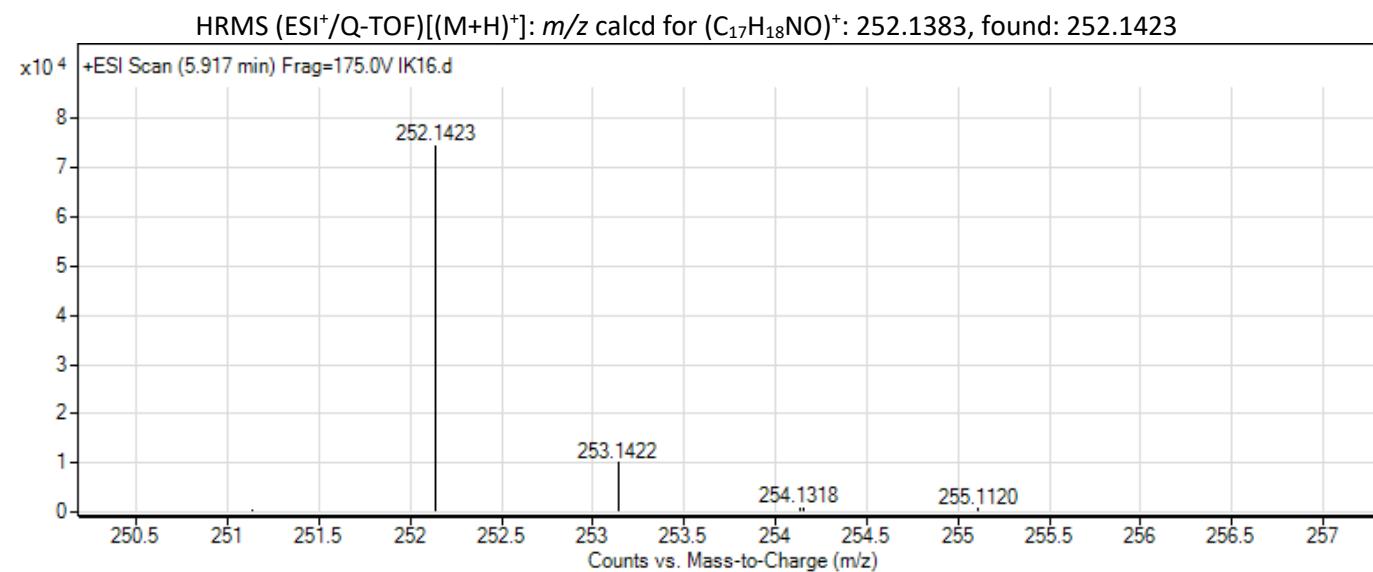
^1H NMR (CDCl_3)

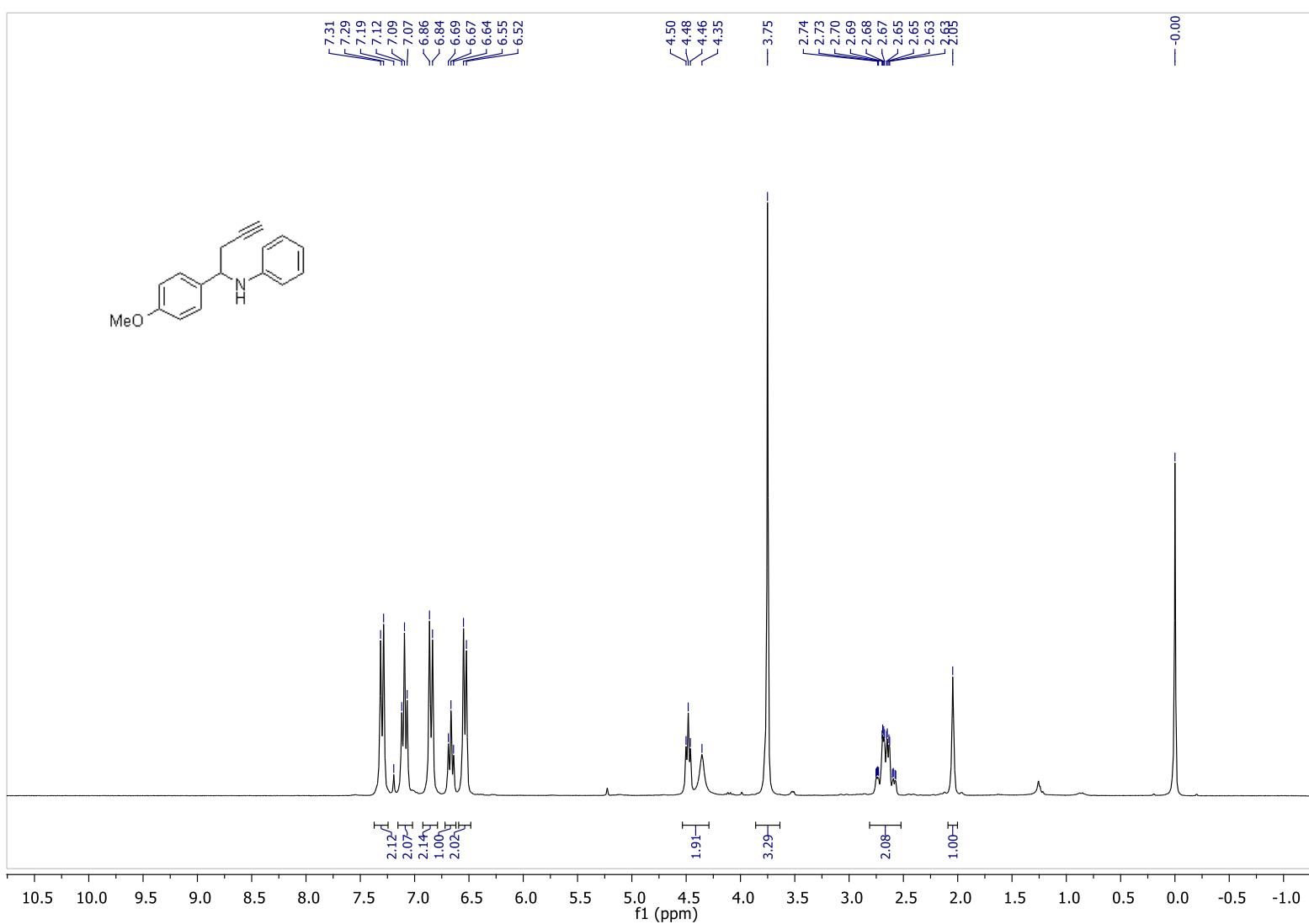
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

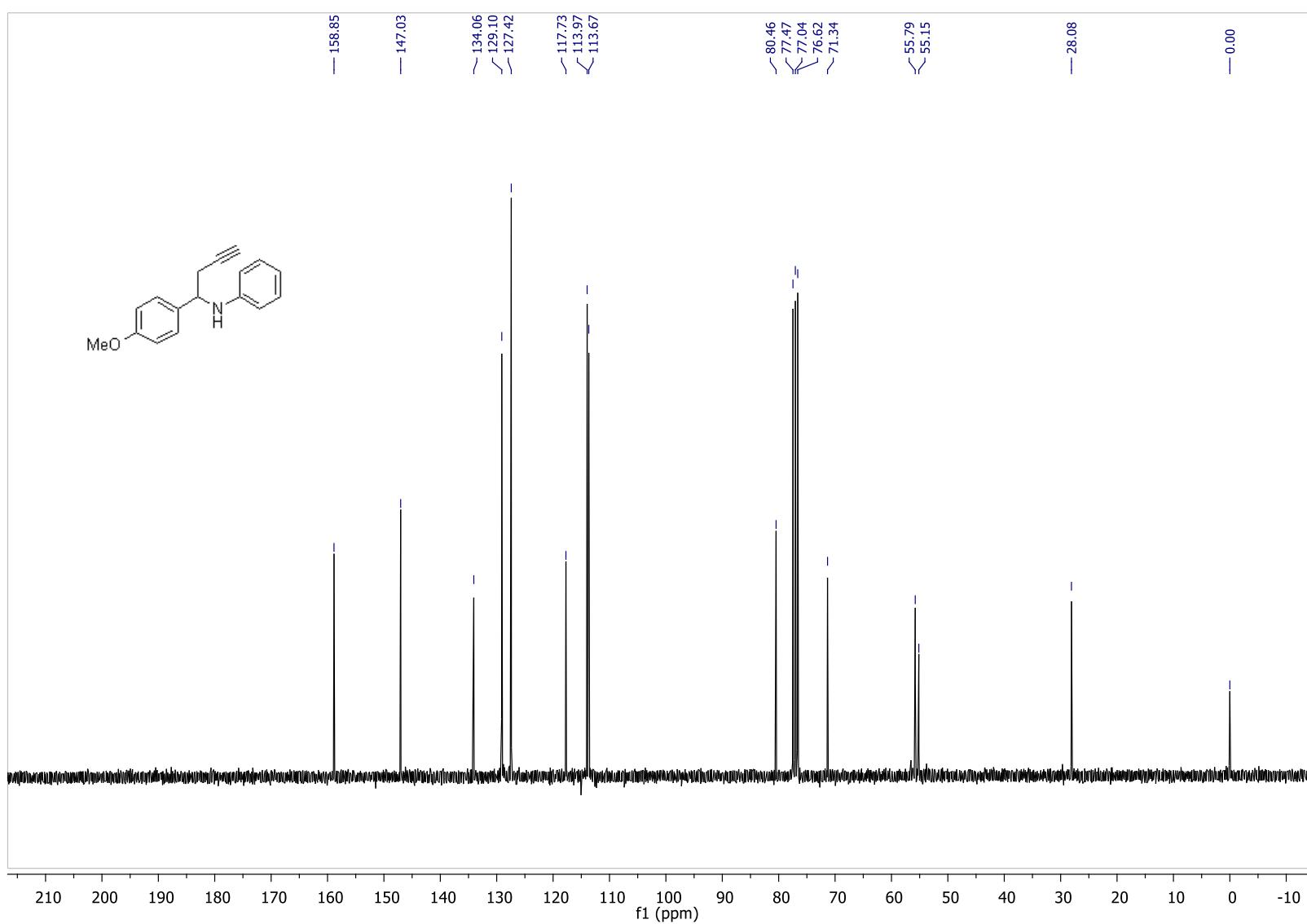
N-(1-(4-bromophenyl)but-3-yn-1-yl)aniline **1i**

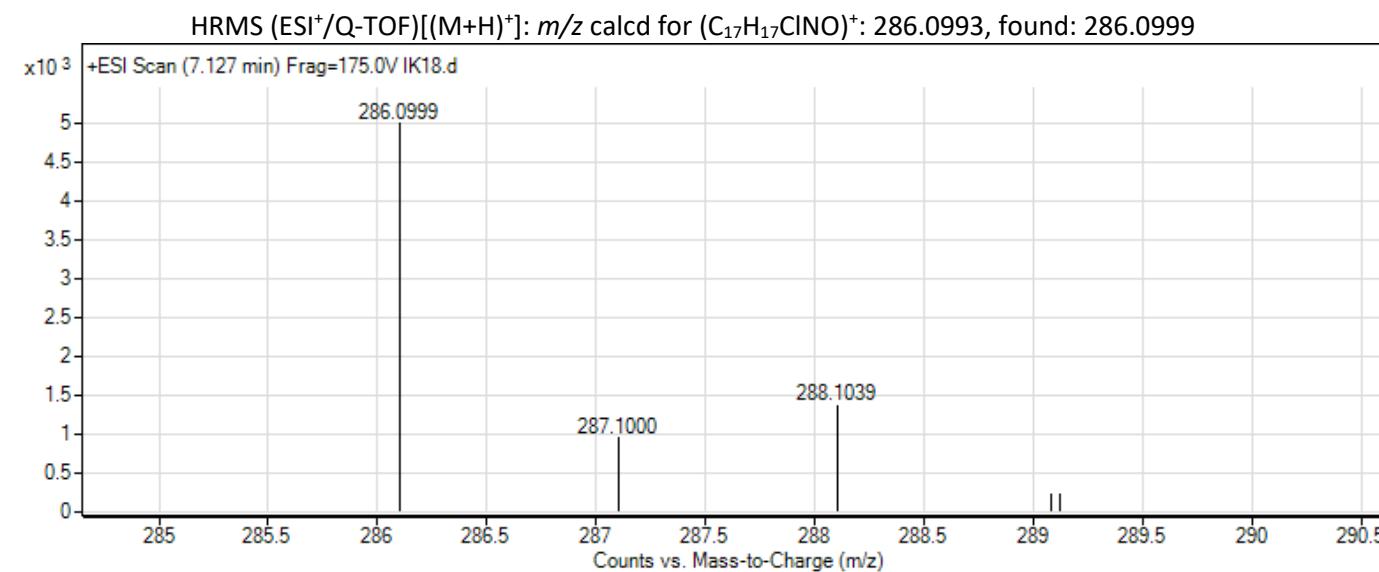
^1H NMR (CDCl_3)

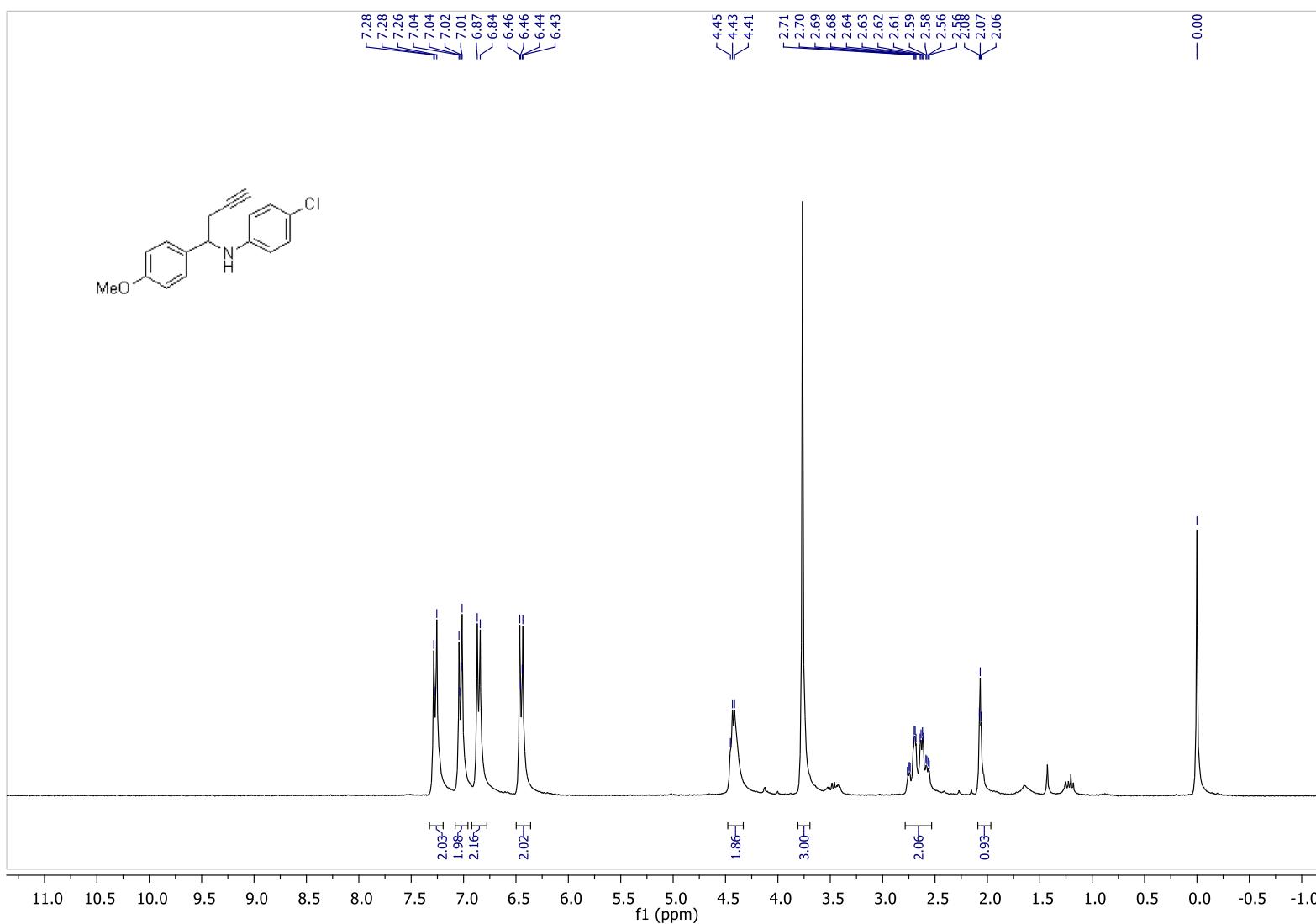
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

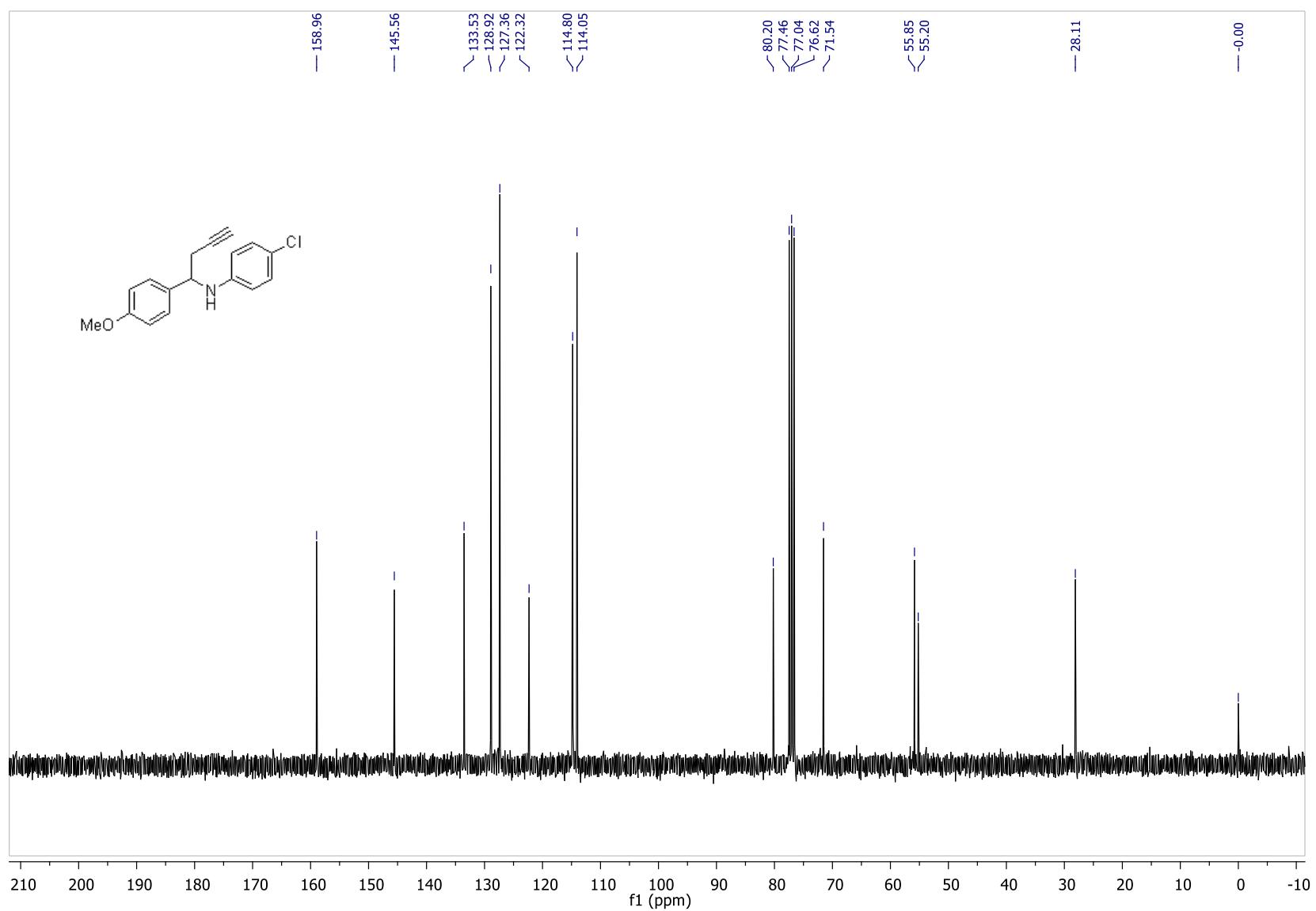
N-(1-(4-Methoxyphenyl)but-3-yn-1-yl)aniline **1j**

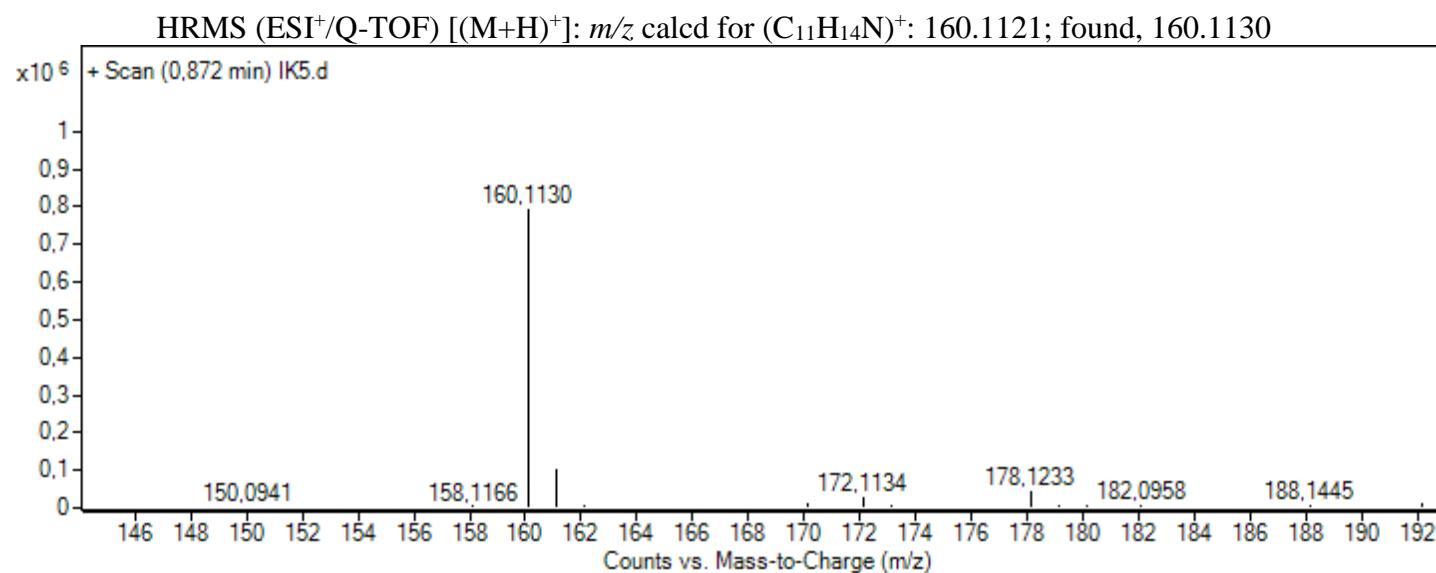
^1H NMR (CDCl_3)

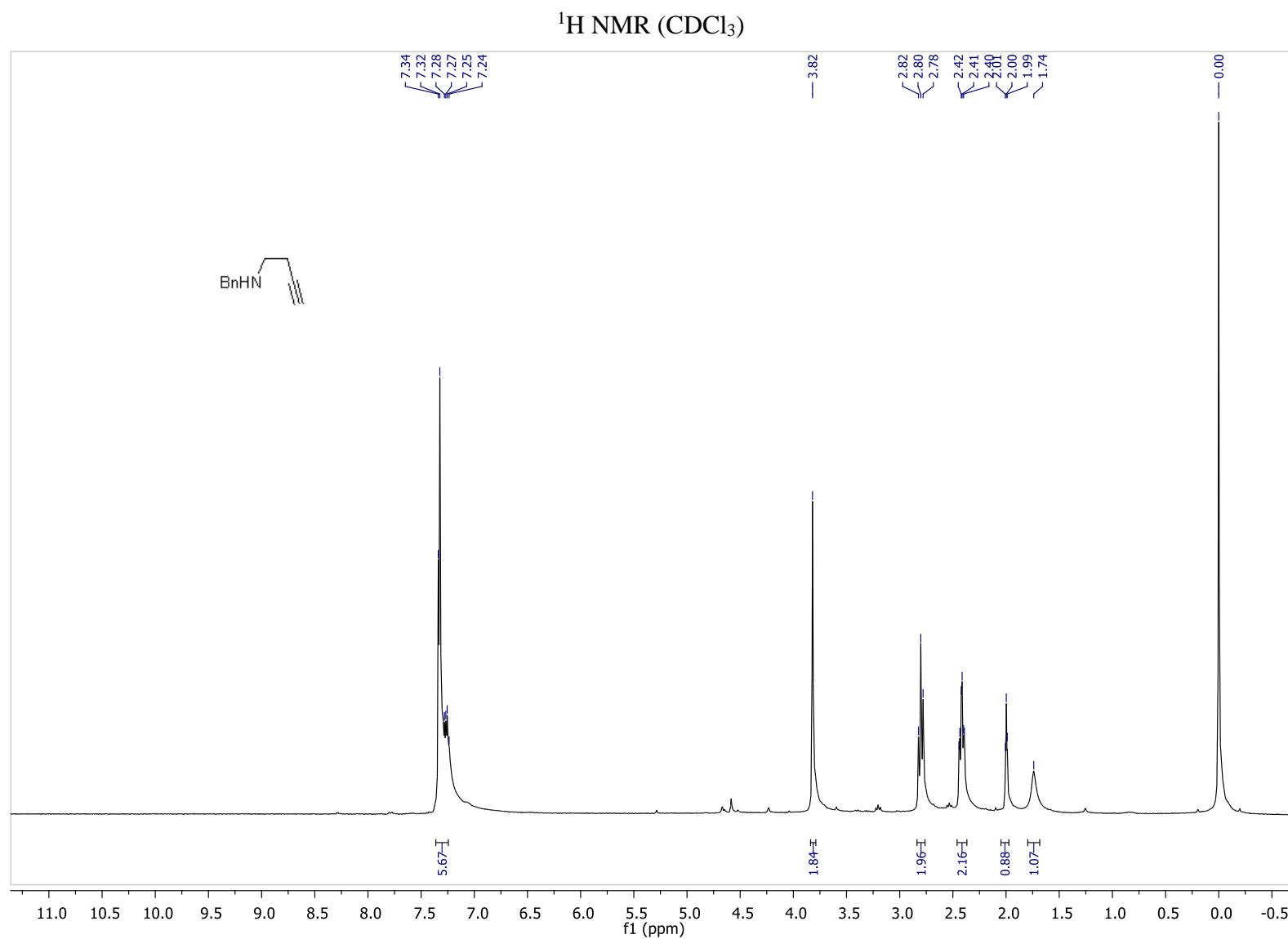
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

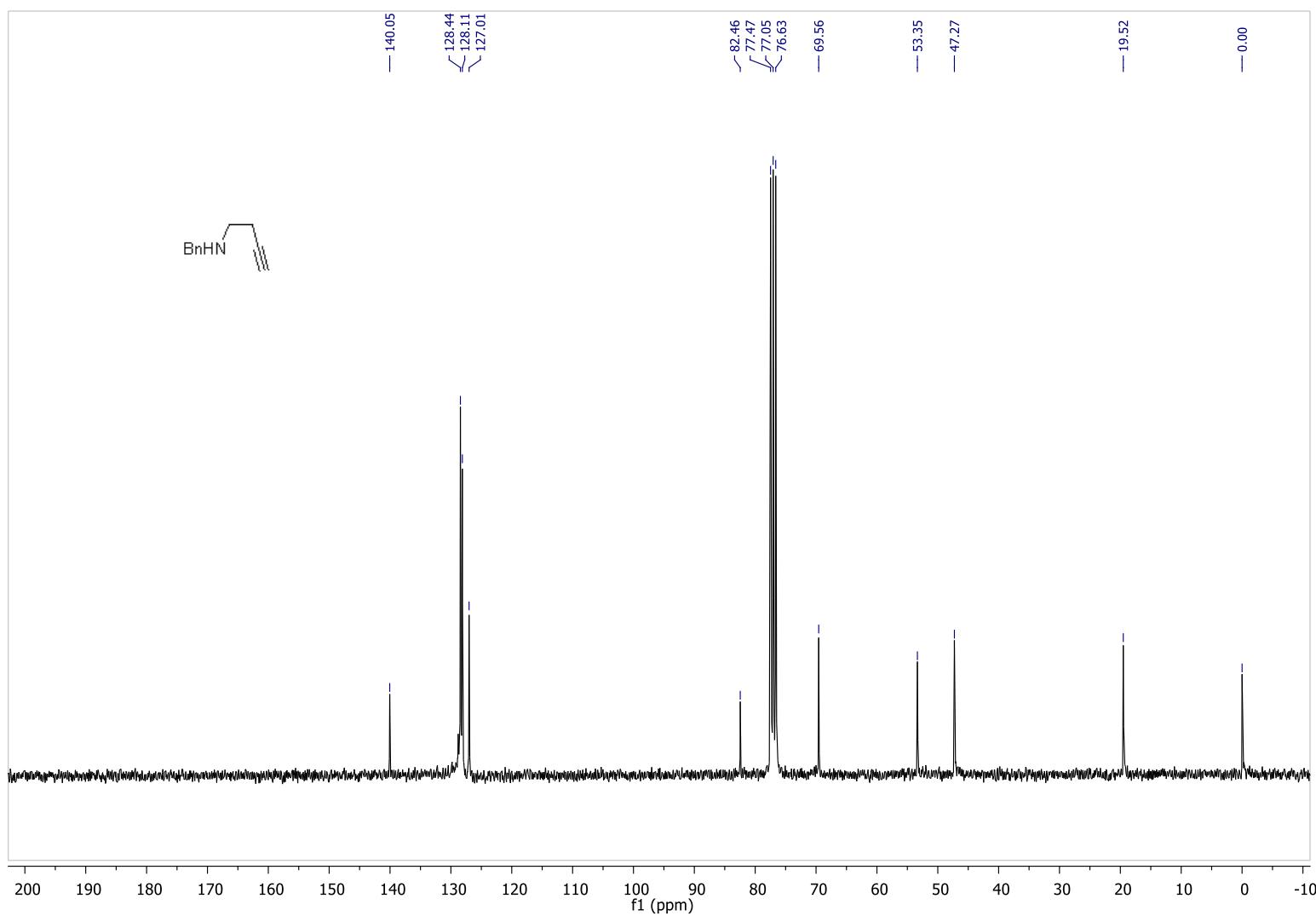
4-chloro-N-(1-(4-methoxyphenyl)but-3-yn-1-yl)aniline **1k**

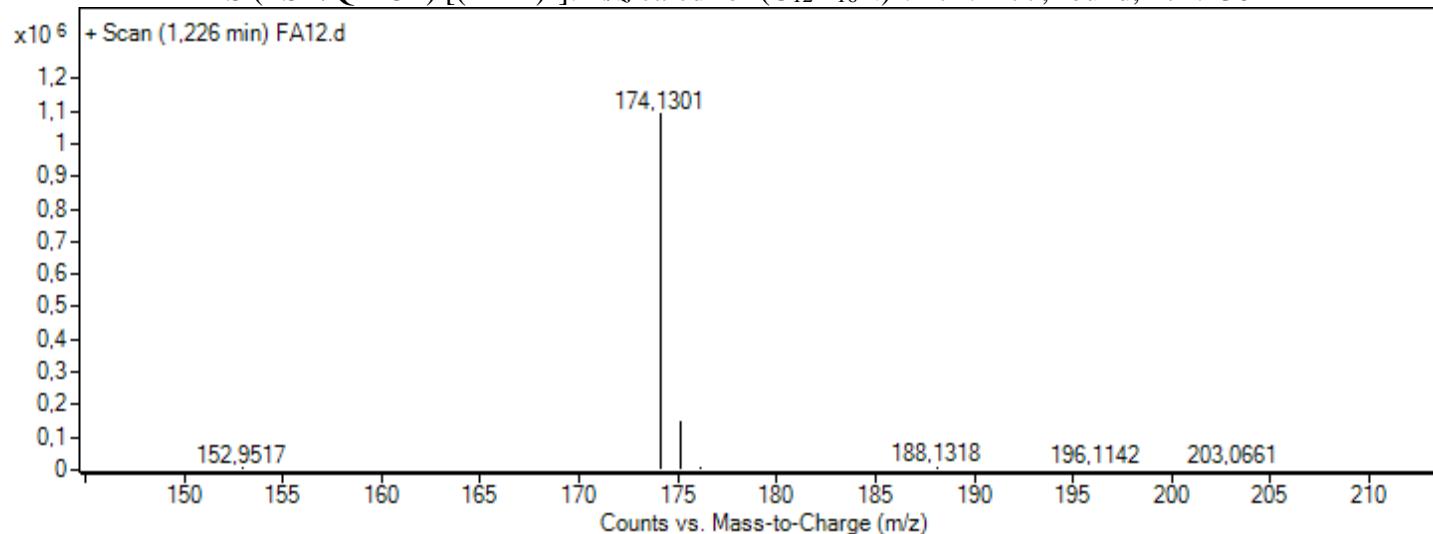
^1H NMR (CDCl_3)

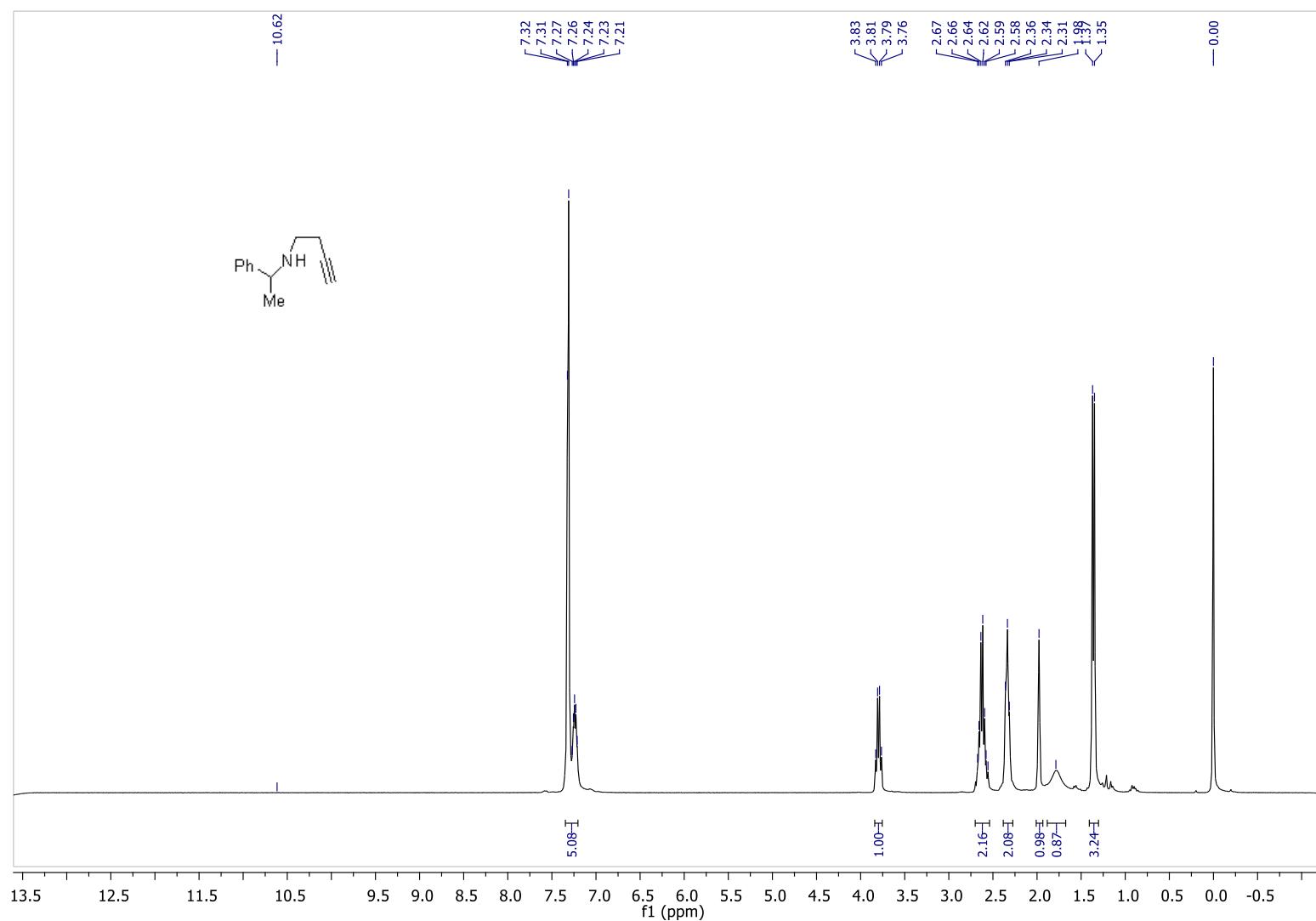
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

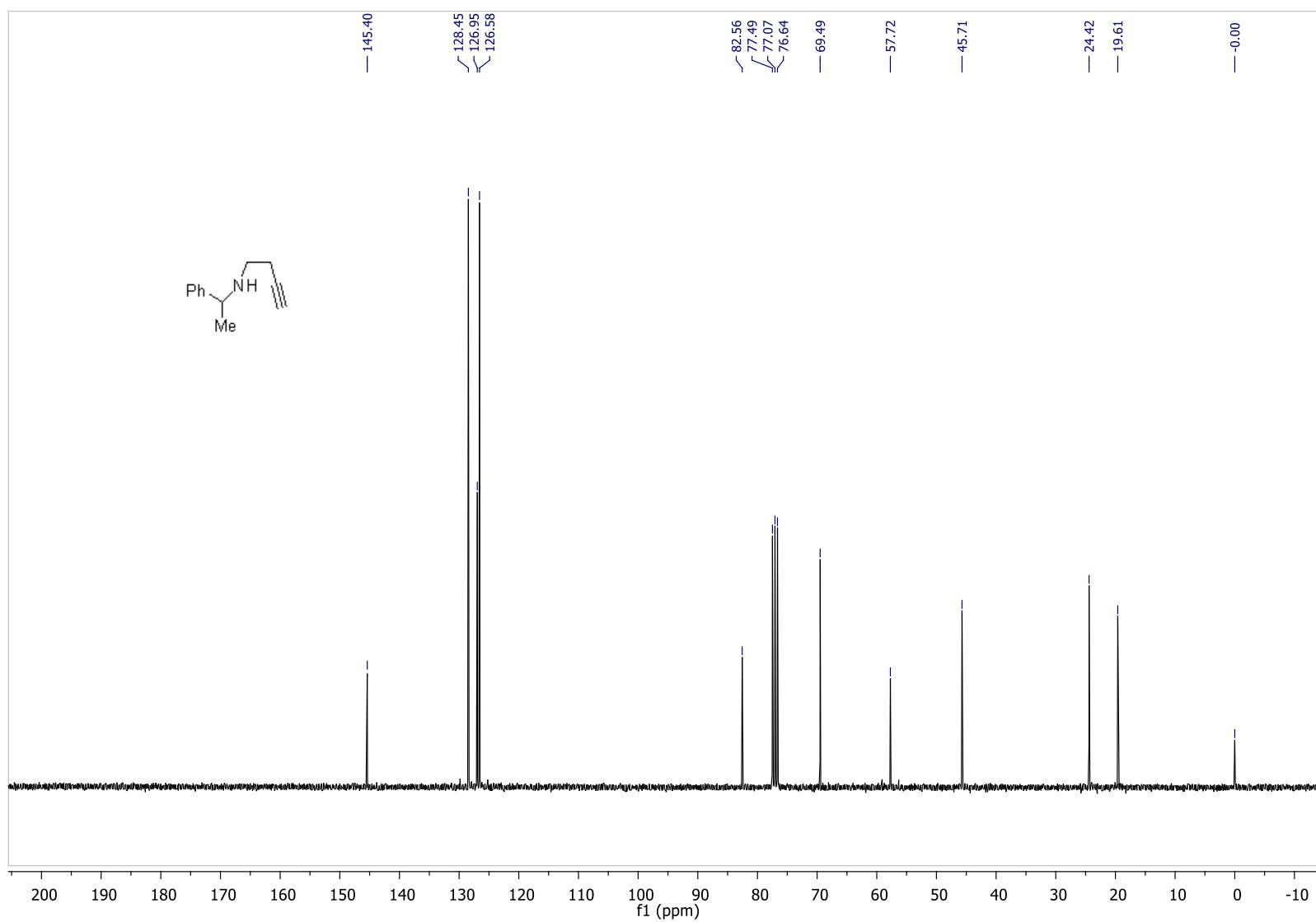
N-Benzylbut-3-yn-1-amine **11**

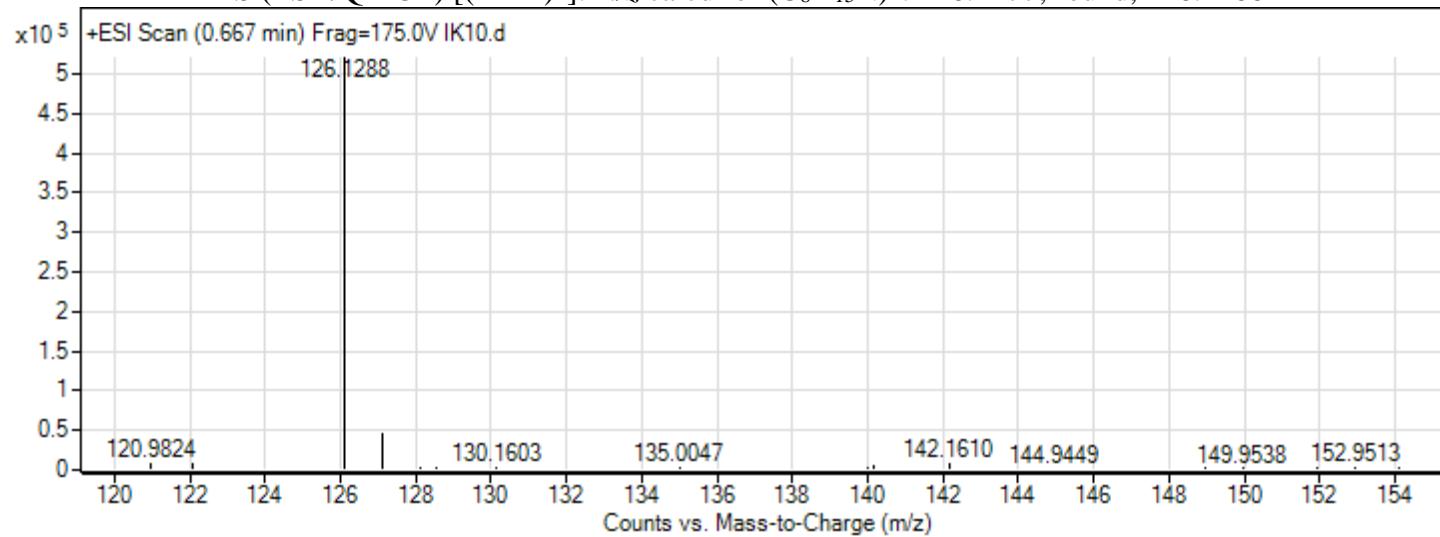


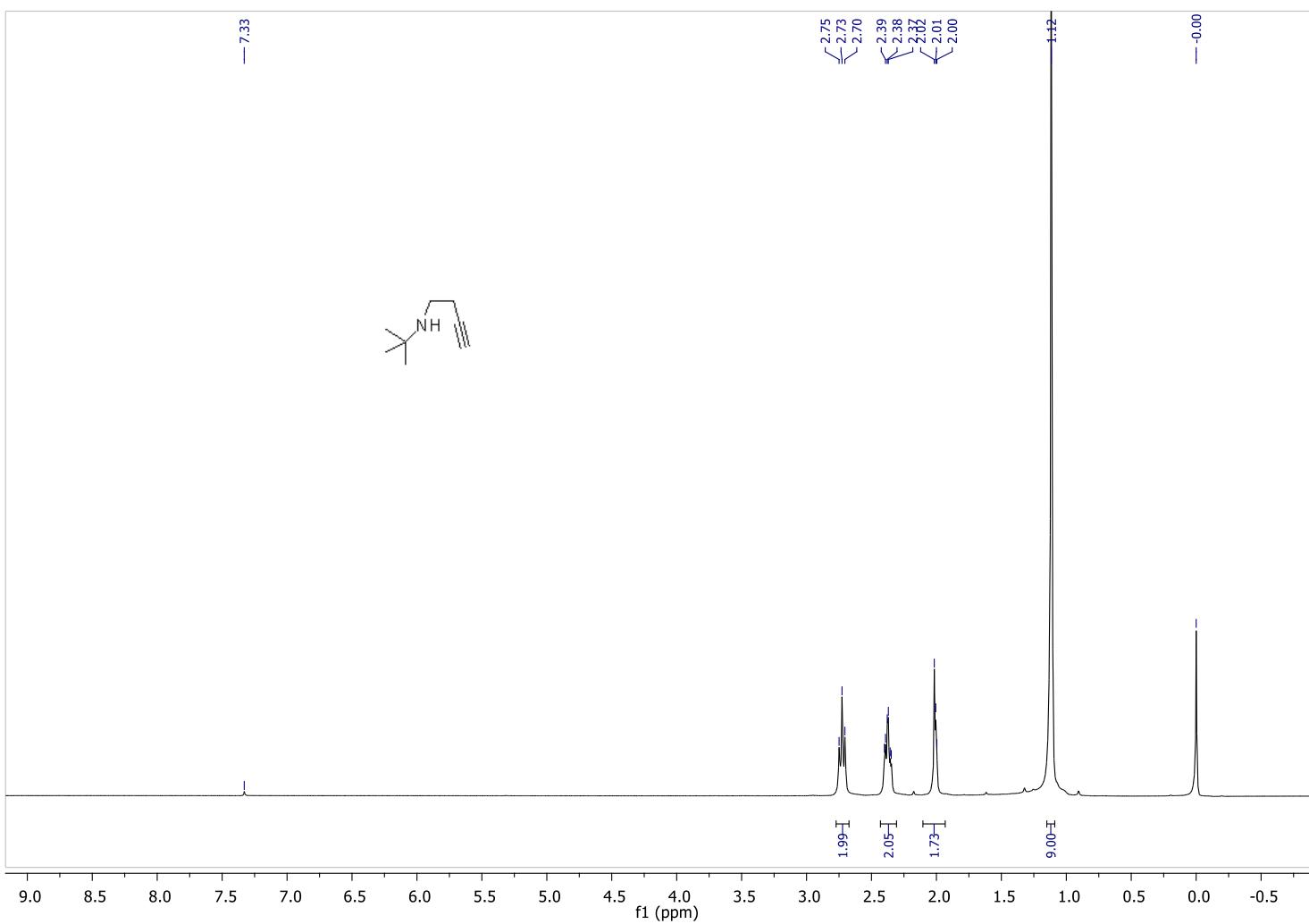
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

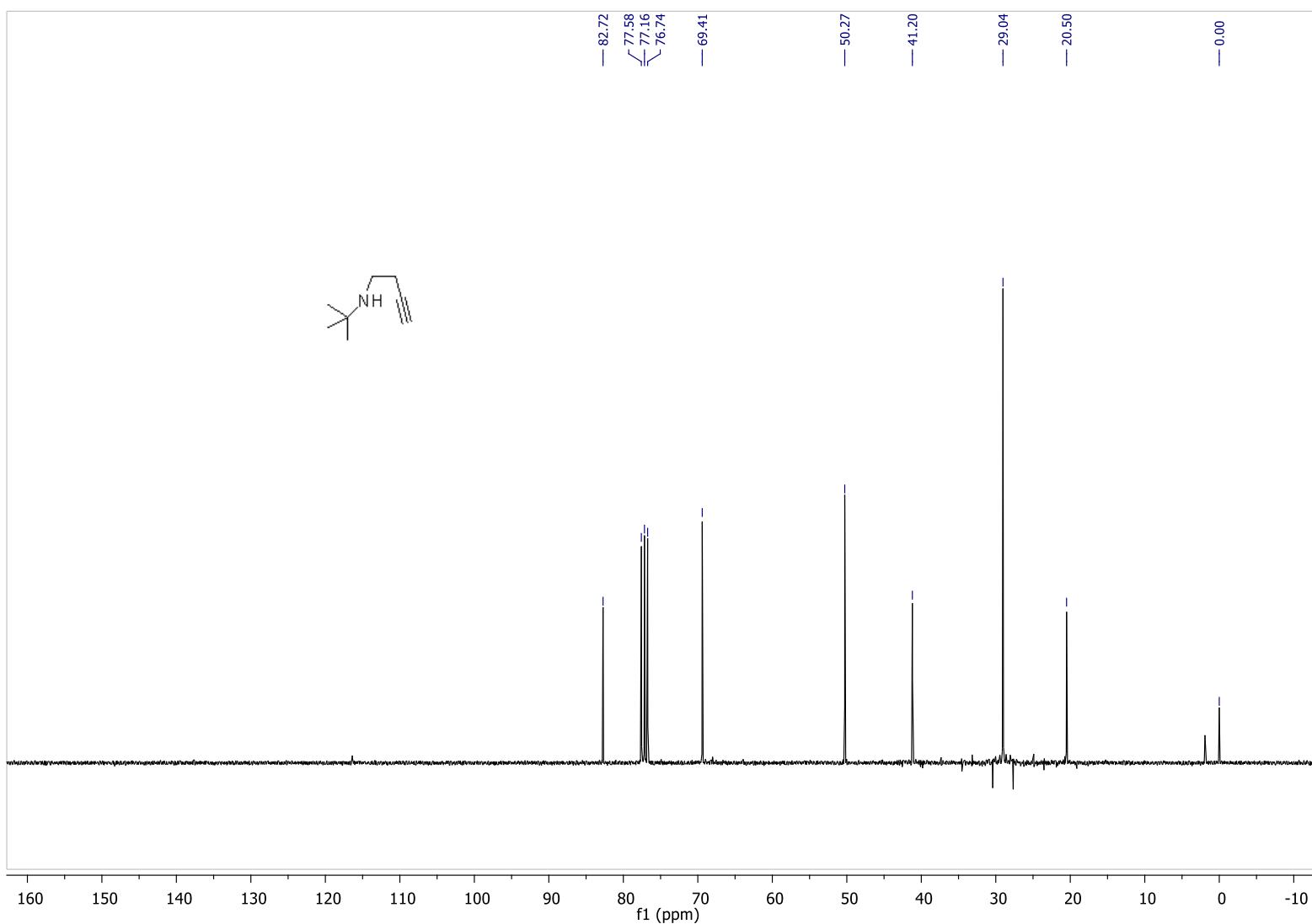
N-(1-Phenylethyl)but-3-yn-1-amine **1m**HRMS (ESI⁺/Q-TOF) [(M+H)⁺]: *m/z* calcd for (C₁₂H₁₆N)⁺: 174.1277; found, 174.1301

^1H NMR (CDCl_3)

$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

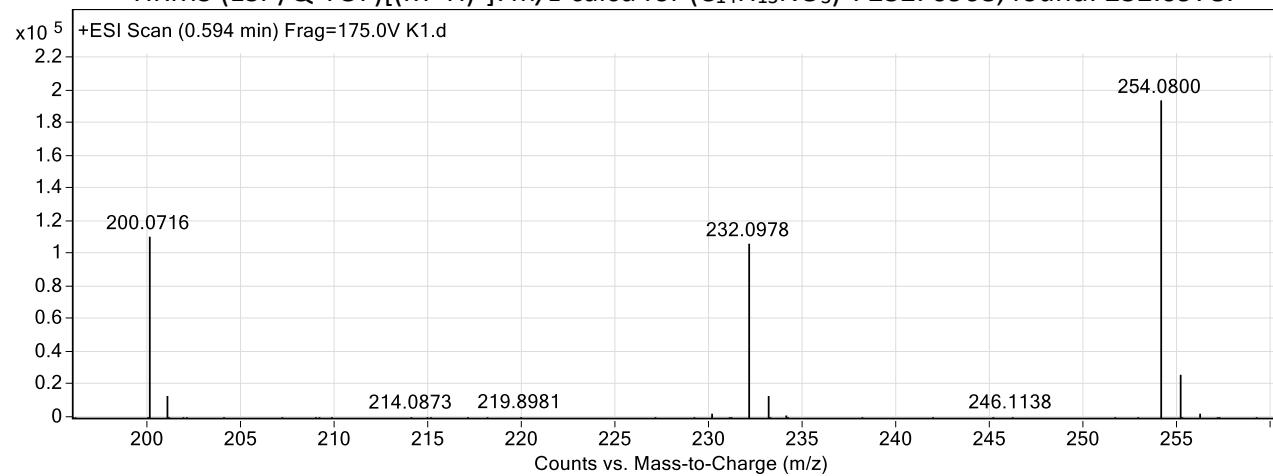
N-t-Butylbut-3-yn-1-amine **1n**HRMS (ESI⁺/Q-TOF) [(M+H)⁺]: *m/z* calcd for (C₈H₁₅N)⁺: 126.1277; found, 126.1288

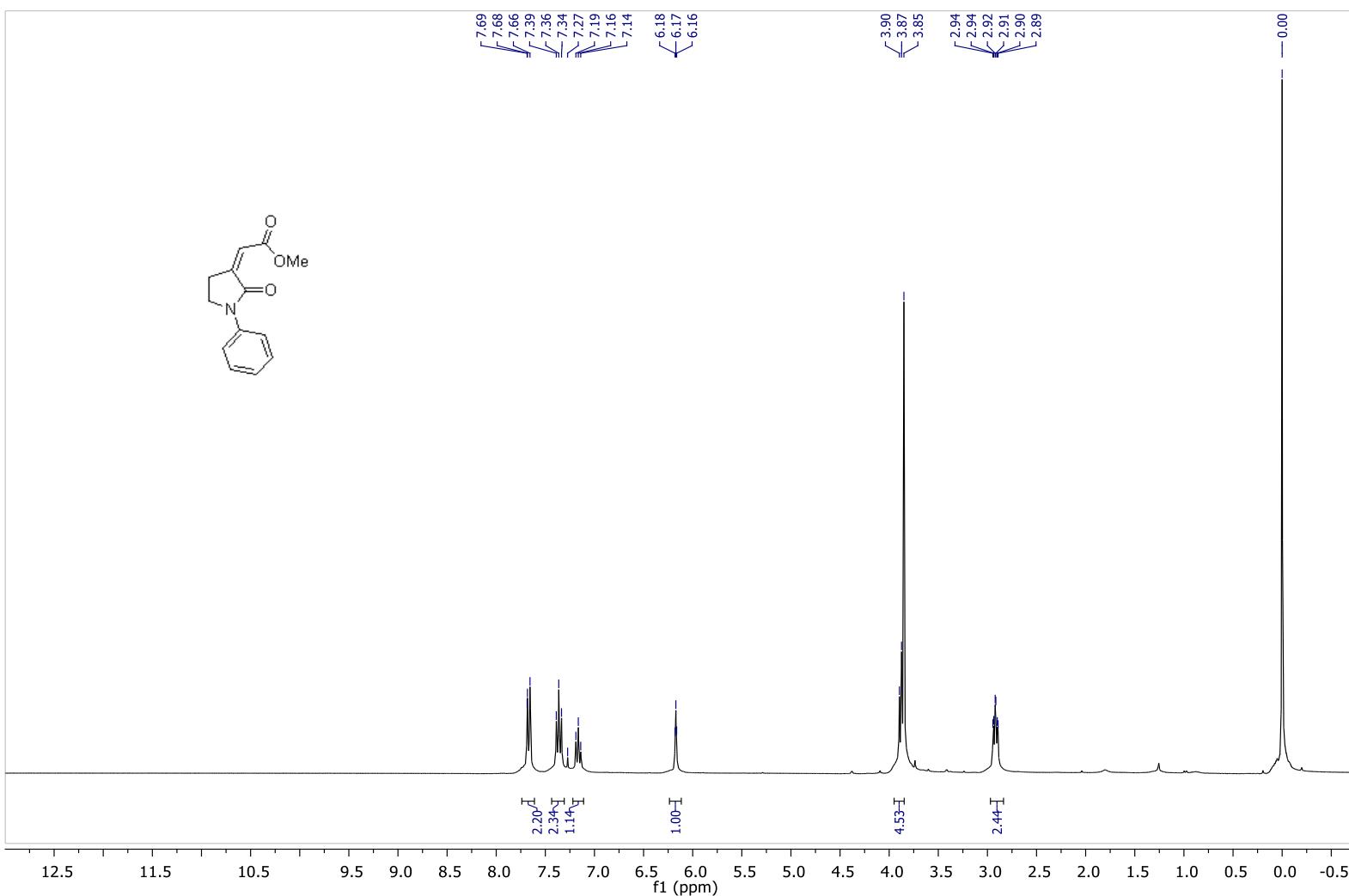
^1H NMR (CDCl_3)

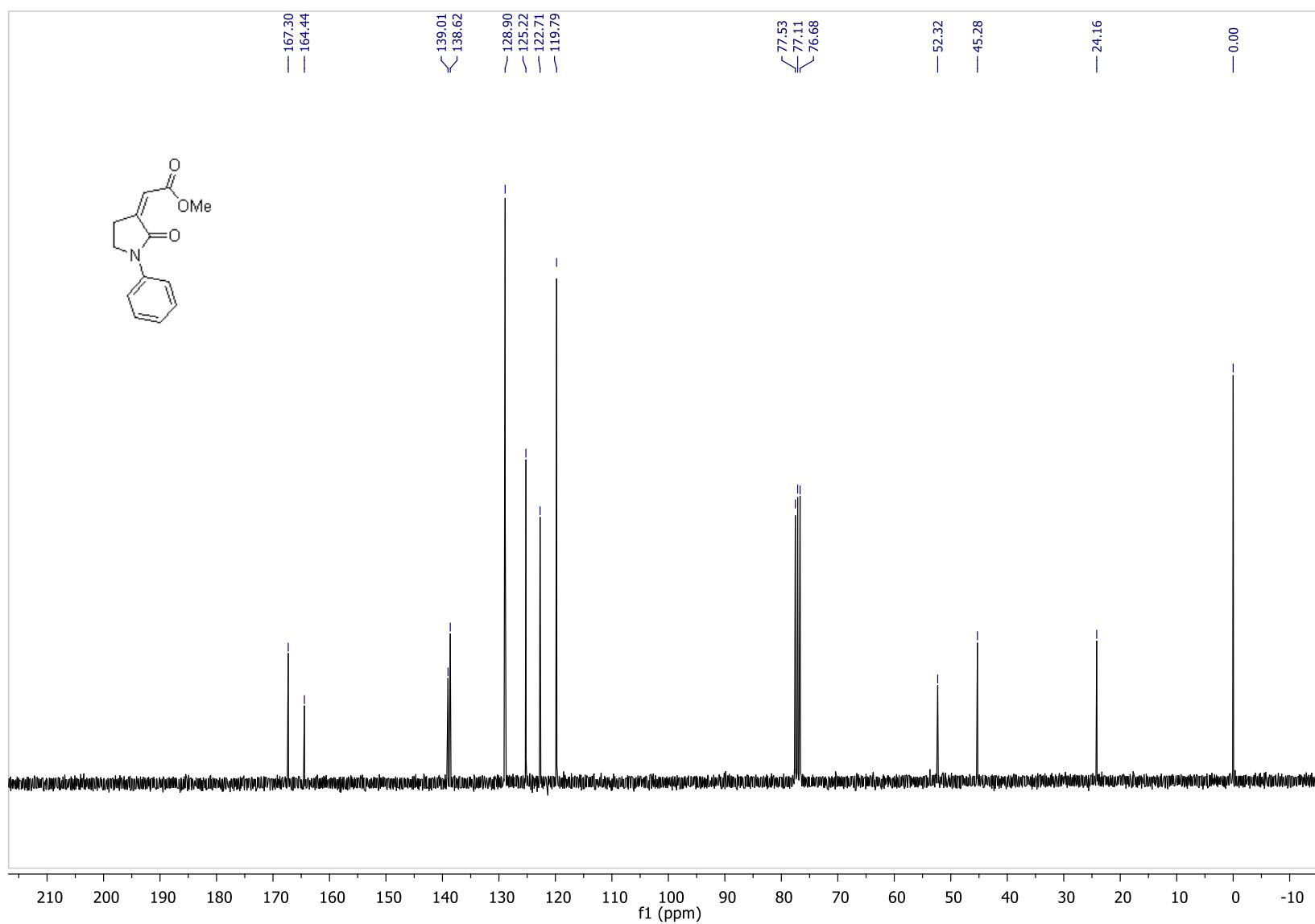
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

(Z)-Methyl 2-(2-oxo-1-phenylpyrrolidin-3-ylidene)acetate **2a**

HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₄H₁₃NO₃)⁺: 232.0968, found: 232.0978.

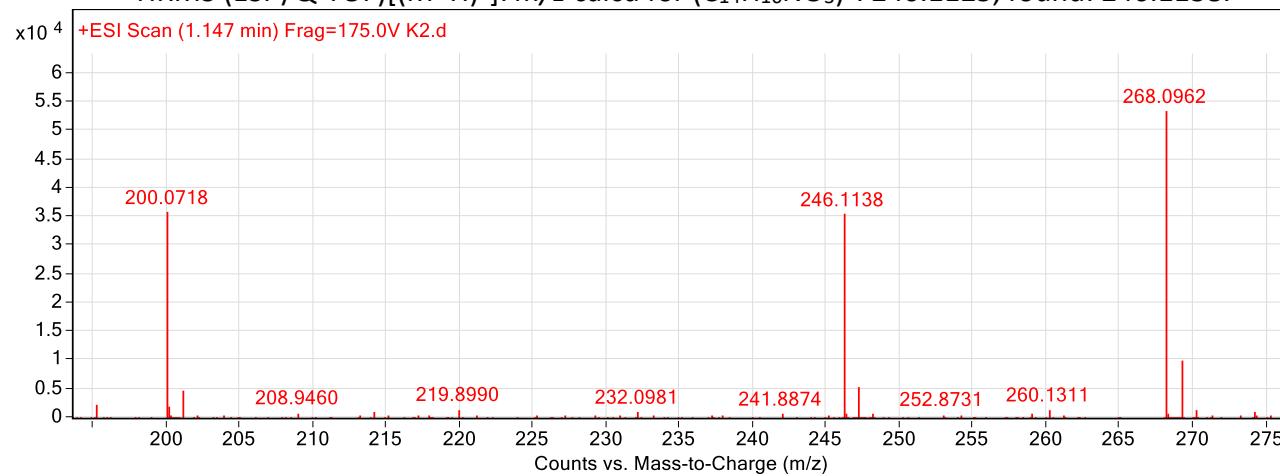


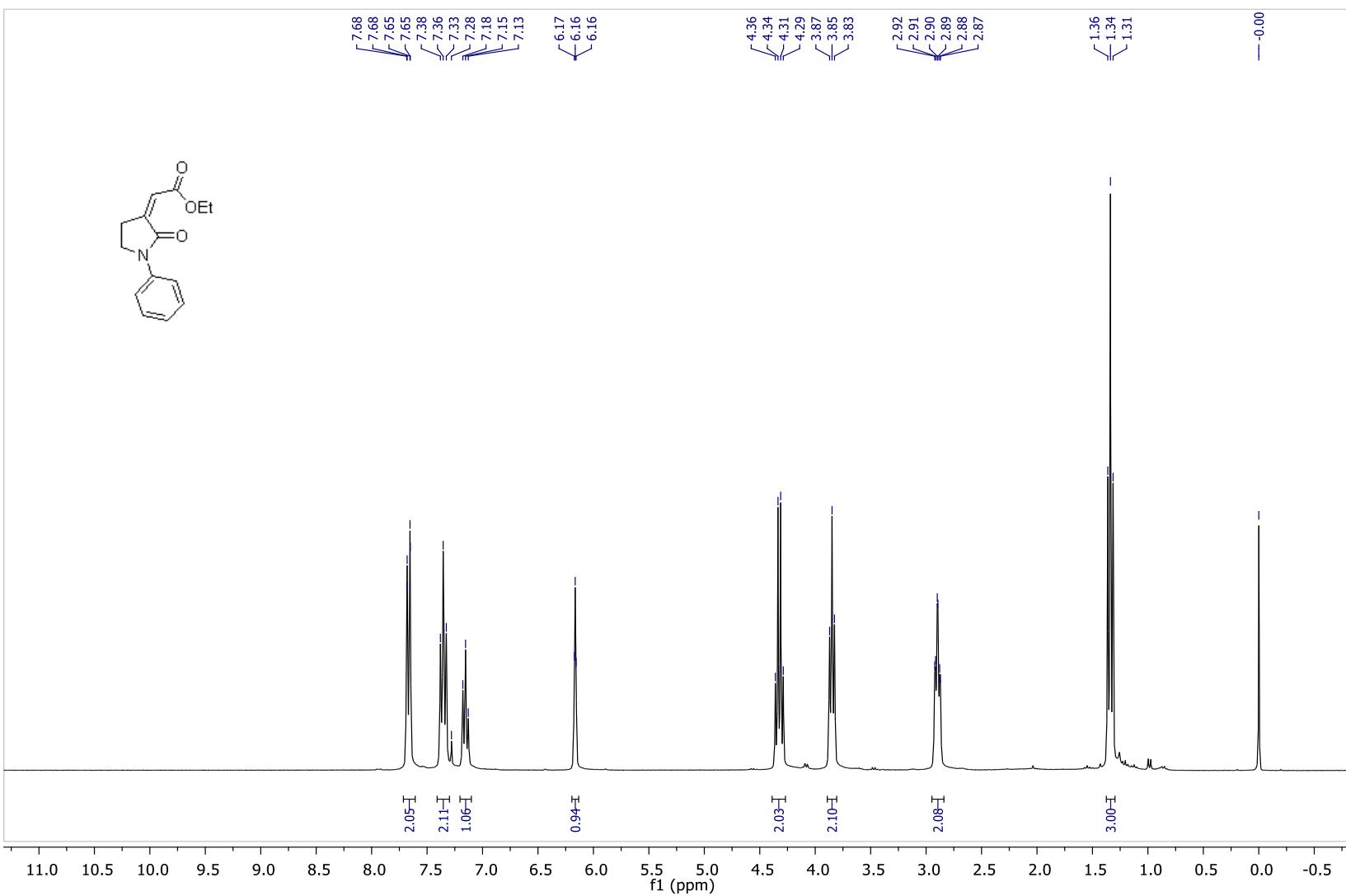
^1H NMR (CDCl_3)

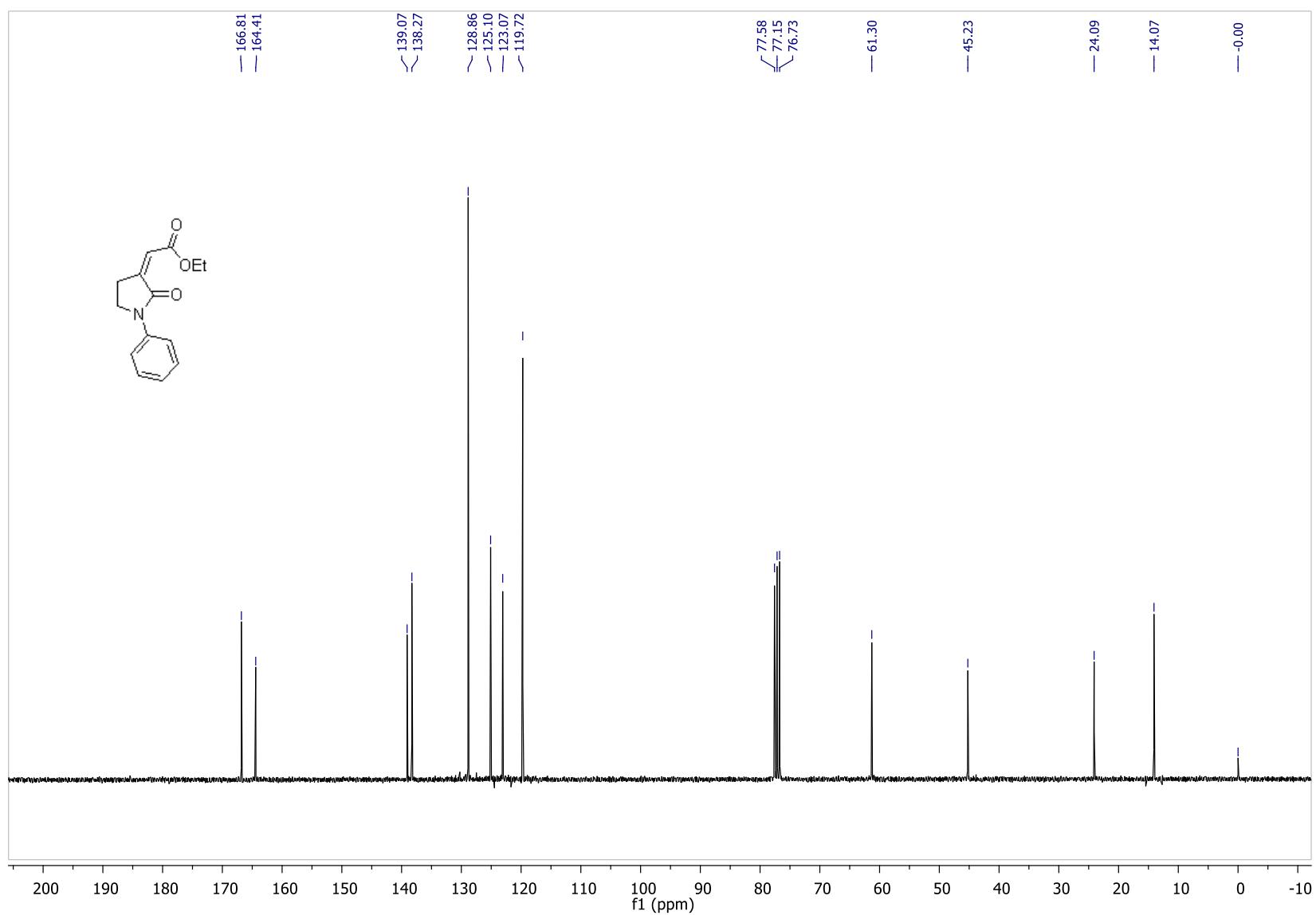
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

(Z)-Ethyl 2-(2-oxo-1-phenylpyrrolidin-3-ylidene)acetate **2a'**

HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₄H₁₆NO₃)⁺: 246.1125, found: 246.1138.

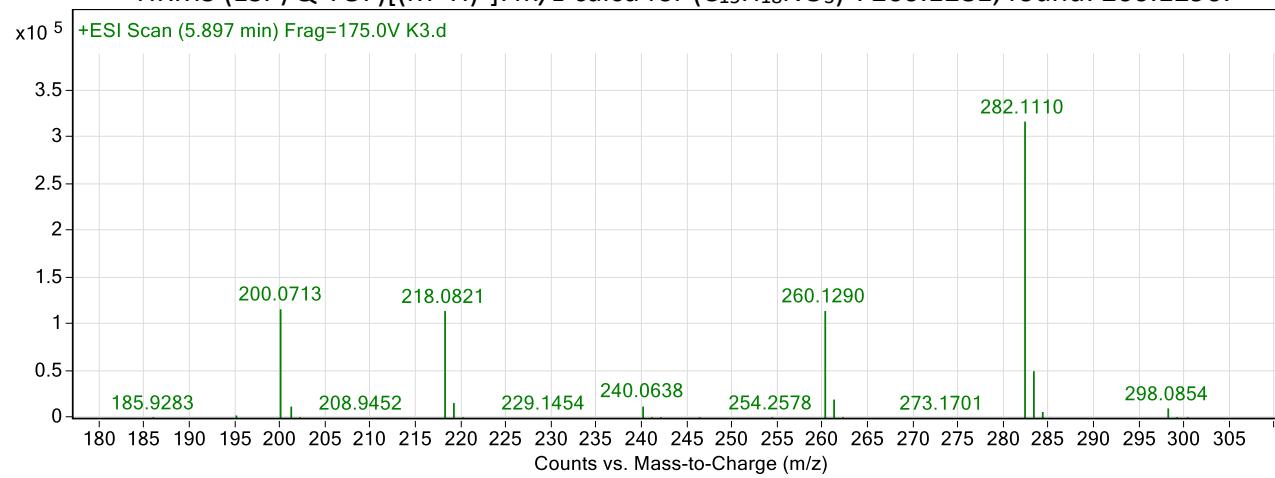


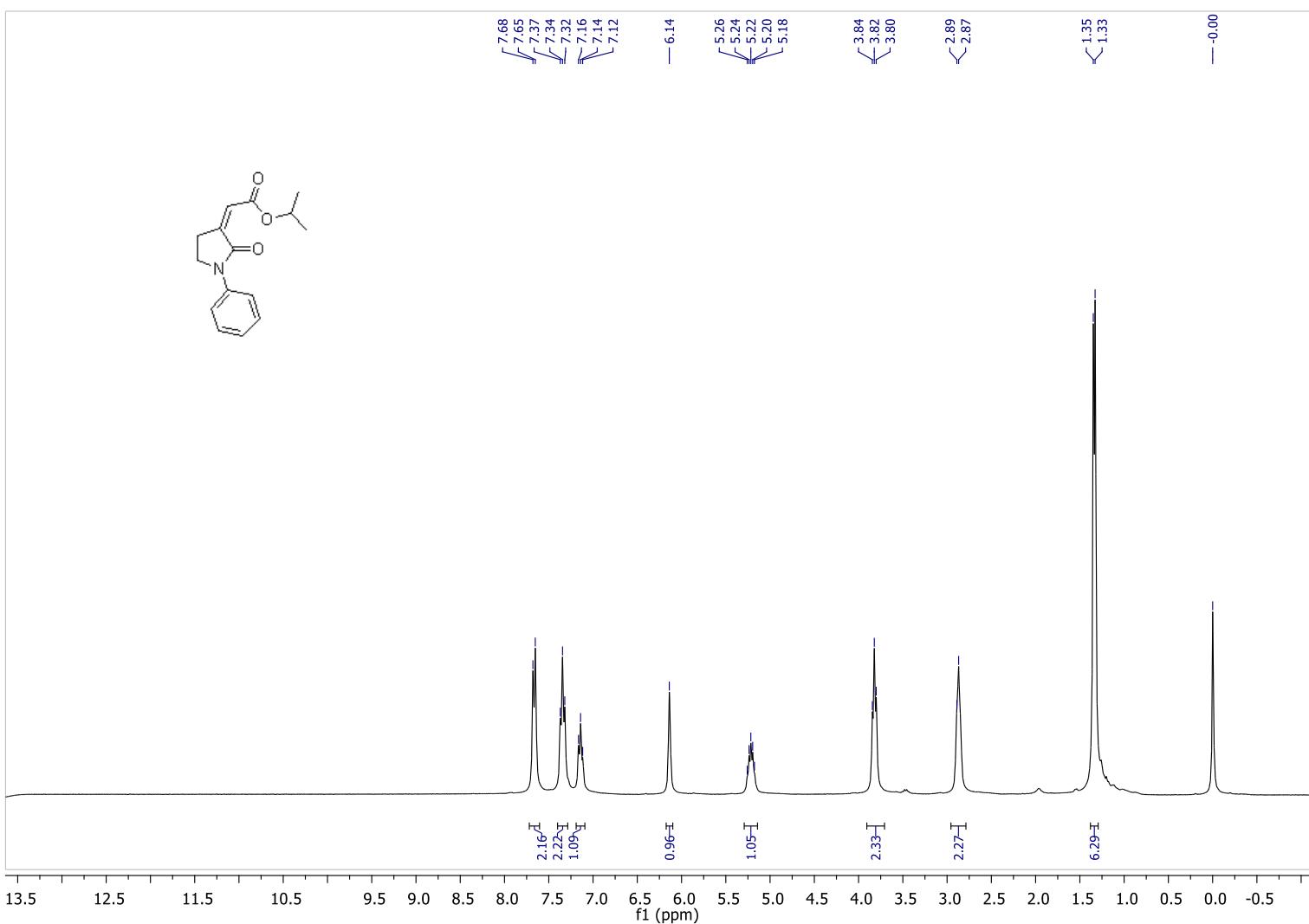
^1H NMR (CDCl_3)

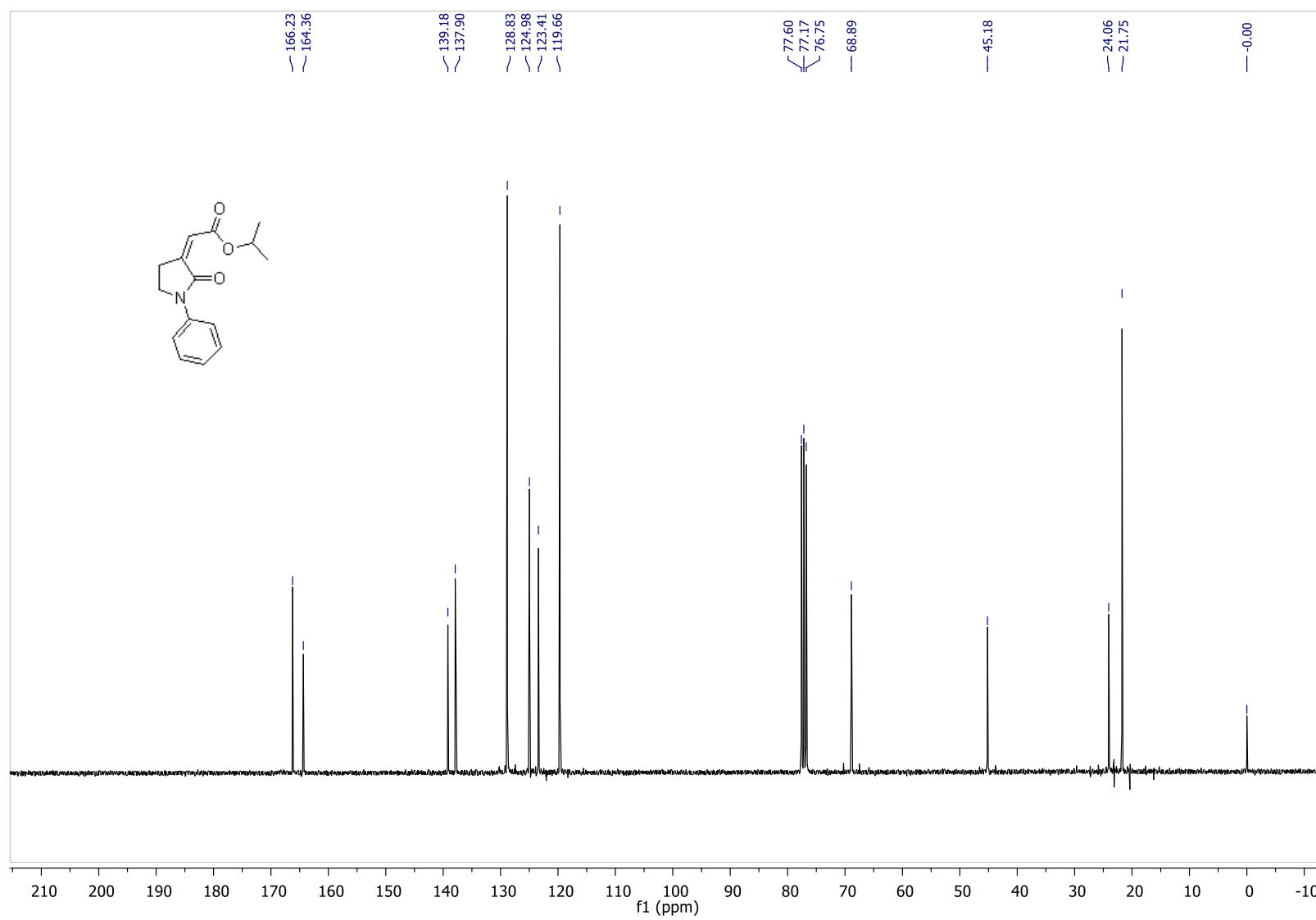
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

(Z)-Isopropyl 2-(2-oxo-1-phenylpyrrolidin-3-ylidene)acetate **2a''**

HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₅H₁₈NO₃)⁺: 260.1281, found: 260.1290.

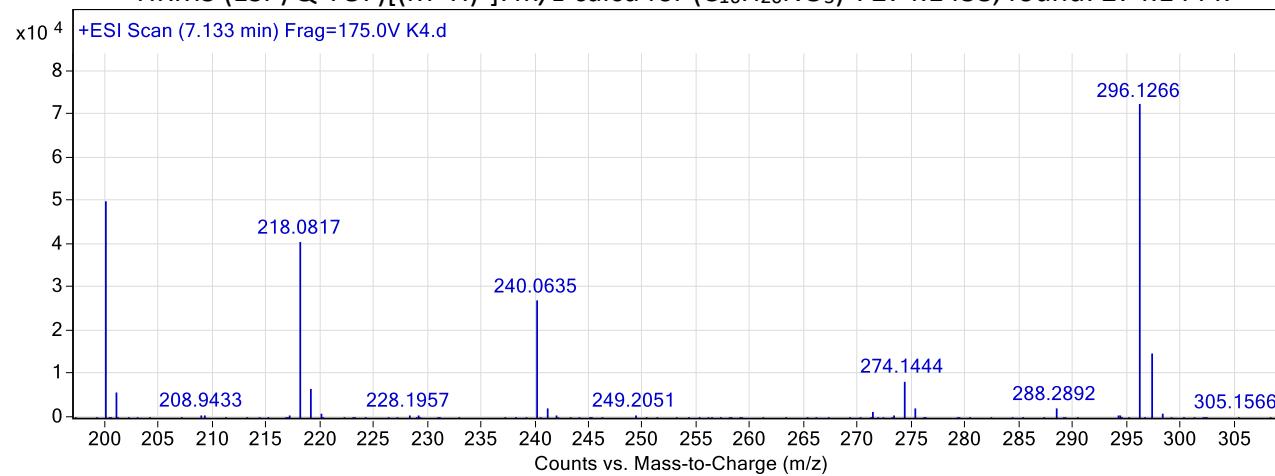


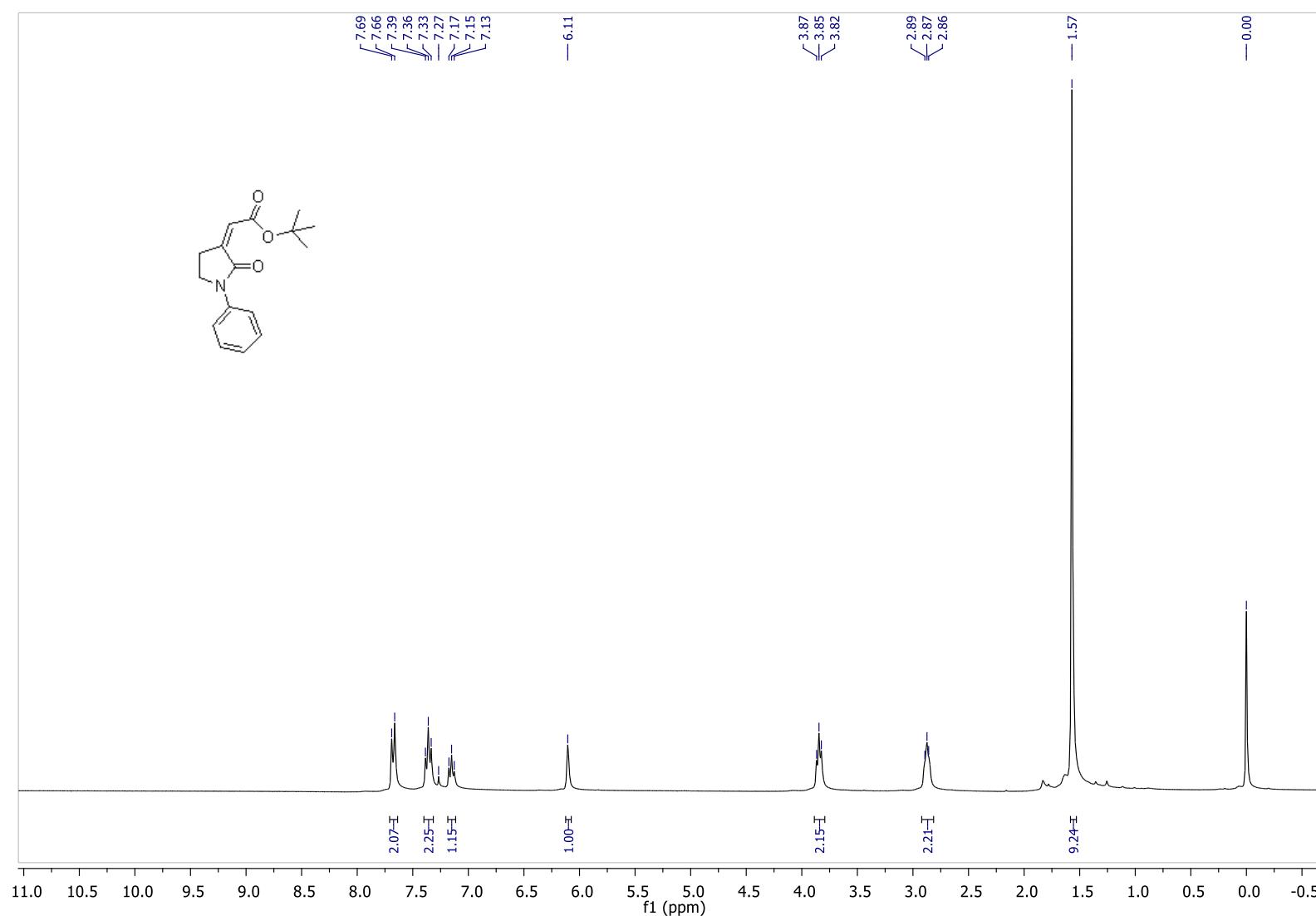
^1H NMR (CDCl_3)

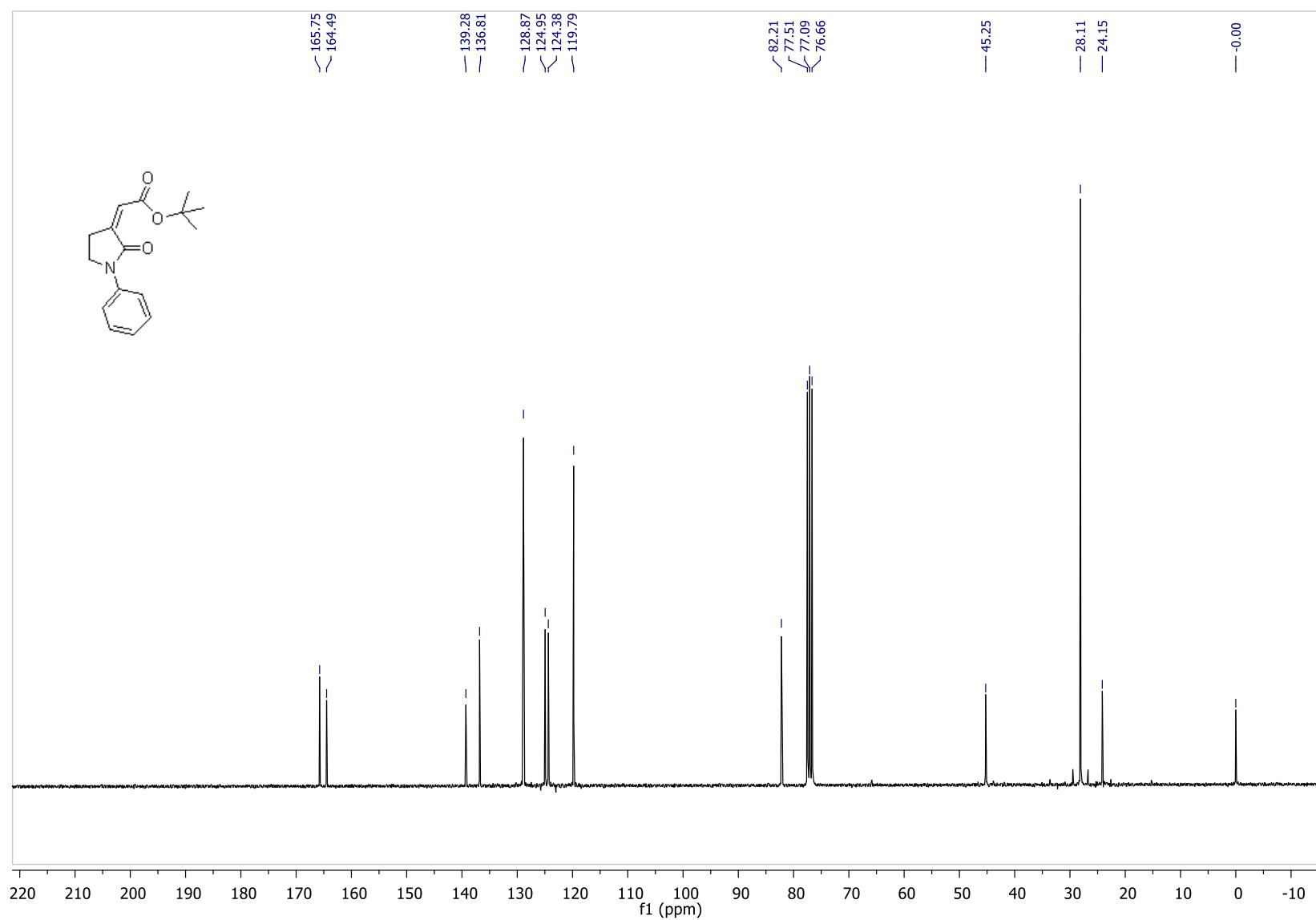
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

(Z)-*tert*-Butyl 2-(2-oxo-1-phenylpyrrolidin-3-ylidene)acetate **2a''**

HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₆H₂₀NO₃)⁺: 274.1438, found: 274.1444.

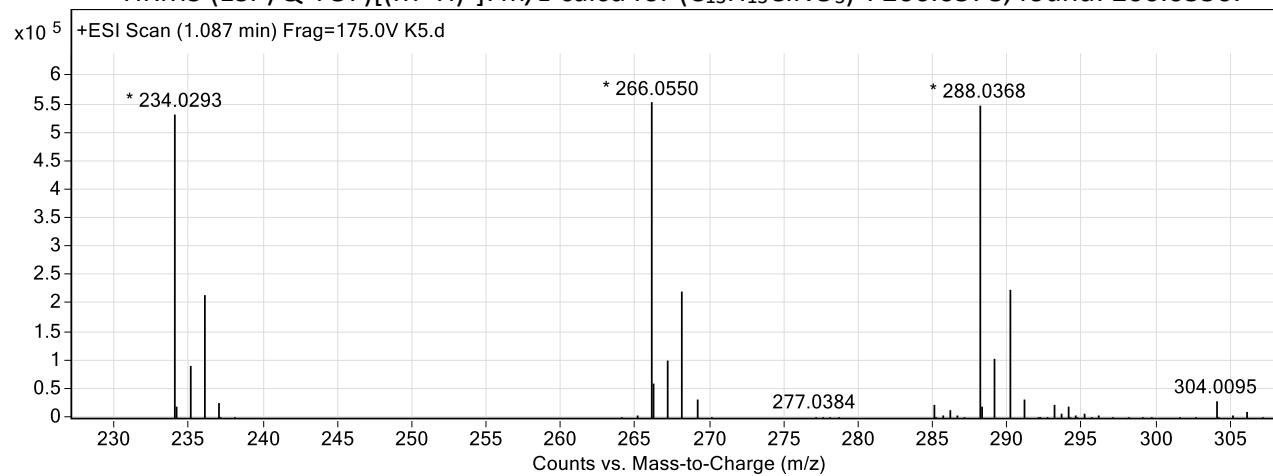


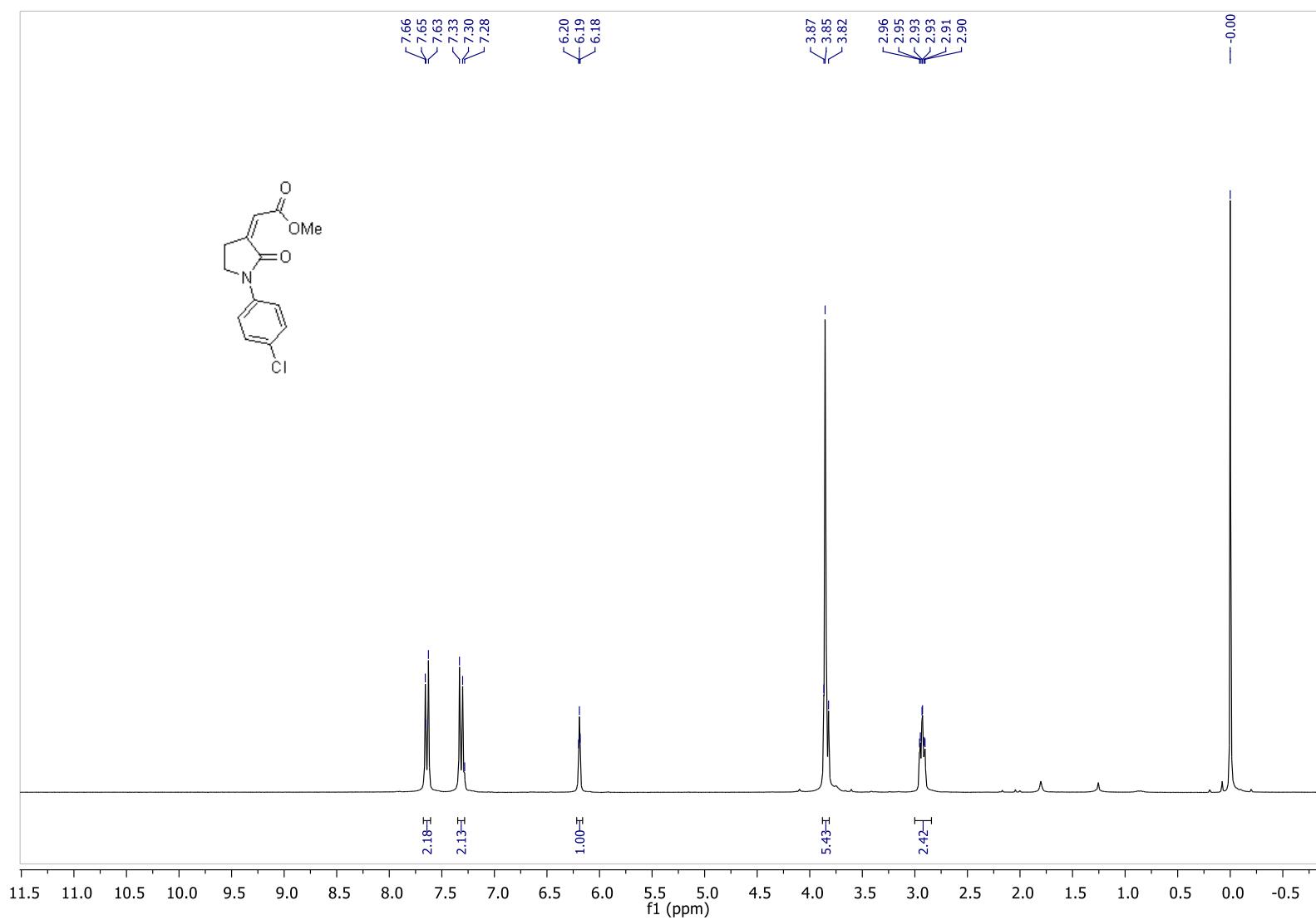
^1H NMR (CDCl_3)

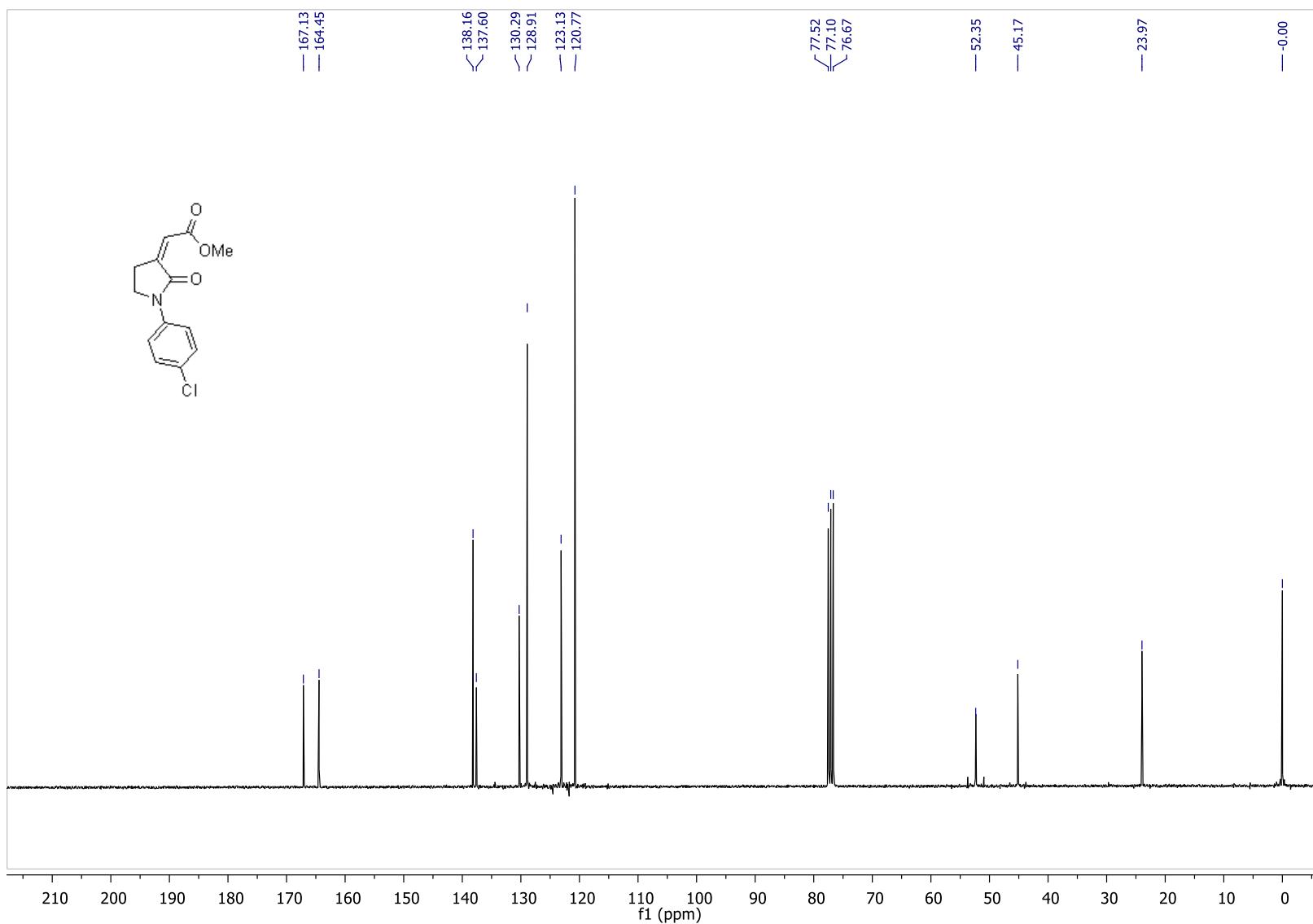
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

(Z)-Methyl 2-(1-(4-chlorophenyl)-2-oxopyrrolidin-3-ylidene)acetate **2b**

HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₃H₁₃ClNO₃)⁺: 266.0578, found: 266.0550.

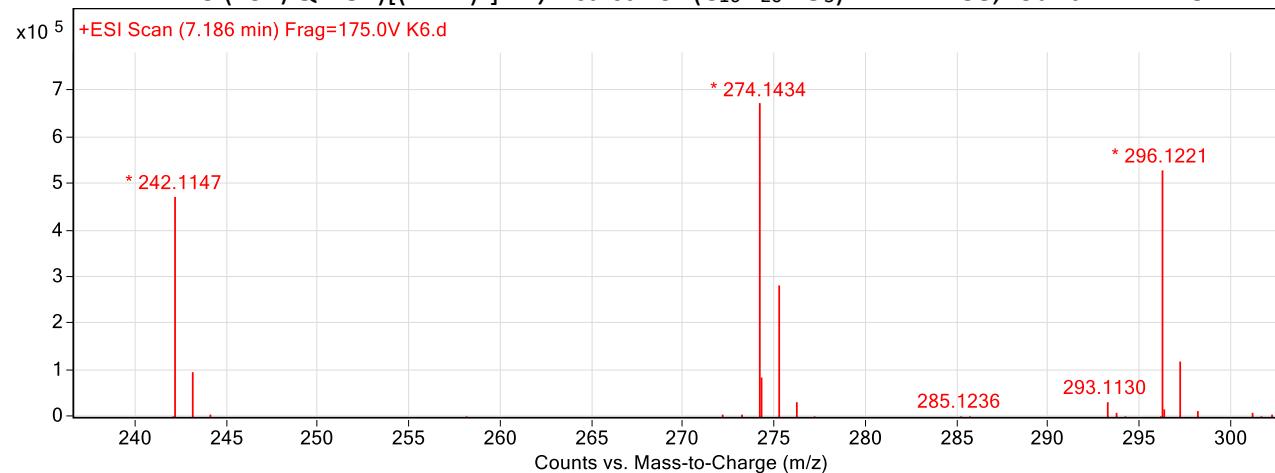


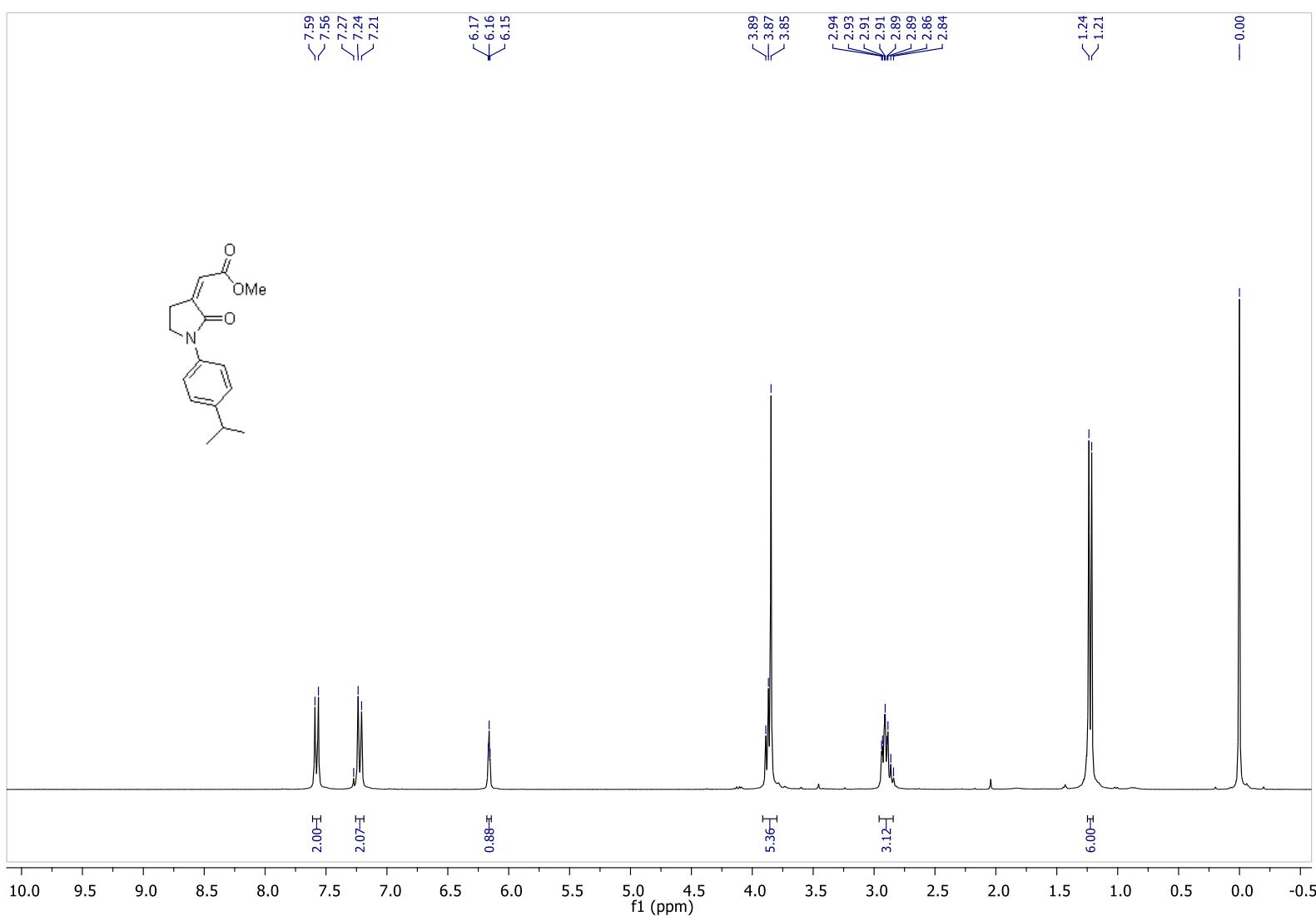
^1H NMR (CDCl_3)

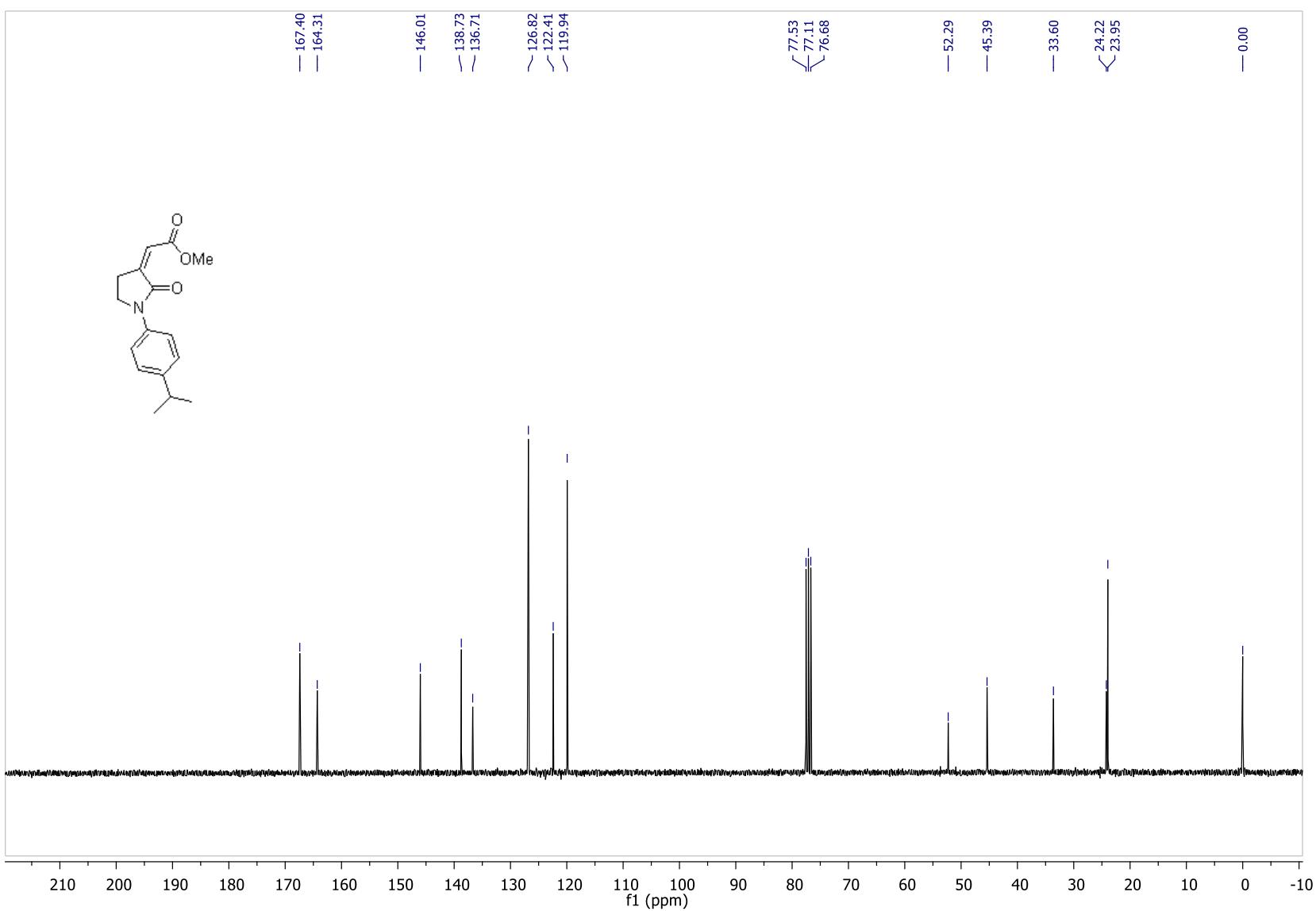
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

(Z)-Methyl 2-(1-(4-isopropylphenyl)-2-oxopyrrolidin-3-ylidene)acetate **2c**

HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₆H₂₀NO₃)⁺: 274.1438, found: 274.1434.

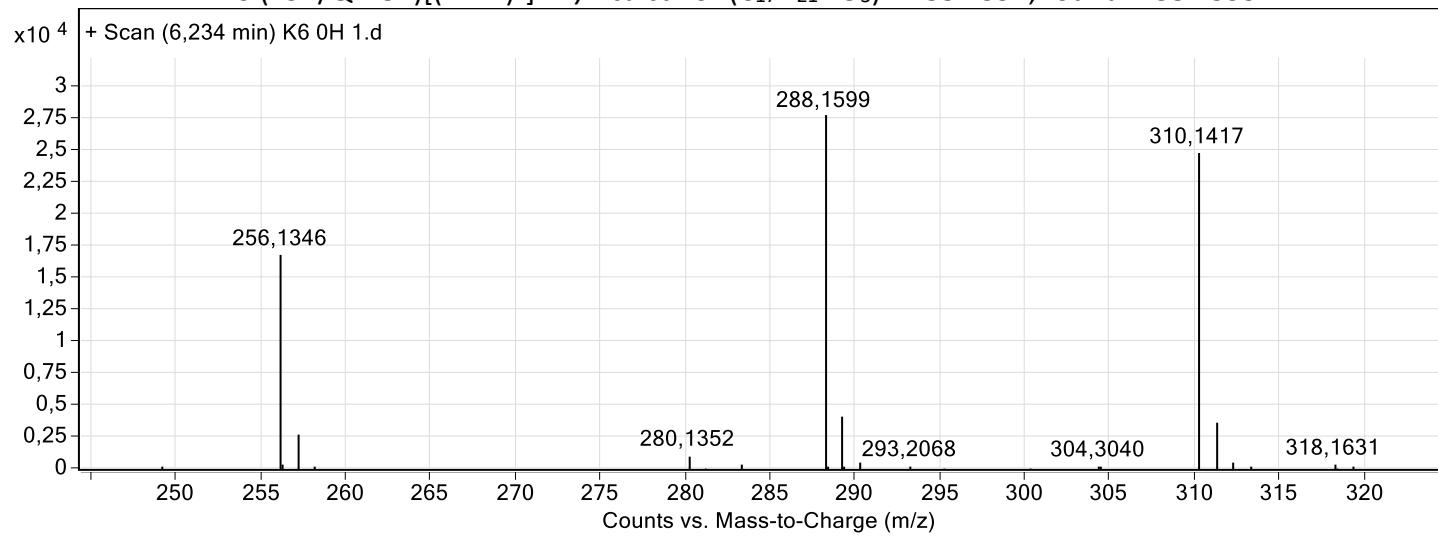


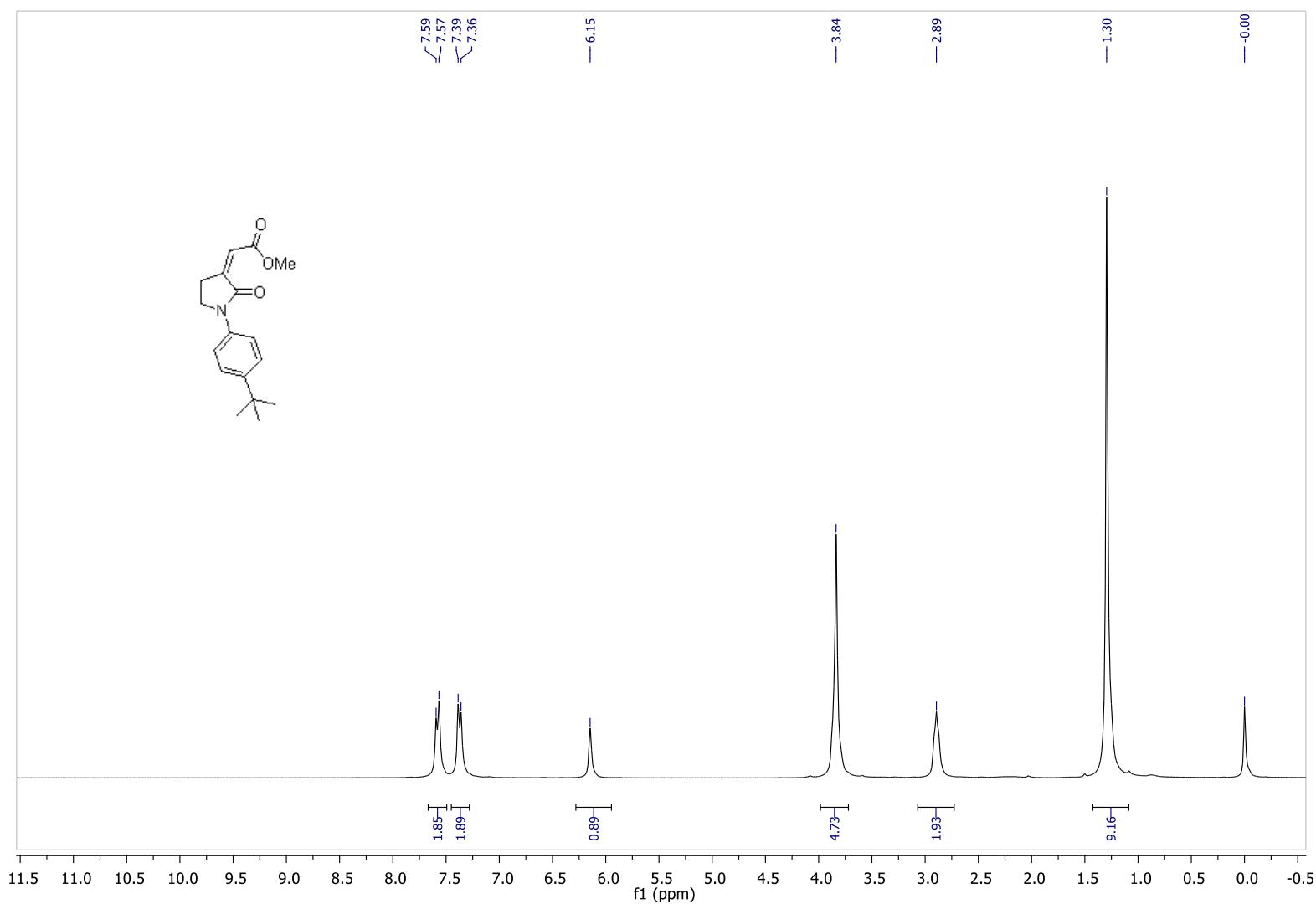
^1H NMR (CDCl_3)

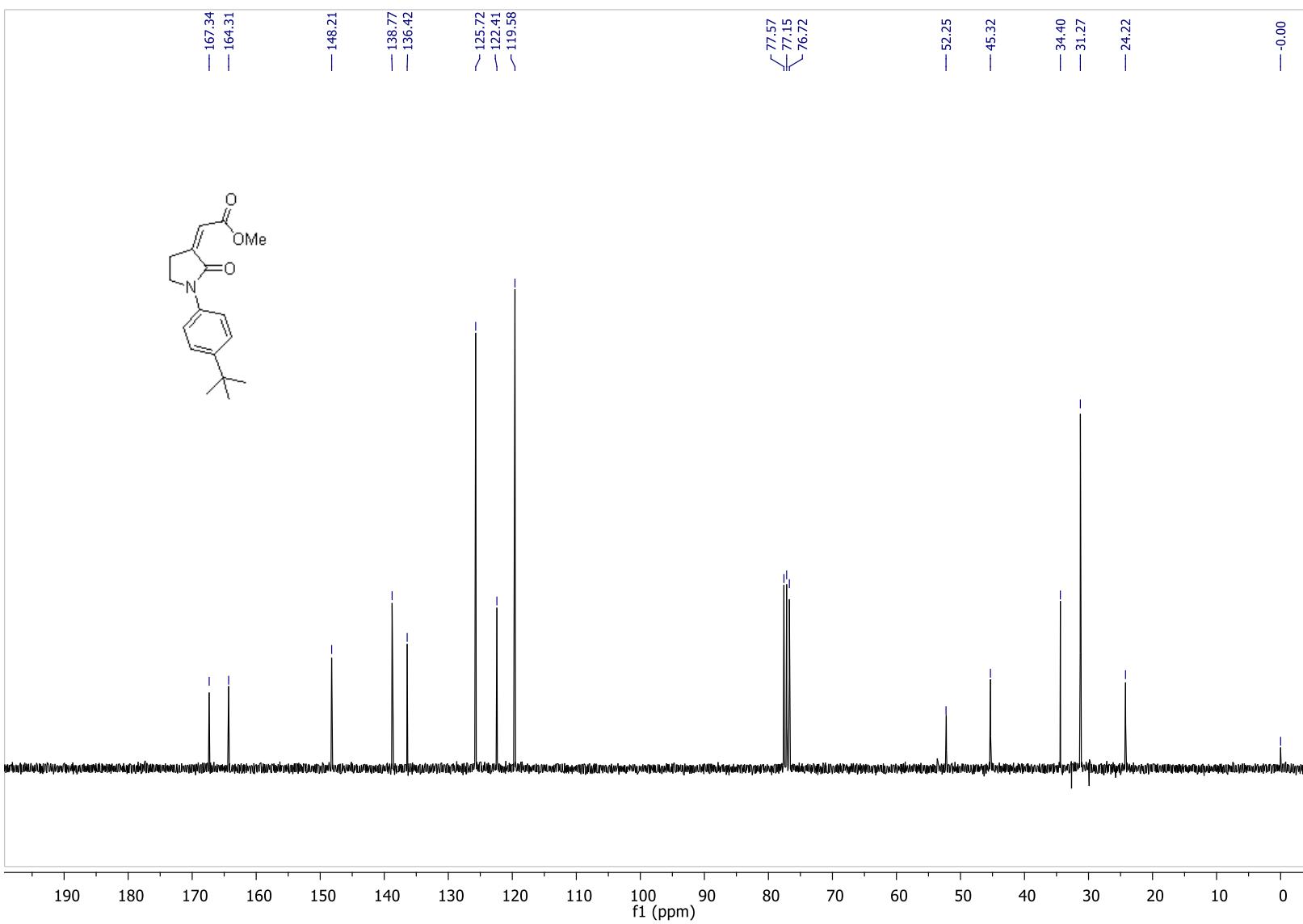
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

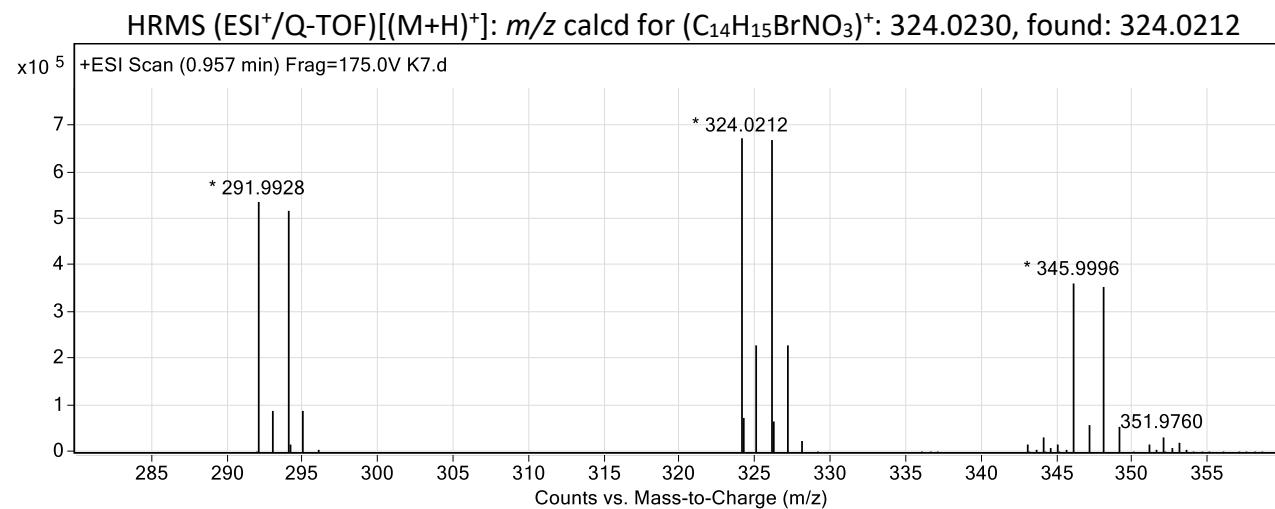
(Z)-Methyl 2-(1-(4-(*tert*-butyl)phenyl)-2-oxopyrrolidin-3-ylidene)acetate **2d**

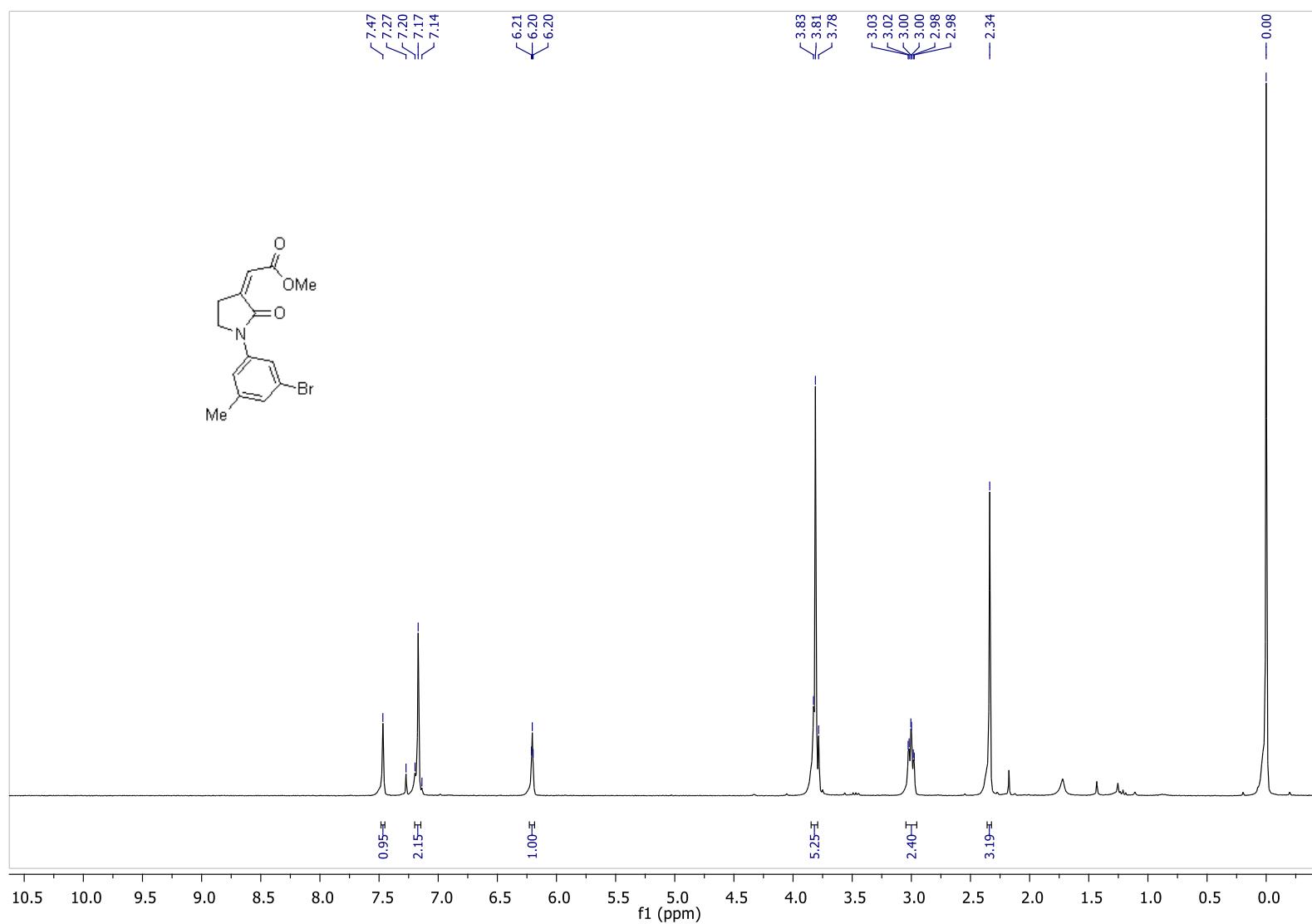
HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₇H₂₁NO₃)⁺: 288.1594, found: 288.1599

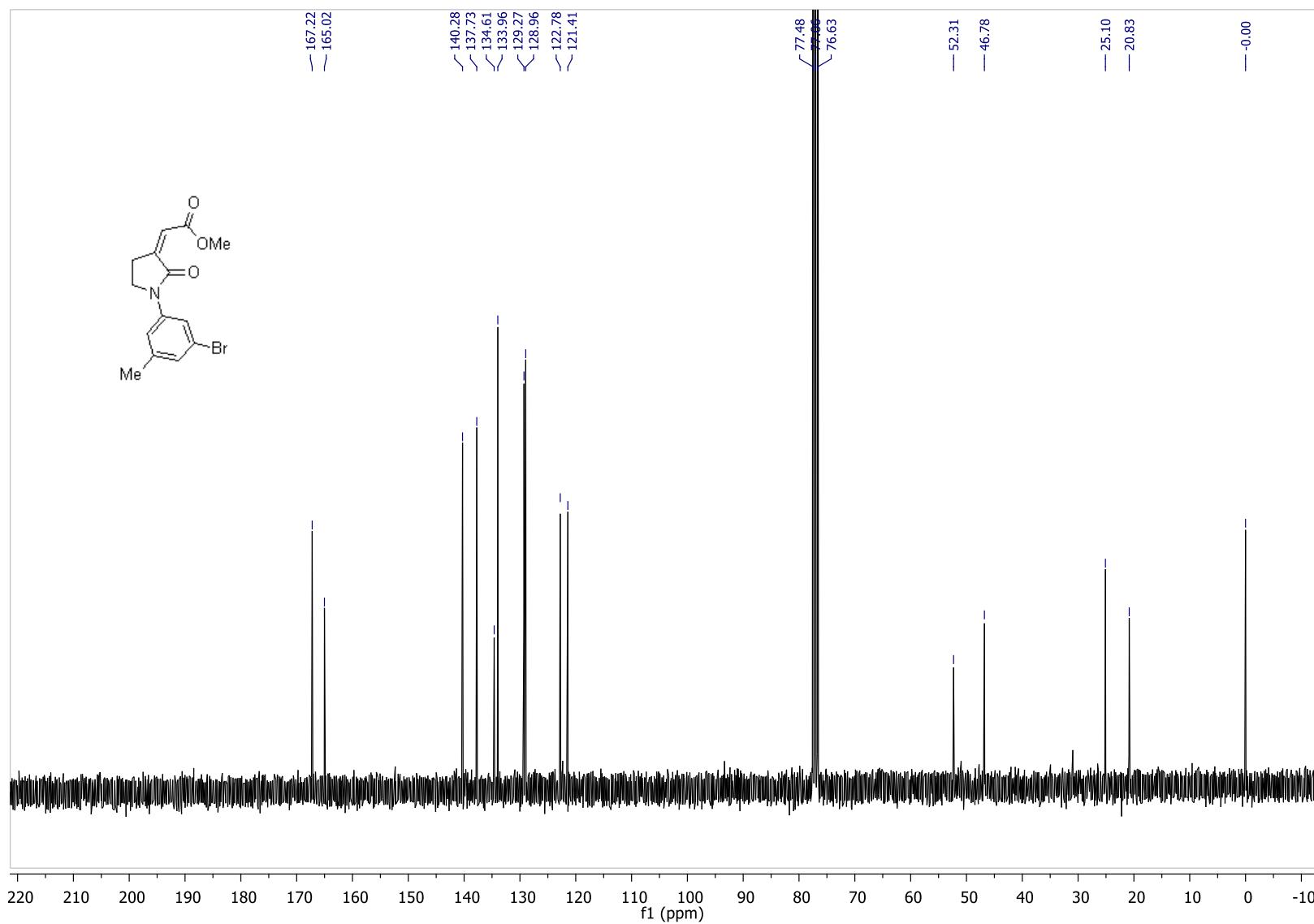


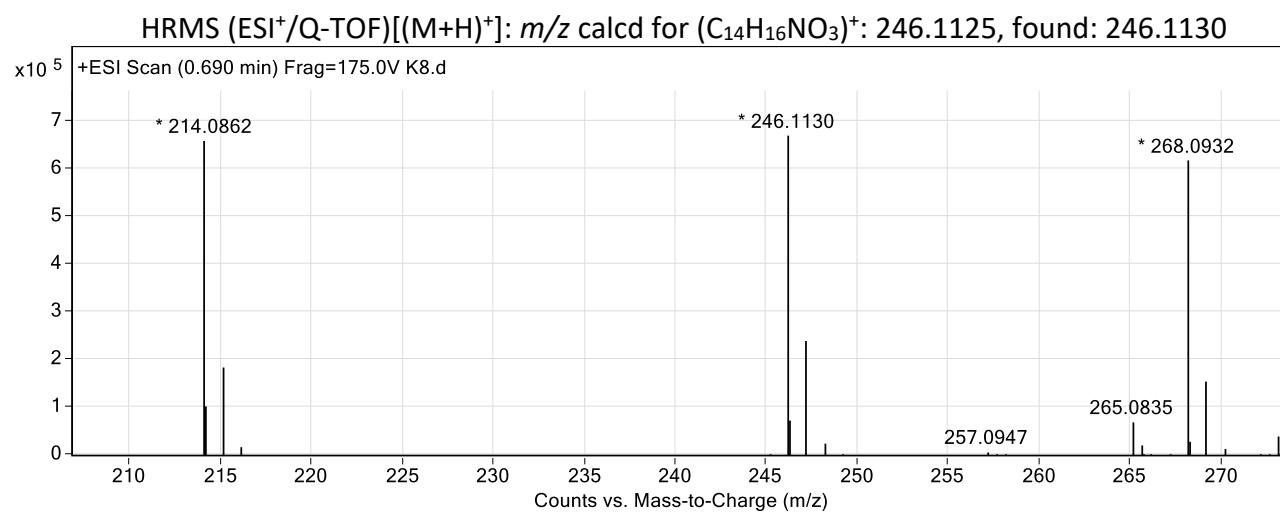
^1H NMR (CDCl_3)

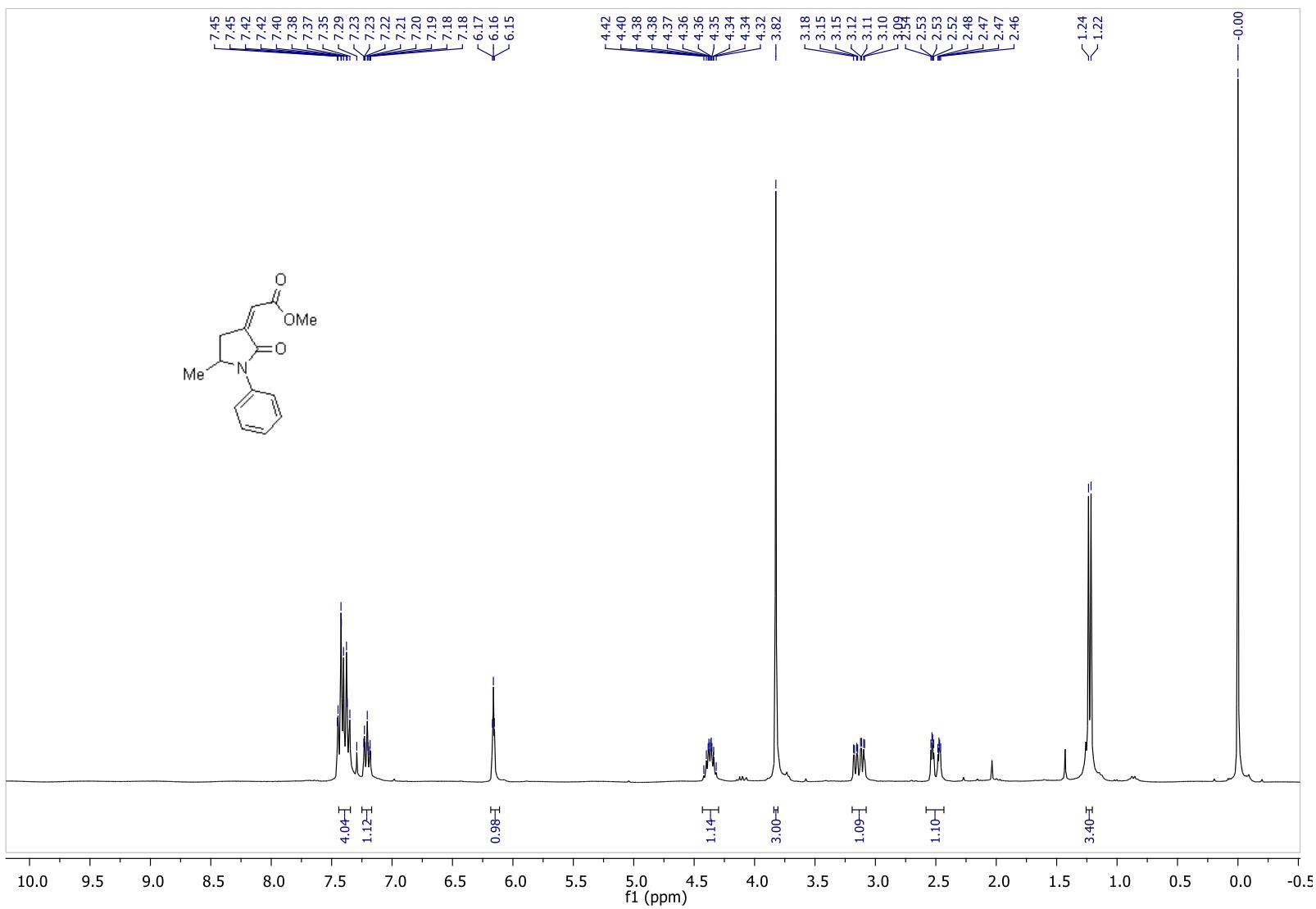
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

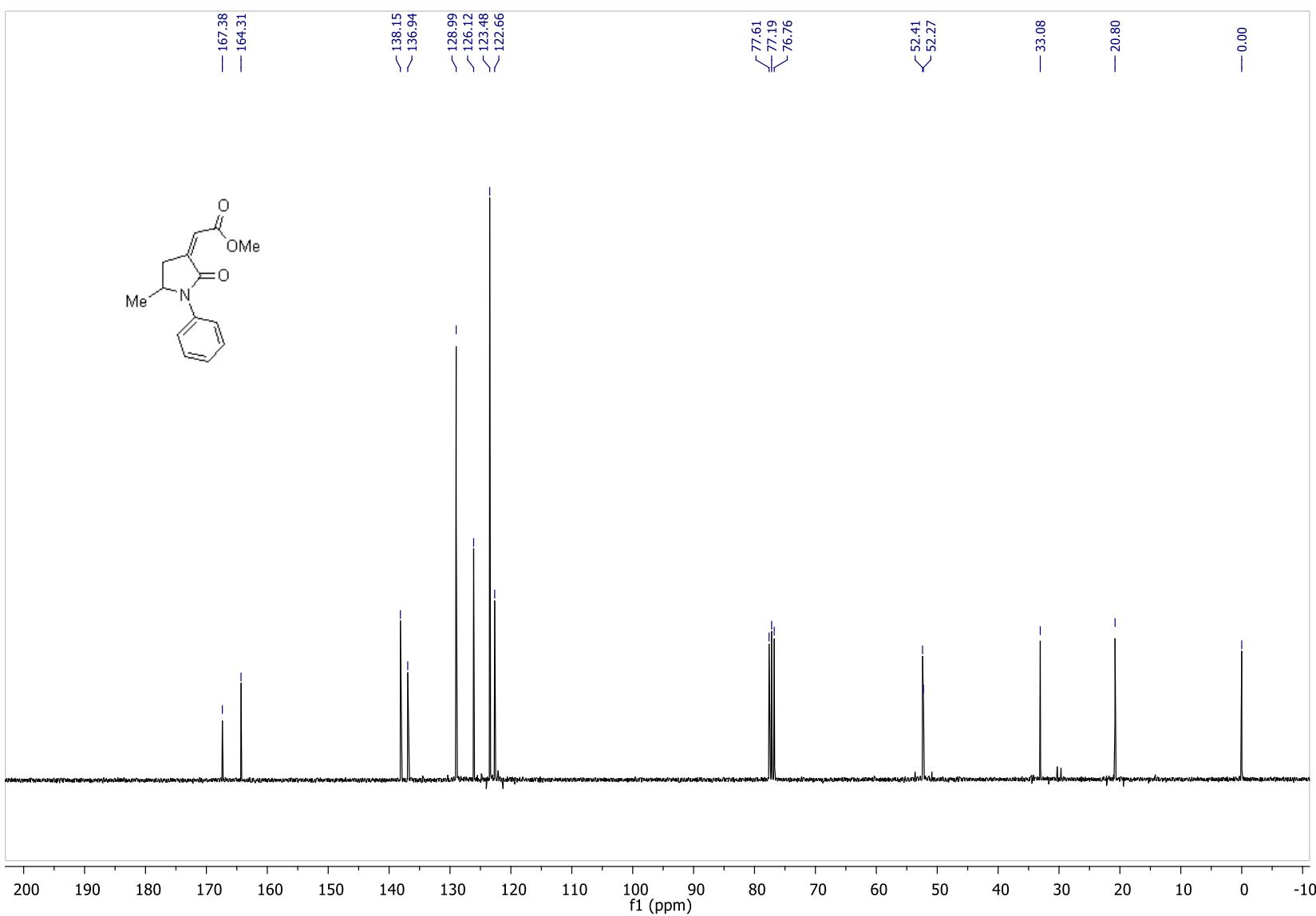
(Z)-Methyl 2-(1-(3-bromo-5-methylphenyl)-2-oxopyrrolidin-3-ylidene)acetate **2e**

¹H NMR (CDCl₃)

$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

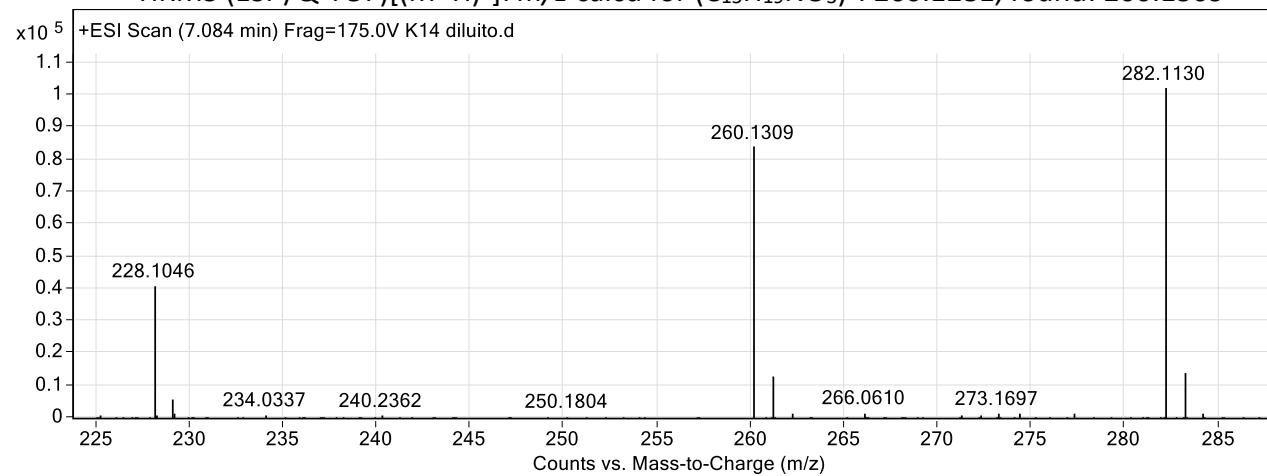
(Z)-Methyl 2-(5-methyl-2-oxo-1-phenylpyrrolidin-3-ylidene)acetate **2f**

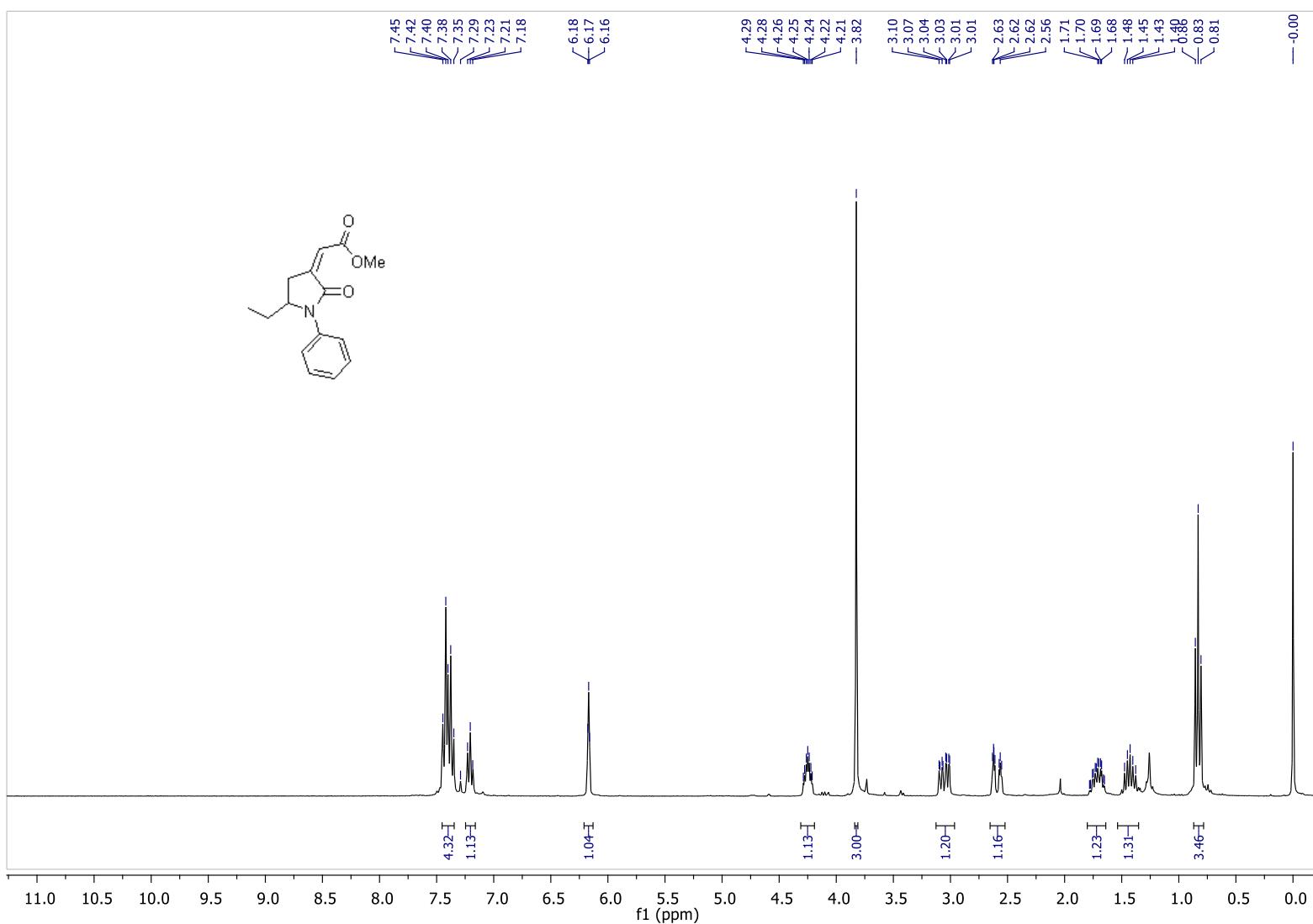
^1H NMR (CDCl_3)

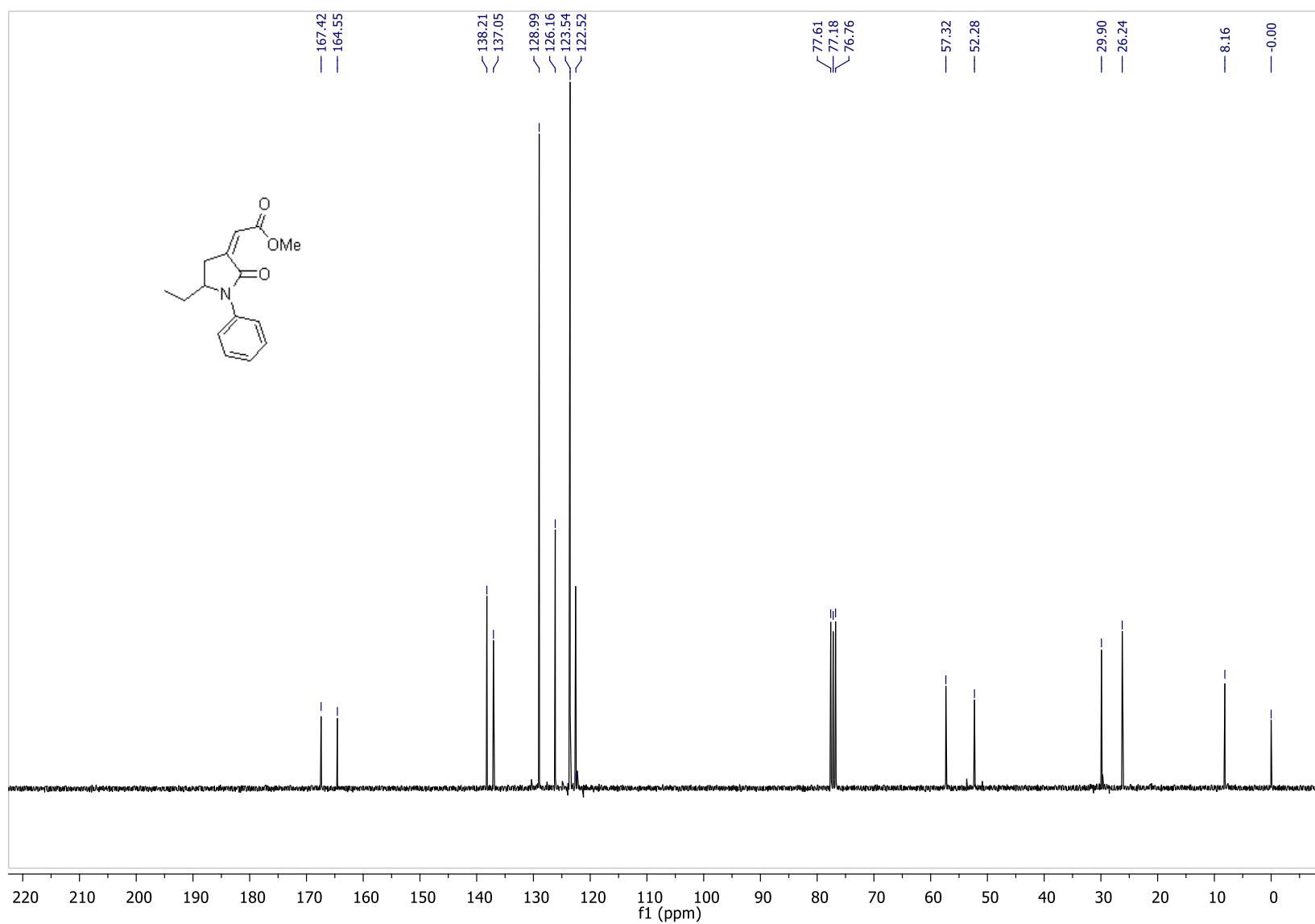
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

(Z)-Methyl 2-(5-ethyl-2-oxo-1-phenylpyrrolidin-3-ylidene)acetate **2g**

HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₅H₁₉NO₃)⁺: 260.1281, found: 260.1309

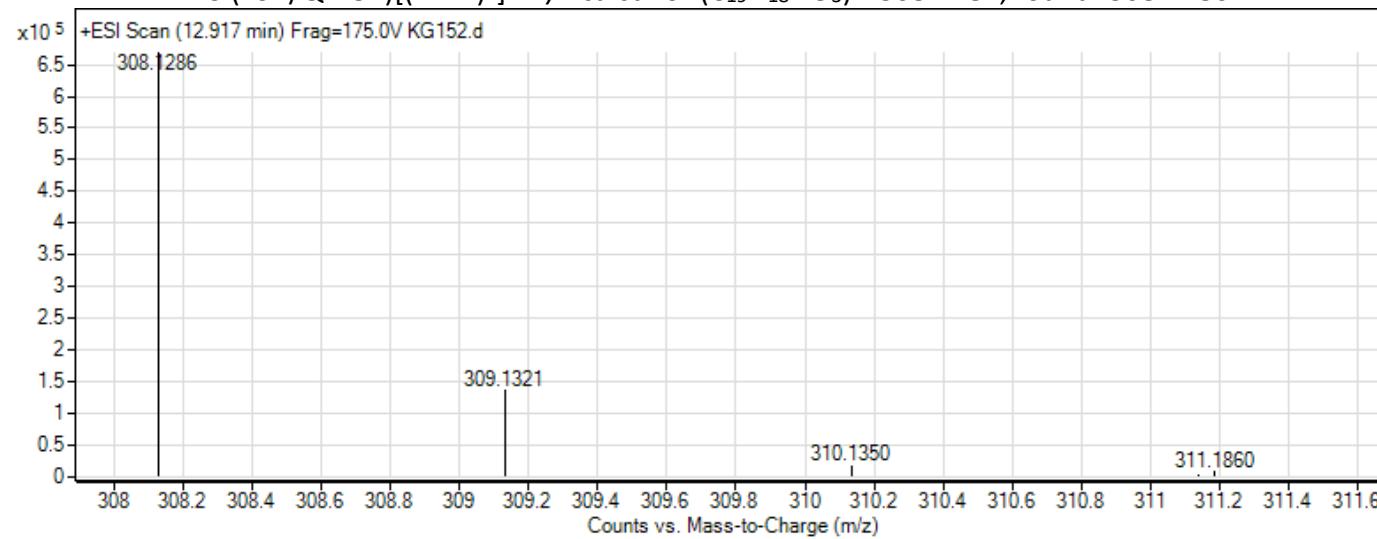


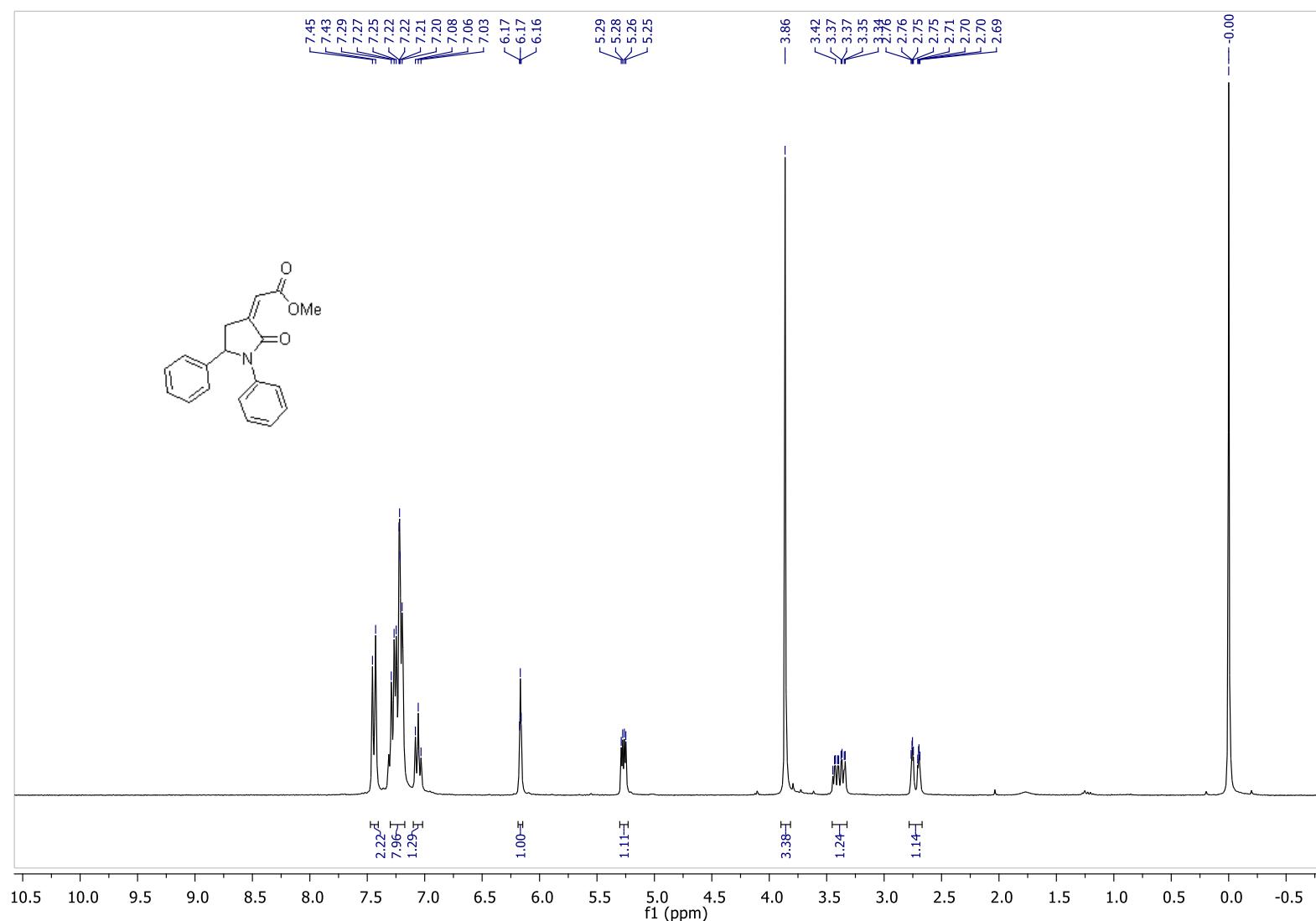
^1H NMR (CDCl_3)

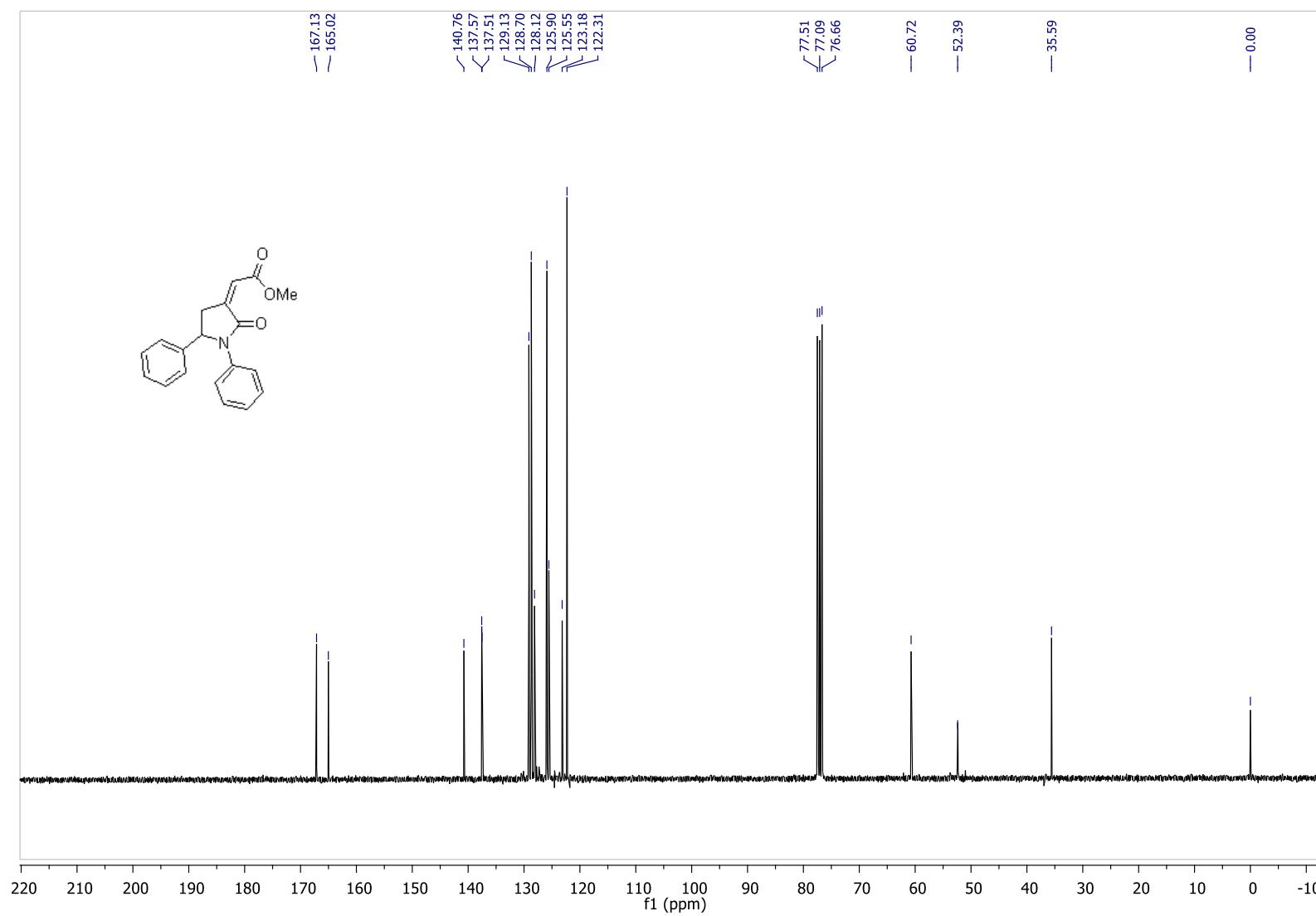
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

(Z)-Methyl 2-(2-oxo-1,5-diphenylpyrrolidin-3-ylidene)acetate **2h**

HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₉H₁₈NO₃)⁺: 308.1281, found: 308.1286

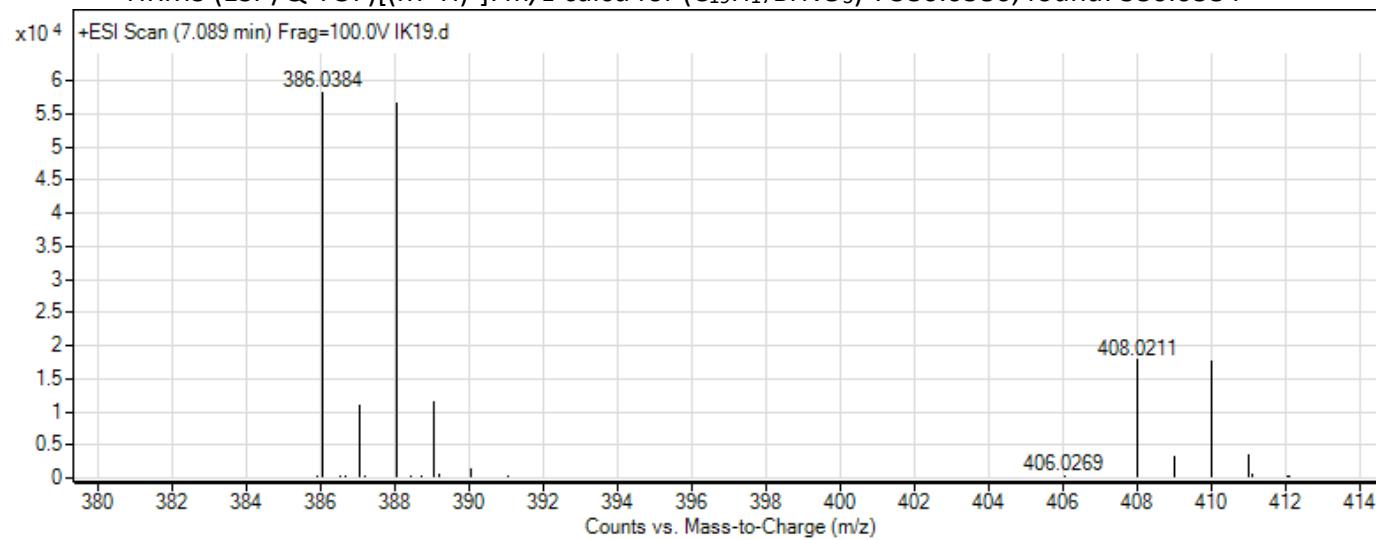


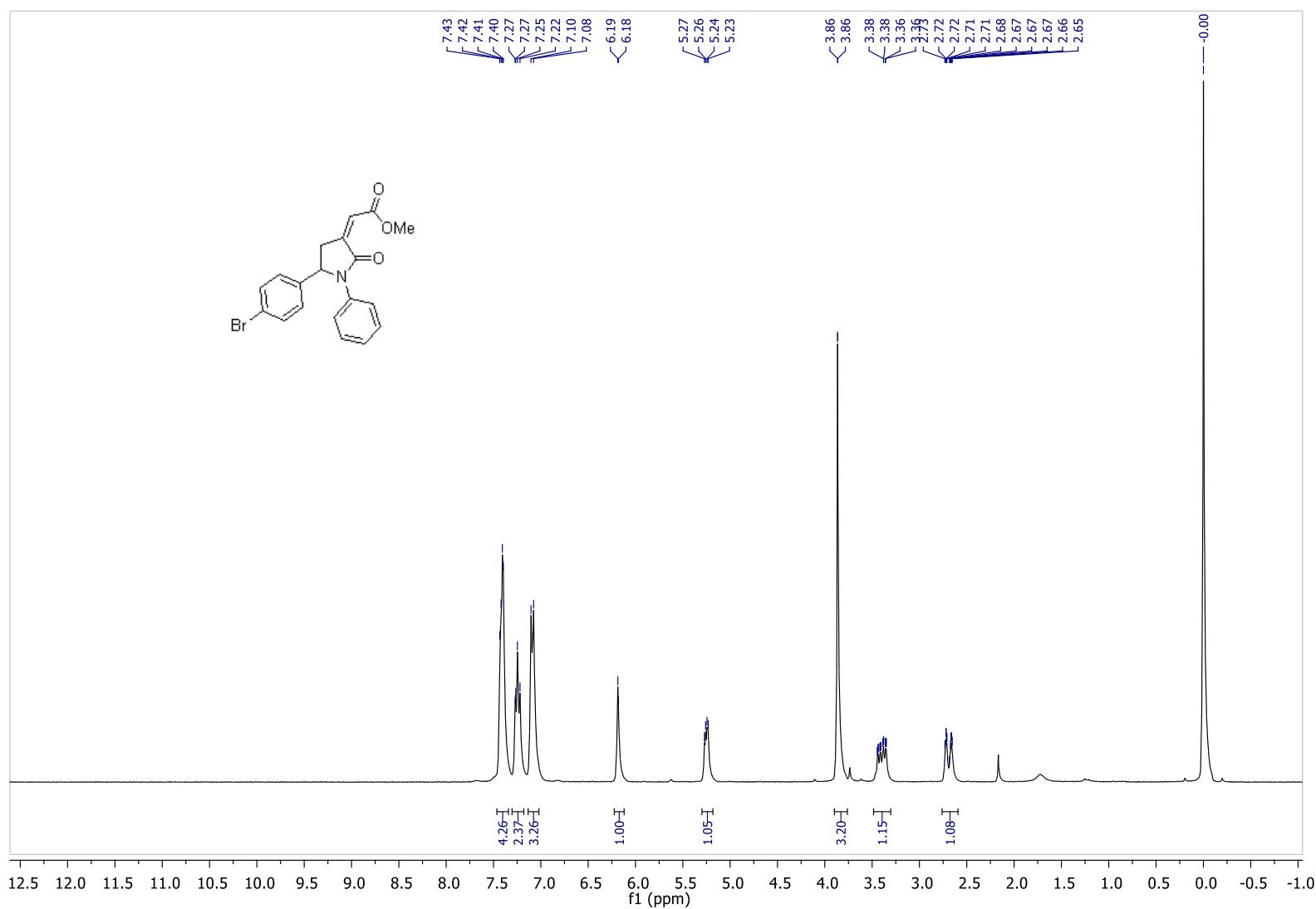
^1H NMR (CDCl_3)

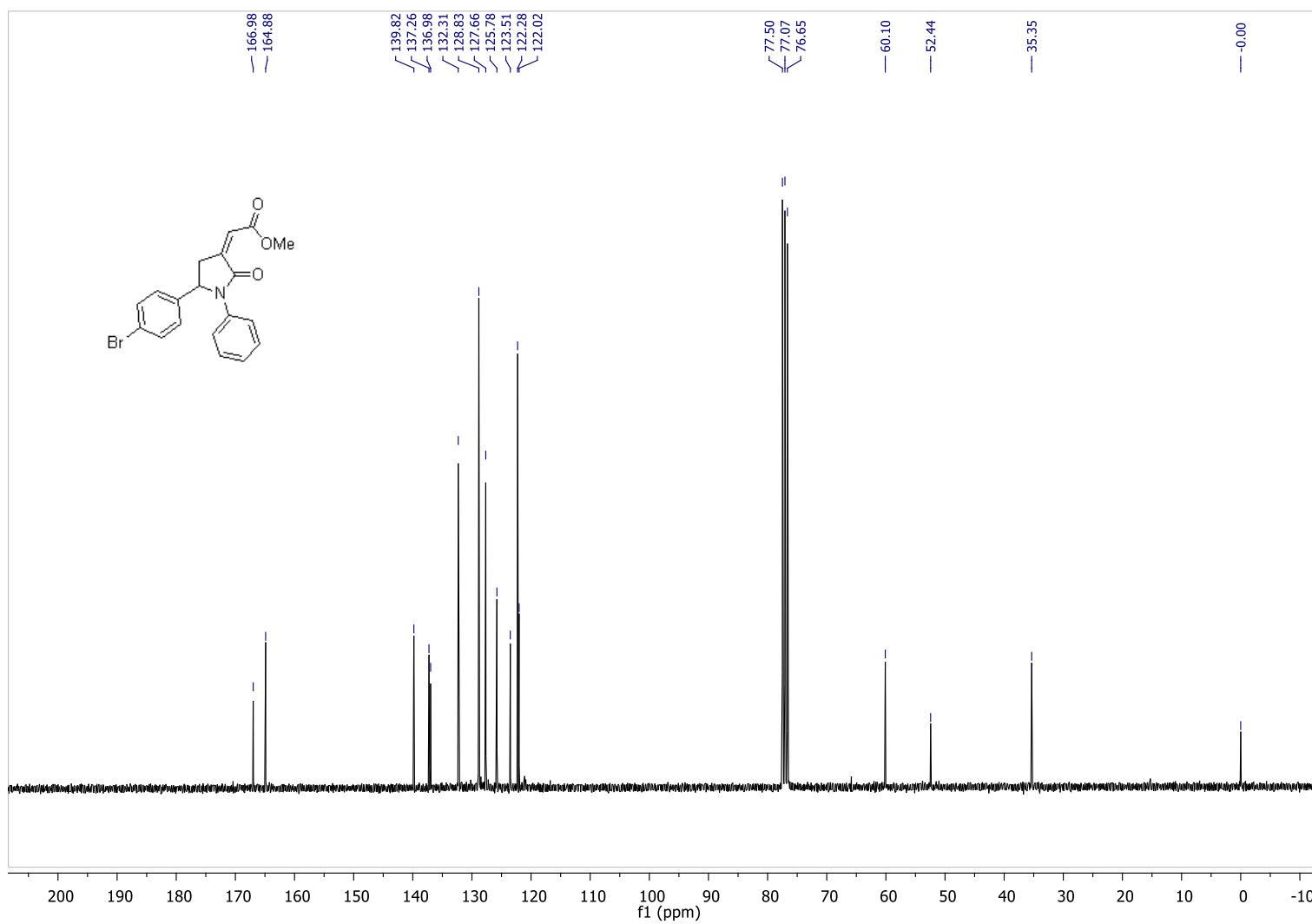
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

(Z)-Methyl 2-(5-(4-bromophenyl)-2-oxo-1-phenylpyrrolidin-3-ylidene)acetate **2i**

HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₉H₁₇BrNO₃)⁺: 386.0386, found: 386.0384

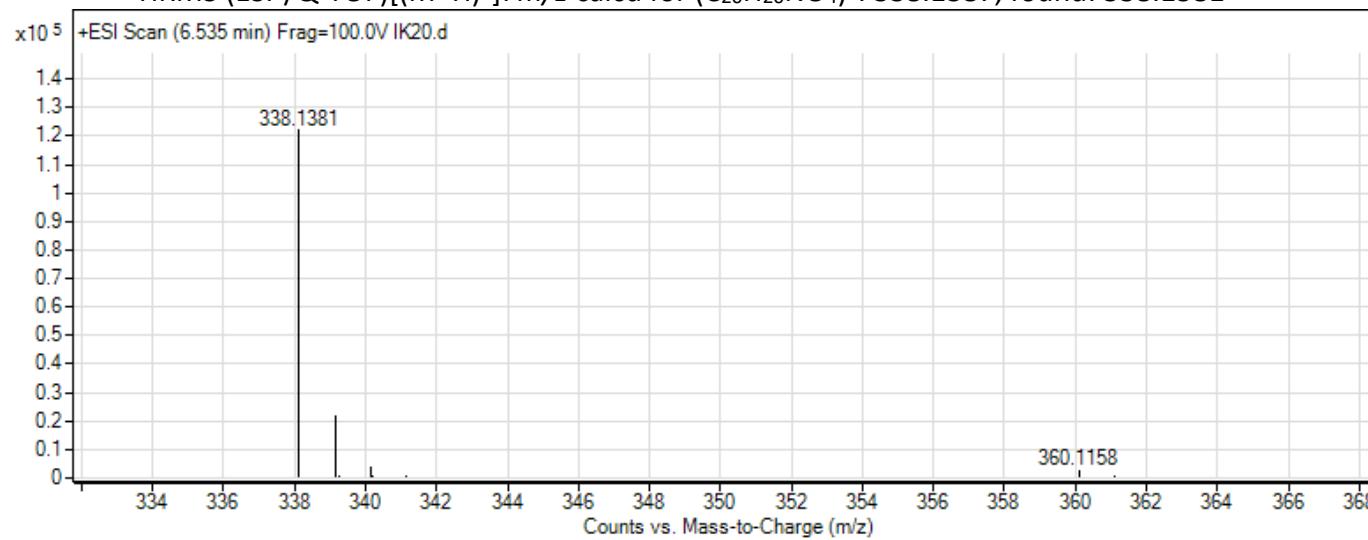


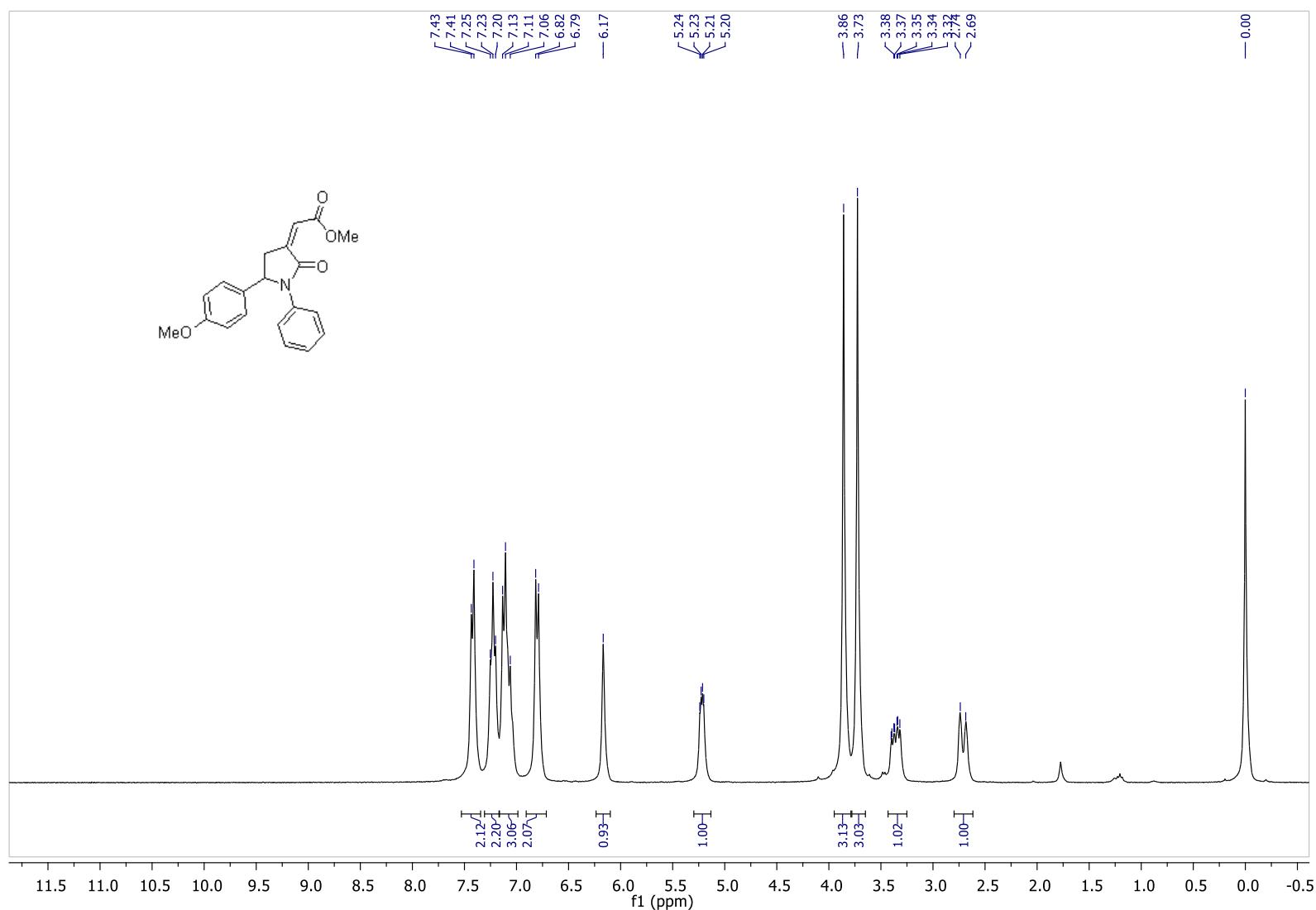
^1H NMR (CDCl_3)

$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

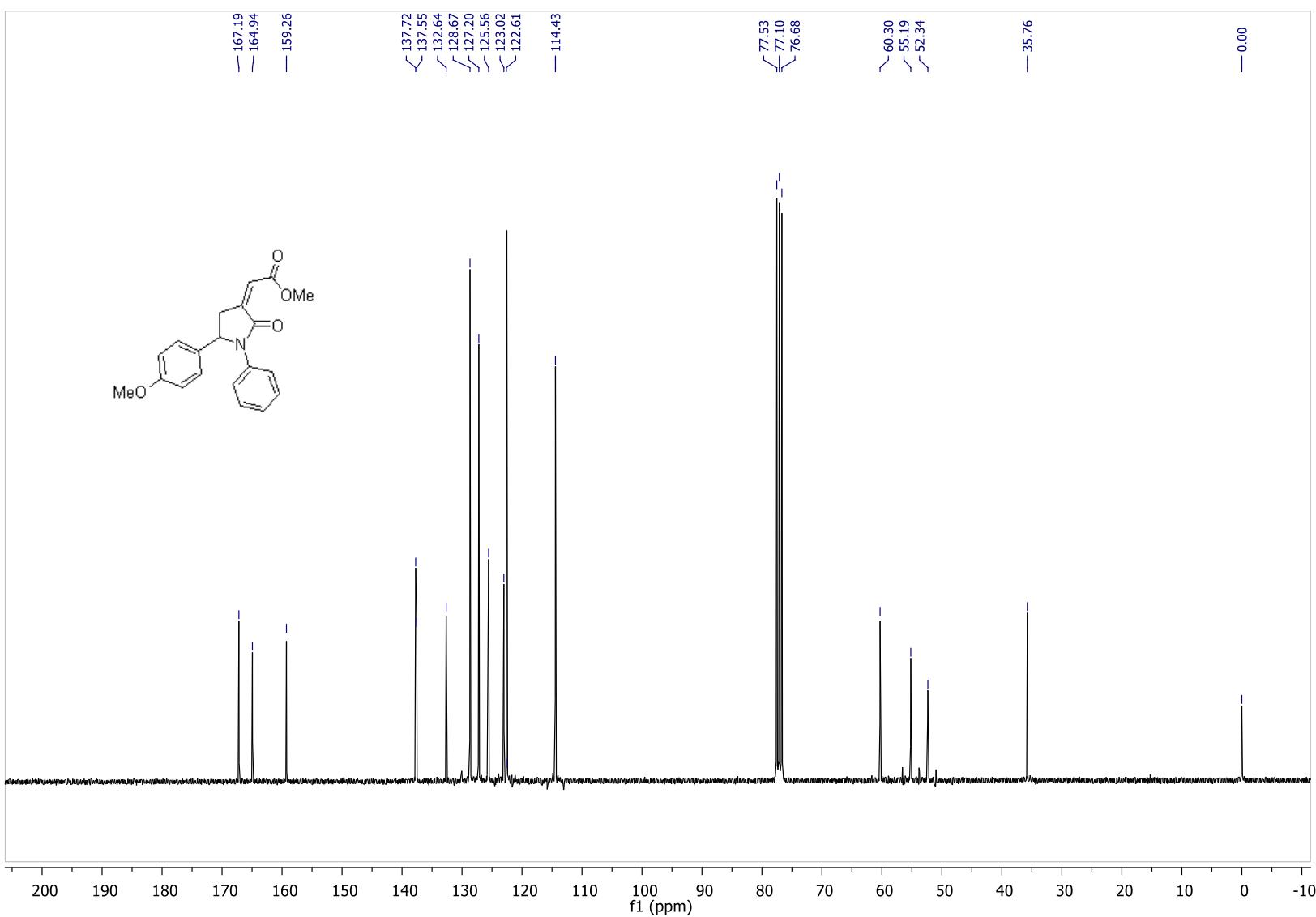
(Z)-Methyl 2-(5-(4-methoxyphenyl)-2-oxo-1-phenylpyrrolidin-3-ylidene)acetate **2j**

HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₂₀H₂₀NO₄)⁺: 338.1387, found: 338.1381



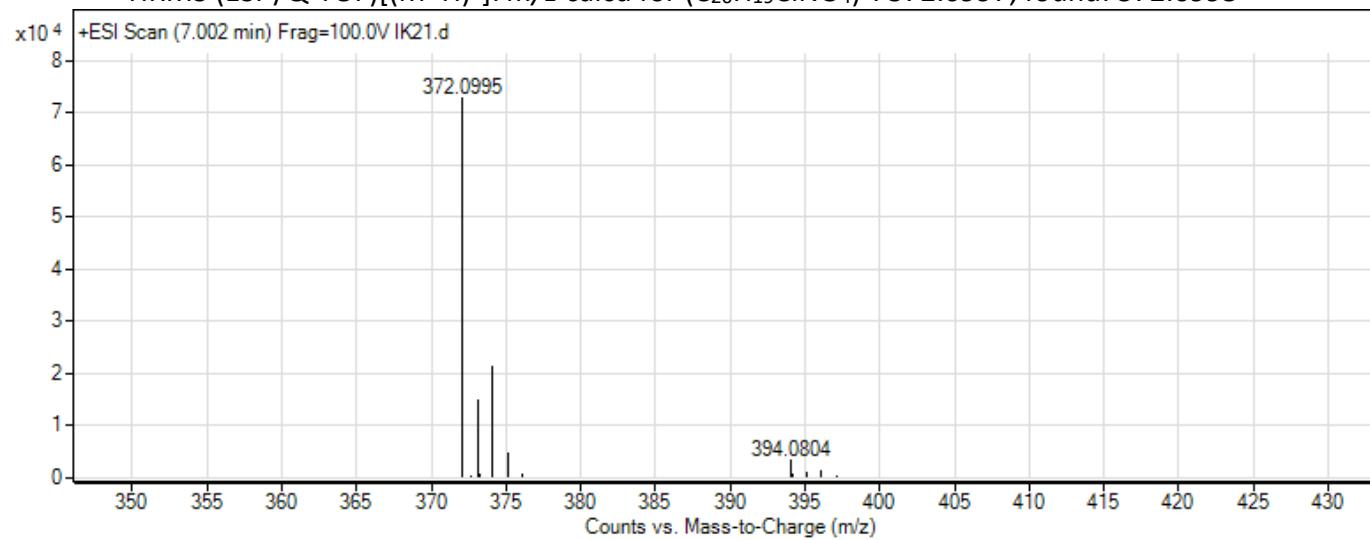
^1H NMR (CDCl_3)

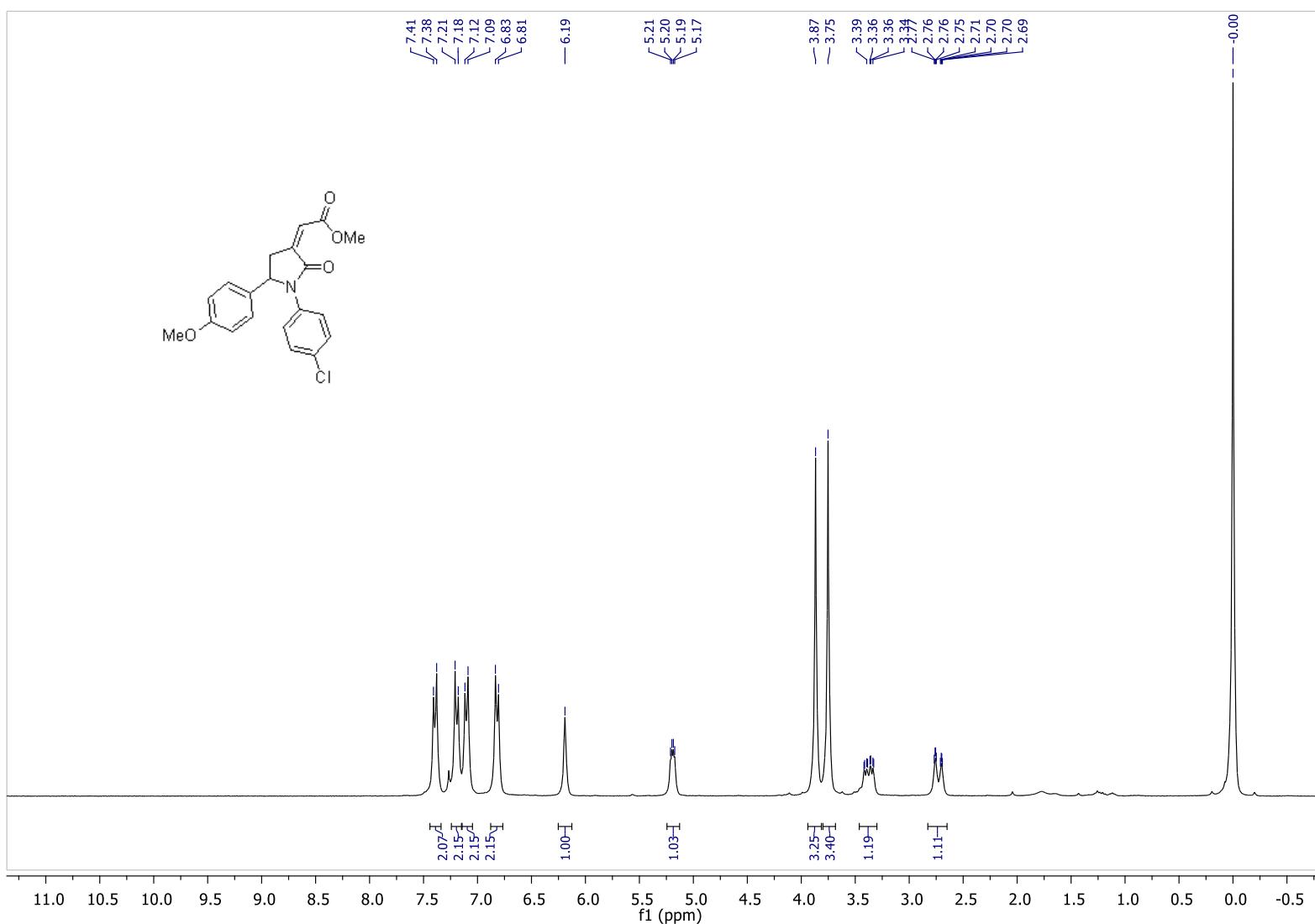
S90

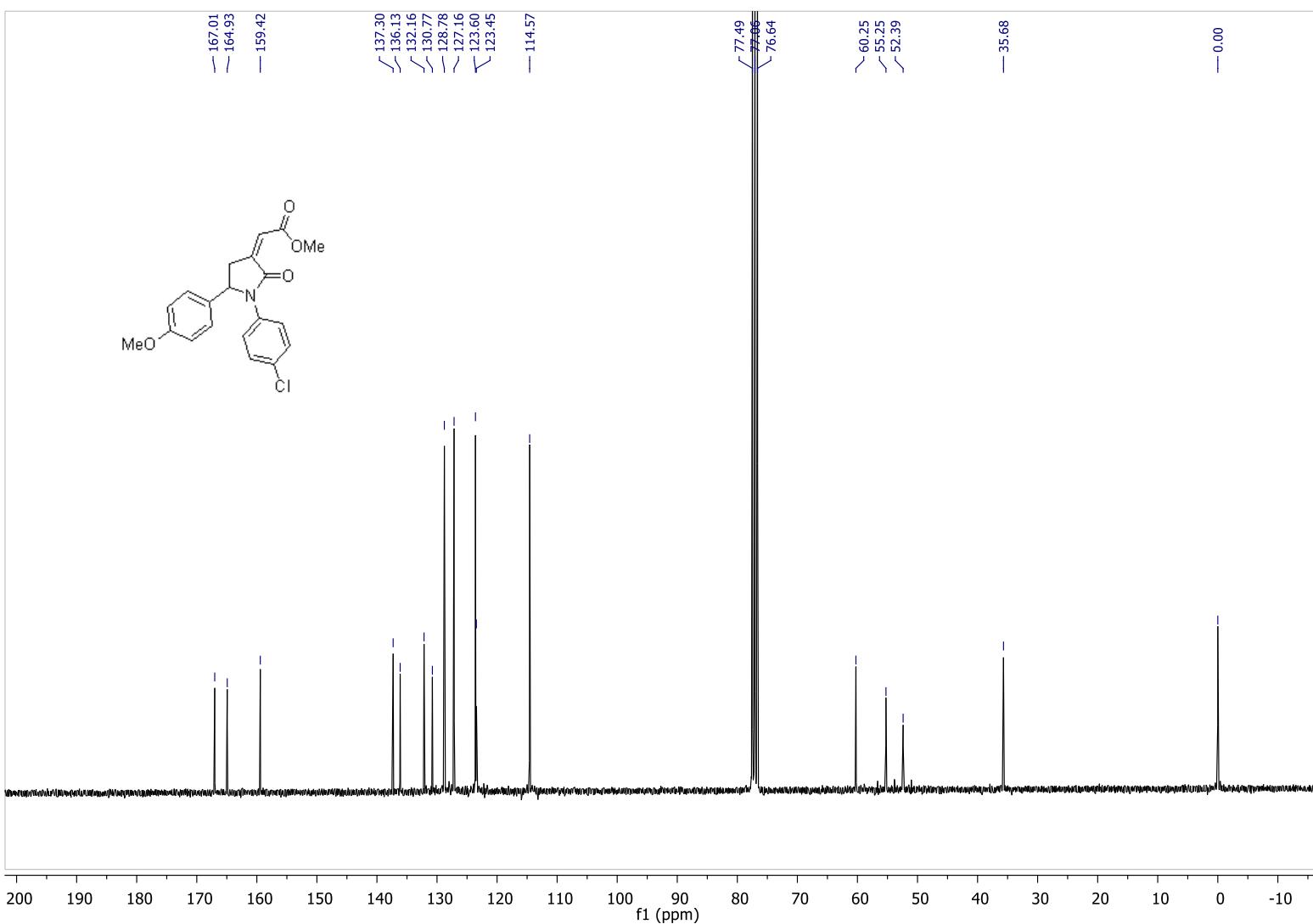
 $^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

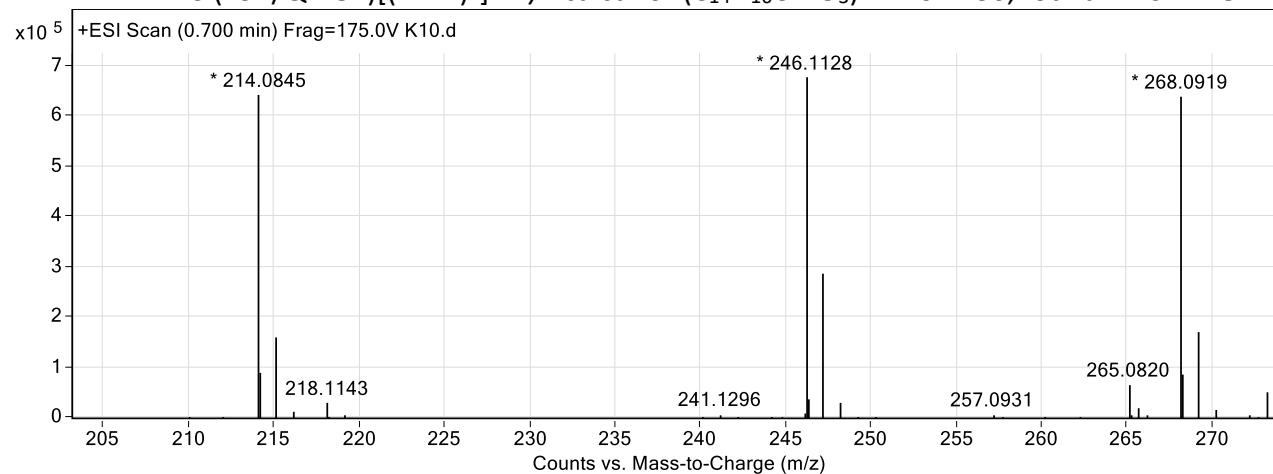
(Z)-Methyl 2-(1-(4-chlorophenyl)-5-(4-methoxyphenyl)-2-oxopyrrolidin-3-ylidene)acetate **2k**

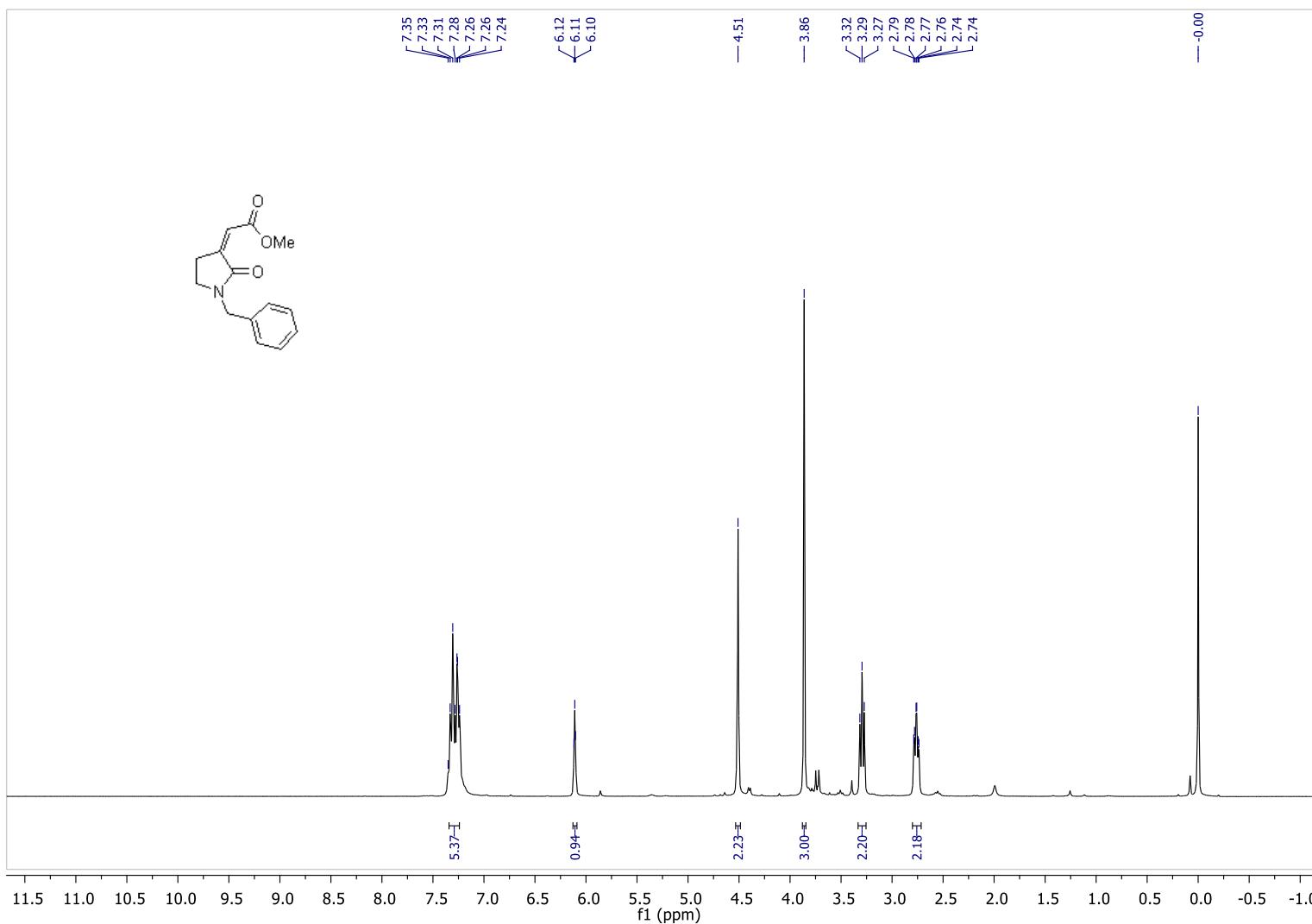
HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₂₀H₁₉ClNO₄)⁺: 372.0997, found: 372.0995

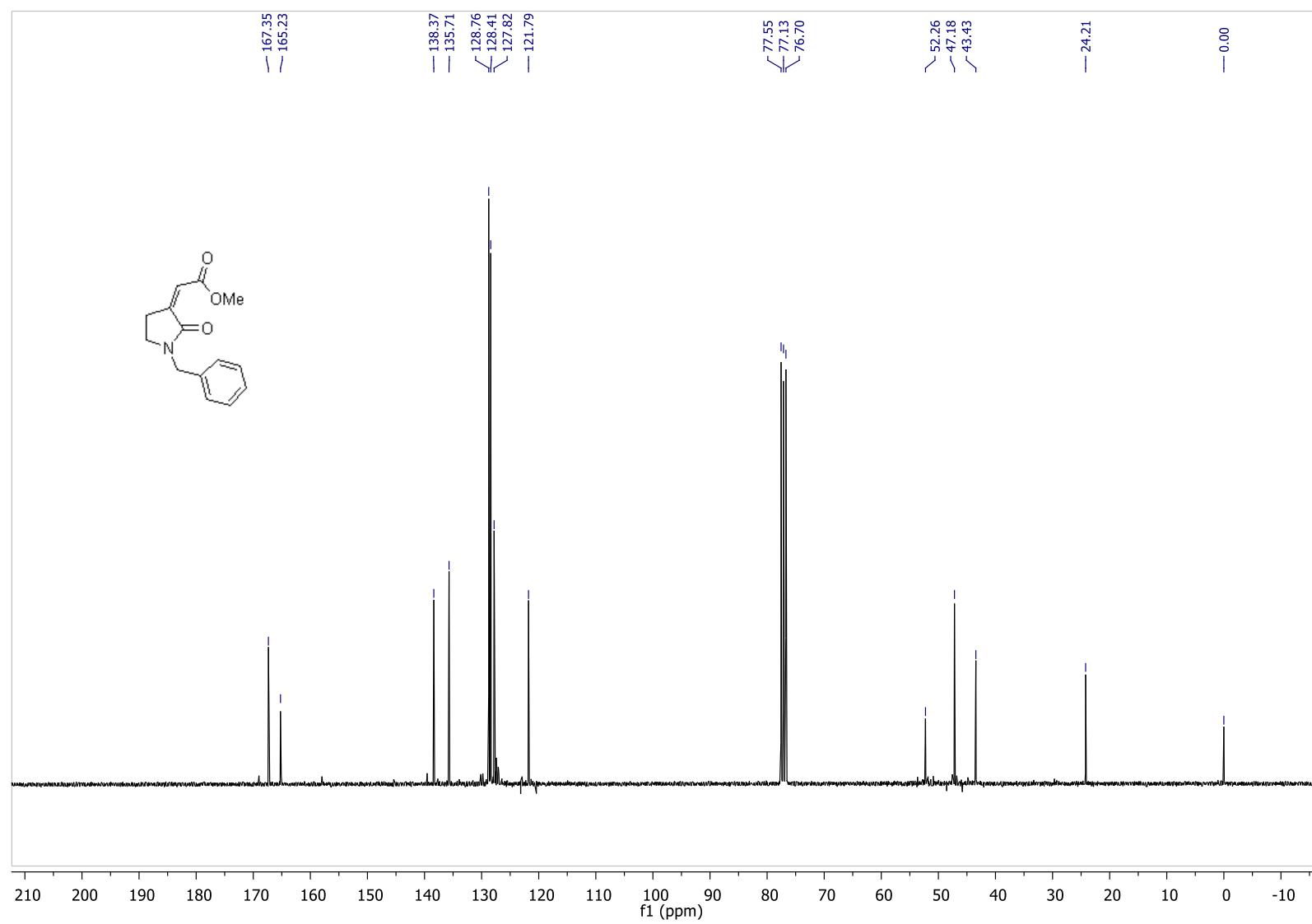


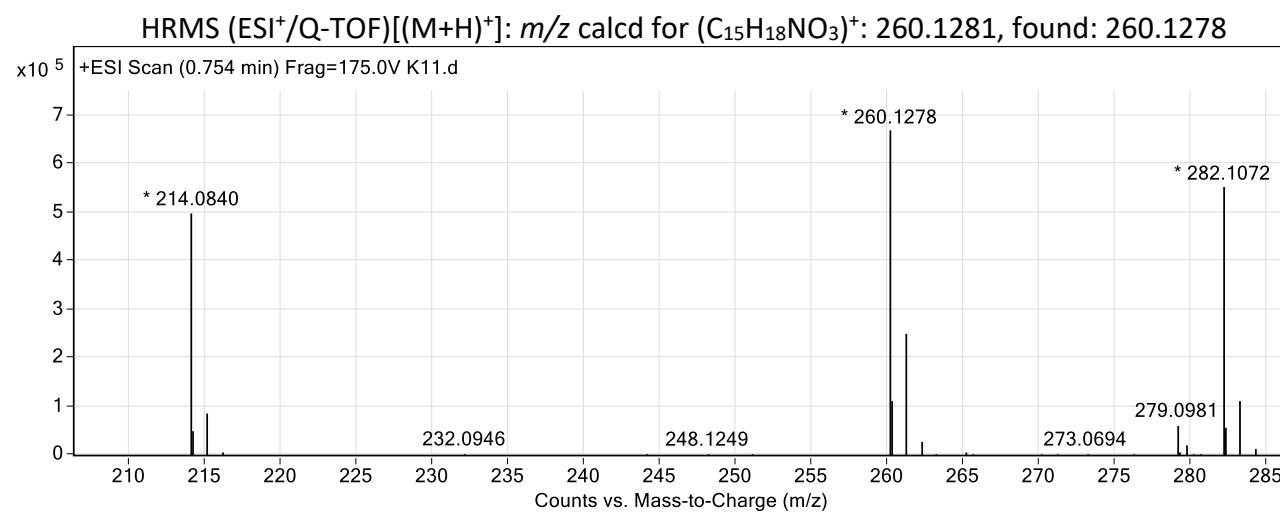
^1H NMR (CDCl_3)

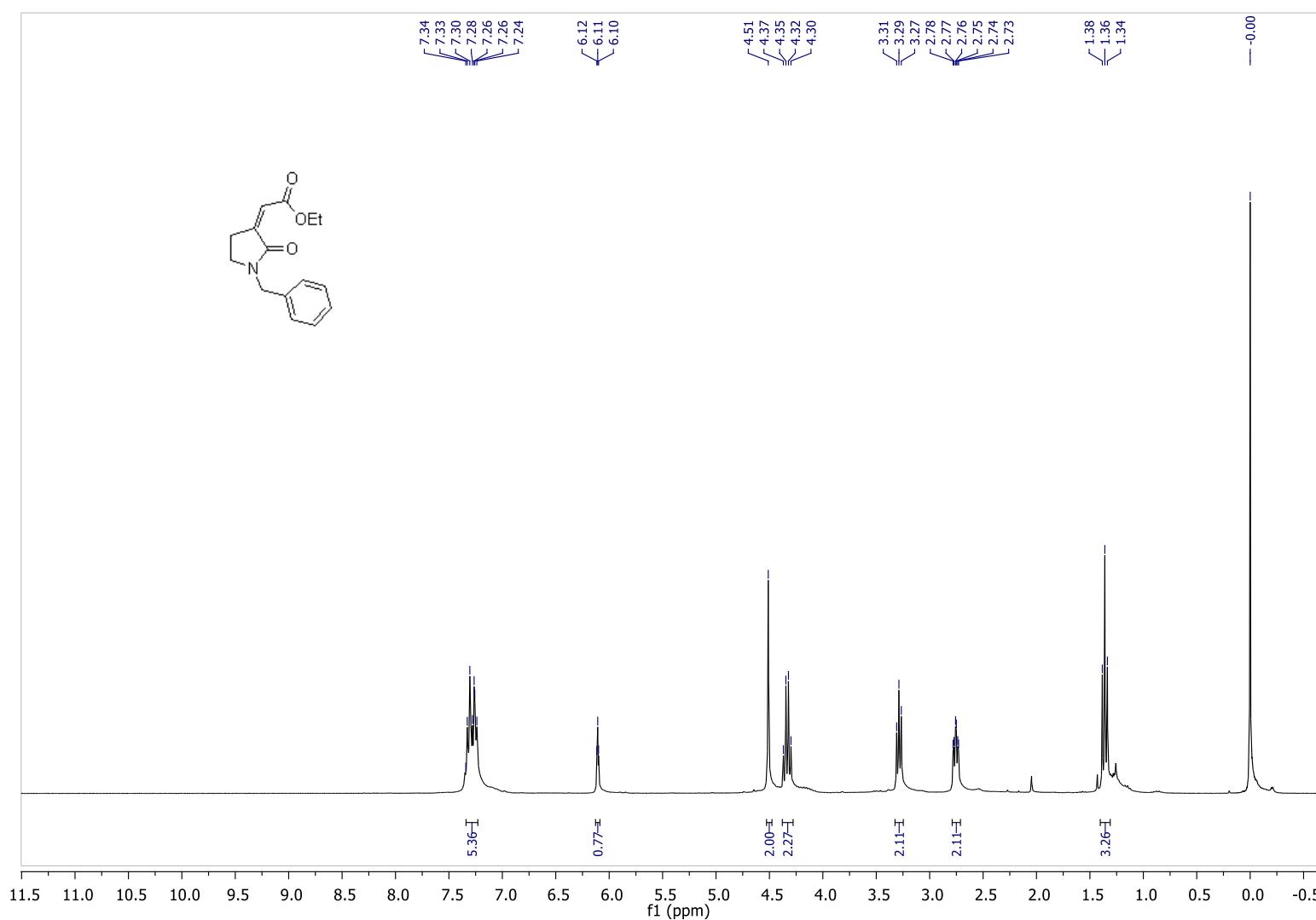
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

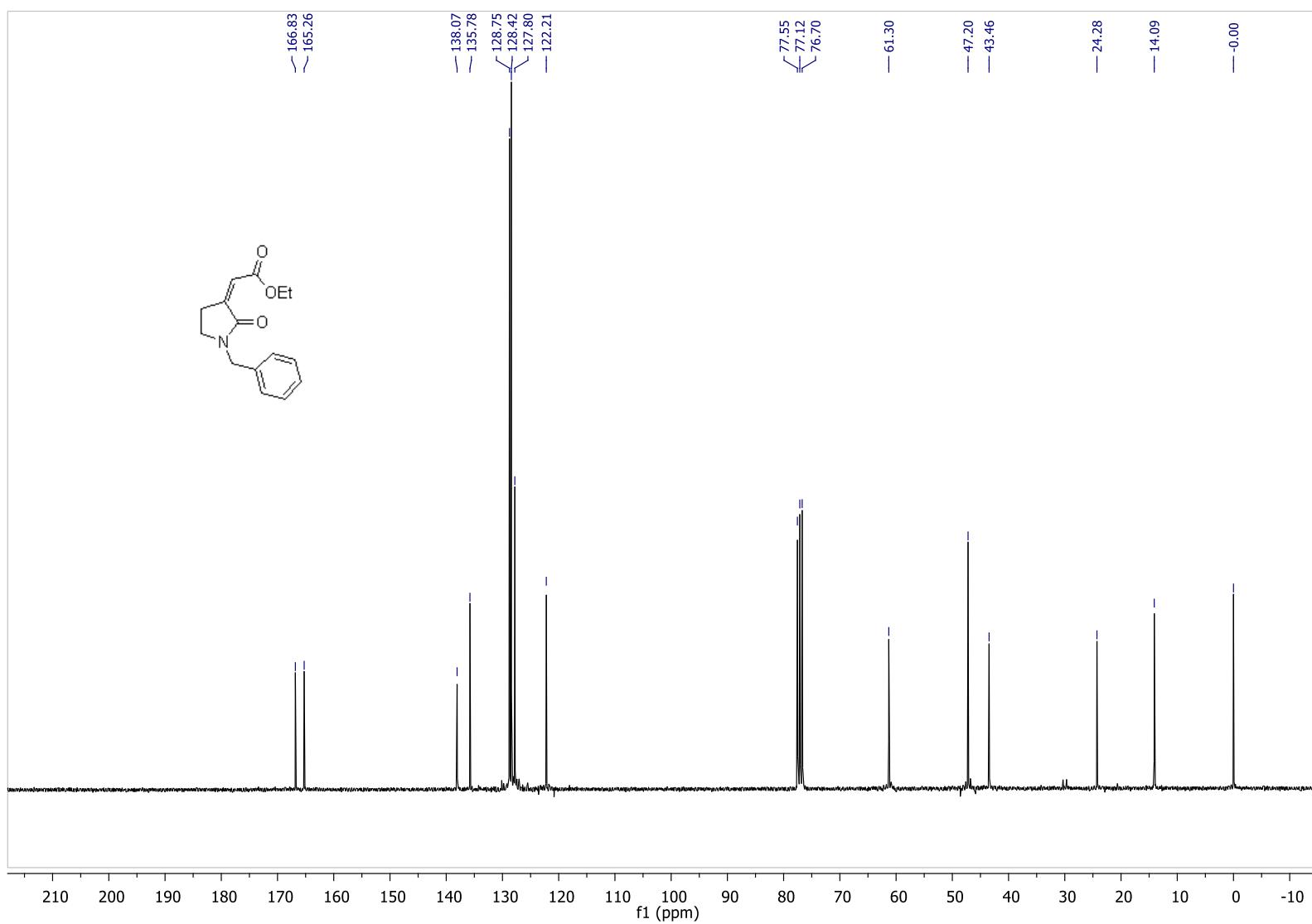
(Z)-Methyl 2-(1-benzyl-2-oxopyrrolidin-3-ylidene)acetate **2I**HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₄H₁₆CINO₃)⁺: 246.1130, found: 246.1128

^1H NMR (CDCl_3)

$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

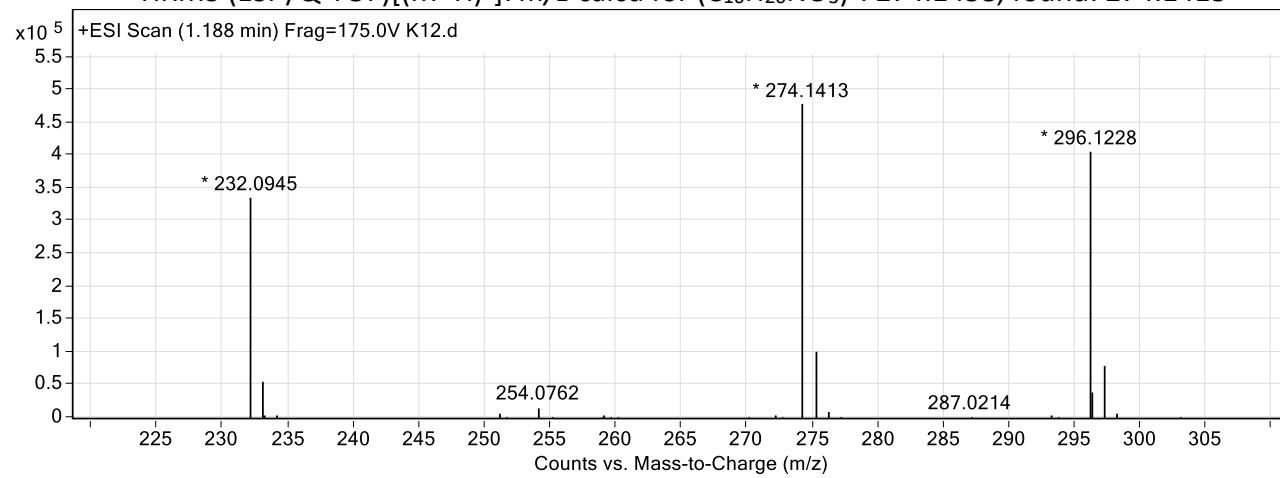
(Z)-Ethyl 2-(1-benzyl-2-oxopyrrolidin-3-ylidene)acetate **2I'**

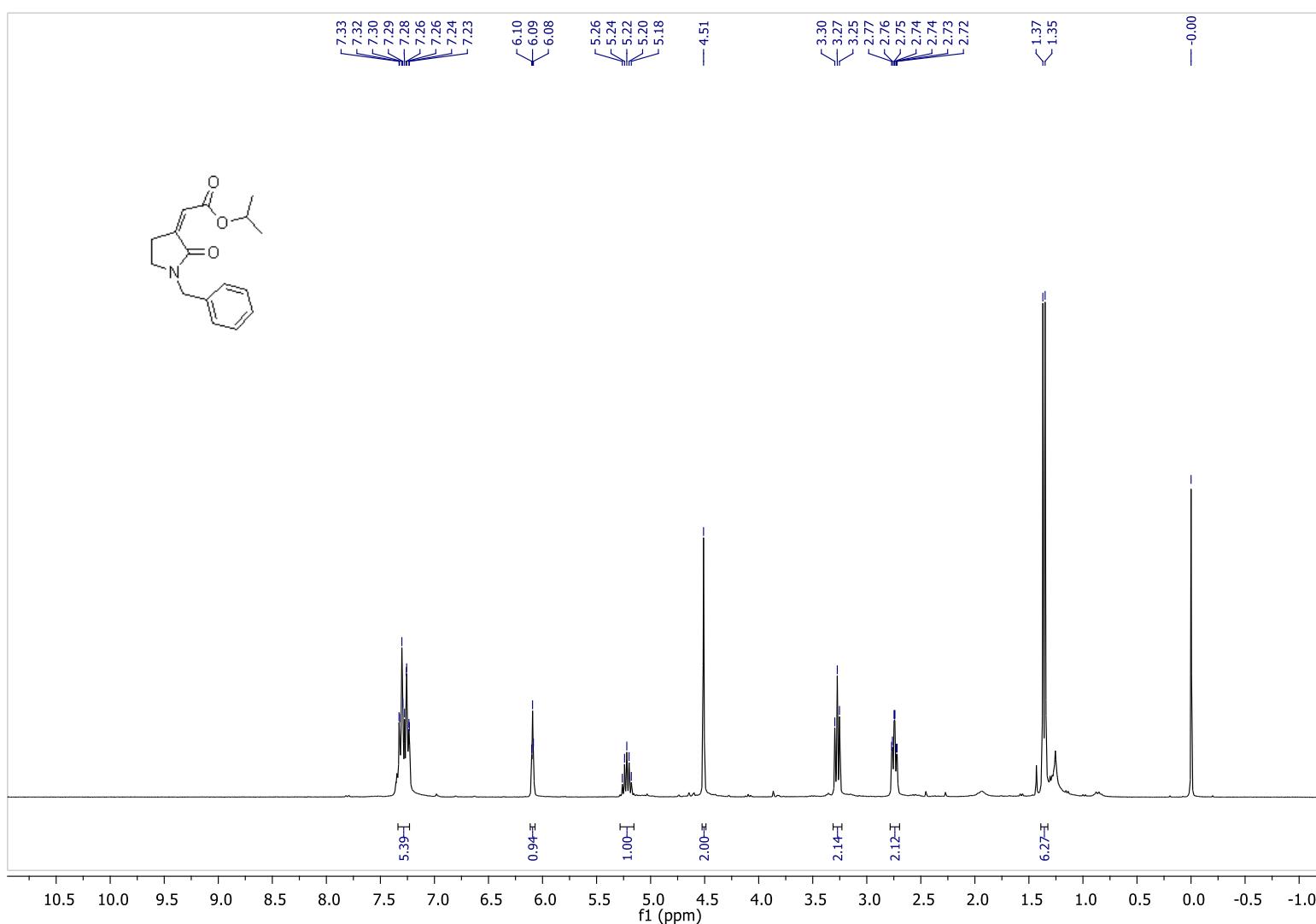
^1H NMR (CDCl_3)

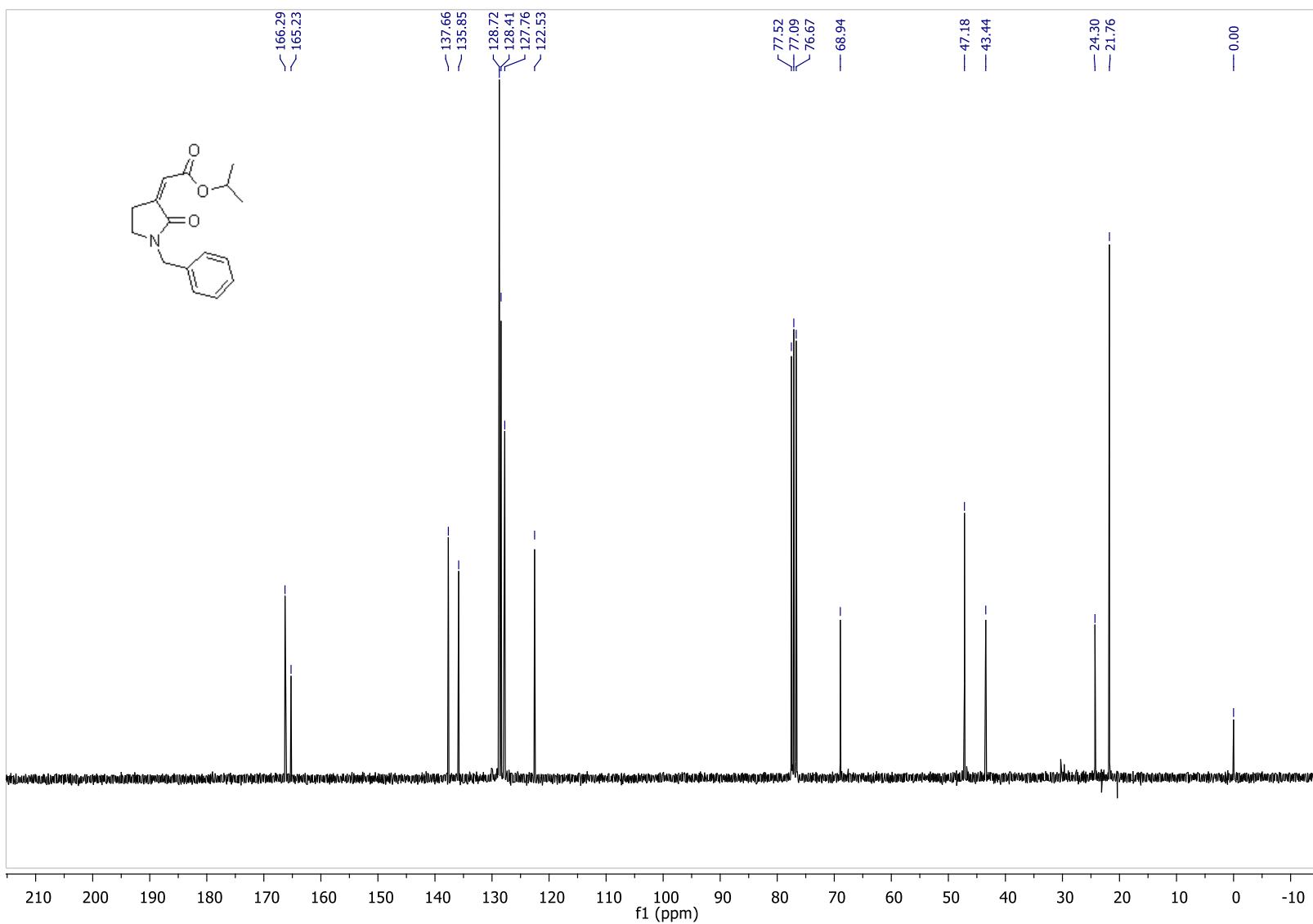
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

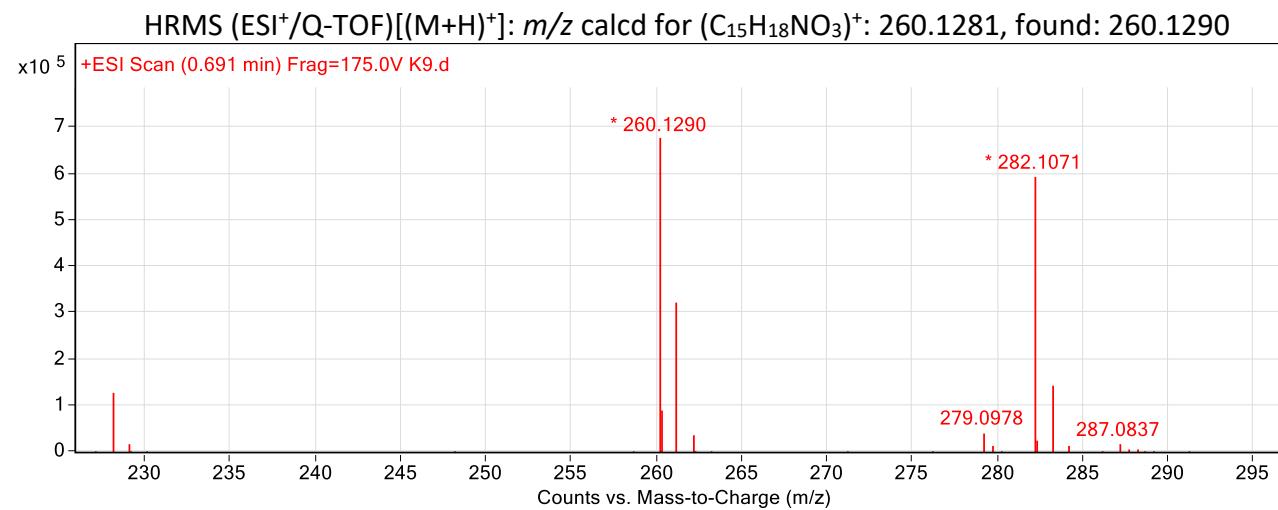
(Z)-Isopropyl 2-(1-benzyl-2-oxopyrrolidin-3-ylidene)acetate **2I''**

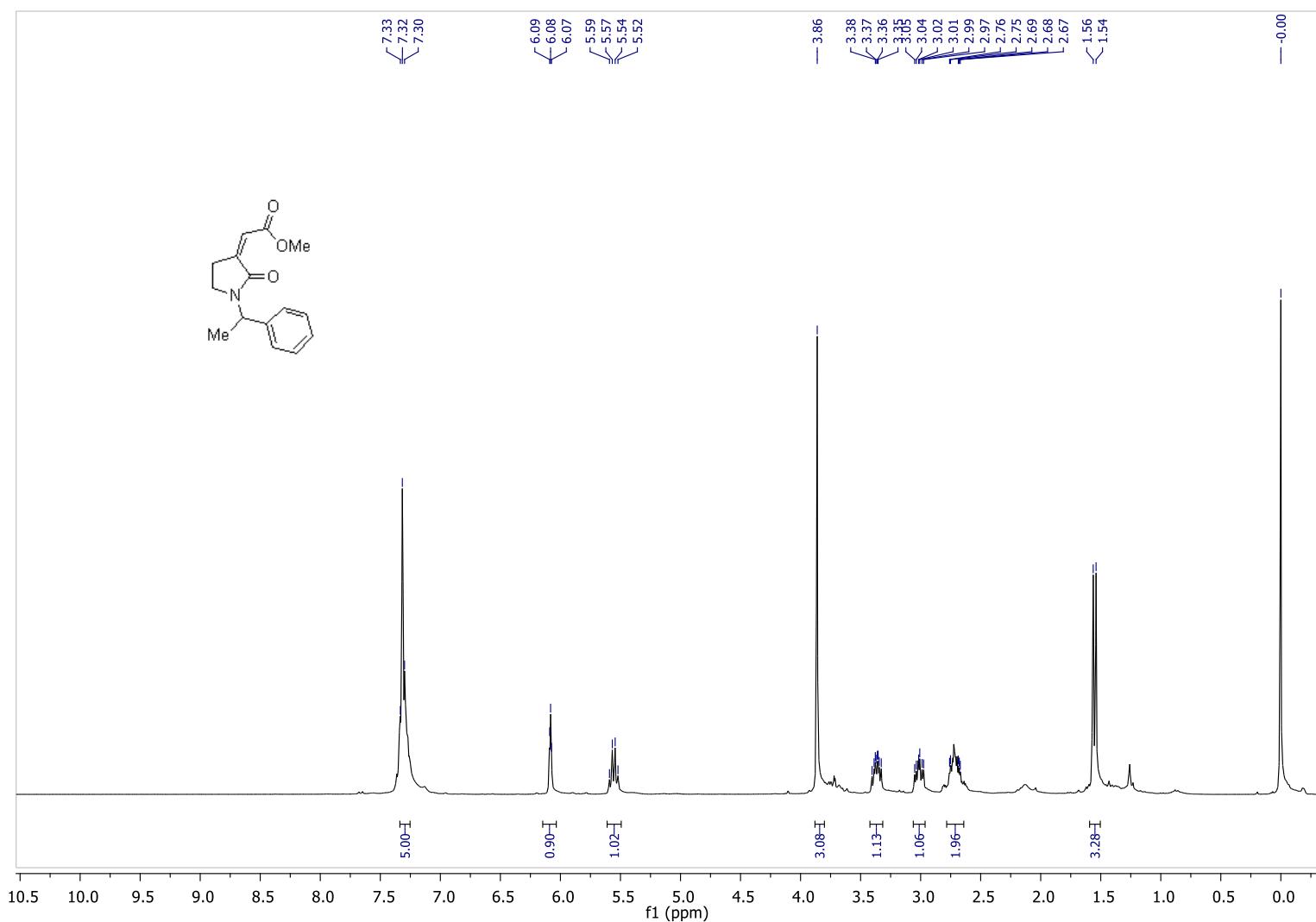
HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₆H₂₀NO₃)⁺: 274.1438, found: 274.1413

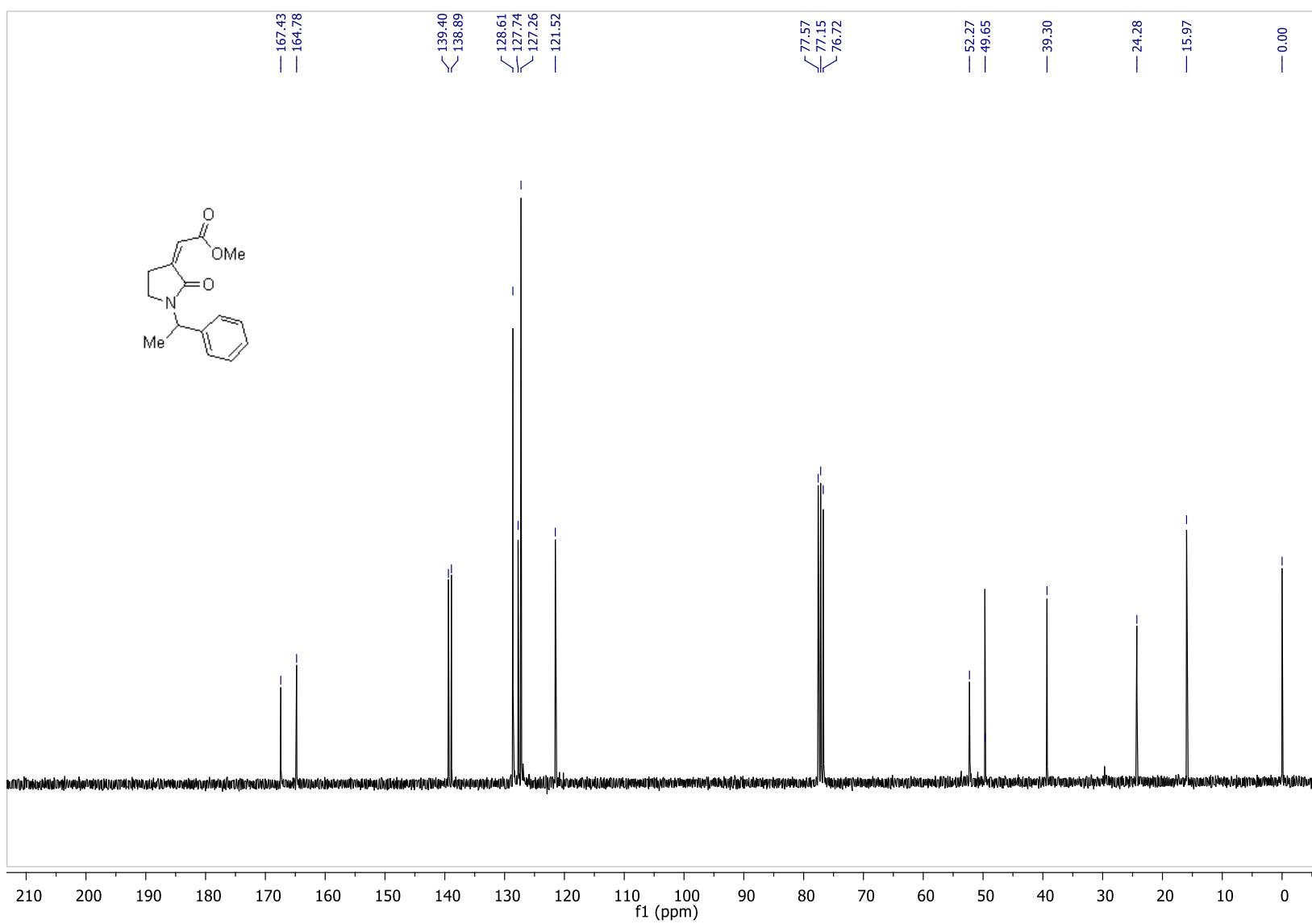


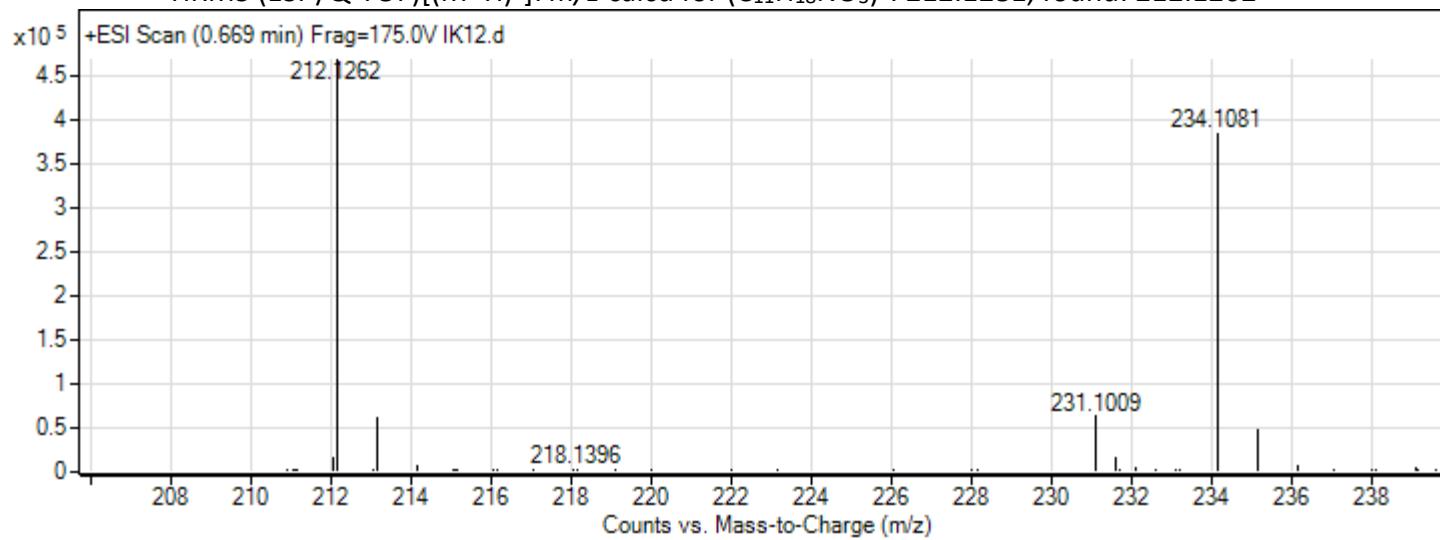
¹H NMR (CDCl₃)

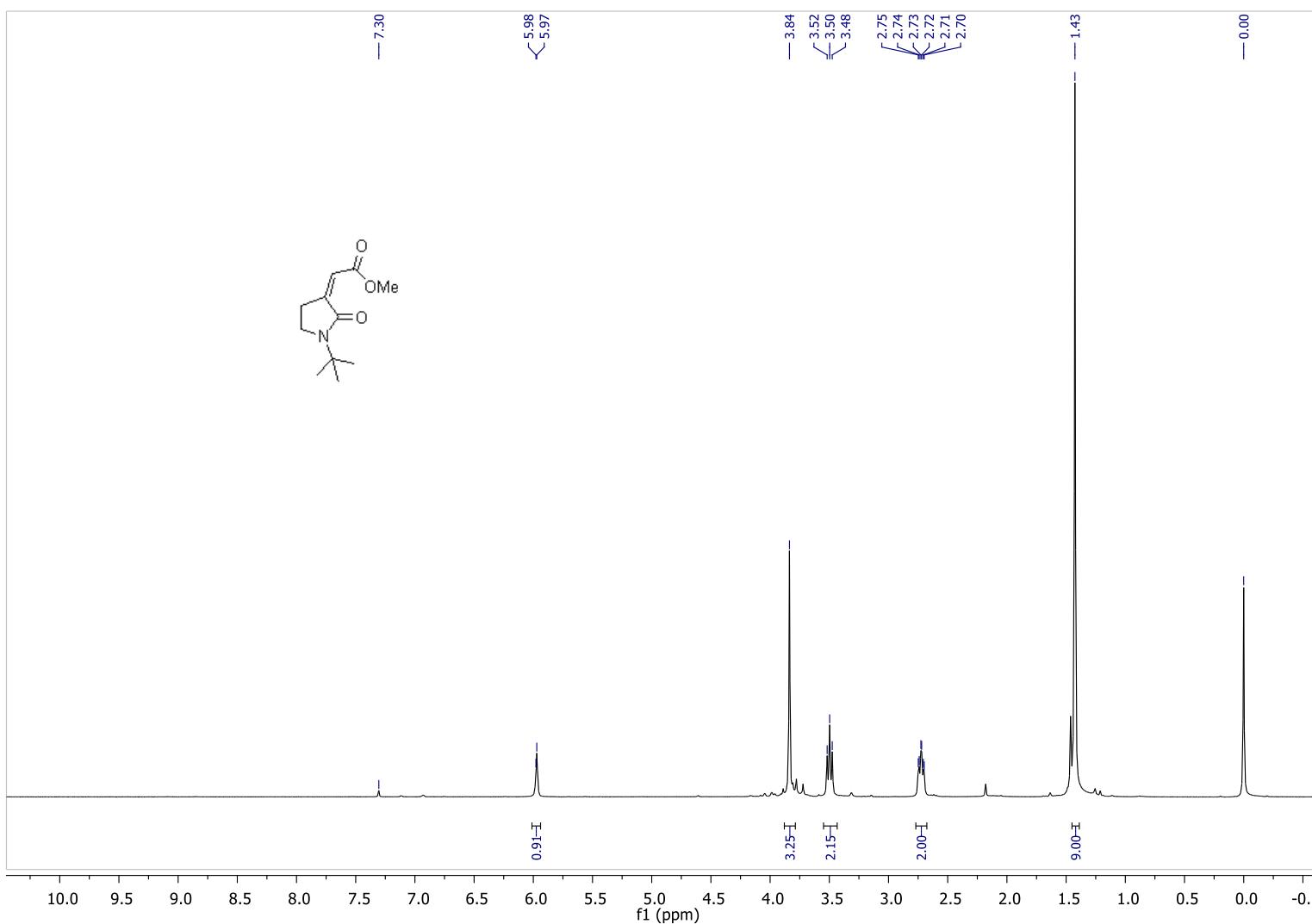
$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

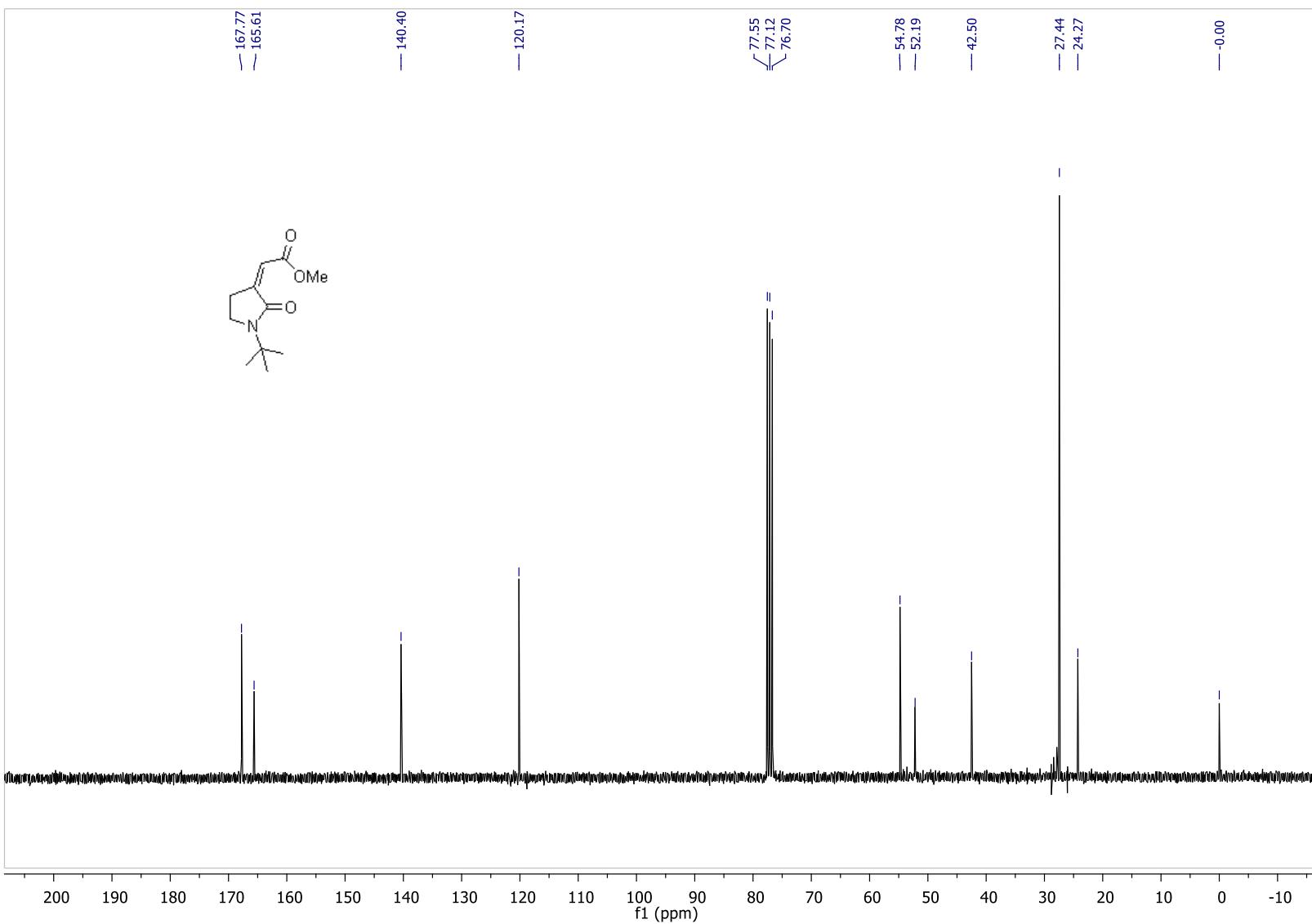
(Z)-Methyl 2-(2-oxo-1-(1-phenylethyl)pyrrolidin-3-ylidene)acetate **2m**

^1H NMR (CDCl_3)

$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

(Z)-Methyl 2-(1-(tert-butyl)-2-oxopyrrolidin-3-ylidene)acetate **2n**HRMS (ESI⁺/Q-TOF)[(M+H)⁺]: *m/z* calcd for (C₁₁H₁₈NO₃)⁺: 212.1281, found: 212.1262

^1H NMR (CDCl_3)

$^{13}\text{C}\{\text{H}\}$ NMR (CDCl_3)

References

- (1) X-ray Crystallographic Information file 2a.cif contains the supplementary crystallographic data for product **2a**, and it is supplied as an independent Supporting Information file for this article. This file can also be obtained free of charge from the Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif (CCDC 1997832).
- (2) Sheldrick, G. M. (Bruker, 2014). SADABS (Version 2008/1). Bruker AXS Inc., Madison, Winsconsin, USA.
- (3) Burla, M.C.; Caliandro, R.; Carrozzini, B.; Cascarano, G.; Cuocci, C.; Giacovazzo, C.; Mallamo, M.; Mazzone, A.; Polidori, G. Crystal structure determination and refinement via SIR2014. *J. Appl. Cryst.* **2015**, *48*, 306-309.
- (4) Sheldrick, G. M. SHELXT - Integrated space-group and crystal-structure determination. *Acta Cryst.* **2015**, *A71*, 3-8.
- (5) X-ray Crystallographic Information file 2b.cif contains the supplementary crystallographic data for product **2b**, and it is supplied as an independent Supporting Information file for this article. This file can also be obtained free of charge from the Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif (CCDC 1997833).