Supporting Information

Cleavage and Reassembly C≡C Bonds of Ynones to Access Highly Functionalized Ketones

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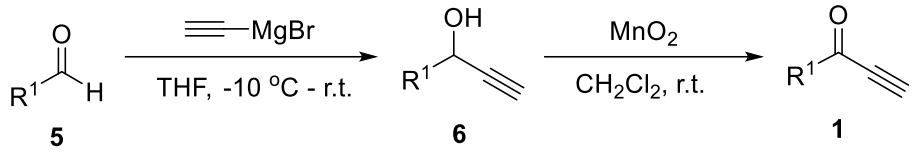
Table of Contents

General methods:

All reactions were carried out in flame or oven-dried glassware under nitrogen atmosphere with freshly distilled dry solvents under anhydrous conditions unless otherwise indicated. Flash column chromatography was performed with silica gel (200 - 300 mesh). Chromatograms were visualized by fluorescence quenching with UV light at 254 nm or by staining with base solution of potassium permanganate and molybdate. NMR spectra were recorded at room temperature on 400 MHz Bruker spectrometers and 400 MHz JEOL spectrometers. The residual solvent signals were taken as the reference (0.00 ppm for ^{1}H NMR spectra and 77.0 ppm for ^{13}C NMR spectra in $CDCl_3$). Chemical shift (δ) is reported in ppm, coupling constants (J) are given in Hz. The following abbreviations classify the multiplicity: s = singlet, d = doublet, t = triplet, m = multiplet, dd = doublet of doublet. HRMS (ESI) spectra were recorded on a Waters Q-Tof premier TM mass spectrometer.

General procedure for ynones 1 and their spectral data:

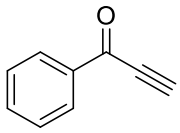


5 6 1

Ethynylmagnesium bromide (0.50 M solution in THF 12 mL, 1.2 equiv.) was added dropwise to a stirred solution of aldehyde **5** (5.0 mmol, 1.0 equiv.) in anhydrous THF (10 mL) at -10 ºC. The mixture was kept stirring at -10 ºC for 20 minutes and warmed to room temperature for additional two hours. Then the reaction mixture was quenched with saturated ammonium chloride (20 mL) and extracted with ethyl acetate for three times. The organic layers was combined and dried with anhydrous $MgSO_4$. It was evaporated under reduced pressure to give intermediate alcohol **6**, which was used in next step directly.

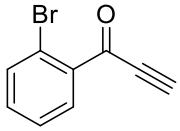
The crude alcohol **6** was dissolved in dichloromethane and treated with activated MnO_2 (10 equiv.). After completion of the reaction (monitored by TLC), the solid was removed by filtration of the reaction mixture through a pad of celite. Then it was extracted with ethyl acetate and dried with anhydrous Na_2SO_4. Then the reaction mixture was concentrated under reduced pressure and purified by column chromatography to give the desired ynones **1**.

1-Phenylprop-2-yn-1-one (1a):



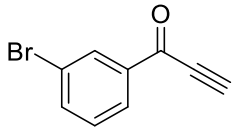
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.38). The product was obtained as yellow solid in 86% yield (559.0 mg), Mp. 44 - 45 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.18 - 8.16 (m, 2H), 7.66 - 7.62 (m, 1H), 7.52 - 7.49 (m, 2H), 3.50 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.4, 136.1, 134.5, 129.7, 128.7, 80.8, 80.2; HRMS (ESI) m/z $[M+H]^+$: Calcd for C_9H_7O: 131.0497. Found: 131.0495.

1-(2-bromophenyl)prop-2-yn-1-one (1b):



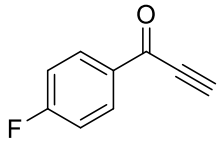
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.60). The product was obtained as yellow oil in 93% yield (967.2 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.13 - 8.11 (d, J = 7.6 Hz, 1H), 7.70 - 7.68 (d, J = 7.6 Hz, 1H), 7.48 - 7.38 (m, 2H), 3.54 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.3, 135.7, 135.1, 133.8, 133.7, 127.3, 121.2, 81.6, 80.7; HRMS (ESI) m/z $[M+H]^+$: Calcd for C_9H_6BrO: 208.9602. Found: 208.9597.

1-(3-bromophenyl)prop-2-yn-1-one (1c):



The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.41). The product was obtained as yellow oil in 89% yield (925.6 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.27 (s, 1H), 8.09 - 8.08 (d, J = 7.6 Hz, 2H), 7.76 - 7.74 (d, J = 8.0 Hz, 2H), 7.41 - 7.37 (m, 1H), 3.53 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 175.8, 137.7, 137.3, 132.4, 130.2, 128.1, 122.9, 81.7, 79.7; HRMS (ESI) m/z $[M+H]^+$: Calcd for C_9H_6BrO: 208.9602. Found: 208.9599.

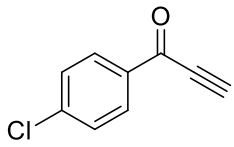
1-(4-Fluorophenyl)prop-2-yn-1-one (1d):



The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.55). The product was obtained as yellow solid in 70% yield (518.0 mg), Mp. 43 - 44 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.21 - 8.17 (m, 2H), 7.19 - 7.15 (t, J = 8.4 Hz, 2H), 3.50 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 175.7, 166.6 (d, J = 256.0 Hz), 132.6

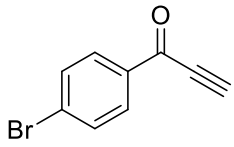
(d, J = 2.9 Hz), 132.3 (d, J = 9.8 Hz), 115.9 (d, J = 22.2 Hz), 81.1, 79.9; ^{19}F NMR (376 MHz, $CDCl_3$) δ -102.2; HRMS (ESI) m/z $[M+H]^+$: Calcd for C_9H_6FO: 149.0403. Found: 149.0403.

1-(4-Chlorophenyl)prop-2-yn-1-one (1e):



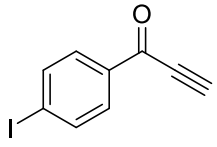
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.58). The product was obtained as yellow solid in 75% yield (615.0 mg), Mp. 94 - 95 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.11 - 8.09 (d, J = 8.4 Hz, 2H), 7.49 - 7.47 (d, J = 8.4 Hz, 2H), 3.49 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.1, 141.2, 134.5, 130.1, 129.1, 81.3, 79.9; HRMS (ESI) m/z $[M+H]^+$: Calcd for C_9H_6ClO: 165.0107. Found: 165.0109.

1-(4-Bromophenyl)prop-2-yn-1-one (1f):



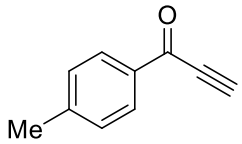
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.73). The product was obtained as yellow solid in 98% yield (1019.2 mg), Mp. 109 - 110 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.03 - 8.00 (d, J = 7.6 Hz, 2H), 7.66 - 7.64 (d, J = 8.0 Hz, 2H), 3.49 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.3, 134.9, 132.1, 131.0, 130.1, 81.4, 79.9; HRMS (ESI) m/z $[M+H]^+$: Calcd for C_9H_6BrO: 208.9602. Found: 208.9607.

1-(4-Iodophenyl)prop-2-yn-1-one (1g):



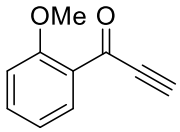
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.68). The product was obtained as yellow solid in 58% yield (742.1 mg), Mp. 117 - 118 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.93 - 7.82 (m, 4H), 3.48 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.6, 138.1, 135.4, 130.8, 103.2, 81.4, 79.9; HRMS (ESI) m/z $[M+H]^+$: Calcd for C_9H_6IO: 256.9463. Found: 256.6466.

1-(*p*-Tolyl)prop-2-yn-1-one (1h):



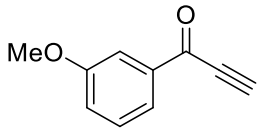
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.54). The product was obtained as yellow solid in 95% yield (684.5 mg), Mp. 35 - 36 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.97 - 7.95 (m, 2H), 7.21 - 7.19 (m, 2H), 3.34 (s, 1H), 2.34 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.0, 145.7, 133.8, 129.8, 129.3, 80.4, 80.3, 21.8; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{10}H_9O$: 145.0653. Found: 145.0655.

1-(2-methoxyphenyl)prop-2-yn-1-one (1i):



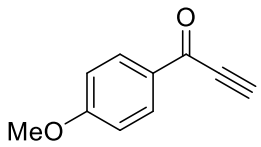
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.58). The product was obtained as yellow oil in 91% yield (728.5 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.07 - 8.05 (m, 1H), 7.57 - 7.53 (m, 1H), 7.06 - 7.00 (m, 2H), 3.93 (s, 3H), 3.39 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.0, 160.0, 135.5, 133.2, 125.7, 120.2, 112.1, 82.1, 79.3, 55.8; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{10}H_9O_2$: 161.0603. Found: 161.0600.

1-(3-Methoxyphenyl)prop-2-yn-1-one (1j):



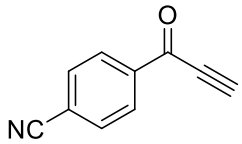
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.58). The product was obtained as yellow oil in 83% yield (664.4 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.81 - 7.79 (m, 1H), 7.64 - 7.63 (m, 1H), 7.43 - 7.39 (t, J = 8 Hz, 1H), 7.20 - 7.17 (m, 1H), 3.87 (s, 3H), 3.45 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.1, 159.7, 137.4, 129.7, 123.0, 121.4, 112.7, 80.7, 80.2, 55.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{10}H_9O_2$: 161.0603. Found: 161.0605.

1-(4-Methoxyphenyl)prop-2-yn-1-one (1k):



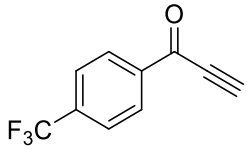
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.45). The product was obtained as yellow solid in 80% yield (640.4 mg), Mp. 72 - 73 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.14 - 8.12 (m, 2H), 6.97 - 6.95 (m, 2H), 3.89 (s, 3H), 3.40 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 175.9, 164.7, 132.1 129.5, 113.9, 80.3, 80.1, 55.6; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{10}H_9O_2$: 161.0603. Found: 161.0600.

4-Propioloylbenzonitrile (1l):



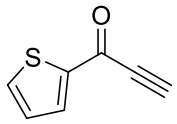
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.34). The product was obtained as yellow solid in 86% yield (666.5 mg), Mp. 99 - 100 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.28 - 8.26 (d, J = 8.8 Hz, 2H), 7.84 - 7.82 (d, J = 8.4 Hz, 2H), 3.62 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 175.7, 138.7, 132.5, 129.9, 117.6, 117.5, 82.7, 79.5; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{10}H_6NO$: 156.0449. Found: 156.0451.

1-(4-(Trifluoromethyl)phenyl)prop-2-yn-1-one (1m):



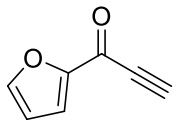
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.68). The product was obtained as yellow solid in 59% yield (584.1 mg), Mp. 65 - 66 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.28 - 8.26 (m, 2H), 7.78 - 7.76 (m, 2H), 3.57 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.2, 138.5, 135.5 (q, J = 32.5 Hz), 129.9, 125.7 (q, J = 3.7 Hz), 123.4 (q, J = 271.4 Hz), 82.1, 79.7; ^{19}F NMR (376 MHz, $CDCl_3$) δ -63.2; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{10}H_6F_3O$: 199.0371. Found: 199.0375.

1-(Thiophen-2-yl)prop-2-yn-1-one (1n):



The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.40). The product was obtained as yellow solid in 84% yield (571.2 mg), Mp. 39 - 40 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.99 - 7.98 (d, J = 3.2 Hz, 1H), 7.77 - 7.76 (d, J = 4.8 Hz, 1H), 7.20 - 7.18 (m, 1H), 3.40 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 169.1, 144.0, 136.2, 136.0, 128.4, 79.8, 79.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for C_7H_5OS: 137.0061. Found: 137.0065.

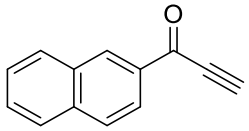
1-(Furan-2-yl)prop-2-yn-1-one (1o):



The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.40). The product was obtained as yellow solid in 77% yield (462.0 mg), Mp. 44 - 45 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.63 (s, 1H), 7.35 (m, 1H), 6.54 (m, 1H), 3.29 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 163.7, 152.6, 148.5, 122.1, 112.8, 79.5, 79.5;

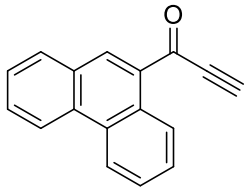
HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_7H_5O_2$: 121.0290. Found: 121.0287.

1-(Naphthalen-2-yl)prop-2-yn-1-one (1p):



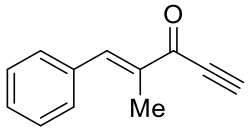
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.63). The product was obtained as yellow solid in 90% yield (810.5 mg), Mp. 91 - 92 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.73 (s, 1H), 8.13 - 8.11 (m, 1H), 8.00 - 7.98 (m, 1H), 7.89 - 7.86 (m, 2H), 7.65 - 7.59 (m, 1H), 7.57 - 7.55 (m, 1H), 3.51 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.3, 136.2, 133.6, 133.3, 132.3, 129.8, 129.2, 128.6, 127.9, 127.0, 123.5, 80.7, 80.3; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{13}H_9O$: 181.0653. Found: 181.0655.

1-(Phenanthren-9-yl)prop-2-yn-1-one (1q):



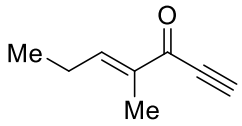
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.55). The product was obtained as yellow solid in 67% yield (770.8 mg), Mp. 89 - 90 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.80 - 9.15 (m, 1H), 8.83 (s, 1H), 8.63 - 8.57 (m, 2H), 7.96 - 7.94 (d, J = 8.0 Hz, 1H), 7.74 - 7.70 (m, 1H), 7.64 - 7.56 (m, 3H), 3.42 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 178.7, 139.0, 133.0, 130.9, 130.7, 130.6, 130.3, 129.7, 128.3, 128.2, 127.5, 127.3, 126.7, 122.8, 122.7, 81.6, 79.5; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{11}O$: 231.0810. Found: 231.0805.

(*E*)-2-Methyl-1-phenylpent-1-en-4-yn-3-one (1r):



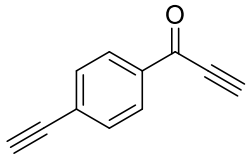
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.69). The product was obtained as yellow solid in 61% yield (518.8 mg), Mp. 43 - 44 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.97 (s, 1H), 7.43 - 7.30 (m, 5H), 3.27 (s, 1H), 2.04 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 180.0, 146.6, 137.6, 135.2, 130.1, 129.4, 128.6, 80.0, 79.7, 12.0; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{12}H_{11}O$: 171.0810. Found: 171.0811.

(*E*)-4-Methylhept-4-en-1-yn-3-one (1s):



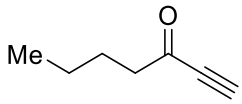
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.80). The product was obtained as yellow oil in 70% yield (427.4 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.20 - 7.17 (t, J = 7.2 Hz, 1H), 3.24 (s, 1H), 2.38 - 2.30 (m, 2H), 1.80 (s, 3H), 1.15 - 1.11 (t, J = 7.6 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 179.6, 153.1, 137.5, 79.6, 79.0, 22.7, 12.7, 10.1; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_8H_{11}O$: 123.0810. Found: 123.0811.

1-(4-Ethynylphenyl)prop-2-yn-1-one (1t):



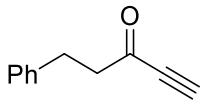
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.60). The product was obtained as yellow solid in 54% yield (415.8 mg), Mp. 97 - 98 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.05 - 8.03 (d, J = 8.0 Hz, 2H), 7.53 - 7.51 (d, J = 8.4 Hz, 2H), 3.41 (s, 1H), 3.24 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.4, 135.7, 132.3, 129.5, 128.3, 82.6, 81.4, 81.3, 80.0; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{11}H_7O$: 155.0497. Found: 155.0501.

Hept-1-yn-3-one (1v):



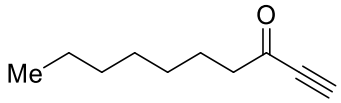
The title compound was prepared according to the general procedure (EA/PE = 1/39, R_f = 0.55). The product was obtained as colorless oil in 35% yield (192.7 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 3.19 (s, 1H), 2.54 - 2.50 (t, J = 7.2 Hz, 2H), 1.63 - 1.55 (m, 2H), 1.33 - 1.24 (m, 2H), 0.87 - 0.83 (t, J = 7.6 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 187.5, 81.3, 78.3, 45.0, 25.7, 21.9, 13.6; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_7H_{11}O$: 111.0810. Found: 111.0814.

5-Phenylpent-1-yn-3-one (1w):



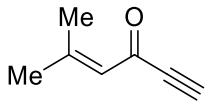
The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.55). The product was obtained as colorless oil in 61% yield (582.3 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.29 - 7.25 (m, 2H), 7.21 - 7.16 (m, 3H), 3.22 (s, 1H), 2.98 - 2.95 (m, 2H), 2.91 - 2.87 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 186.1, 139.8, 128.4, 128.2, 126.2, 81.2, 78.9, 46.7, 29.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{11}H_{11}O$: 159.0810. Found: 159.0812.

Dec-1-yn-3-one (1x):



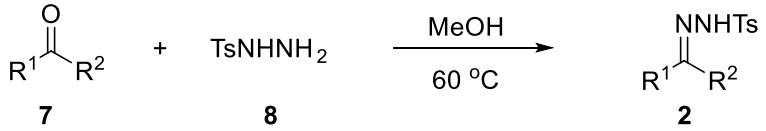
The title compound was prepared according to the general procedure (EA/PE = 1/39, R_f = 0.35). The product was obtained as colorless oil in 47% yield (357.4 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 3.20 (s, 1H), 2.60 - 2.57 (t, J = 7.6 Hz, 2H), 1.70 - 1.66 (m, 2H), 1.31 - 1.28 (m, 8H), 0.90 - 0.87 (t, J = 6.8 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 187.6, 81.4, 78.2, 45.4, 31.5, 28.9, 28.8, 23.7, 22.5, 14.0; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{10}H_{17}O$: 153.1279. Found: 153.1277.

5-Methylhex-4-en-1-yn-3-one (1z):



The title compound was prepared according to the general procedure (EA/PE = 1/9, R_f = 0.80). The product was obtained as colorless oil in 83% yield (448.7 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 6.20 (m, 1H), 3.18 (s, 1H), 2.23 (d, J = 0.8 Hz, 3H), 1.96 (d, J = 1.2 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 175.5, 159.7, 125.2, 83.6, 76.7, 27.8, 21.2; HRMS (ESI) m/z $[M+H]^+$: Calcd for C_7H_9O: 109.0653. Found: 109.0650.

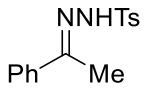
General procedure for synthesis of *N*-tosylhydrazones:



7 **8** **2**

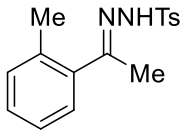
The carbonyl compound **7** (10 mmol, 1.0 equiv.) was added to a solution of $TsNHNH_2$ **8** (10 mmol, 1.0 equiv.) in methanol (20 mL) at 60 ºC. The reaction mixture was then stirred at 60 °C until 7 was consumed completely (monitor by TLC). The solution was cooled down to 0 °C, at which point the product precipitated out of solution in most cases (precipitation can be induced by addition of pentane). The precipitate was collected by vacuum filtration and washed with pentane to give the pure *N*-tosylhydrazones **2**.

(*E*)-4-Methyl-*N*'-(1-phenylethylidene)benzenesulfonohydrazide (2a):



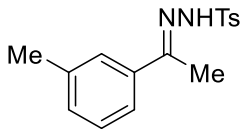
The title compound was prepared according to the general procedure. The product was obtained as white solid in 95% yield (2737.0 mg), Mp. 128 - 129 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.25 (s, 1H), 7.95 - 7.93 (m, 2H), 7.64 - 7.62 (m, 2H), 7.33 - 7.30 (m, 5H), 2.39 (s, 3H), 2.16 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 152.7, 144.1, 137.2, 135.3, 129.5, 129.4, 128.2, 128.0, 126.2, 21.5, 13.5; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{15}H_{17}N_2O_2S$: 289.1011. Found: 289.1014.

(*E*)-4-Methyl-*N′*-(1-(*o*-tolyl)ethylidene)benzenesulfonohydrazide (2b):



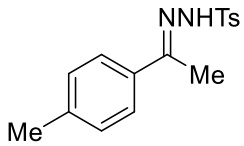
The title compound was prepared according to the general procedure. The product was obtained as white solid in 95% yield (2870.0 mg), Mp. 115 - 116 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.28 (s, 1H), 7.91 - 7.89 (m, 2H), 7.32 - 7.30 (m, 2H), 7.22 - 7.18 (m, 1H), 7.15 - 7.09 (m, 3H), 2.42 (s, 3H), 2.14 (s, 3H), 2.14 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 155.6, 144.1, 138.5, 135.7, 135.5, 130.8, 129.6, 128.5, 128.1, 128.0, 125.6, 21.6, 20.3, 17.4, 17.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{16}H_{19}N_2O_2S$: 303.1167. Found: 303.1171.

(*E*)-4-Methyl-*N′*-(1-(*m*-tolyl)ethylidene)benzenesulfonohydrazide (2c):



The title compound was prepared according to the general procedure. The product was obtained as white solid in 93% yield (2808.5 mg), Mp. 120 - 121 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.51 (s, 1H), 7.96 - 7.94 (m, 2H), 7.42 (s, 2H), 7.28 - 7.27 (m, 2H), 7.19 - 7.13 (m, 2H), 2.36 (s, 3H), 2.31 (s, 3H), 2.14 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 153.0, 143.9, 137.6, 137.1, 135.2, 130.1, 129.4, 128.0, 127.9, 126.8, 123.3, 21.4, 21.3, 13.6; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{16}H_{19}N_2O_2S$: 303.1167. Found: 303.1168.

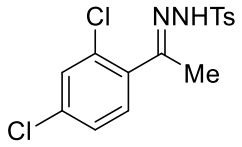
(*E*)-4-Methyl-*N′*-(1-(*p*-tolyl)ethylidene)benzenesulfonohydrazide (2d):



The title compound was prepared according to the general procedure. The product was obtained as white solid in 91% yield (2749.1 mg), Mp. 157 - 158 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.02 (s, 1H), 7.93 - 7.91 (m, 2H), 7.55 - 7.53 (m, 2H), 7.31 - 7.30 (m,

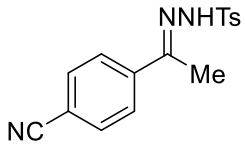
2H), 7.14 - 7.12 (m, 2H), 2.40 (s, 3H), 2.34 (s, 3H), 2.14 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 152.8, 144.0, 139.6, 135.3, 134.4, 129.5, 128.9, 128.0, 126.1, 21.6, 21.2, 13.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{16}H_{19}N_2O_2S$: 303.1167. Found: 303.1168.

(*E*)-*N'*-(1-(2,4-Dichlorophenyl)ethylidene)-4-methylbenzenesulfonohydrazide (2i):



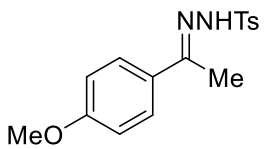
The title compound was prepared according to the general procedure. The product was obtained as white solid in 98% yield (3488.4 mg), Mp. 164 - 165 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.30 (s, 1H), 7.89 - 7.87 (m, 2H), 7.34 - 7.32 (m, 3H), 7.21 - 7.19 (m, 1H), 7.16 - 7.14 (m, 1H), 2.44 (s, 3H), 2.17 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 152.5, 144.3, 136.6, 135.2, 132.9, 131.1, 129.6, 128.0, 127.1, 21.6, 17.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{15}H_{15}Cl_2N_2O_2S$: 357.0231. Found: 357.0233.

(*E*)-*N*'-(1-(4-cyanophenyl)ethylidene)-4-methylbenzenesulfonohydrazide (2l):



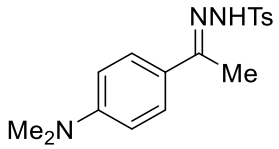
The title compound was prepared according to the general procedure. The product was obtained as white solid in 88% yield (2755.3 mg), Mp. 189 - 190 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.16 (s, 1H), 7.92 - 7.90 (m, 2H), 7.75 - 7.73 (m, 2H), 7.64 - 7.62 (m, 2H), 7.35 - 7.33 (m, 2H), 2.43 (s, 3H), 2.17 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 149.9, 144.6, 141.3, 135.0, 132.1, 129.7, 128.0, 126.7, 118.5, 112.8, 21.6, 13.2; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{16}H_{16}N_3O_2S$: 314.0963. Found: 314.0961.

(*E*)-*N*'-(1-(4-methoxyphenyl)ethylidene)-4-methylbenzenesulfonohydrazide (2m):



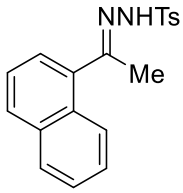
The title compound was prepared according to the general procedure. The product was obtained as white solid in 89% yield (2831.1 mg), Mp. 156 - 157 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.97 (s, 1H), 7.94 - 7.92 (m, 2H), 7.60 - 7.58 (m, 2H), 7.32 - 7.30 (m, 2H), 6.85 - 6.83 (m, 2H), 3.80 (s, 3H), 2.40 (s, 3H), 2.13 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 160.7, 152.7, 144.0, 135.4, 129.8, 129.5, 128.1, 127.7, 113.6, 55.3, 21.6, 13.3; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{16}H_{19}N_2O_3S$: 319.1116. Found: 319.1118.

(*E*)-*N*'-(1-(4-(Dimethylamino)phenyl)ethylidene)-4-methylbenzenesulfonohydrazide (2n):



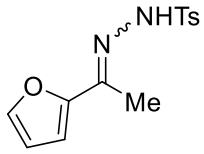
The title compound was prepared according to the general procedure. The product was obtained as white solid in 85% yield (2814.4 mg), Mp. 158 - 159 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.93 - 7.91 (m, 2H), 7.57 - 7.55 (m, 2H), 7.53 (s, 1H), 7.31 - 7.29 (m, 2H), 6.64 - 6.62 (m, 2H), 2.97 (s, 6H), 2.40 (s, 3H), 2.10 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 153.6, 151.3, 143.8, 135.5, 129.5, 128.1, 127.4, 124.8, 111.4, 40.1, 21.6, 13.0; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{22}N_3O_2S$: 332.1433. Found: 332.1433.

(*E*)-4-Methyl-*N*'-(1-(naphthalen-1-yl)ethylidene)benzenesulfonohydrazide (2o):



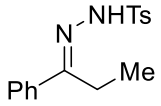
The title compound was prepared according to the general procedure. The product was obtained as white solid in 45% yield (1521.5 mg), Mp. 163 - 164 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.95 - 7.91 (m, 3H), 7.85 - 7.79 (m, 3H), 7.48 - 7.40 (m, 2H), 7.35 - 7.33 (m, 4H), 2.46 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 154.6, 144.2, 136.5, 135.5, 133.8, 130.3, 129.7, 129.5, 128.4, 128.3, 126.4, 126.2, 125.9, 125.4, 124.9, 21.6, 18.1; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{19}H_{19}N_2O_2S$: 339.1167. Found: 339.1168.

***N'*-(1-(Furan-2-yl)ethylidene)-4-methylbenzenesulfonohydrazide (2p):**



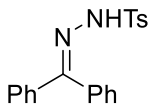
The title compound was prepared according to the general procedure. The product was obtained as white solid in a *cis* : *trans* of 1 : 1 in 87% yield (2419.5 mg), Mp. 139 - 140 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.49 (s, 0.39H), 7.92 - 7.86 (m, 2H), 7.59 - 7.43 (m, 1H), 7.32 - 7.30 (m, 2H), 6.70 - 6.69 (m, 1H), 6.52 - 6.40 (m, 1H), 2.42 & 2.41 (s, 3H), 2.18 & 2.10 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 151.1, 148.7, 145.5, 144.1, 144.1, 144.0, 144.0, 138.2, 135.5, 135.2, 129.5, 129.5, 128.0, 127.9, 114.7, 111.7, 111.5, 110.7, 21.6, 21.6, 20.9, 12.8; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{13}H_{15}N_2O_3S$: 278.0803. Found: 278.0806.

(*E*)-4-Methyl-*N*'-(1-phenylpropylidene)benzenesulfonohydrazide (2r):



The title compound was prepared according to the general procedure. The product was obtained as white solid in 88% yield (2649.7 mg), Mp. 109 - 110 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.80 - 7.78 (m, 2H), 7.44 - 7.40 (m, 2H), 7.36 (s, 1H), 7.33 - 7.31 (m, 2H), 7.08 - 7.06 (m, 2H), 2.51 - 2.46 (q, J = 7.6 Hz, 2H), 2.44 (s, 3H), 1.04 - 1.00 (t, J = 7.6 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 159.1, 143.9, 135.2, 132.8, 129.7, 129.5, 129.4, 127.8, 126.6, 31.4, 21.6, 10.6; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{16}H_{19}N_2O_2S$: 302.1167. Found: 302.1169.

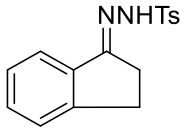
***N*'-(Diphenylmethylene)-4-methylbenzenesulfonohydrazide (2s):**



The title compound was prepared according to the general procedure. The product was obtained as white solid in 88% yield (3080.9 mg), Mp. 134 - 135 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.87 - 7.85 (m, 2H), 7.56 - 7.51 (m, 4H), 7.45 - 7.43 (m, 2H), 7.34 -

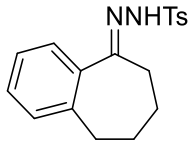
7.28 (m, 5H), 7.13 - 7.11 (m, 2H), 2.43 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 154.2, 144.1, 136.4, 135.4, 131.1, 130.1, 129.8, 129.7, 129.6, 128.2, 128.1, 127.9, 127.5, 21.6; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{20}H_{19}N_2O_2S$: 351.1167. Found: 351.1167.

(*E*)-N'-(2,3-dihydro-1H-inden-1-ylidene)-4-methylbenzenesulfonohydrazide (2u):



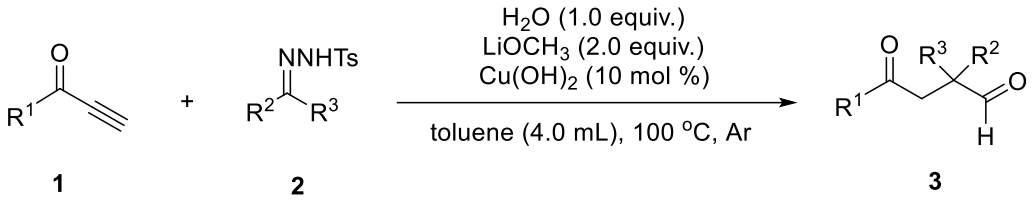
The title compound was prepared according to the general procedure. The product was obtained as white solid in 60% yield (1800.6 mg), Mp. 183 - 184 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.94 - 7.92 (m, 2H), 7.76 (s, 1H), 7.71 - 7.69 (m, 1H), 7.34 - 7.30 (m, 3H), 7.27 - 7.21 (m, 2H), 3.07 - 3.04 (m, 2H), 2.64 - 2.65 (m, 2H), 2.40 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 162.3, 148.3, 144.0, 137.0, 135.4, 130.9, 129.5, 128.0, 127.0, 125.3, 122.1, 28.4, 26.6, 21.6; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{16}H_{17}N_2O_2S$: 301.1011. Found: 301.1011.

(*E*)-4-Methyl-*N*'-(6,7,8,9-tetrahydro-5*H*-benzo[7]annulen-5-ylidene)benzenesulfonohydrazide (2w):



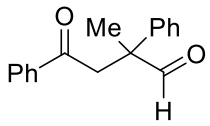
The title compound was prepared according to the general procedure. The product was obtained as white solid in 68% yield (2231.1 mg), Mp. 139 - 140 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.26 (s, 1H), 7.85 - 7.83 (m, 2H), 7.24 - 7.22 (m, 2H), 7.19 - 7.15 (m, 2H), 7.11 - 7.07 (m, 1H), 6.97 - 6.95 (m, 1H), 2.56 - 2.52 (t, J = 6.8 Hz, 2H), 2.35 - 2.32 (m, 5H), 1.65 - 1.58 (m, 2H), 1.50 - 1.44 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 160.5, 144.0, 138.7, 137.2, 135.3, 129.5, 129.4, 128.4, 128.0, 127.9, 126.4, 31.1, 27.4, 25.3, 21.5, 20.7; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{21}N_2O_2S$: 329.1324. Found: 329.1328.

General procedure for 1,4-ketoaldehydes and their spectral data:



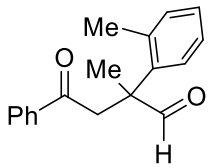
A mixture of **1** (0.20 mmol, 1.0 equiv.), **2** (0.40 mmol, 2.0 equiv.), $Cu(OH)_2$ (0.02 mmol, 10 mol %), $LiOCH_3$ (0.40 mmol, 2.0 equiv.), H_2O (3.6 μL, 1.0 equiv.) and toluene (4.0 mL) was sealed in a Schlenk tube under argon protection at 100 °C and the mixture was stirred for 24 h or until the **1** was consumed completely. Then the reaction mixture was concentrated under reduced pressure and purified by flash column chromatography to give the desired product **3**.

2-Methyl-4-oxo-2,4-diphenylbutanal (3aa):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.45). The product was obtained as yellow oil in 94% yield (47.4 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.73 (s, 1H), 7.94 - 7.92 (m, 2H), 7.57 - 7.54 (m, 2H), 7.45 - 7.42 (m, 2H), 7.39 - 7.34 (m, 4H), 7.29 -7.25 (m, 1 H), 3.75 (d, J = 17.6 Hz, 1H), 3.70 (d, J = 17.6 Hz, 1H), 1.68 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.6, 197.3, 139.9, 136.9, 133.5, 129.0, 128.7, 128.2, 127.5, 127.0, 51.9, 46.2, 20.9; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{17}O_2$: 253.1229. Found: 253.1228.

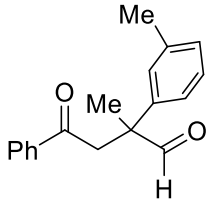
2-Methyl-4-oxo-4-phenyl-2-(*o*-tolyl)butanal (3ab):



The title compound was prepared according to the general procedure for reaction in 24

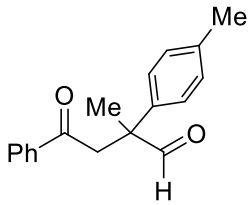
h (EA/PE = 1/9, R_f = 0.60). The product was obtained as yellow oil in yield: 68% (36.2 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.92 (s, 1H), 7.89 - 7.87 (m, 2H), 7.56 - 7.53 (m, 1H), 7.44 - 7.40 (m, 2H), 7.27 - 7.24 (m, 1H), 7.20 - 7.16 (m, 3H), 3.84 (d, J = 17.2 Hz, 1H), 3.71 (d, J = 17.2 Hz, 1H), 2.31 (s, 3H), 1.65 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 204.0, 197.6, 138.5, 136.9, 135.7, 133.2, 132.4, 128.6, 128.0, 127.6, 127.5, 126.3, 52.6, 45.8, 22.0, 21.5; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{19}O_2$: 267.1385. Found: 267.1387.

2-Methyl-4-oxo-4-phenyl-2-(*m*-tolyl)butanal (3ac):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.47). The product was obtained as yellow oil in 81% yield (43.2 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.71 (s, 1H), 7.95 - 7.93 (m, 2H), 7.58 - 7.54 (m, 1H), 7.46 - 7.43 (m, 2H), 7.28 - 7.24 (m, 1H), 7.14 - 7.12 (m, 2 H), 7.10 - 7.08 (m, 1H), 3.74 (d, J = 17.6 Hz, 1H), 3.39 (d, J = 17.6 Hz, 1H), 2.34 (s, 3H), 1.67 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.4, 197.3, 139.5, 138.5, 136.7, 133.3, 128.7, 128.6, 128.1, 128.0, 127.6, 123.8, 51.6, 45.9, 21.6, 20.5; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{19}O_2$: 267.1385. Found: 267.1385.

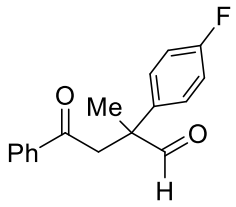
2-Methyl-4-oxo-4-phenyl-2-(*p*-tolyl)butanal (3ad):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.39). The product was obtained as yellow oil in 91% yield (48.5 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.69 (s, 1H), 7.94 - 7.93 (m, 2H), 7.57 - 7.54 (m,

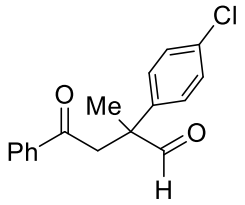
1H), 7.46 - 7.42 (m, 2H), 7.24 - 7.16 (m, 4H), 3.73 (d, J = 17.6 Hz, 1H), 3.68 (d, J = 17.6 Hz, 1H), 2.32 (s, 3H), 1.65 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.4, 197.2, 137.1, 136.8, 136.5, 133.3, 129.6, 128.5, 128.0, 126.7, 51.4, 45.9, 20.9, 20.6; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{19}O_2$: 267.1385. Found: 267.1385.

2-(4-Fluorophenyl)-2-methyl-4-oxo-4-phenylbutanal (3ae):



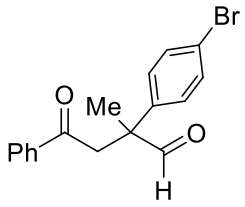
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.57). The product was obtained as yellow oil in 78% yield (42.2 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.70 (s, 1H), 7.94 - 7.92 (m, 2H), 7.59 - 7.57 (m, 1H), 7.56 - 7.55 (m, 1H), 7.47 - 7.43 (m, 2H), 7.32 - 7.29 (m, 2H), 7.08 - 7.04 (m, 2H), 3.71 (s, 2H), 1.66 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.2, 197.0, 163.1, 160.7, 136.6, 135.3, 133.5, 128.6 (J = 8.0 Hz), 128.3 (J = 59.8 Hz) , 115.7 (J = 21.1 Hz), 51.2, 46.1, 20.8; ^{19}F NMR (376 MHz, $CDCl_3$) δ -114.9; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{16}FO_2$: 271.1134. Found: 271.1134.

2-(4-Chlorophenyl)-2-methyl-4-oxo-4-phenylbutanal (3af):



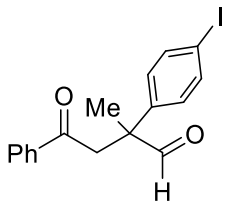
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.48). The product was obtained as yellow oil in 69% yield (34.5 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.70 (s, 1H), 7.94 - 7.92 (m, 2H), 7.59 - 7.56 (m, 1H), 7.47 - 7.43 (m, 2H), 7.34 - 7.26 (m, 4H), 3.71 (s, 2H), 1.66 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.0, 196.8, 138.2, 136.5, 133.5, 133.4, 129.0, 128.6, 128.3, 128.0, 51.3, 46.0, 20.8; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{16}ClO_2$: 287.0839. Found: 287.0840.

2-(4-Bromophenyl)-2-methyl-4-oxo-4-phenylbutanal (3ag):



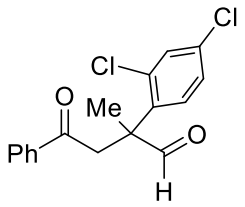
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.31). The product was obtained as yellow oil in 86% yield (56.8 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.71 (s, 1H), 7.94 - 7.92 (d, 2H), 7.59 - 7.55 (m, 1H), 7.50 - 7.43 (m, 4H), 7.22 - 7.20 (m, 2H), 3.71 (s, 2H), 1.65 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.0, 196.8, 138.8, 136.4, 133.5, 131.9, 128.6, 128.0, 121.5, 51.4, 46.0, 20.8; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{16}BrO_2$: 331.0334. Found: 331.0332.

2-(4-Iodophenyl)-2-methyl-4-oxo-4-phenylbutanal (3ah):



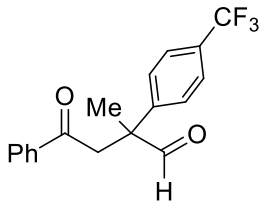
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.52). The product was obtained as yellow oil in 69% yield (52.2 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.70 (s, 1H), 7.94 - 7.92 (m, 2H), 7.69 - 7.67 (m, 2H), 7.59 - 7.56 (m, 1H), 7.47 - 7.43 (m, 2H), 7.09 - 7.07 (m, 2H), 3.71 (s, 2H), 1.64 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.0, 196.8, 139.5, 137.9, 136.4, 133.5, 128.9, 128.6, 128.0, 51.4, 45.9, 20.8; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{16}IO_2$: 379.0195. Found: 379.0198.

2-(2,4-Dichlorophenyl)-2-methyl-4-oxo-4-phenylbutanal (3ai):



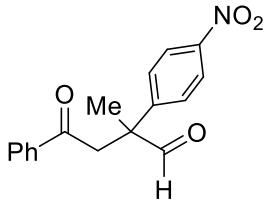
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.58). The product was obtained as yellow oil in 99% yield (63.4 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.89 (s, 1H), 7.91 - 7.89 (m, 2H), 7.58 - 7.55 (m, 1H), 7.46 - 7.42 (m, 2H), 7.39 - 7.34 (m, 2H), 7.28 - 7.25 (m, 1H), 3.91 (d, J = 18.4 Hz, 1H), 3.86 (d, J = 18.4 Hz, 1H), 1.67 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.9, 197.3, 137.1, 136.6, 134.2, 133.5, 133.5, 130.7, 130.5, 128.7, 128.0, 127.4, 52.1, 44.4, 21.3; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{15}Cl_2O_2$: 321.0449. Found: 321.0448.

2-Methyl-4-oxo-4-phenyl-2-(4-(trifluoromethyl)phenyl)butanal (3aj):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.48). The product was obtained as yellow oil in 53% yield (53.9 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.78 (s, 1H), 7.95 - 7.93 (m, 2H), 7.64 - 7.62 (m, 2H), 7.59 - 7.57 (m, 1H), 7.48 - 7.44 (m, 4H), 3.79 (d, J = 18.4 Hz, 1H), 3.75 (d, J = 18.4 Hz, 1H), 1.70 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.0, 196.7, 144.1, 136.4, 133.6, 129.4, 128.7, 128.1, 127.3, 125.8 (q, J = 42.6 Hz), 51.8, 46.2, 21.2; ^{19}F NMR (376 MHz, $CDCl_3$) δ -62.7; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{16}F_3O_2$: 321.1102. Found: 321.1101.

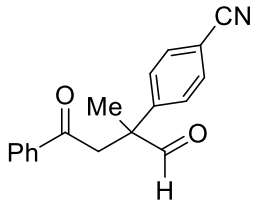
2-Methyl-2-(4-nitrophenyl)-4-oxo-4-phenylbutanal (3ak):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.50). The product was obtained as yellow oil in 32% yield (19.0 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.80 (s, 1H), 8.24 - 8.22 (m, 2H), 7.95 - 7.93 (m, 2H), 7.62 - 7.56 (m, 1H), 7.54 - 7.52 (m, 2H), 7.49 - 7.45 (m, 2H), 3.80 (s, 2H), 1.72 (s,

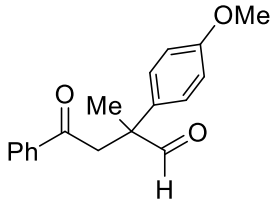
3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 200.5, 196.3, 147.5, 136.2, 133.8, 128.8, 128.1, 128.0, 123.9, 52.0, 46.4, 21.5; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{16}NO_4$: 298.1079. Found: 298.1077.

4-(2-Methyl-1,4-dioxo-4-phenylbutan-2-yl)benzonitrile (3al):



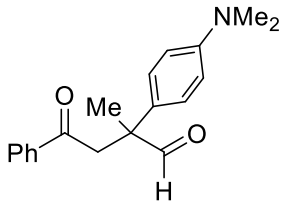
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.15). The product was obtained as yellow oil in 56% yield (31.1 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.77 (s, 1H), 7.94 - 7.92 (m, 2H), 7.68 - 7.66 (m, 2H), 7.62 - 7.58 (m, 1H), 7.49 - 7.46 (m, 4H), 3.77 (s, 2H), 1.69 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 200.6, 196.4, 145.5, 136.2, 133.7, 132.5, 128.8, 128.1, 127.8, 118.4, 111.3, 52.0, 46.2, 21.2; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{16}NO_2$: 278.1181. Found: 278.1183.

2-(4-Methoxyphenyl)-2-methyl-4-oxo-4-phenylbutanal (3am):



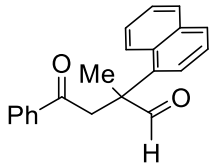
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.60). The product was obtained as yellow oil in 90% yield (50.8 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.65 (s, 1H), 7.94 - 7.92 (m, 2H), 7.58 - 7.54 (m, 1H), 7.46 - 7.44 (m, 2H), 7.26 - 7.24 (m, 2H), 6.91 - 7.89 (m, 2H), 3.79 (s, 3H), 3.69 (s, 2H), 1.66 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.2, 197.3, 158.7, 136.8, 133.3, 131.2, 128.6, 128.1, 114.3, 55.2, 51.1, 45.9, 20.5; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{19}O_3$: 283.1334. Found: 283.1332.

2-(4-(Dimethylamino)phenyl)-2-methyl-4-oxo-4-phenylbutanal (3an):



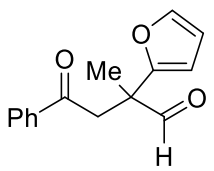
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.20). The product was obtained as yellow oil in 87% yield (51.4 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.54 (s, 1H), 7.88 - 7.86 (m, 2H), 7.52 - 7.47 (m, 1H), 7.40 - 7.36 (m, 2H), 7.16 - 7.14 (m, 2H), 6.73 (s, 2H), 3.63 (d, J = 17.6 Hz, 1H), 3.58 (d, J = 17.6 Hz, 1H), 2.89 (s, 6H), 1.58 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.1, 197.6, 137.0, 133.2, 130.5, 128.6, 128.1, 127.7, 112.8, 110.6, 51.0, 45.7, 40.5, 20.1; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{19}H_{22}NO_2$: 296.1651. Found: 296.1647.

2-Methyl-2-(naphthalen-1-yl)-4-oxo-4-phenylbutanal (3ao):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.80). The product was obtained as yellow oil in 58% yield (35.1 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 10.07 (s, 1H), 7.89 - 7.87 (m, 1H), 7.81 - 7.77 (m, 4H), 7.52 - 7.39 (m, 5H), 7.35 - 7.32 (m, 2H), 4.07 (d, J = 16.8 Hz, 1H), 3.91 (d, J = 16.8 Hz, 1H), 1.82 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 204.8, 197.8, 136.8, 136.1, 134.7, 133.2, 130.9, 129.8, 128.9, 128.5, 128.0, 126.5, 125.8, 125.4, 125.3, 124.1, 52.8, 46.4, 22.7; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{21}H_{19}O_2$: 303.1385. Found: 303.1382.

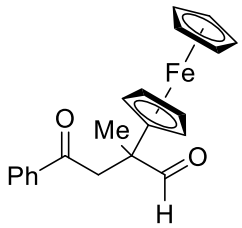
2-(Furan-2-yl)-2-methyl-4-oxo-4-phenylbutanal (3ap):



The title compound was prepared according to the general procedure for reaction in 24

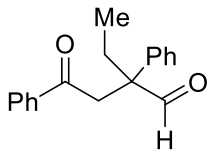
h (EA/PE = 1/9, R_f = 0.51). The product was obtained as yellow oil in 64% yield (31.0 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.74 (s, 1H), 7.96 - 7.94 (m, 2H), 7.60 - 7.56 (m, 1H), 7.48 - 7.44 (m, 2H), 7.38 - 7.37 (m, 1H), 6.36 - 6.34 (m, 1H), 6.25 - 6.24 (m, 1H), 3.81 (d, J = 17.6 Hz, 1H), 3.67 (d, J = 17.6 Hz, 1H), 1.61 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 199.0, 196.9, 153.4, 142.6, 136.5, 133.4, 128.6, 128.1, 110.6, 107.1, 49.1, 44.3, 19.2; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{15}H_{15}O_3$: 243.1021. Found: 243.1023.

2-(Ferrocenyl)-2-methyl-4-oxo-4-phenylbutanal (3aq):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.56). The product was obtained as yellow oil in 38% yield (27.4 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.86 (s, 1H), 7.92 - 7.90 (m, 2H), 7.58 - 7.54 (m, 1H), 7.46 - 7.43 (m, 2H), 4.22 (t, J = 2.0 Hz, 2H), 4.21 (s, 5H), 4.10 - 4.08 (m, 1H), 4.07-4.05 (m, 1H), 3.60 (d, J = 17.6 Hz, 1H), 3.46 (d, J = 17.6 Hz, 1H), 1.59 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 200.0, 197.4, 136.6, 133.3, 128.6, 128.1, 88.4, 68.8, 68.6, 68.3, 66.2, 65.8, 48.3, 46.0, 20.5; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{21}H_{21}FeO_2$: 361.0891. Found: 361.0890.

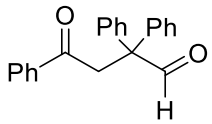
2-Ethyl-4-oxo-2,4-diphenylbutanal (3ar):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.46). The product was obtained as yellow oil in 63% yield (33.6 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.85 (s, 1H), 7.97 - 7.95 (m, 2H), 7.60 - 7.56 (m, 1H), 7.50 - 7.44 (m, 2H), 7.39 - 7.35 (m, 2H), 7.31 - 7.27 (m, 4H), 3.79 (s, 2H), 2.36 - 2.26 (m, 1H), 2.11 - 2.01 (m, 1H), 0.79 - 0.75 (m, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ

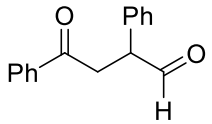
202.8, 197.3, 158.5, 138.5, 136.5, 133.4, 128.8, 128.6, 128.1, 127.3, 127.2, 55.7, 42.2, 27.3, 8.6; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{19}O_2$: 267.1385. Found: 267.1385.

4-oxo-2,2,4-Triphenylbutanal (3as):



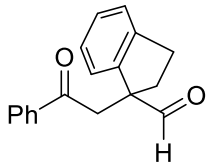
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.53). The product was obtained as yellow oil in 91% yield (57.2 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 10.19 (s, 1H), 7.92 - 7.90 (m, 2H), 7.55 - 7.53 (m, 1H), 7.43 - 7.39 (m, 2H), 7.39 - 7.30 (m, 4H), 7.27 - 7.23 (m, 6H), 4.18 (s, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 199.6, 196.2, 140.4, 136.4, 133.3, 128.7, 128.6, 128.5, 128.1, 127.2, 60.7, 46.5; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{22}H_{19}O_2$: 315.1385. Found: 315.1388.

4-oxo-2,4-Diphenylbutanal (3at):



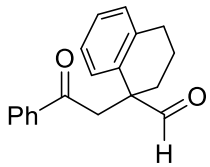
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.41). The product was obtained as yellow oil in 30% yield (14.3 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.81 (s, 1H), 7.99 - 7.97 (m, 2H), 7.59 - 7.55 (m, 1H), 7.48 - 7.44 (m, 2H), 7.41 - 7.39 (m, 2H), 7.38 - 7.34 (m, 1H), 7.32 - 7.29 (m, 2H), 4.48 - 4.45 (m, 1 H), 3.99 - 3.93 (m, 1H), 3.26 - 3.20 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 199.0, 197.3, 136.3, 135.3, 133.3, 129.2, 129.1, 128.6, 128.1, 127.9, 53.6, 39.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{16}H_{15}O_2$: 239.1072. Found: 239.1074.

1-(2-oxo-2-Phenylethyl)-2,3-dihydro-1H-indene-1-carbaldehyde (3au):



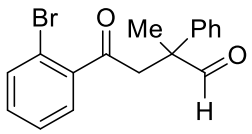
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.65). The product was obtained as yellow oil in 44% yield (23.2 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.71 (s, 1H), 7.88 - 7.86 (m, 2H), 7.51 - 7.47 (m, 1H), 7.39 - 7.36 (m, 2H), 7.24 - 7.15 (m, 4H), 3.90 (d, J = 18.0 Hz, 1H), 3.36 (d, J = 18.0 Hz, 1H), 3.08 - 2.98 (m, 2H), 2.88 - 2.81 (m, 1H), 2.20 - 2.13 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.2, 197.3, 144.8, 141.1, 136.4, 128.6, 128.4, 128.1, 127.0, 125.2, 124.5, 60.6, 46.2, 33.0, 30.9; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{17}O_2$: 265.1229. Found: 265.1227.

1-(2-oxo-2-Phenylethyl)-1,2,3,4-tetrahydronaphthalene-1-carbaldehyde (3av):



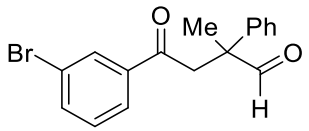
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.56). The product was obtained as yellow oil in 45% yield (25.1 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.76 (s, 1H), 7.94 - 7.92 (m, 2H), 7.58 - 7.54 (m, 1H), 7.46 - 7.42 (m, 2H), 7.26 - 7.25 (m, 1H), 7.20 - 7.16 (m, 3H), 3.78 (d, J = 18.0 Hz, 1H), 3.59 (d, J = 18.0 Hz, 1H), 2.88 - 2.84 (m, 2H), 2.44 - 2.38 (m, 1H), 2.23 - 2.16 (m, 1H), 1.99 - 1.82 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.5, 197.1, 138.7, 136.9, 133.6, 133.2, 130.0, 128.6, 128.2, 128.0, 127.2, 126.5, 51.2, 47.1, 29.9, 28.7, 19.3; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{19}H_{19}O_2$: 279.1385. Found: 279.1383.

4-(2-Bromophenyl)-2-methyl-4-oxo-2-phenylbutanal (3ba):



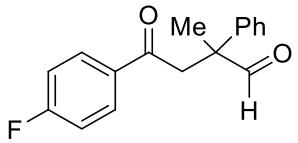
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.44). The product was obtained as yellow oil in 40% yield (26.5 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.57 (s, 1H), 7.51 - 7.48 (m, 1H), 7.27 - 7.23 (m, 2H), 7.22 - 7.21 (m, 3H), 7.20 - 7.17 (m, 3H), 3.60 (d, J = 17.6 Hz, 1H), 3.55 (d, J = 17.6 Hz, 1H), 1.66 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.5, 200.7, 141.5, 138.8, 133.6, 131.6, 128.9, 128.5, 127.5, 127.4, 126.9, 118.4, 52.4, 49.6, 19.9; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{16}BrO_2$: 331.0334. Found: 331.0331.

4-(3-Bromophenyl)-2-methyl-4-oxo-2-phenylbutanal (3ca):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.60). The product was obtained as yellow oil in 66% yield (43.6 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.69 (s, 1H), 8.03 (s, 1H), 7.85 - 7.83 (m, 1H), 7.69 - 7.66 (m, 1H), 7.40 - 7.32 (m, 2H), 7.31 - 7.26 (m, 4H), 3.69 (d, J = 17.6 Hz, 1H), 3.64 (d, J = 17.6 Hz, 1H), 1.70 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.2, 195.8, 139.2, 138.4, 136.1, 131.1, 130.2, 128.9, 127.5, 126.8, 126.5, 122.9, 51.8, 45.9, 20.3; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{16}BrO_2$: 331.0334. Found: 331.0331.

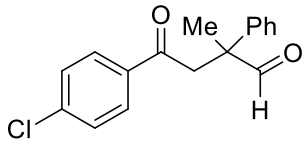
4-(4-Fluorophenyl)-2-methyl-4-oxo-2-phenylbutanal (3da):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.56). The product was obtained as yellow oil in 91% yield (49.2 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.71 (s, 1H), 7.93 (m, 2H), 7.39 - 7.31 (m, 2H), 7.30 - 7.26 (m, 1H), 7.10 - 7.08 (m, 2H), 3.86 (s, 3H), 3.71 (d, J = 17.2 Hz, 1H), 3.66 (d, J = 17.2 Hz, 1H), 1.68 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.4, 195.6, 167.0, 164.5, 133.1 (J = 11.2), 130.8, 130.7, 128.9, 127.4, 126.8, 115.8, 115.6, 51.8, 45.8, 20.5;

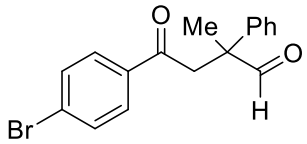
^{19}F NMR (376 MHz, $CDCl_3$) δ -104.5; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{16}FO_2$: 271.1134. Found: 271.1140.

4-(4-Chlorophenyl)-2-methyl-4-oxo-2-phenylbutanal (3ea):



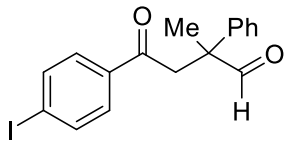
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.59). The product was obtained as yellow solid in 68% yield (38.9 mg), Mp. 70 - 71 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.70 (s, 1H), 7.87 - 7.85 (m, 2H), 7.43 - 7.36 (m, 4H), 7.33 - 7.28 (m, 3H), 3.70 (d, J = 18.0 Hz, 1H), 3.64 (d, J = 18.0 Hz, 1H), 1.69 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.3, 196.0, 139.8, 139.4, 135.0, 129.5, 129.0, 128.9, 127.5, 126.8, 51.8, 45.9, 20.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{16}ClO_2$: 287.0839. Found: 287.0837.

4-(4-Bromophenyl)-2-methyl-4-oxo-2-phenylbutanal (3fa):



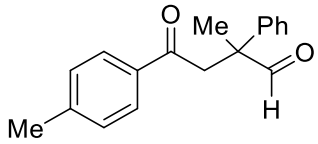
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.60). The product was obtained as yellow solid in 89% yield (58.7 mg), Mp. 70 - 71 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.69 (s, 1H), 7.79 - 7.77 (m, 2H), 7.58 - 7.56 (m, 2H), 7.39 - 7.35 (m, 2H), 7.32 - 7.28 (m, 3H), 3.69 (d, J = 18.0 Hz, 1H), 3.64 (d, J = 18.0 Hz, 1H), 1.68 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.2, 196.2, 139.3, 135.4, 131.8, 129.5, 128.9, 128.5, 127.4, 126.8, 51.8, 45.8, 20.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{16}BrO_2$: 331.0334. Found: 331.0329.

4-(4-Iodophenyl)-2-methyl-4-oxo-2-phenylbutanal (3ga):



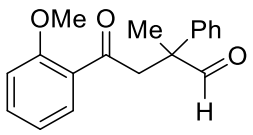
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.61). The product was obtained as yellow solid in 77% yield (58.3 mg), Mp. 69 - 70 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.62 (s, 1H), 7.74 - 7.72 (m, 2H), 7.56 - 7.54 (m, 2H), 7.32 - 7.30 (m, 2H), 7.25 - 7.18 (m, 3H), 3.61 (d, J = 18.0 Hz, 1H), 3.55 (d, J = 18.0 Hz, 1H), 1.61 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.3, 196.5, 139.3, 137.9, 135.9, 129.4, 128.9, 127.5, 126.8, 101.4, 51.8, 45.8, 20.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{16}BrO_2$: 379.0195. Found: 379.0199.

2-Methyl-4-oxo-2-phenyl-4-(p-tolyl)butanal (3ha):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.58). The product was obtained as yellow oil in 81% yield (43.1 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.75 (s, 1H), 7.85 - 7.83 (m, 2H), 7.39 - 7.34 (m, 4H), 7.29 - 7.23 (m, 3H), 3.74 (d, J = 17.6 Hz, 1H), 3.68 (d, J = 17.6 Hz, 1H), 2.40 (s, 3H), 1.66 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.6, 196.8, 144.3, 139.8, 134.3, 129.3, 128.9, 128.2, 127.3, 126.9, 51.8, 45.9, 21.6, 20.8; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{19}O_2$: 267.1340. Found: 267.1343.

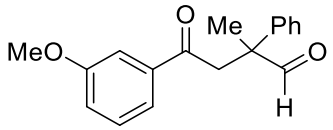
4-(2-Methoxyphenyl)-2-methyl-4-oxo-2-phenylbutanal (3ia):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.51). The product was obtained as yellow oil in 80% yield (45.1 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.79 (s, 1H), 7.65 - 7.62 (m, 1H), 7.49 - 7.45 (m,

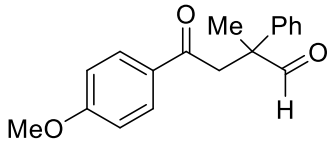
1H), 7.38 - 7.31 (m, 5H), 7.00 - 6.95 (m, 2H), 3.93 (s, 3H), 3.80 (s, 2H), 1.60 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.9, 199.6, 158.6, 140.2, 133.9, 130.4, 128.8, 127.9, 127.1, 126.9, 120.8, 111.4, 55.5, 52.1, 51.3, 21.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{19}O_3$: 283.1334. Found: 283.1336.

4-(3-Methoxyphenyl)-2-methyl-4-oxo-2-phenylbutanal (3ja):



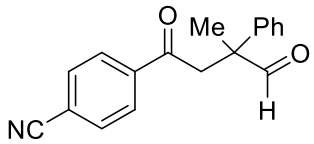
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.49). The product was obtained as yellow oil in 83% yield (46.9 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.73 (s, 1H), 7.54 - 7.52 (m, 1H), 7.45 - 7.44 (m, 1H), 7.40 - 7.23 (m, 5H), 7.30 - 7.26 (m, 1H), 7.12 - 7.09 (m, 1H), 3.83 (s, 3H), 3.74 (d, J = 18.0 Hz, 1H), 3.69 (d, J = 18.0 Hz, 1H), 1.68 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.5, 197.0, 159.7, 139.6, 138.0, 129.6, 128.9, 127.4, 126.8, 120.7, 120.0, 112.0, 55.4, 51.8, 46.1, 20.7; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{19}O_3$: 283.1334. Found: 283.1331.

4-(4-Methoxyphenyl)-2-methyl-4-oxo-2-phenylbutanal (3ka):



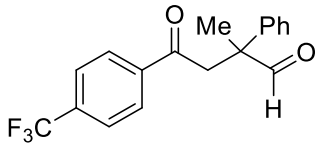
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.22). The product was obtained as yellow oil in 80% yield (45.1 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.75 (s, 1H), 7.93 - 7.91 (m, 2H), 7.39 - 7.32 (m, 4H), 7.29 - 7.26 (m, 1H), 6.93 - 6.90 (m, 2H), 3.86 (s, 3H), 3.71 (d, J = 17.6 Hz, 1H), 3.67 (d, J = 17.6 Hz, 1H), 1.65 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.6, 195.7, 163.7, 139.9, 130.4, 129.8, 128.9, 127.3, 126.8, 113.7, 55.5, 51.8, 45.8, 20.9; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{19}O_3$: 283.1334. Found: 283.1331.

4-(4-Isocyanophenyl)-2-methyl-4-oxo-2-phenylbutanal (3la):



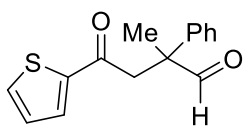
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.27). The product was obtained as yellow solid in 64% yield (41.1 mg), Mp. 127 - 128 ºC. ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.65 (s, 1H), 7.99 - 7.97 (m, 2H), 7.74 - 7.73 (m, 2H), 7.40 - 7.36 (m, 2H), 7.31 - 7.28 (m, 3H), 3.69 (d, J = 17.6 Hz, 1H), 3.64 (d, J = 17.6 Hz, 1H), 1.73 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 200.9, 196.0, 139.7, 138.9, 132.4, 129.0, 128.4, 127.6, 126.8, 117.8, 116.4, 51.9, 46.0, 20.0; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{16}NO_2$: 278.1181. Found: 278.1180.

2-Methyl-4-oxo-2-phenyl-4-(4-(trifluoromethyl)phenyl)butanal (3ma):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.67). The product was obtained as yellow oil in 70% yield (44.9 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.69 (s, 1H), 8.02 - 8.00 (m, 2H), 7.71 - 7.69 (m, 2H), 7.40 - 7.37 (m, 2H), 7.33 - 7.28 (m, 3H), 3.73 (d, J = 17.6 Hz, 1H), 3.67 (d, J = 17.6 Hz, 1H), 1.72 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.1, 196.4, 139.5, 139.2, 134.5 (q, J = 32.7, 129.0, 128.4, 127.6, 126.9, 125.7 (q, J = 4.0 Hz), 123.5 (d, J = 271.1 Hz), 51.9, 46.1, 20.2; ^{19}F NMR (376 MHz, $CDCl_3$) δ -63.0; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{16}F_3O_2$: 321.1102. Found: 321.1104.

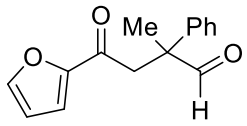
2-Methyl-4-oxo-2-phenyl-4-(thiophen-2-yl)butanal (3na):



The title compound was prepared according to the general procedure for reaction in 24

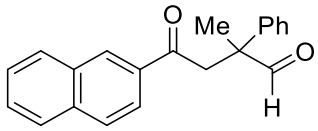
h (EA/PE = 1/9, R_f = 0.50). The product was obtained as yellow oil in 95% yield (49.1 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.68 (s, 1H), 7.69 - 7.68 (m, 1H), 7.64 - 7.63 (d, J = 5.2, 1H), 7.40 - 7.36 (m, 2H), 7.33 - 7.26 (m, 3H), 7.12 - 7.09 (m, 1H), 3.65 (d, J = 16.8 Hz, 1H), 3.60 (d, J = 16.8 Hz, 1H), 1.68 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.0, 190.2, 144.1, 139.2, 134.2, 132.2, 128.9, 128.1, 127.5, 126.9, 52.0, 46.2, 20.2; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{15}H_{15}O_2S$: 259.0793. Found: 259.0794.

4-(Furan-2-yl)-2-methyl-4-oxo-2-phenylbutanal (3oa):



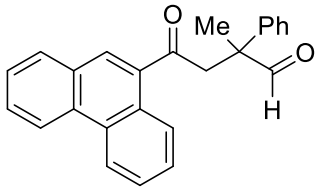
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.41). The product was obtained as yellow oil in 94% yield (45.6 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.67 (s, 1H), 7.54 (s, 1 H), 7.38-7.35 (m, 2H), 7.32 - 7.25 (m, 3H), 7.14 - 7.13 (m, 1H), 6.51 - 6.50 (m, 1H), 3.57 (d, J = 16.8 Hz, 1H), 3.52 (d, J = 16.8 Hz, 1H), 1.66 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 200.9, 186.5, 152.7, 146.5, 139.2, 128.9, 127.5, 126.9, 117.3, 112.4, 52.0, 45.1, 20.2; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{15}H_{15}O_3$: 253.1021. Found: 253.1021.

2-Methyl-4-(naphthalen-2-yl)-4-oxo-2-phenylbutanal (3pa):



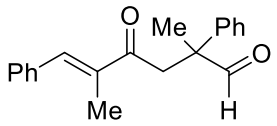
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.57). The product was obtained as yellow oil in 64% yield (38.7 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.78 (s, 1H), 8.45 (s, 1H), 7.99 - 7.93 (m, 2), 7.88 - 7.86 (m, 2H), 7.65 - 7.55 (m, 2H), 7.39 - 7.38 (m, 4H), 7.28 - 7.27 (m, 1H), 3.87 (s, 2H), 1.72 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.5, 197.2, 139.7, 135.6, 134.1, 132.4, 129.9, 129.6, 128.9, 128.6, 128.5, 127.8, 127.4, 126.9, 126.8, 123.7, 51.9, 46.1, 20.7; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{21}H_{19}O_2$: 303.1385. Found: 303.1383.

2-Methyl-4-oxo-4-(phenanthren-9-yl)-2-phenylbutanal (3qa):



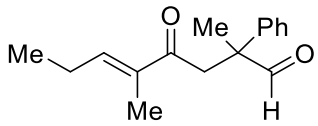
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.50). The product was obtained as yellow oil in 80% yield (56.4 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.73 (s, 1H), 8.61 - 8.56 (m, 2H), 8.31 - 8.29 (m, 1H), 7.97 (s, 1H), 7.83 - 7.81 (m, 1H), 7.67 - 7.64 (m, 1H), 7.61 - 7.50 (m, 3H), 7.29 - 7.28 (m, 4H), 7.21 - 7.18 (m, 1H), 3.79 (s, 2H), 1.70 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 202.0, 201.5, 139.6, 135.2, 131.9, 130.8, 129.9, 129.8, 129.5, 129.1, 129.0, 128.1, 127.6, 127.6, 127.36, 127.3, 127.1, 126.6, 122.9, 122.8, 52.6, 49.6, 20.8; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{25}H_{21}O_2$: 353.1542. Found: 353.1540.

(*E*)-2,5-Dimethyl-4-oxo-2,6-diphenylhex-5-enal (3ra):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.60). The product was obtained as yellow oil in 79% yield (46.1 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.73 (s, 1H), 7.51 (s, 1H), 7.40 - 7.30 (m, 10H), 3.60 (d, J = 17.2 Hz, 1H), 3.55 (d, J = 17.2 Hz, 1H), 2.00 (s, 3H), 1.62 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.7, 199.4, 139.9, 139.4, 137.4, 135.6, 129.7, 128.9, 128.7, 128.4, 127.3, 126.9, 51.9, 45.3, 20.8, 13.0; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{20}H_{21}O_2$: 293.1542. Found: 293.1542.

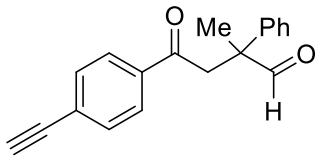
(*E*)-2,5-Dimethyl-4-oxo-2-phenyloct-5-enal (3sa):



The title compound was prepared according to the general procedure for reaction in 24

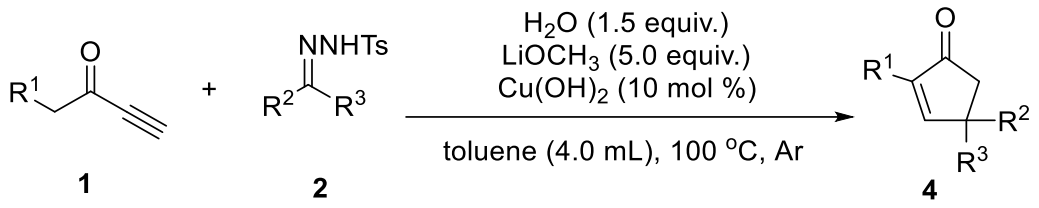
h (EA/PE = 1/9, R_f = 0.67). The product was obtained as colorless oil in 42% yield (20.5 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.70 (s, 1H), 7.38 - 7.35 (m, 2H), 7.29 - 7.26 (m, 3H), 6.64 - 6.61 (m, 1H), 3.46 (d, J = 17.2 Hz, 1H), 3.40 (d, J = 17.2 Hz, 1H), 2.27 - 2.19 (m, 2H), 1.72 (s, 3H), 1.57 (s, 3H), 1.06 (t, J = 7.6 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.8, 198.9, 145.1, 140.1, 136.9, 128.8, 127.2, 126.8, 51.7, 44.9, 22.4, 20.8, 13.0, 11.0; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{16}H_{21}O_2$: 245.1542. Found: 245.1544.

4-(4-Ethynylphenyl)-2-methyl-4-oxo-2-phenylbutanal (3ta):



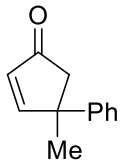
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/9, R_f = 0.50). The product was obtained as yellow oil in 33% yield (18.3 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.63 (s, 1H), 7.81 - 7.79 (m, 2H), 7.48 - 7.46 (m, 2H), 7.32 - 7.28 (m, 2H), 7.25 - 7.21 (m, 3H), 3.64 (d, J = 17.6 Hz, 1H), 3.59 (d, J = 17.6 Hz, 1H), 3.18 (s, 1H), 1.62 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.3, 196.4, 139.4, 136.4, 132.3, 128.9, 127.9, 127.5, 127.1, 126.8, 82.6, 80.6, 51.8, 46.0, 20.5; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{19}H_{17}O_2$: 277.1229. Found: 277.1227.

General procedure for cyclopentenones and their spectral data:



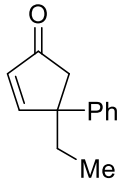
A mixture of **1** (0.20 mmol, 1.0 equiv.), **2** (0.40 mmol, 2.0 equiv.), $Cu(OH)_2$ (0.02 mmol, 10 mol %), $LiOCH_3$ (1.0 mmol, 5.0 equiv.), H_2O (5.4 μL, 1.5 equiv.) and toluene (4.0 mL) was sealed in a Schlenk tube under argon protection at 100 °C and the mixture was stirred for 24 h or until the **1** was consumed completely. Then the reaction mixture was concentrated under reduced pressure and purified by flash column chromatography to give the desired product **4**.

4-Methyl-4-phenylcyclopent-2-en-1-one (4ua):



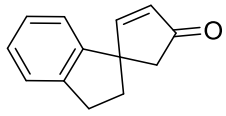
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/19, R_f = 0.50). The product was obtained as yellow oil in 81% yield (27.9 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.70 - 7.68 (d, J = 5.6 Hz, 1H), 7.37 - 7.33 (m, 2H), 7.28 - 7.25 (m, 3H), 6.22 - 6.21 (d, J = 5.6 Hz, 1H), 2.66 (d, J = 18.8 Hz, 1H), 2.56 (d, J = 18.8 Hz, 1H), 1.64 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 209.8, 171.3, 145.2, 131.7, 128.7, 126.8, 125.7, 51.9, 48.2, 27.2; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{12}H_{13}O$: 173.0966. Found: 173.0966.

4-Ethyl-4-phenylcyclopent-2-en-1-one (4ub):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/19, R_f = 0.55). The product was obtained as yellow oil in 64% yield (23.8 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.86 - 7.84 (d, J = 6.0 Hz, 1H), 7.37 - 7.33 (m, 2H), 7.26 - 7.23 (m, 3H), 6.27 (d, J = 5.6 Hz, 1H), 2.63 (d, J = 0.4 Hz, 2H), 2.04 - 1.91 (m, 2H), 0.83 (t, J = 7.6 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 209.2, 169.2, 144.7, 132.9, 128.7, 126.6, 126.2, 52.6, 49.2, 33.9, 9.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{13}H_{15}O$: 187.1123. Found: 187.1120.

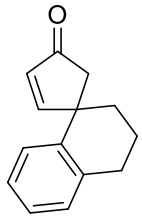
2',3'-Dihydrospiro[cyclopentane-1,1'-inden]-2-en-4-one (4uc):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/19, R_f = 0.70). The product was obtained as yellow oil in 51% yield (18.8

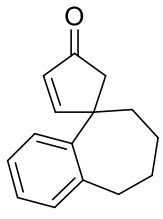
mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.51 (d, J = 5.2 Hz, 1H), 7.29 - 7.26 (m, 1H), 7.23 - 7.18 (m, 2H), 6.99 - 6.97 (m, 1H), 6.19 (d, J = 5.6 Hz, 1H), 3.07 - 3.03 (m, 2H), 2.65 (d, J = 18.8 Hz, 1H), 2.56 (d, J = 18.8 Hz, 1H), 2.40 - 2.32 (m, 1H), 2.22 - 2.16 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 209.9, 169.4, 146.1, 143.1, 131.9, 127.6, 127.1, 124.8, 122.5, 56.8, 49.3, 39.0, 30.9; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{13}H_{13}O$: 185.0966. Found: 185.0968.

3',4'-Dihydro-2'H-spiro[cyclopentane-1,1'-naphthalen]-2-en-4-one (4ud):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/19, R_f = 0.55). The product was obtained as yellow oil in 49% yield (19.5 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.55 (d, J = 5.6 Hz, 1H), 7.15 - 7.13 (m, 3H), 7.00 - 6.98 (m, 1H), 6.26 - 6.25 (d, J = 5.6 Hz, 1H), 2.88 - 2.85 (m, 2H), 2.58 (d, J = 16.0 Hz, 1H), 2.51 (d, J = 16.0 Hz, 1H), 2.05 - 1.92 (m, 2H), 1.88 - 1.75 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 210.4, 172.0, 139.1, 136.4, 132.1, 129.5, 127.1, 126.8, 126.6, 52.7, 48.6, 36.1, 29.7, 20.6; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{14}H_{16}O$: 199.1123. Found: 199.1124.

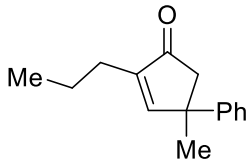
6,7,8,9-Tetrahydrospiro[benzo[7]annulene-5,1'-cyclopentan]-2'-en-4'-one (4ue):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/19, R_f = 0.40). The product was obtained as yellow oil in 42% yield (17.9 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.94 (d, J = 6.0 Hz, 1H), 7.13 - 7.11 (m, 2H), 7.08

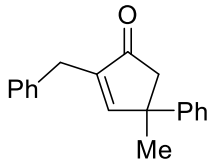
- 7.07 (m, 1H), 6.99 - 6.97 (m, 1H), 6.21 (d, J = 5.6 Hz, 1H), 2.98 (d, J = 18.4 Hz, 1H), 2.44 (d, J = 18.4 Hz, 1H), 2.91 - 2.77 (m, 2H), 2.04 - 1.97 (m, 2H), 1.92 - 1.86 (m, 2H), 1.75 - 1.72 (m, 1H), 1.49 - 1.40 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 209.4, 172.6, 145.5, 142.3, 131.1, 129.9, 127.0, 126.2, 125.9, 52.5, 46.1, 37.4, 36.6, 28.7, 27.4; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{15}H_{17}O$: 213.1279. Found: 213.1279.

4-Methyl-4-phenyl-2-propylcyclopent-2-en-1-one (4va):



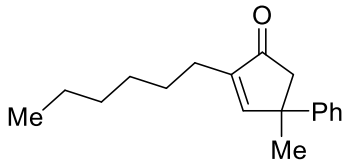
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/19, R_f = 0.65). The product was obtained as yellow oil in 57% yield (24.5 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.28 - 7.24 (m, 2H), 7.21 - 7.14 (m, 4H), 2.62 (d, J = 18.4 Hz, 1H), 2.52 (d, J = 18.4 Hz, 1H), 2.17 - 2.13 (m, 2H), 1.56 - 1.46 (m, 5H), 0.91 - 0.87 (t, J = 7.6 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 209.5, 164.5, 146.2, 143.7, 128.6, 126.5, 125.7, 52.5, 45.5, 27.6,26.5, 21.0, 13.9; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{15}H_{19}O$: 215.1436. Found: 215.1437.

4-Methyl-2,4-diphenylcyclopent-2-en-1-one (4xa):



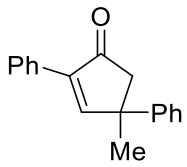
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/19, R_f = 0.68). The product was obtained as yellow oil in 56% yield (29.4 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.33 - 7.29 (m, 4H), 7.25 - 7.19 (m, 6H), 7.13 - 7.12 (m, 1H), 3.16 - 3.52 (m, 2H), 2.73 (d, J = 18.4 Hz, 1H), 2.63 (d, J = 18.4 Hz, 1H), 1.57 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 208.5, 165.5, 145.9, 143.5, 138.5, 128.9, 128.62, 128.59, 126.6, 126.4, 125.7; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{19}H_{19}O$: 263.1436. Found: 263.1439.

2-Hexyl-4-methyl-4-phenylcyclopent-2-en-1-one (4wa):



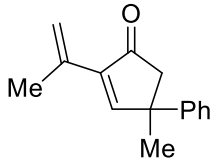
The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/19, R_f = 0.70). The product was obtained as yellow oil in 55% yield (28.2 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.35 - 7.32 (m, 2H), 7.27 - 7.21 (m, 4H), 2.69 (d, J = 18.8 Hz, 1H), 2.59 (d, J = 18.8 Hz, 1H), 2.26 - 2.22 (m, 2H), 1.60 (s, 3H), 1.56 - 1.50 (m, 2H), 1.37 - 1.26 (m, 6H), 0.90 - 0.87 (t, J = 6.8 Hz, 6H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 209.5, 164.3, 146.2, 144.0, 128.6, 126.5, 125.7, 52.5, 45.5, 31.5, 29.0, 27.7, 27.6, 24.5, 22.5, 14.0; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{25}O$: 257.1905. Found: 257.1901.

4-Methyl-2,4-diphenylcyclopent-2-en-1-one (4ya):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/19, R_f = 0.65). The product was obtained as yellow oil in 35% yield (17.4 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.72 (s, 1H), 7.71 - 7.68 (m, 2H), 7.35 - 7.32 (m, 2H), 7.30 - 7.25 (m, 5H), 7.20 - 7.16 (m, 1H), 2.82 (d, J = 18.8 Hz, 1H), 2.71 (d, J = 18.8 Hz, 1H), 1.60 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 207.1, 165.3, 145.7, 140.7, 131.1, 128.8, 128.6, 128.5, 127.3, 126.8, 125.8, 53.4, 45.0, 27.3; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{18}H_{17}O$: 249.1729. Found: 249.1727.

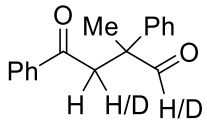
4-Methyl-4-phenyl-2-(prop-1-en-2-yl)cyclopent-2-en-1-one (4za):



The title compound was prepared according to the general procedure for reaction in 24 h (EA/PE = 1/19, R_f = 0.65). The product was obtained as colorless oil in 52% yield (22.1 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.43 (s, 1H), 7.36 - 7.32 (m, 2H), 7.29 - 7.22 (m, 3H), 6.16 (s, 1H), 5.25 (s, 1H), 2.79 (d, J = 18.4 Hz, 1H), 2.67 (d, J = 18.4 Hz, 1H), 1.98 (s, 3H), 1.63 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 207.3, 164.4, 145.9, 140.2, 134.1, 128.7, 126.7, 125.8, 117.8, 53.6, 44.3, 27.4, 22.0; HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{15}H_{17}O$: 213.1279. Found: 213.1281.

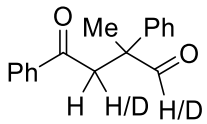
Spectral data of mechanistic investigations

3aa-I:



The title compound was prepared under standard conditions with D_2O (1.0 equiv.) instead of H_2O. The product was obtained as yellow oil in 71% yield (35.8 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.66 (s, 0.65H), 7.87 - 7.85 (m, 2H), 7.51 - 7.47 (m, 1H), 7.32 - 7.35 (m, 2H), 7.32 - 7.26 (m, 4H), 7.22 -7.18 (m, 1 H), 3.71 - 3.62 (m, 1.12H), 1.60 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.5, 197.3, 197.2, 139.6, 136.7, 133.4, 128.9, 128.6, 128.1, 127.4, 126.8, 51.8, 51.7, 46.0, 20.7, 20.6. HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{15}D_2O_2$: 255.1354. Found: 255.1350.

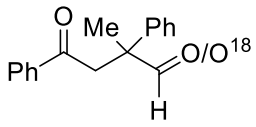
3aa-II:



The title compound was prepared under standard conditions with **2a'** instead of **2a**. The product was obtained as yellow oil in 69% yield (34.8 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.73 (m, 0.82H), 7.94 - 7.92 (m, 2H), 7.58 - 7.54 (m, 1H), 7.46 - 7.42 (m, 2H), 7.39 - 7.32 (m, 4H), 7.30 -7.27 (m, 1 H), 3.78 - 3.68 (m, 1.51H), 1.68 - 1.66 (d, J = 6.8 Hz, 2.39H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.5, 201.5, 201.5, 197.2, 139.6, 136.7, 136.7,

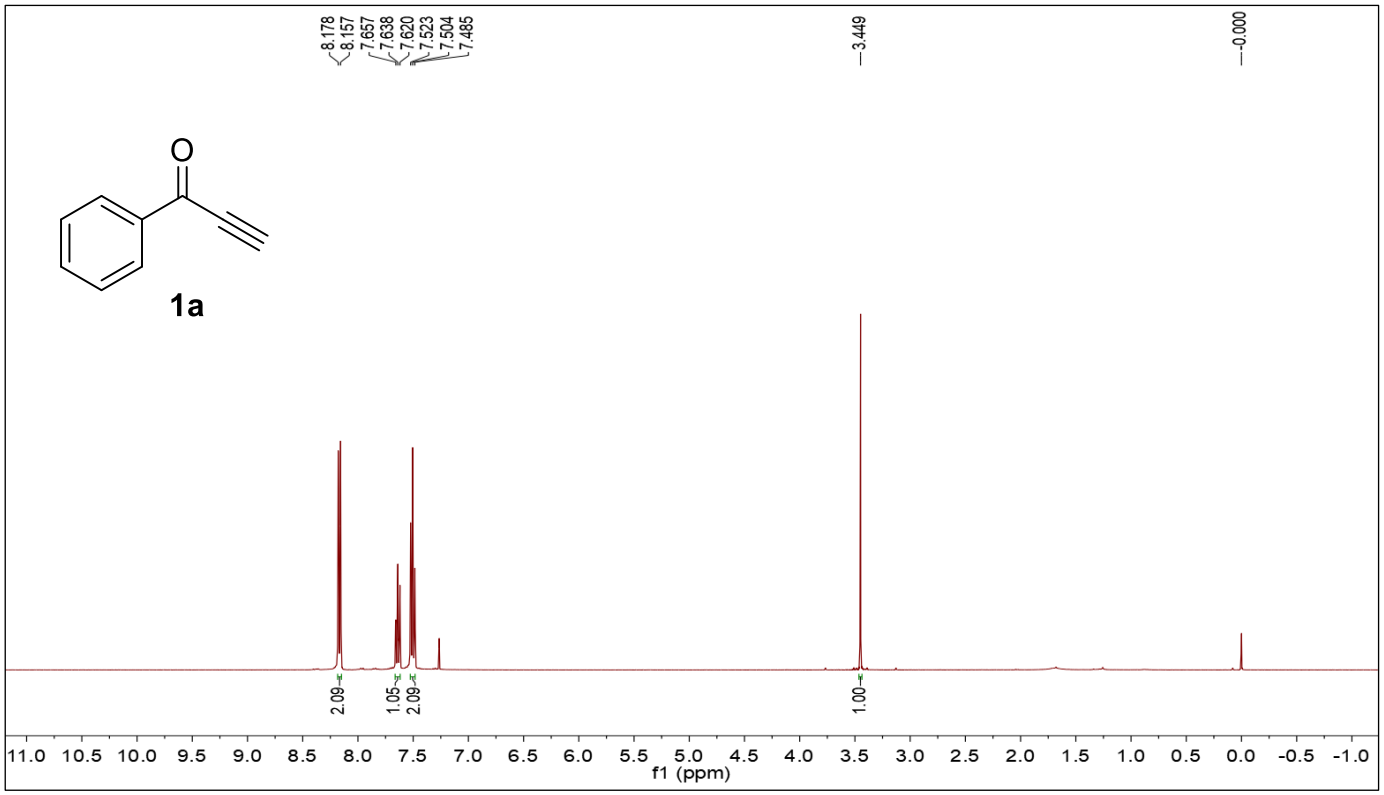
133.3, 128.9, 128.6, 128.1, 127.3, 126.8, 51.7, 51.7, 51.6, 46.0, 46.0, 20.7, 20.2. HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{15}D_2O_2$: 255.1354. Found: 255.1350.

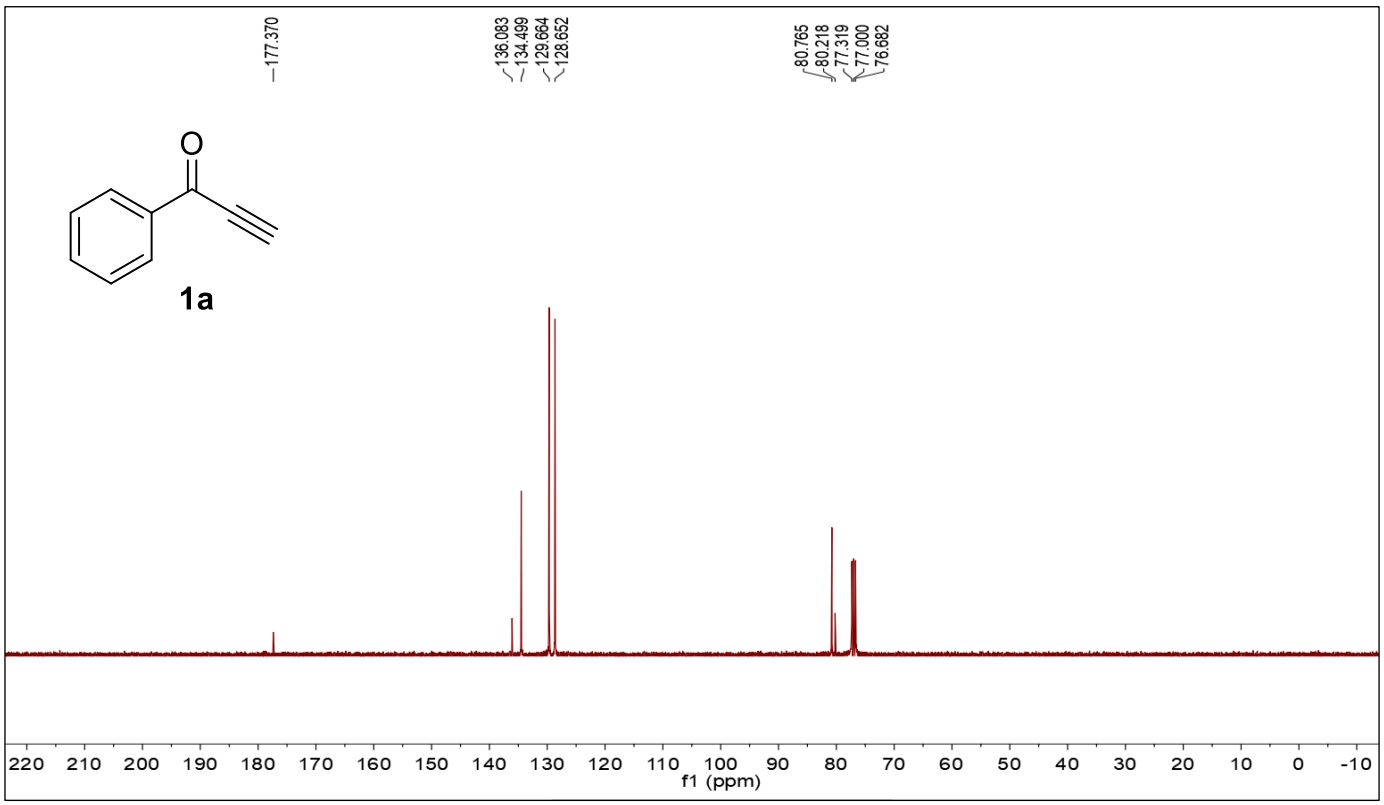
3aa-III:

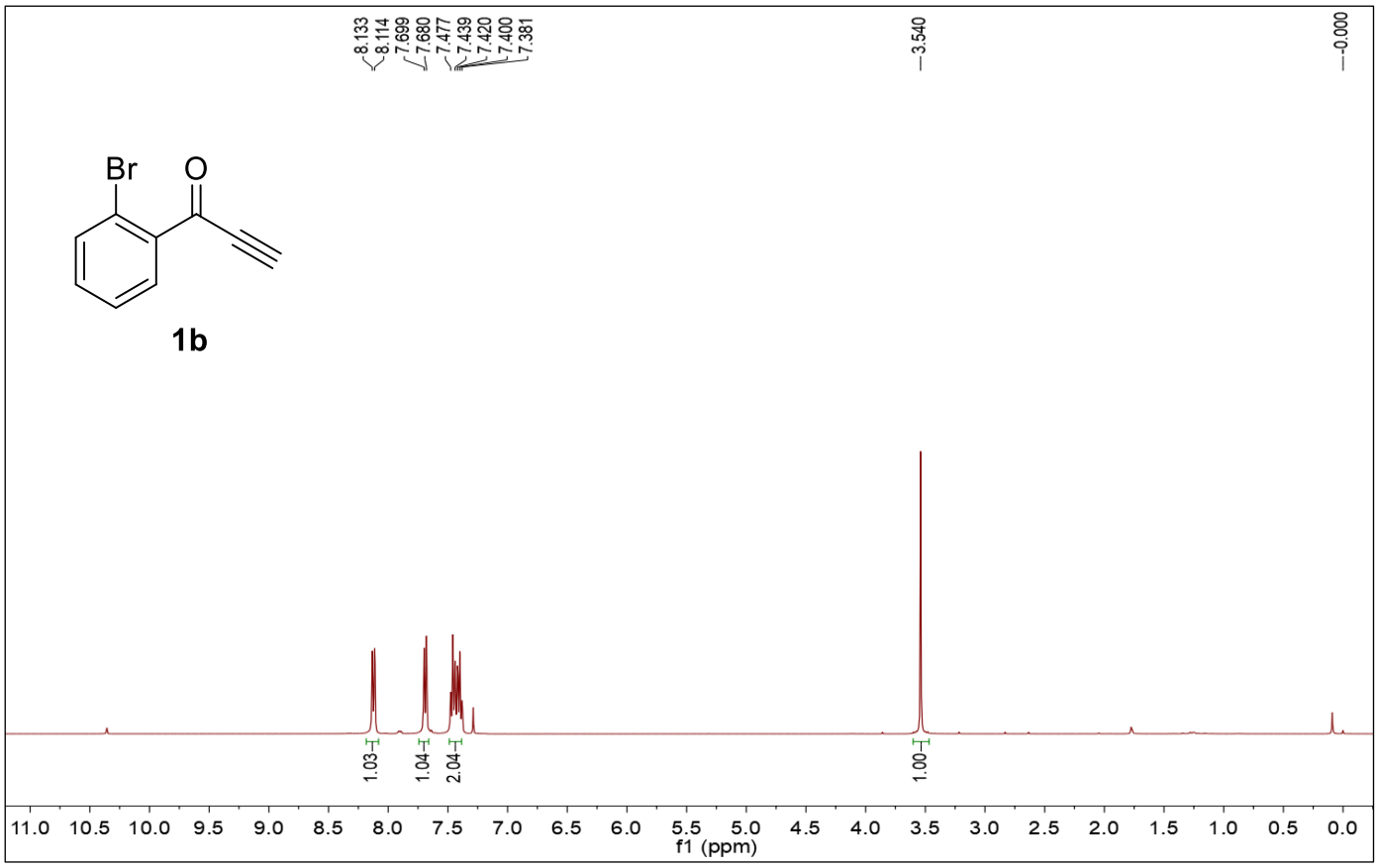


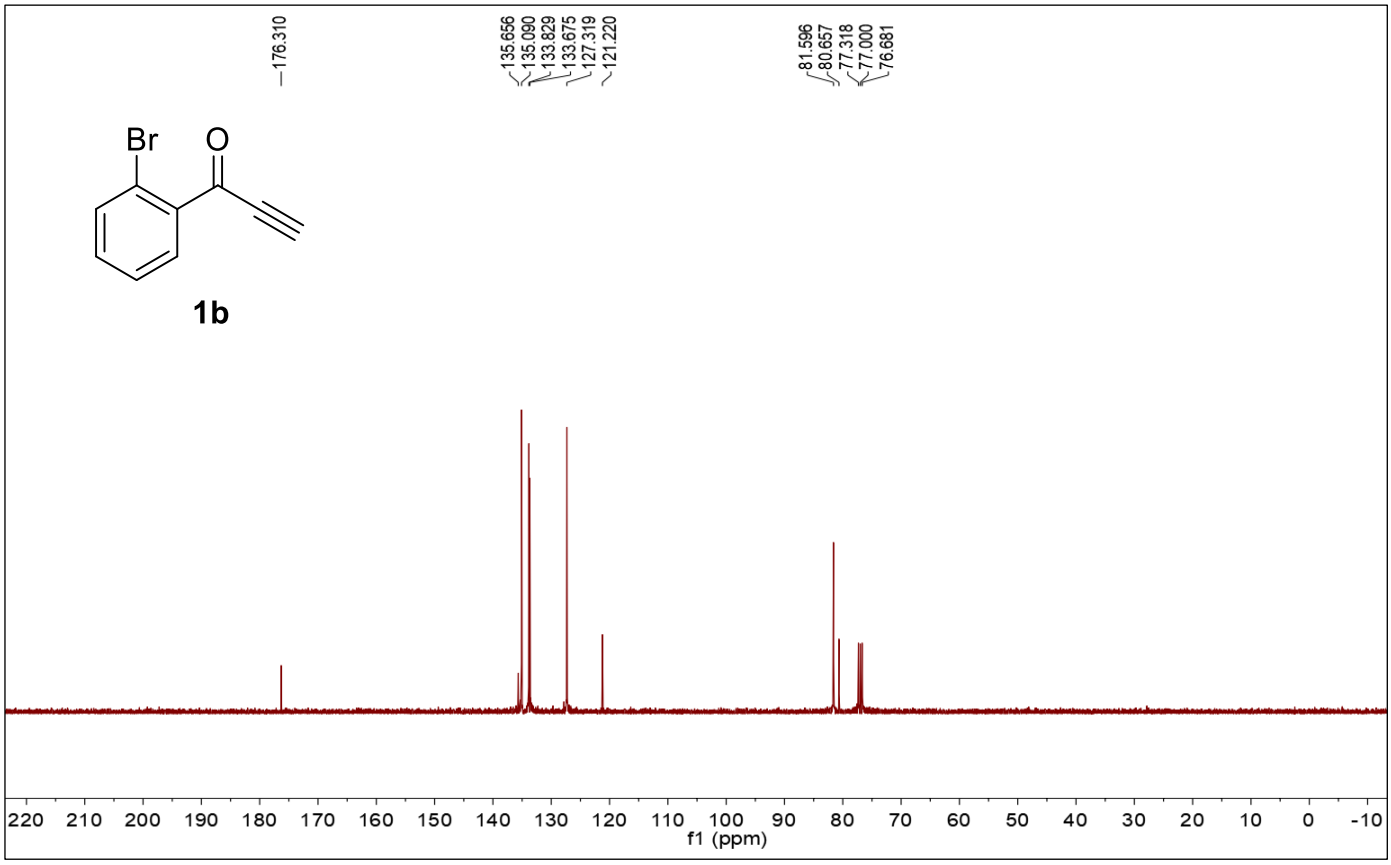
The title compound was prepared under standard conditions with $H_2{}^{18}O$ (5.0 equiv.) instead of H_2O. The product was obtained as yellow oil in 56% yield (28.5 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.66 (s, 0.90H), 7.87 - 7.85 (m, 2H), 7.51 - 7.47 (m, 1H), 7.39 - 7.35 (m, 2H), 7.32 - 7.25 (m, 4H), 7.22 -7.18 (m, 1 H), 3.68 (d, J = 17.6 Hz, 1H), 3.63 (d, J = 17.6 Hz, 1H), 1.61 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.5, 201.4, 197.2, 139.7, 136.7, 133.3, 128.9, 128.6, 128.1, 127.4, 126.8, 51.8, 46.0, 20.7. HRMS (ESI) m/z $[M+H]^+$: Calcd for $C_{17}H_{17}O^{18}O$: 255.1271. Found: 255.1267; HRMS (ESI) m/z $[M+Na]^+$: Calcd for $C_{17}H_{16}NaO^{18}O$: 277.1090. Found: 277.1086.

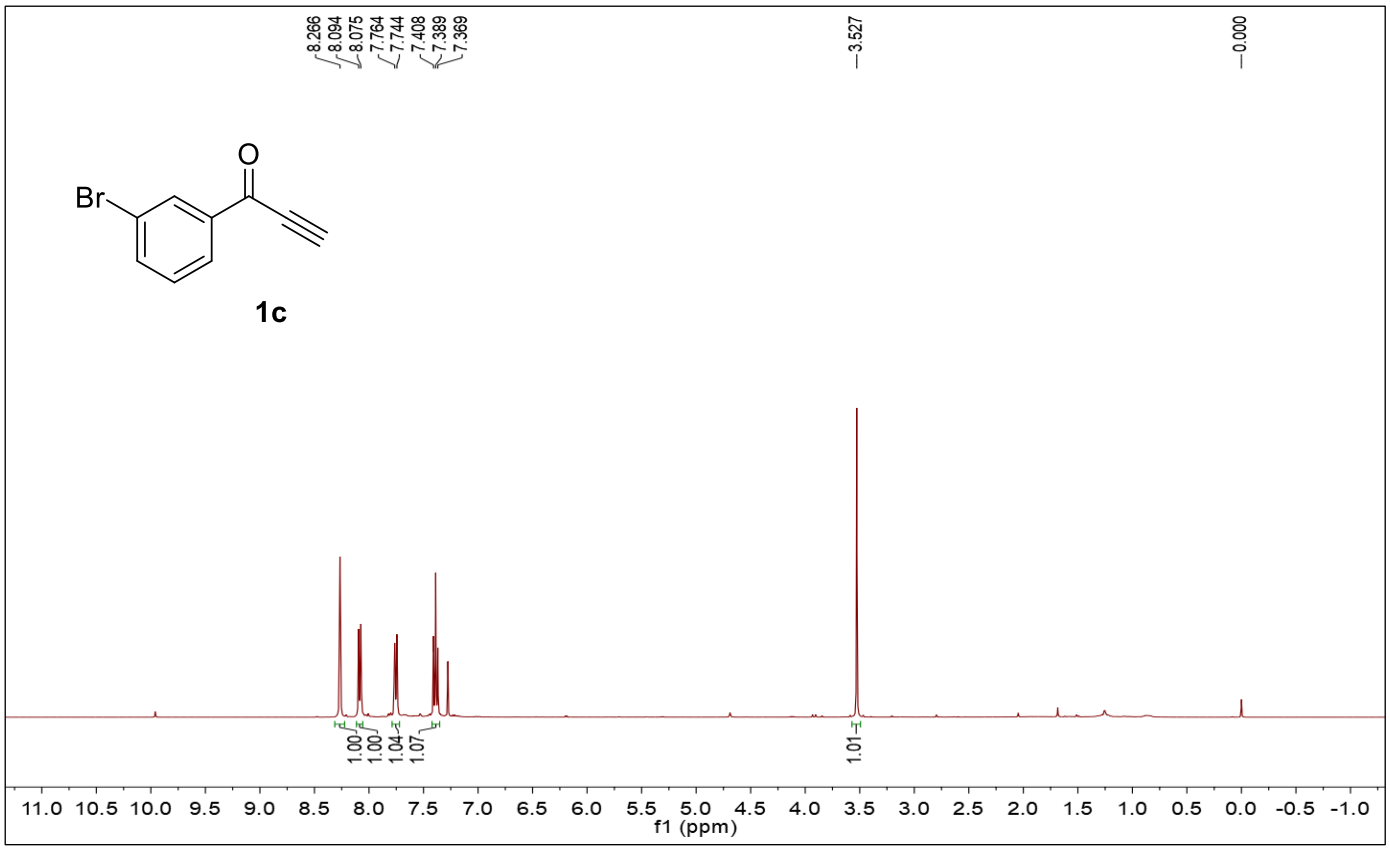
^{1}H and ^{13}C NMR spectra of ynones

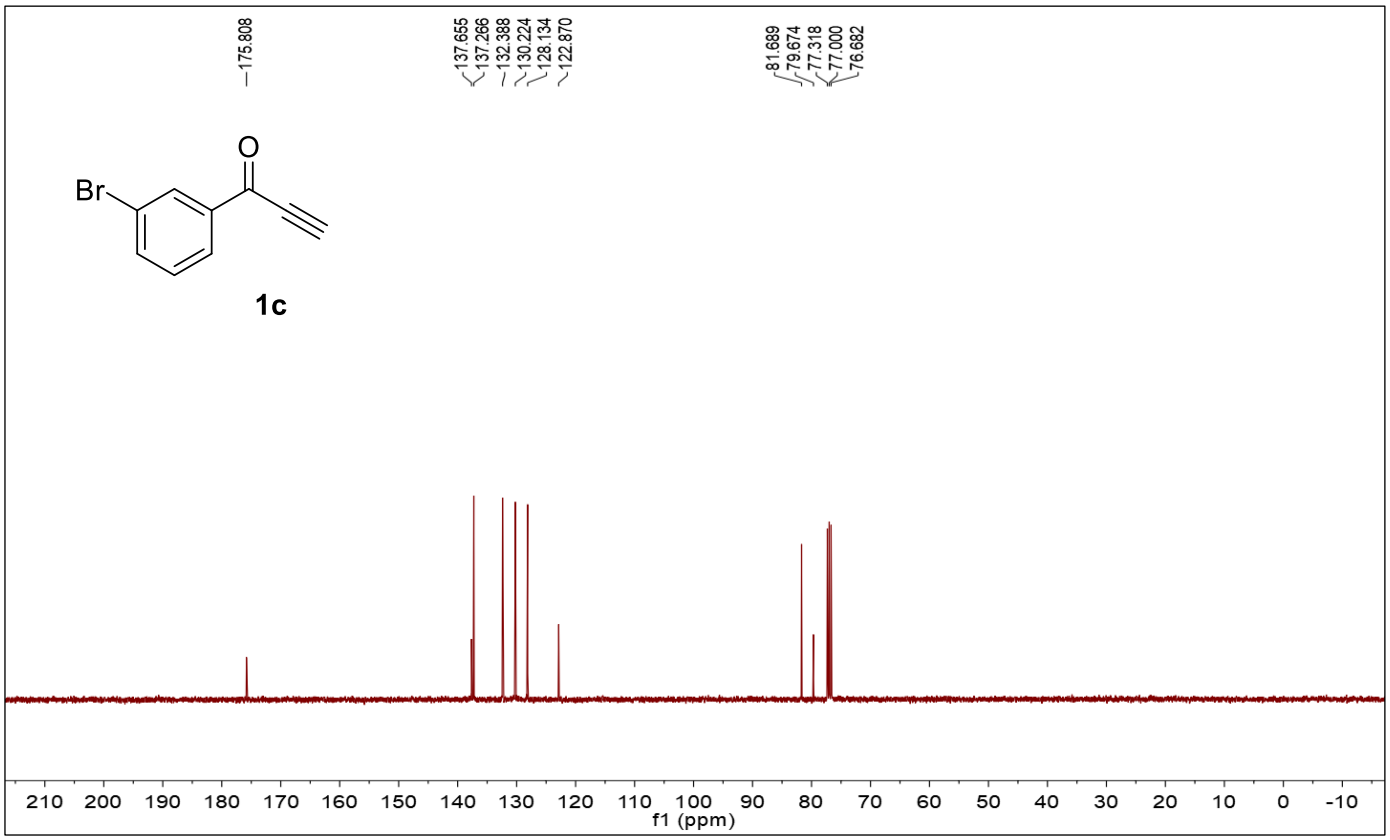

8.178
8.157
7.657
7.638
7.620
7.523
7.504
7.485
3.449
0.000
O
1a
2.09
1.05
2.09
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

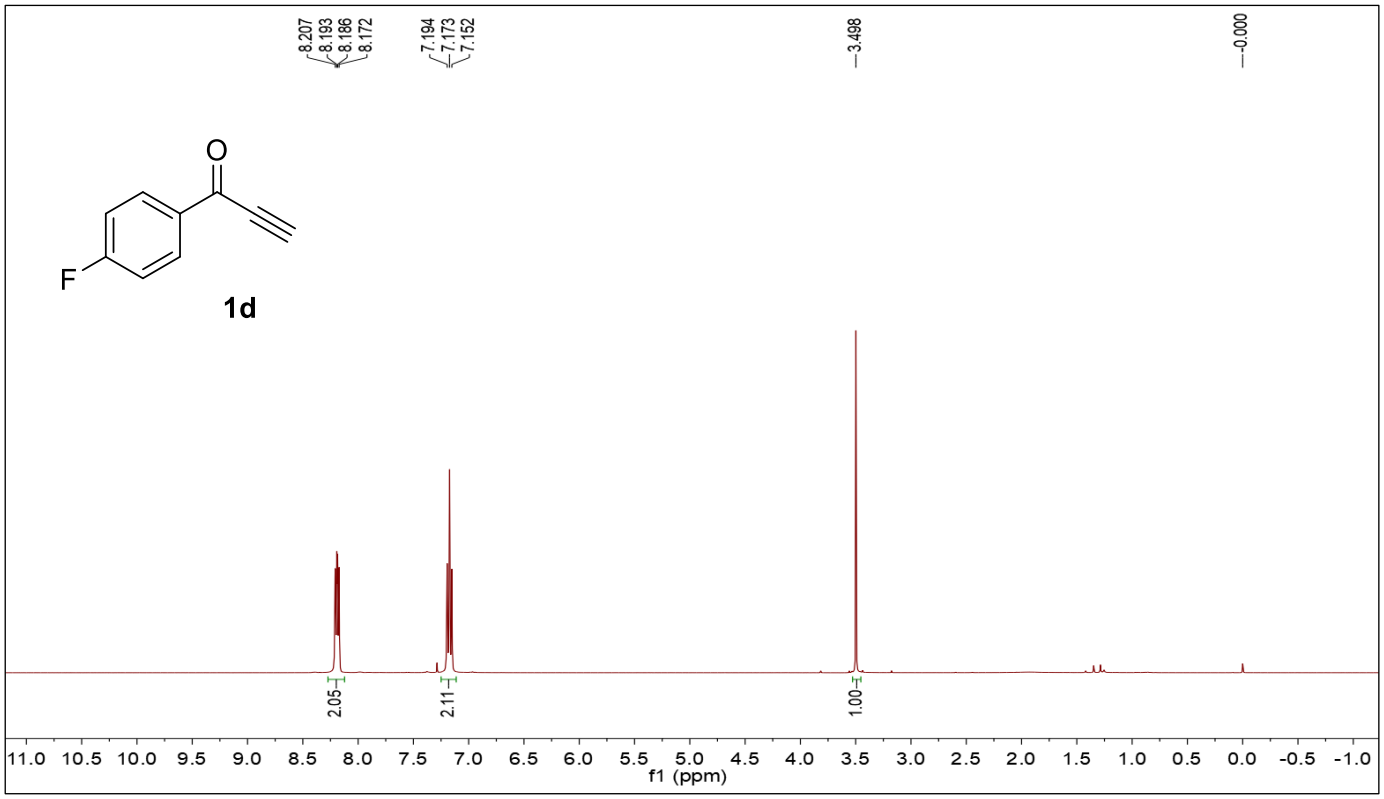

177.370
136.083
134.499
129.664
128.652
80.765
80.218
77.319
77.000
76.682
O
1a
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

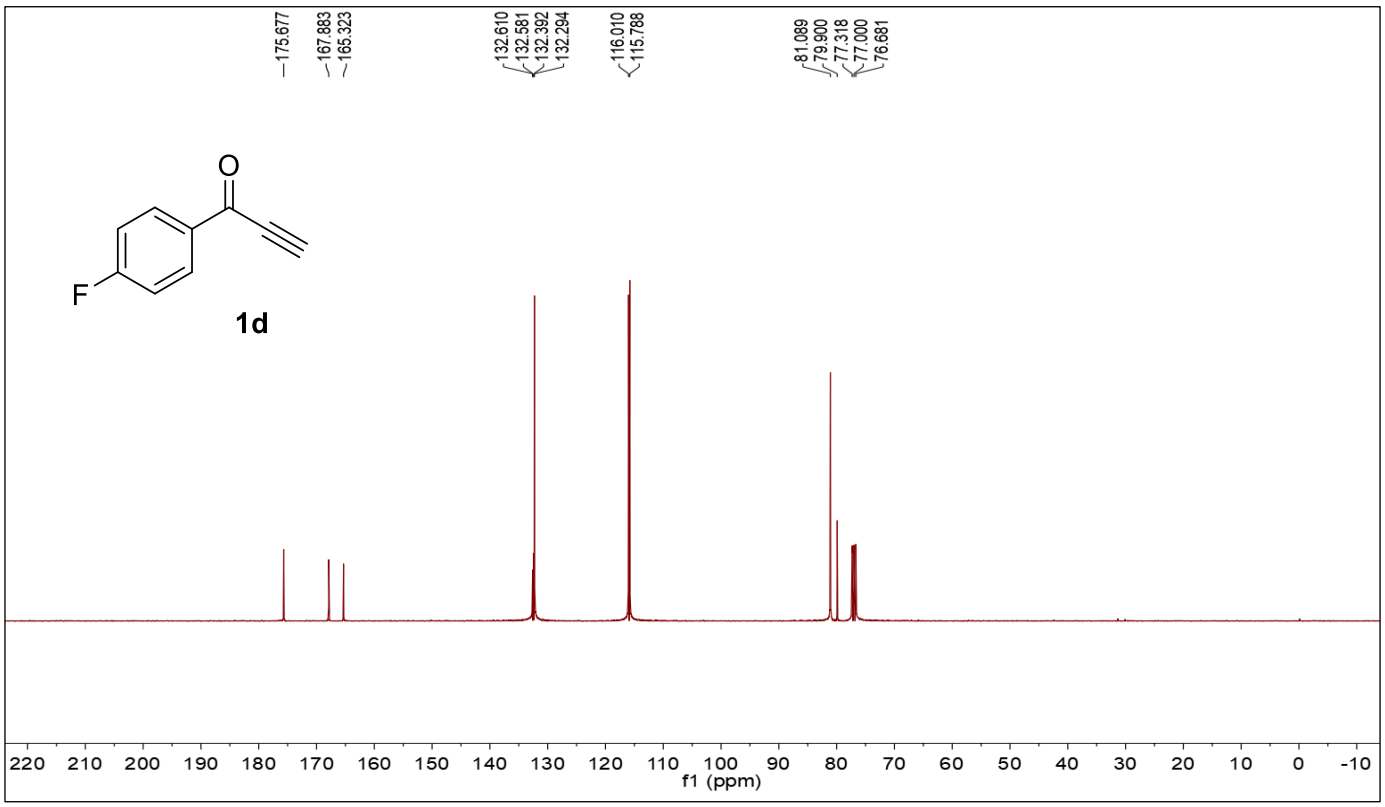

8.133
8.114
7.699
7.680
7.477
7.439
7.420
7.400
7.381
3.540
0.000
Br
O
1b
1.03
1.04
2.04
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

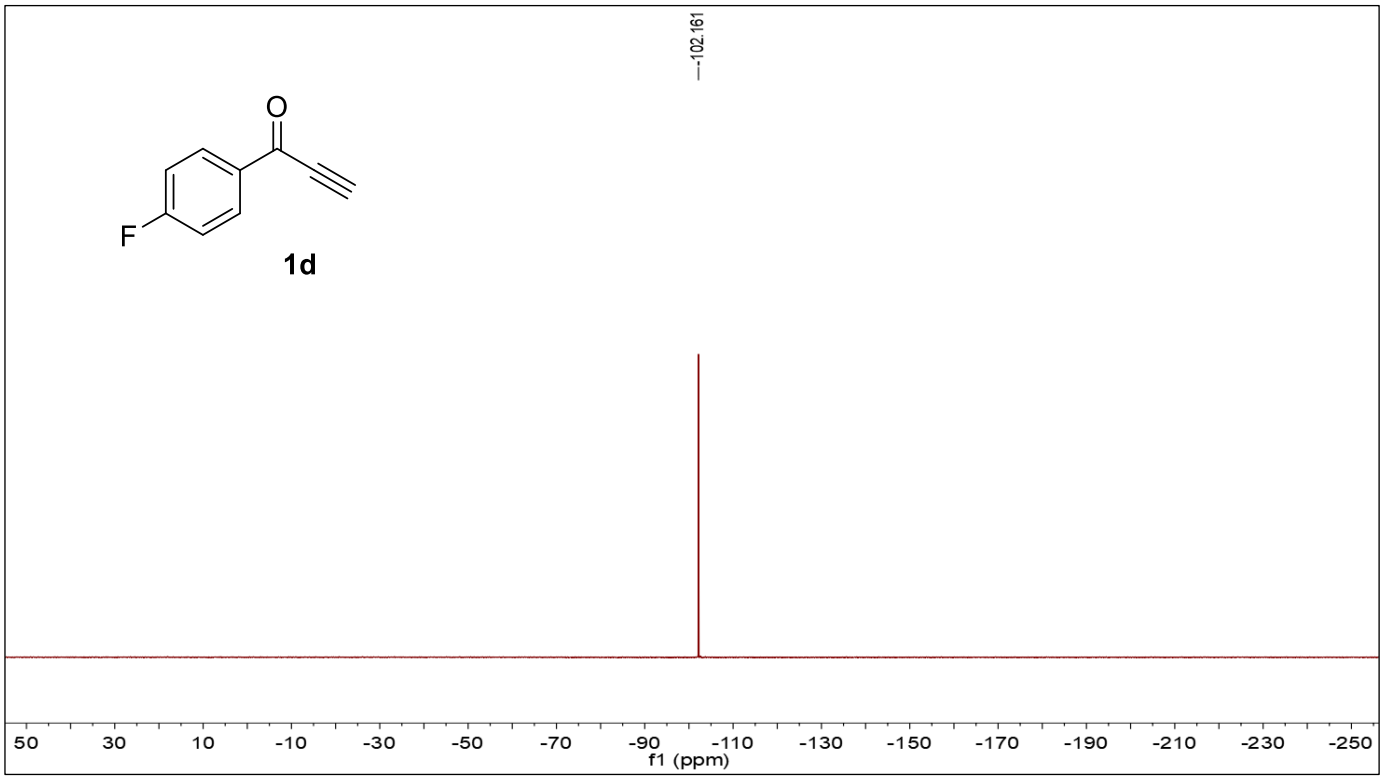

176.310
135.656
135.090
133.829
133.675
127.319
121.220
81.596
80.657
77.318
77.000
76.681
Br
O
1b
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

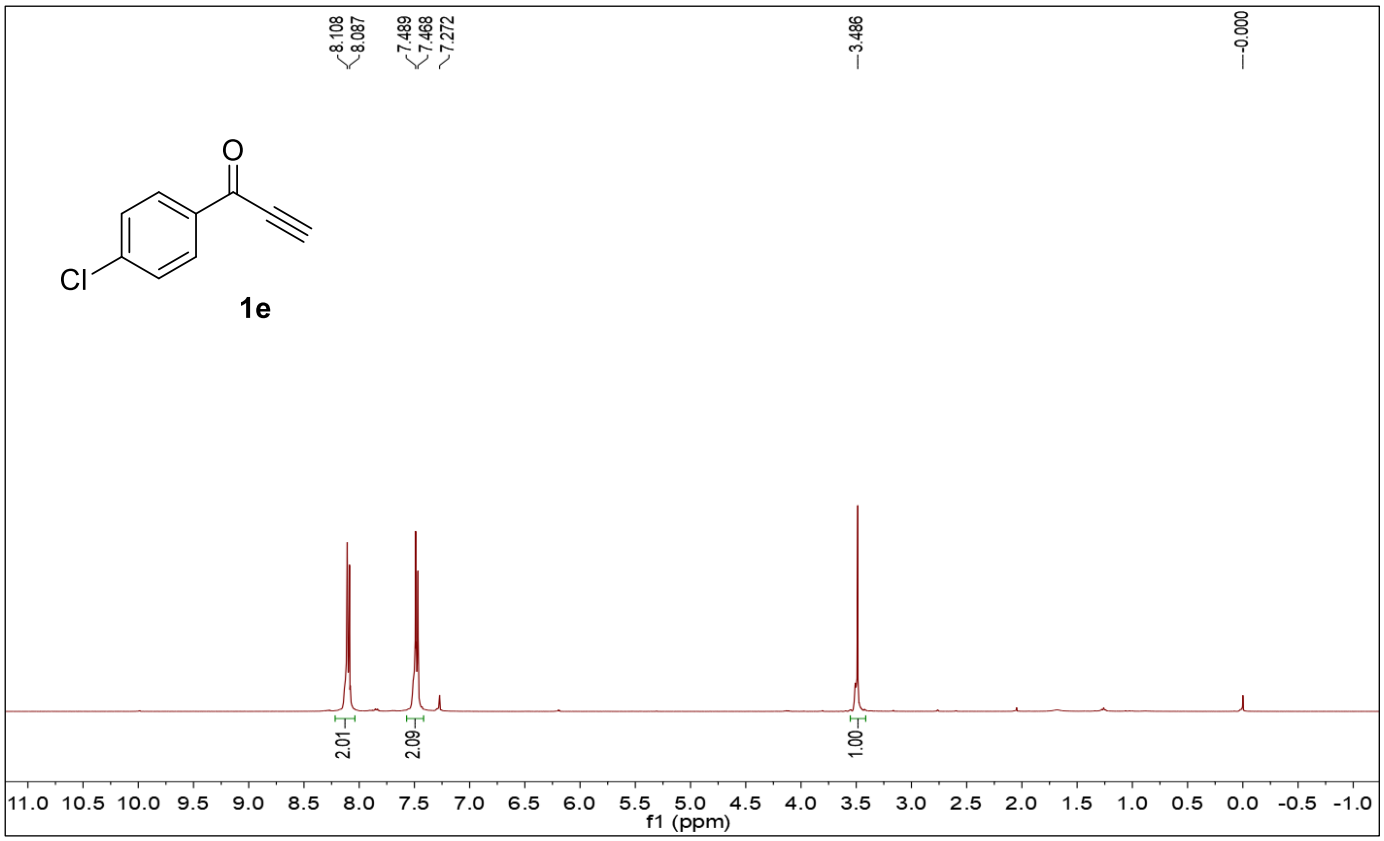

8.266
8.094
8.075
7.764
7.744
7.408
7.389
7.369
3.527
0.000
Br
O
1c
1.00
1.00
1.04
1.07
1.01
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

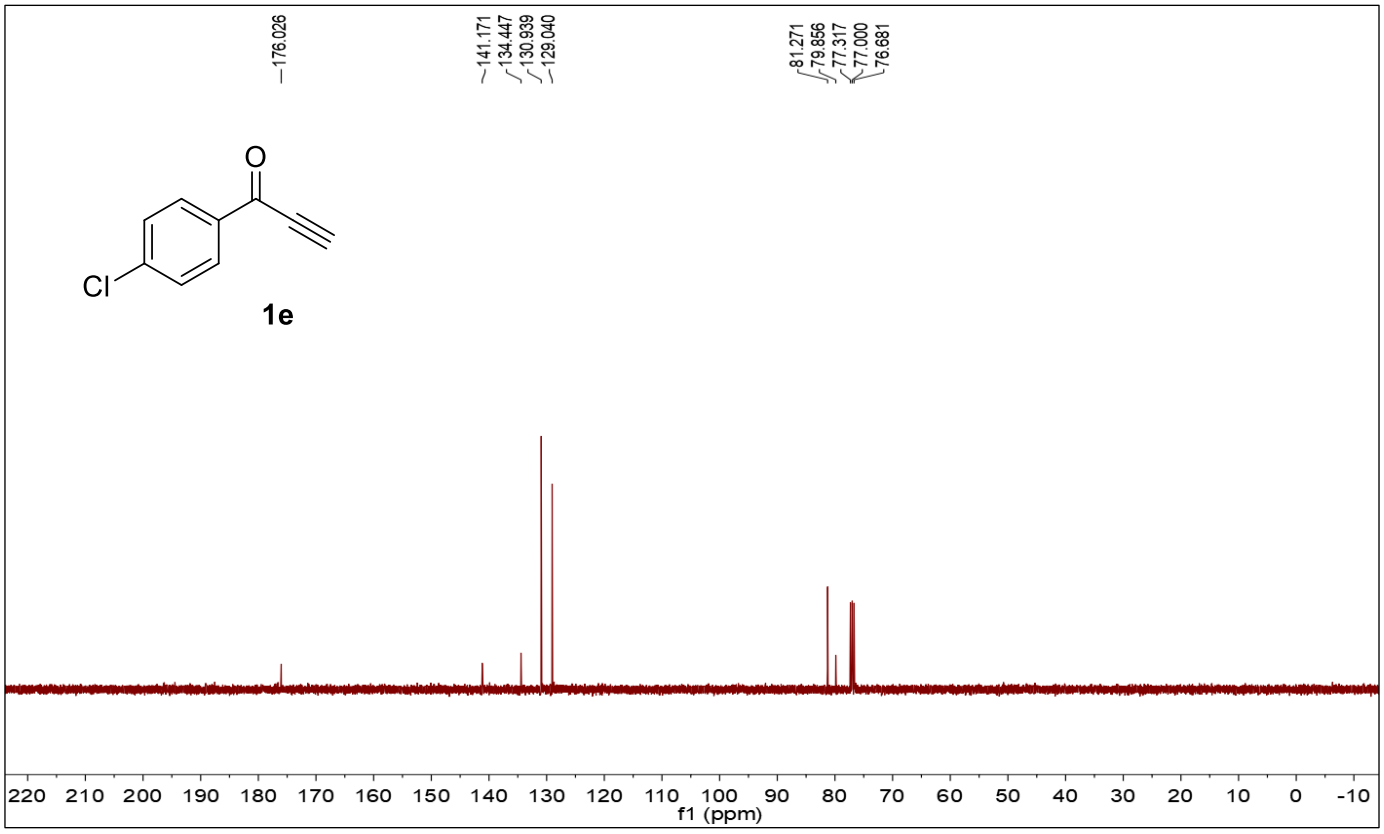

175.808
137.655
137.266
132.388
130.224
128.134
122.870
81.689
79.674
77.318
77.000
76.682
Br
O
1c
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

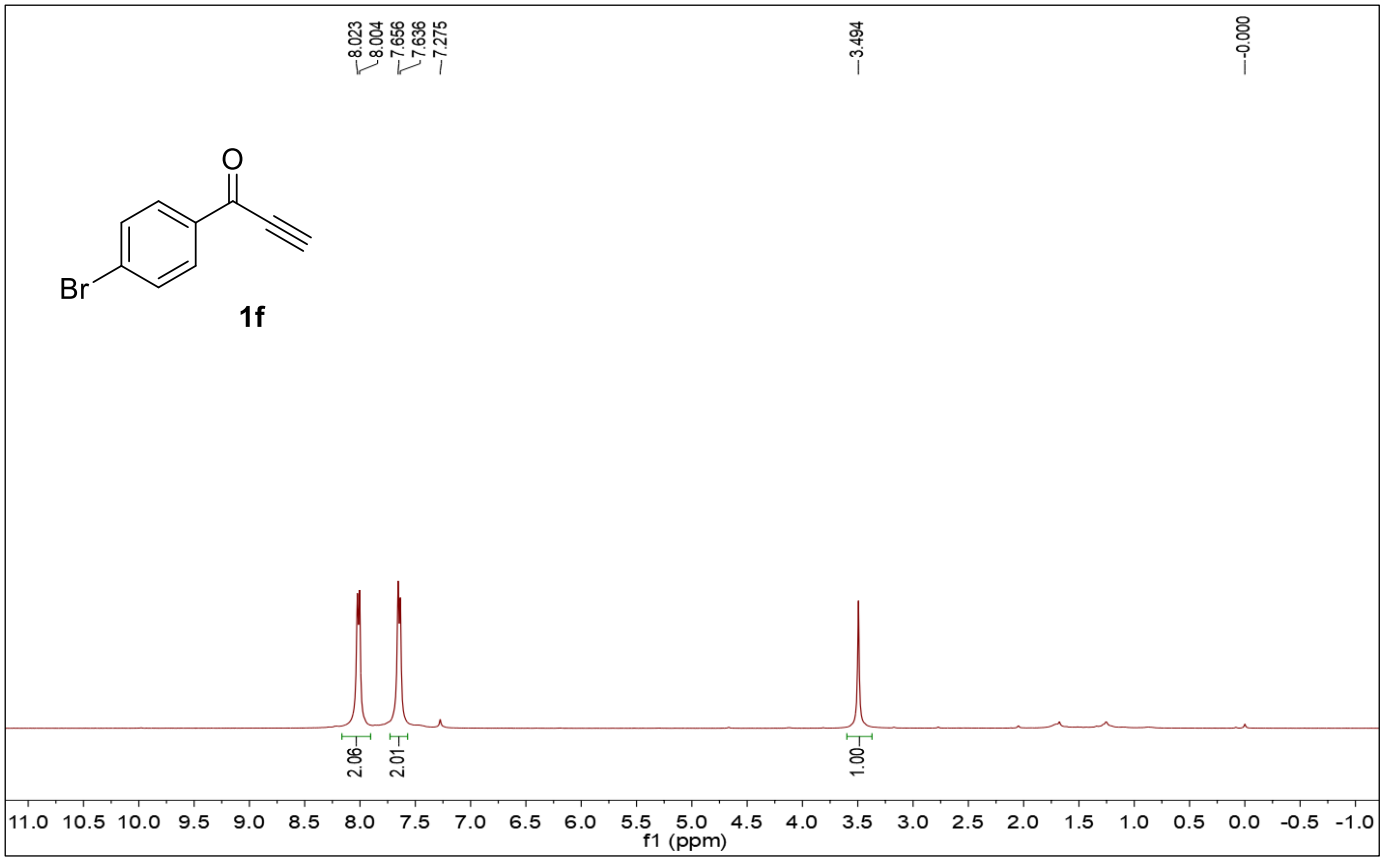

8.207
8.193
8.186
8.172
7.194
7.173
7.152
3.498
0.000
O
F
1d
2.05
2.11
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


175.677
167.883
165.323
132.610
132.581
132.392
132.294
116.010
115.788
81.089
79.900
77.318
77.000
76.681
O
F
1d
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


-102.161
O
F
1d
50
30
10
-10
-30
-50
-70
-90
-110
-130
-150
-170
-190
-210
-230
-250
f1 (ppm)


8.108
8.087
7.489
7.468
7.272
3.486
0.000
O
Cl
1e
2.01
2.09
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


176.026
141.171
134.447
130.939
129.040
81.271
79.856
77.317
77.000
76.681
O
Cl
1e
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


8.023
8.004
7.656
7.636
7.275
3.494
0.000
O
Br
1f
2.06
2.01
1.00
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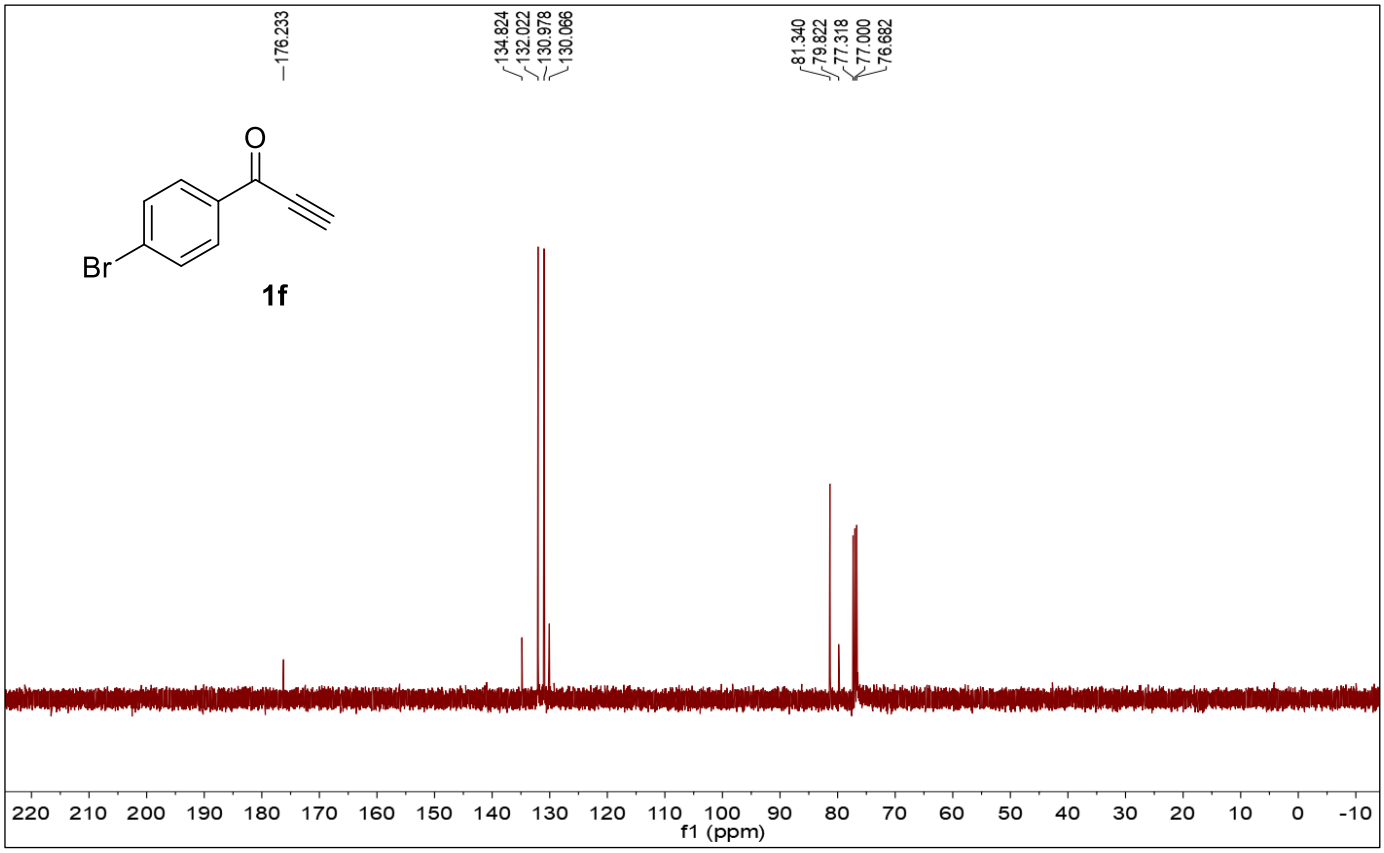
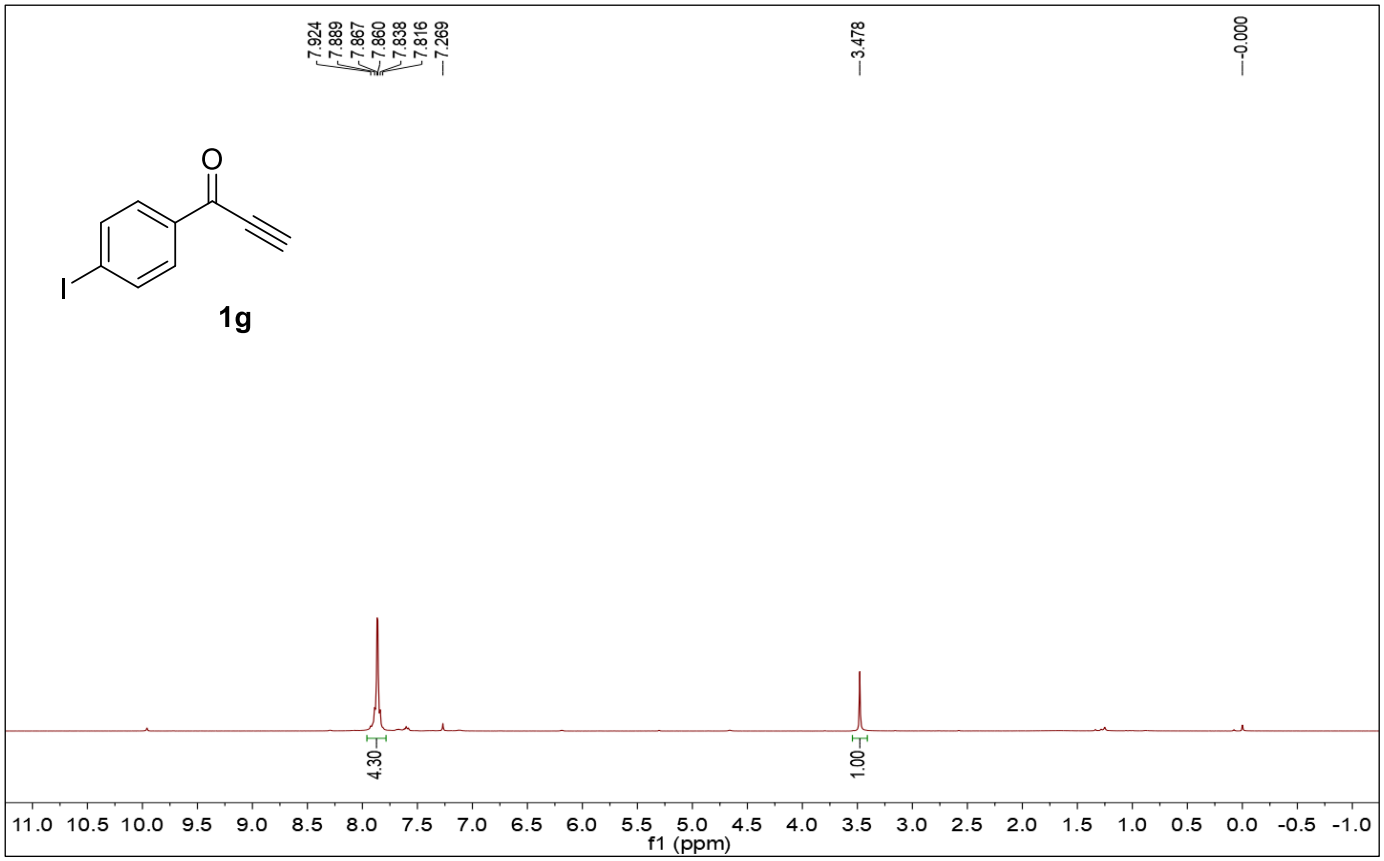
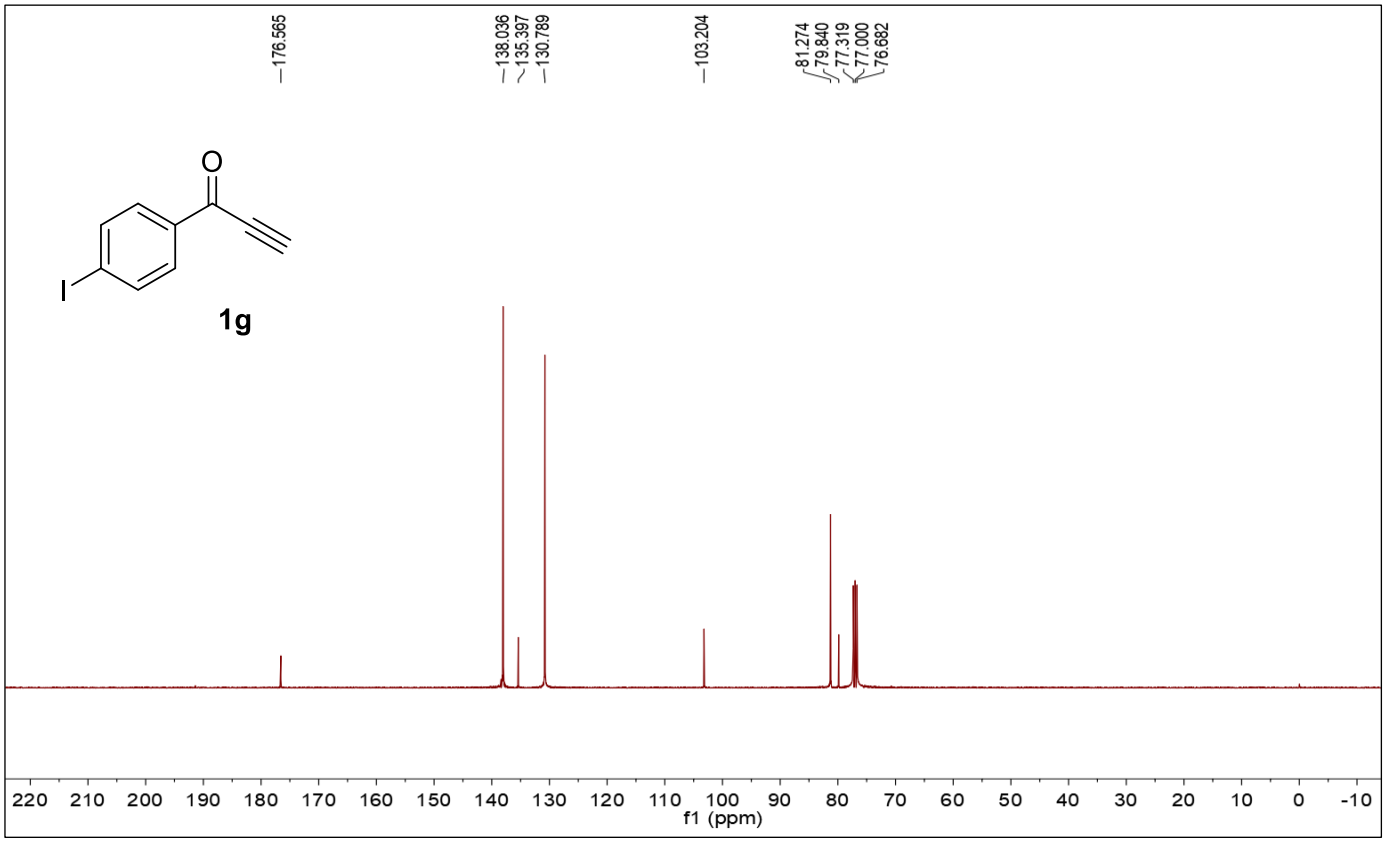
f1 (ppm)


176.233
134.824
132.022
130.978
130.066
81.340
79.822
77.318
77.000
76.682
O
Br
1f
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


7.924
7.889
7.867
7.860
7.838
7.816
7.269
3.478
0.000
O
I
1g
4.30
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


176.565
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135.397
130.789
103.204
81.274
79.840
77.319
77.000
76.682
O
I
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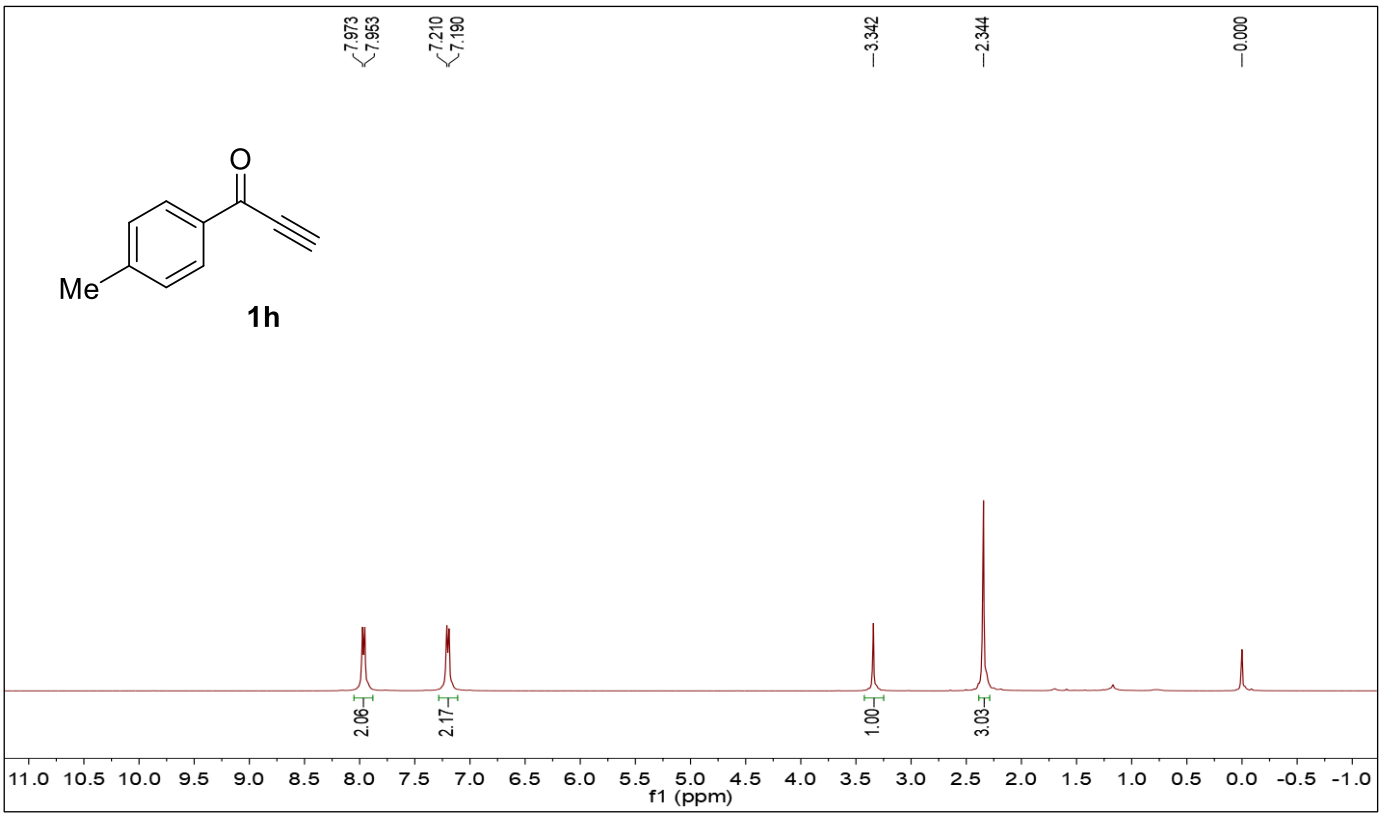
f1 (ppm)


7.973
7.953
7.210
7.190
3.342
2.344
0.000
O
Me
1h
2.06
2.17
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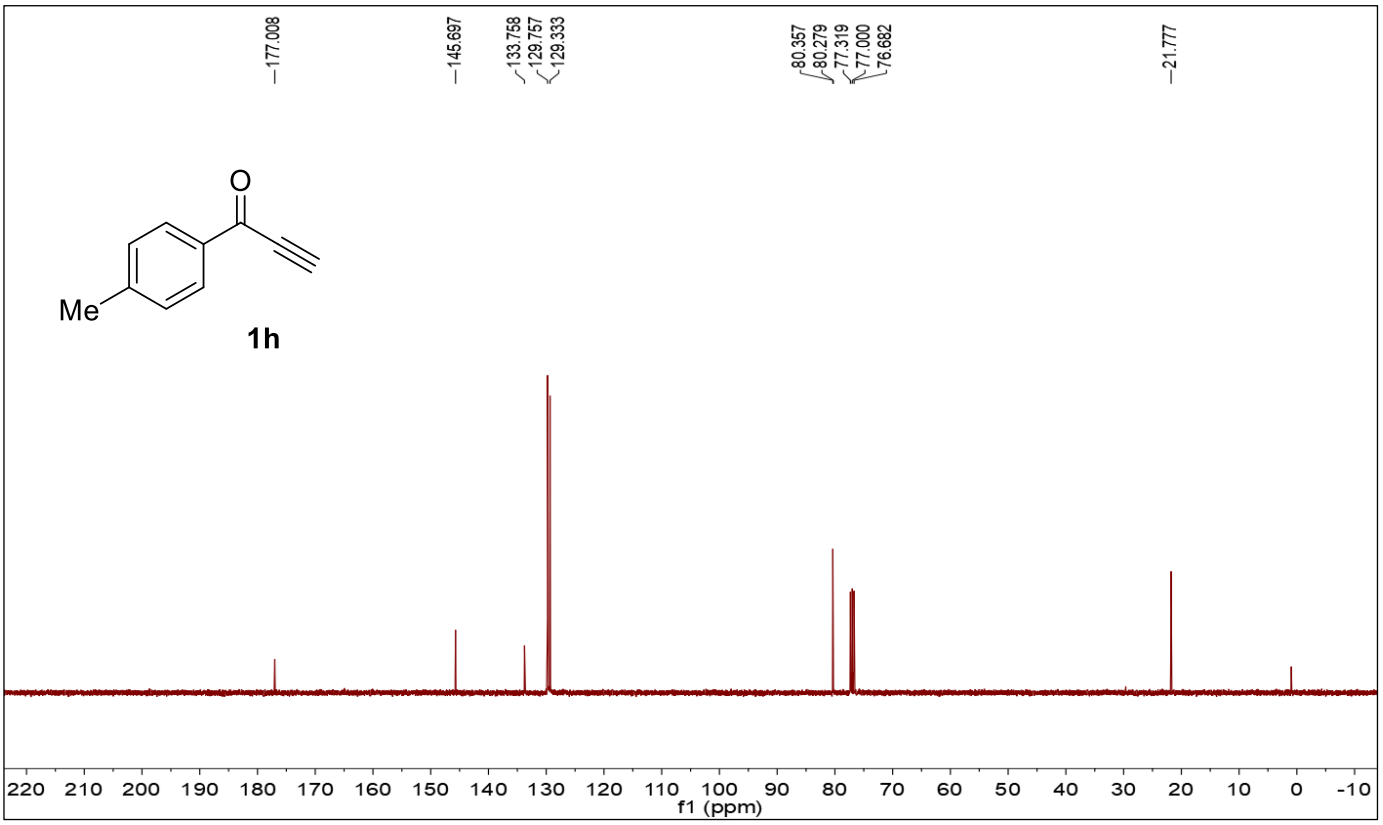
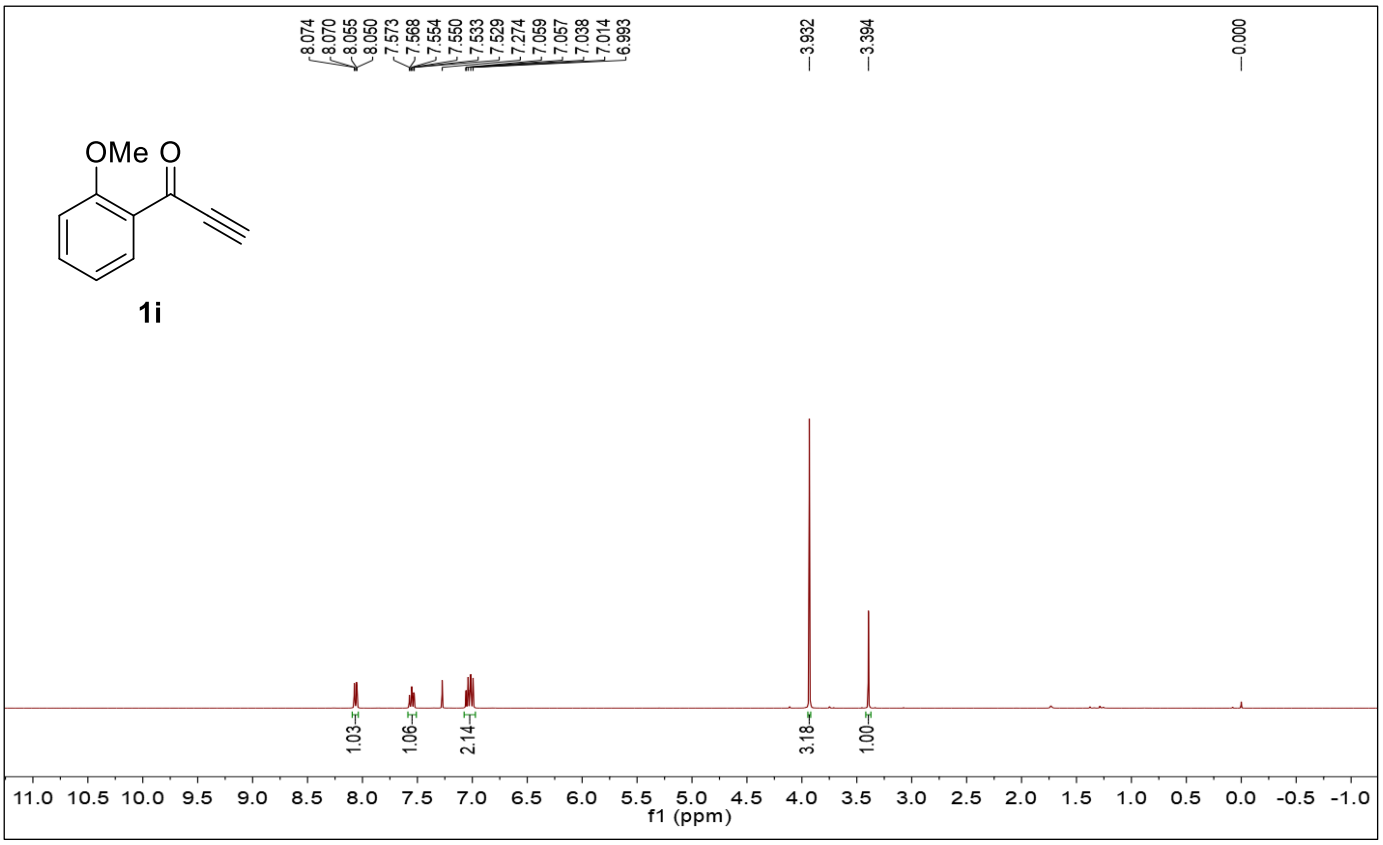
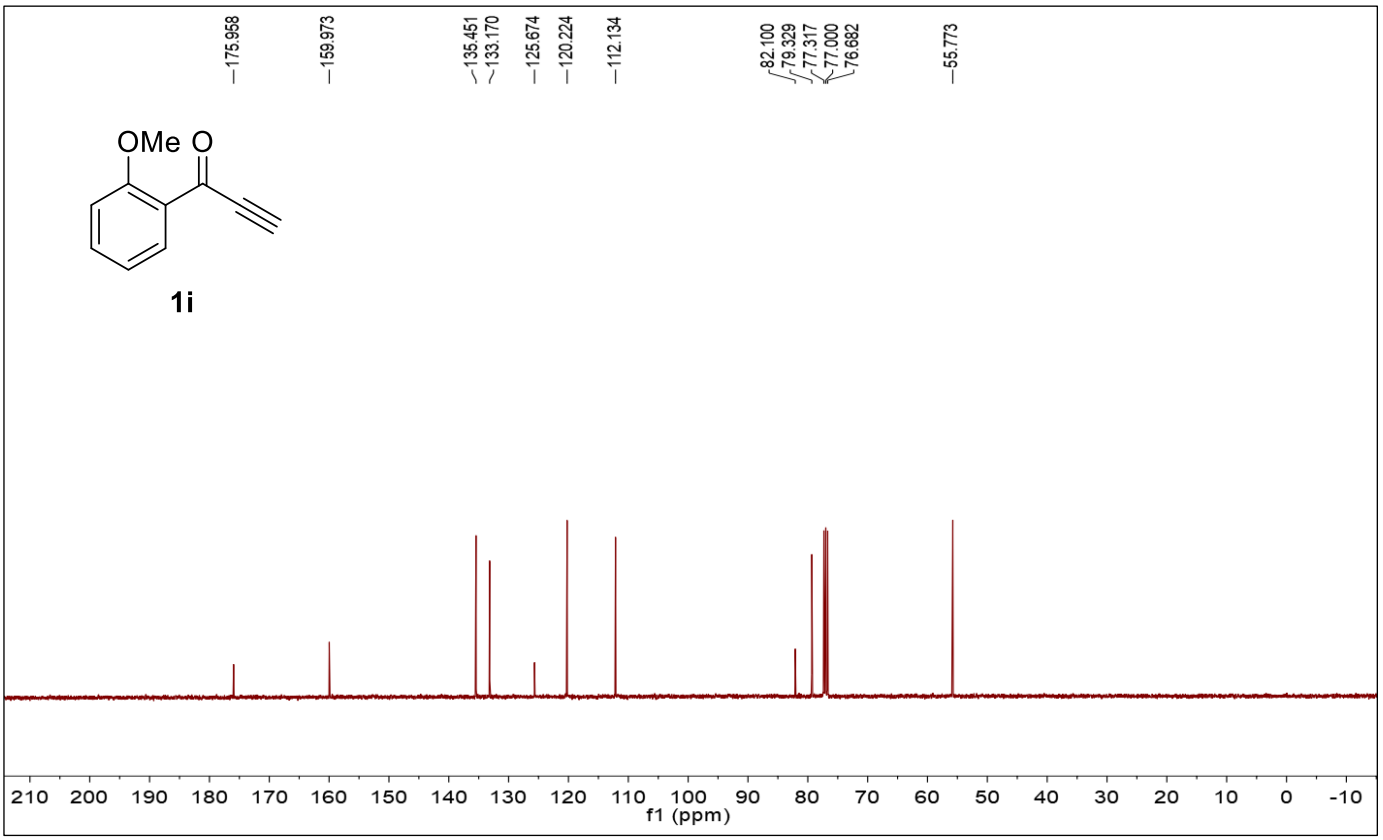
3.03
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


177.008
145.697
133.758
129.757
129.333
80.357
80.279
77.319
77.000
76.682
21.777
O
Me
1h
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


8.074
8.070
8.055
8.050
7.573
7.568
7.554
7.550
7.533
7.529
7.274
7.059
7.057
7.038
7.014
6.993
3.932
3.394
0.000
OMe O
1i
1.03
1.06
2.14
3.18
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


175.958
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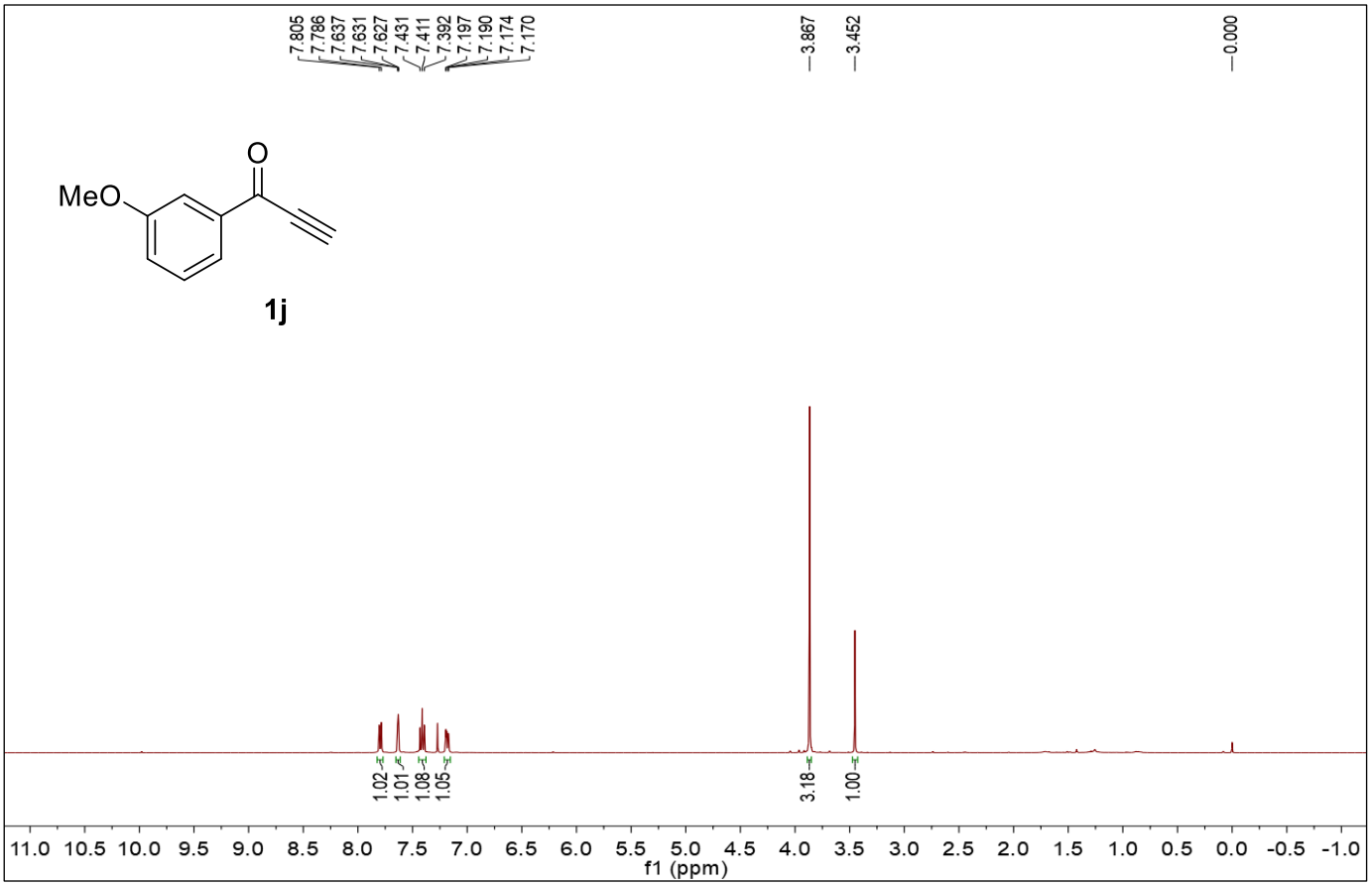
133.170
125.674
120.224
112.134
82.100
79.329
77.317
77.000
76.682
55.773
OMe O
1i
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


7.805
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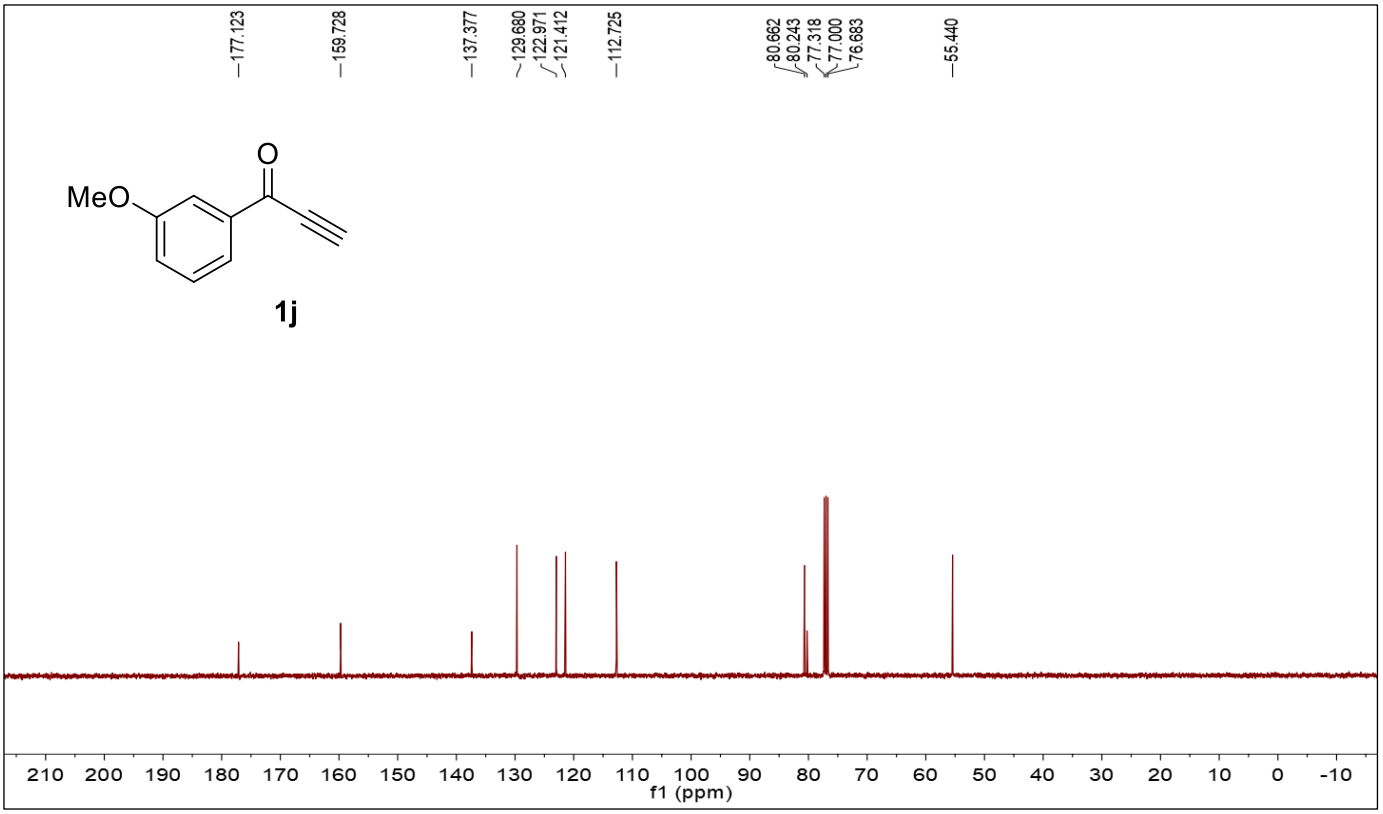
7.637
7.631
7.627
7.431
7.411
7.392
7.197
7.190
7.174
7.170
3.867
3.452
0.000
MeO
O
1j
1.02
1.01
1.08
1.05
3.18
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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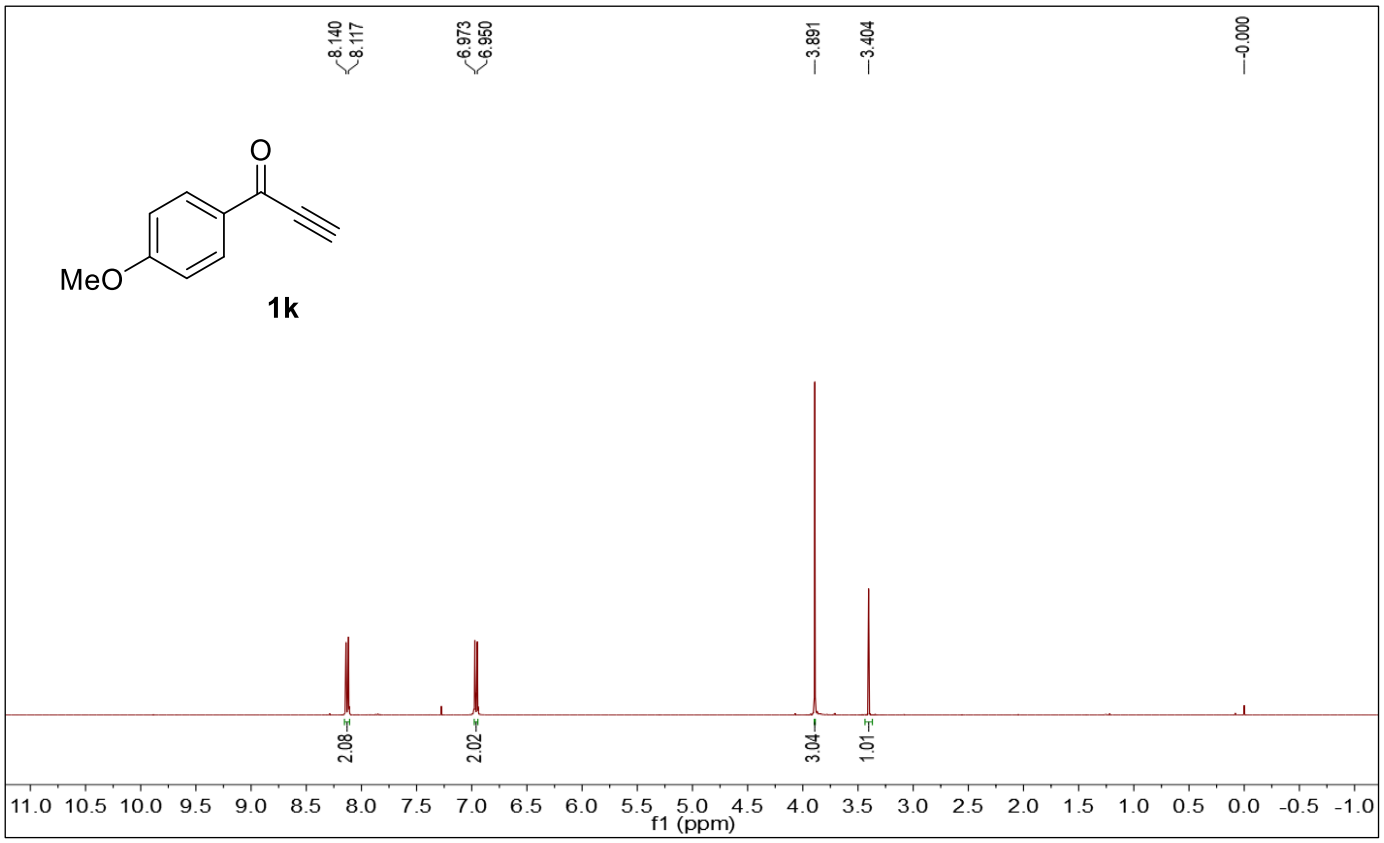
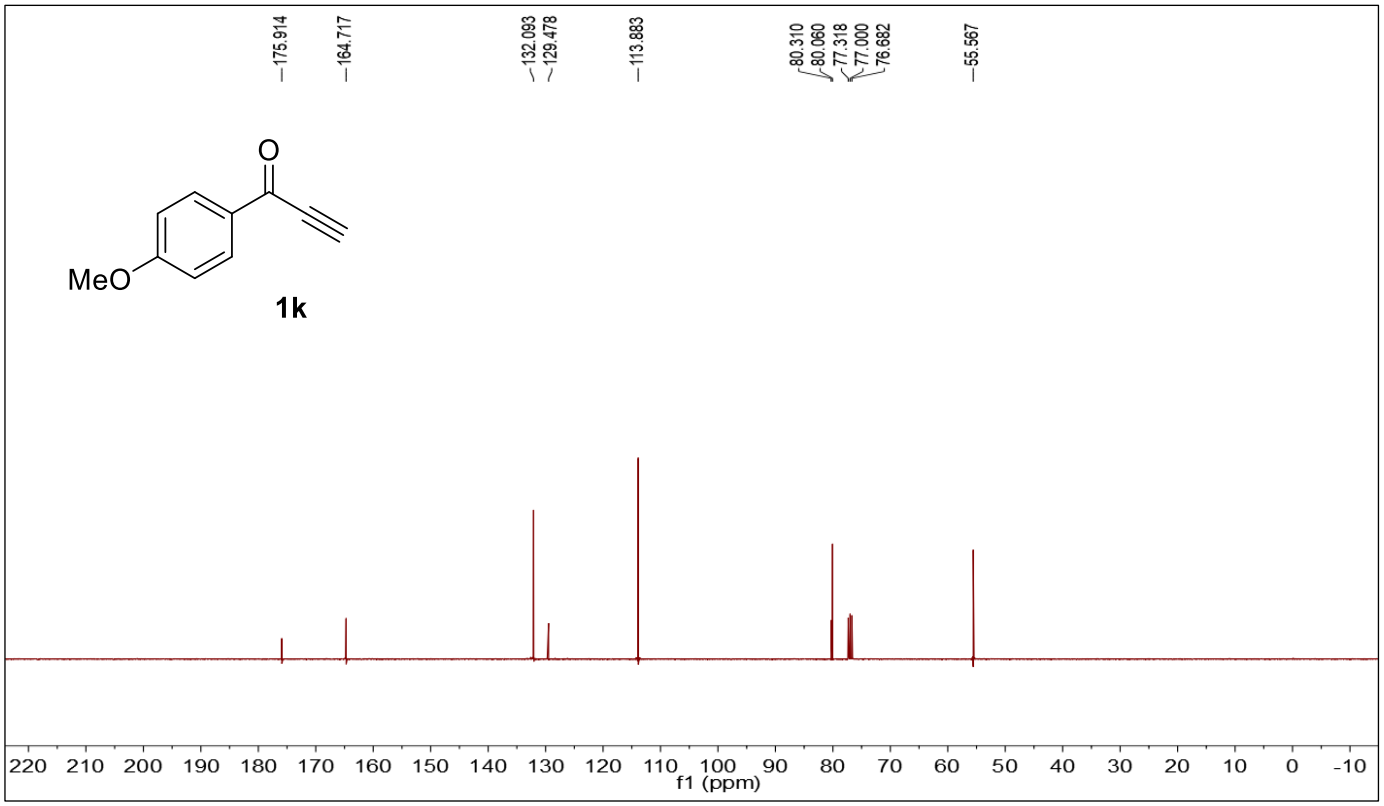
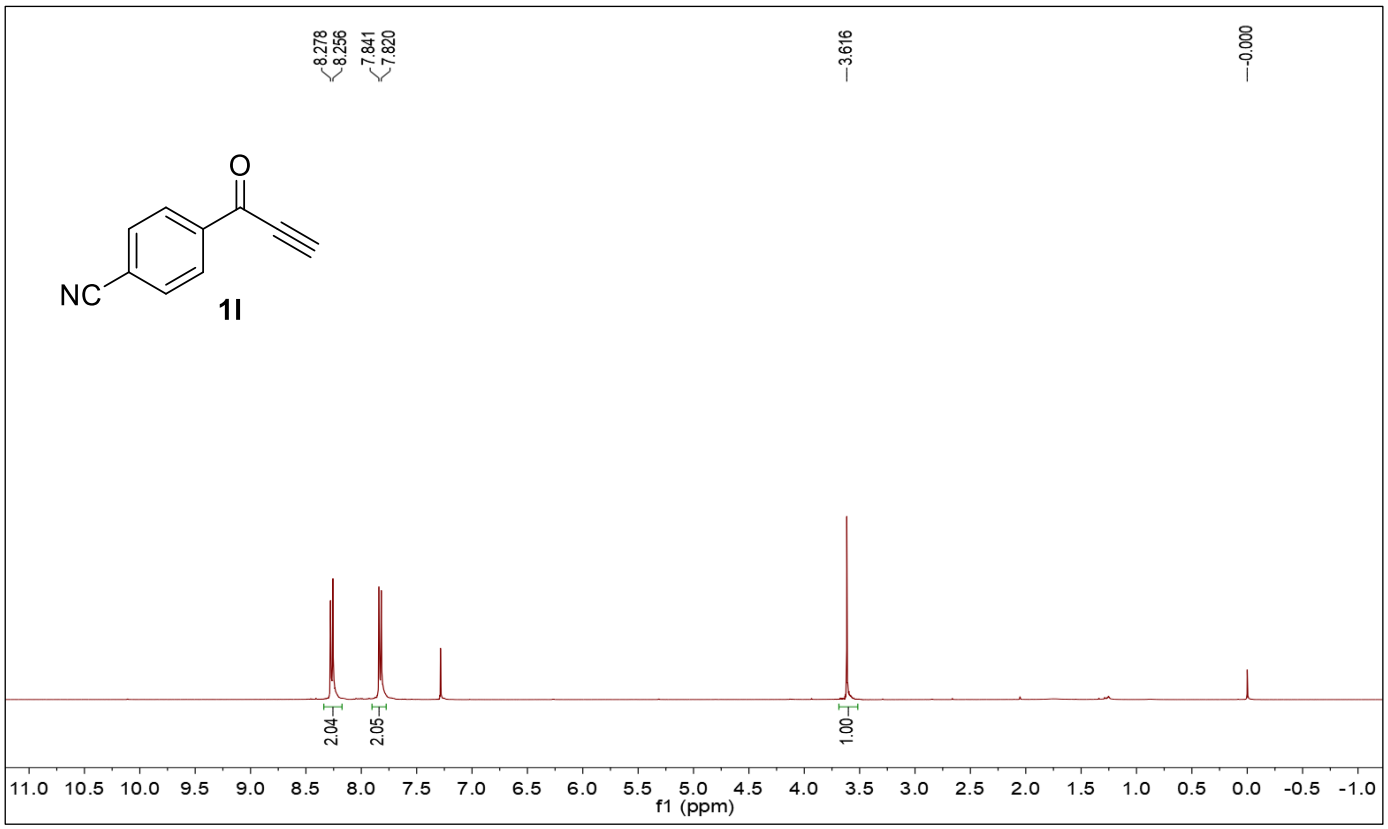
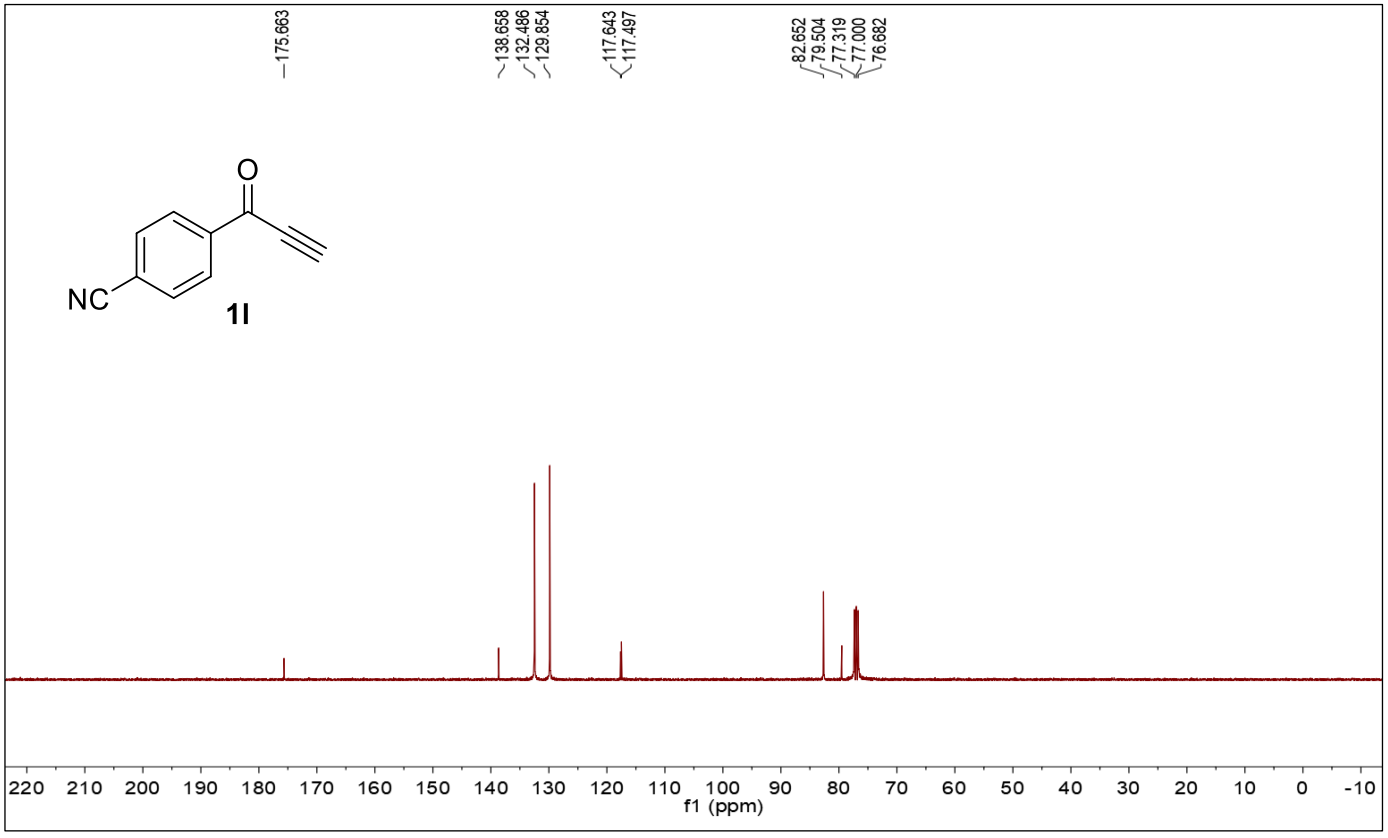
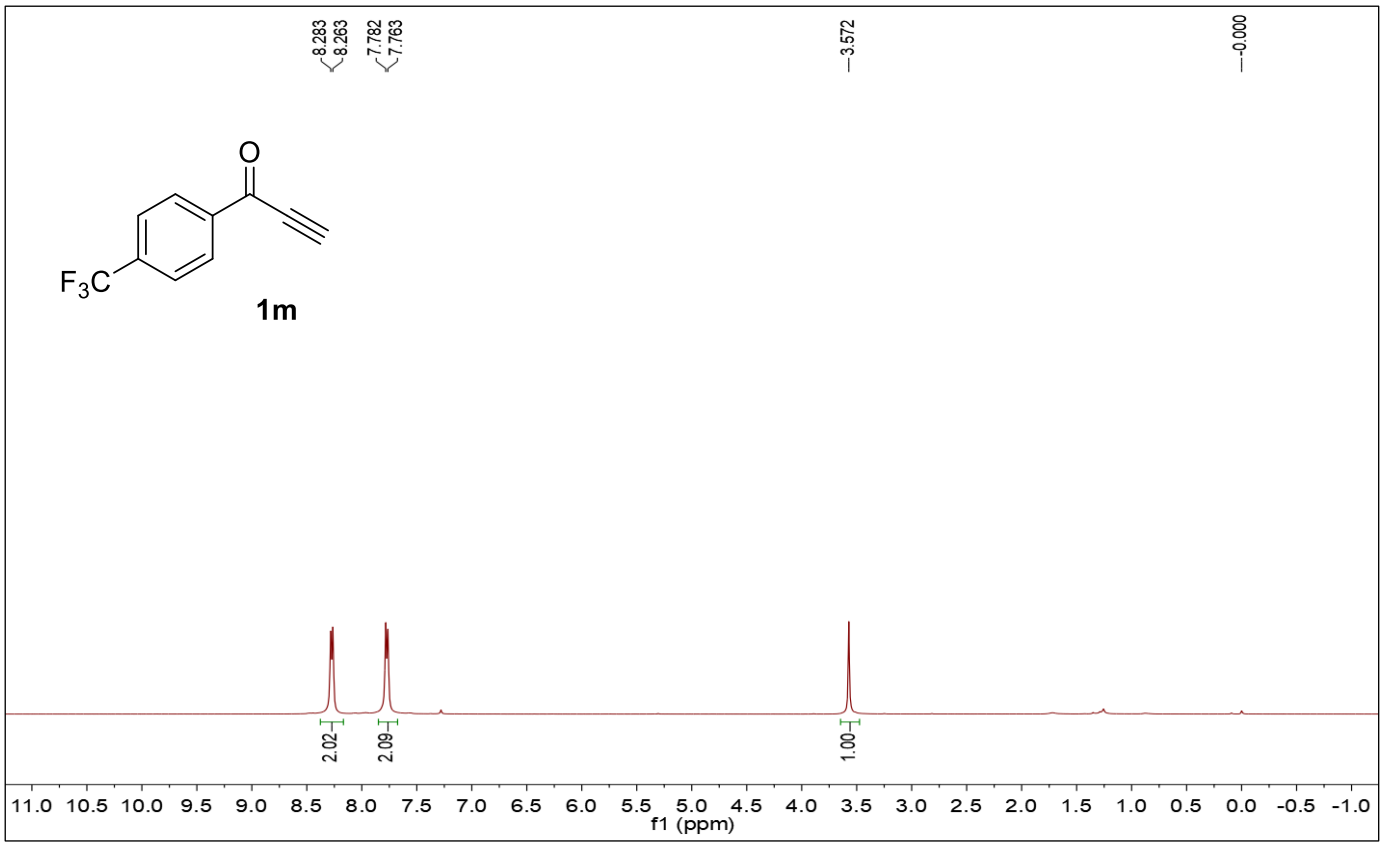
112.725
80.662
80.243
77.318
77.000
76.683
55.440
MeO
O
1j
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

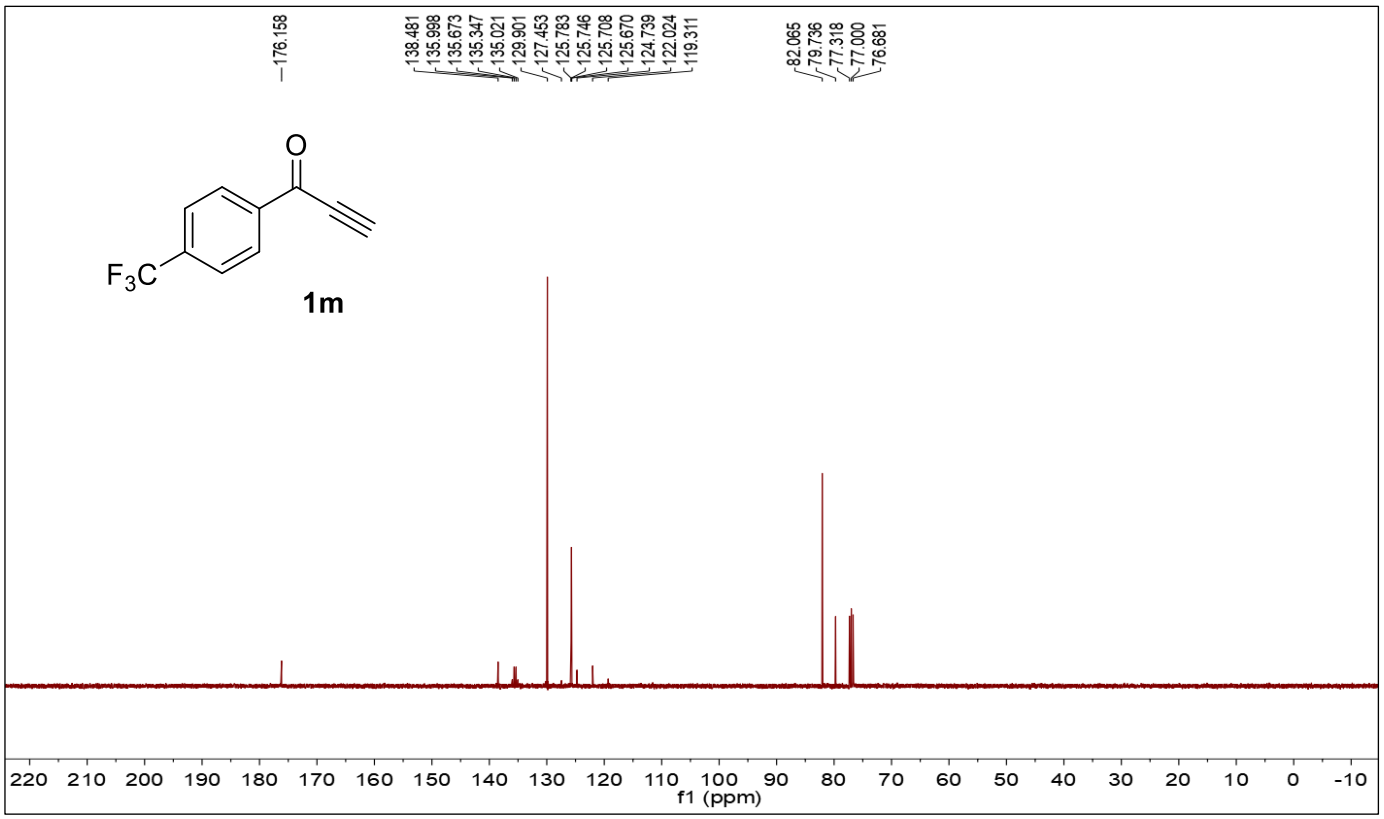

8.140
8.117
6.973
6.950
3.891
3.404
0.000
O
MeO
1k
2.08
2.02
3.04
1.01
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

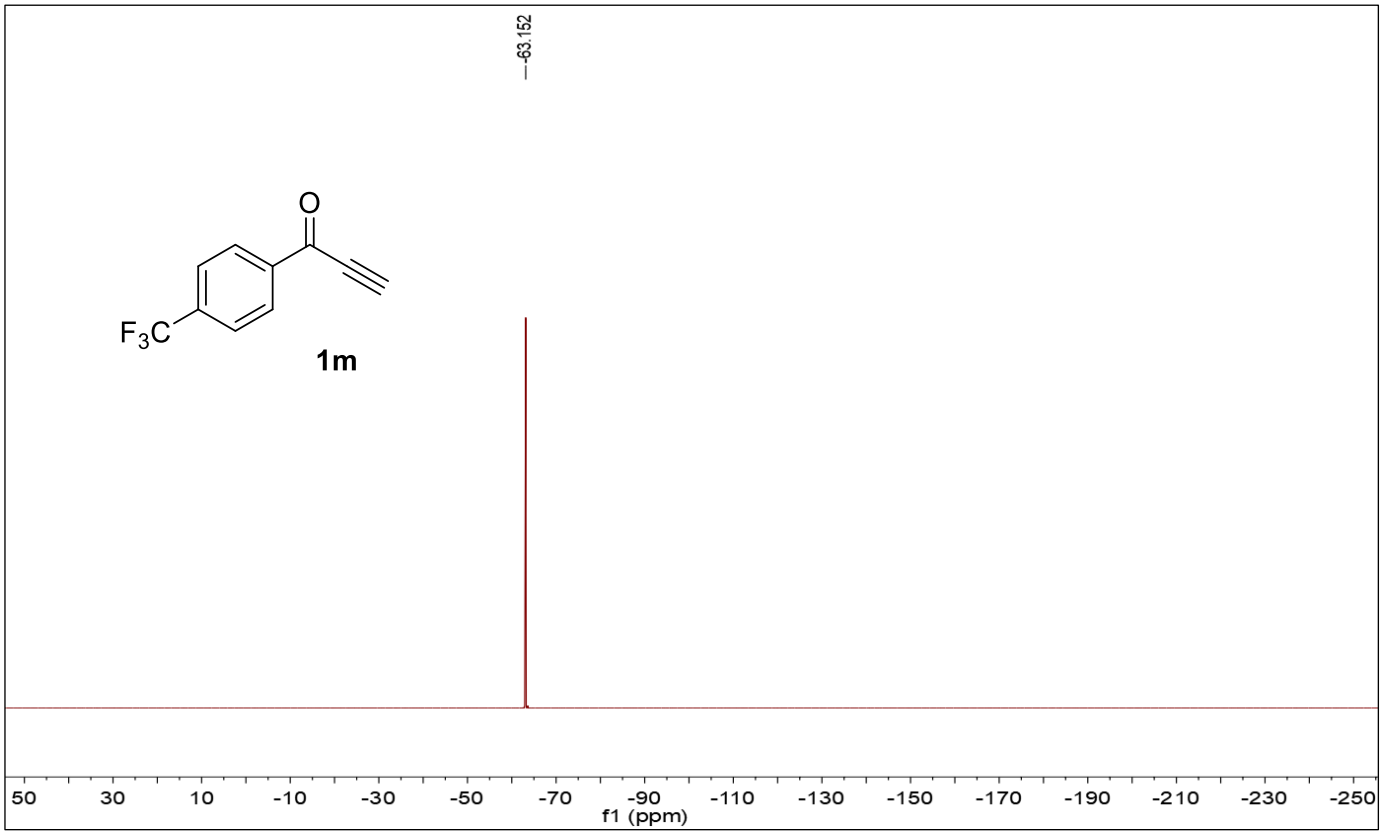

175.914
164.717
132.093
129.478
113.883
80.310
80.060
77.318
77.000
76.682
55.567
O
MeO
1k
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

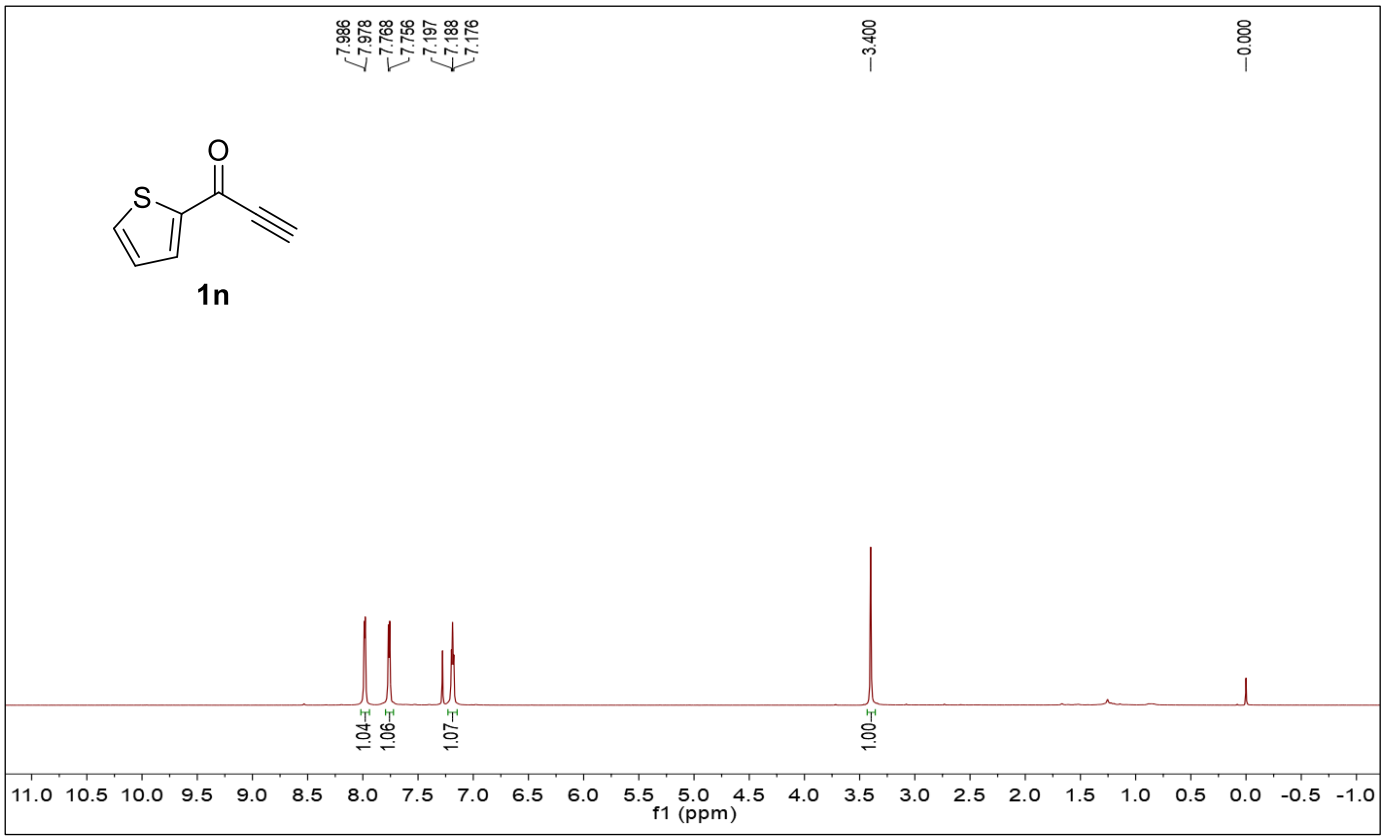

8.278
8.256
7.841
7.820
3.616
0.000
O
NC
1l
2.04
2.05
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

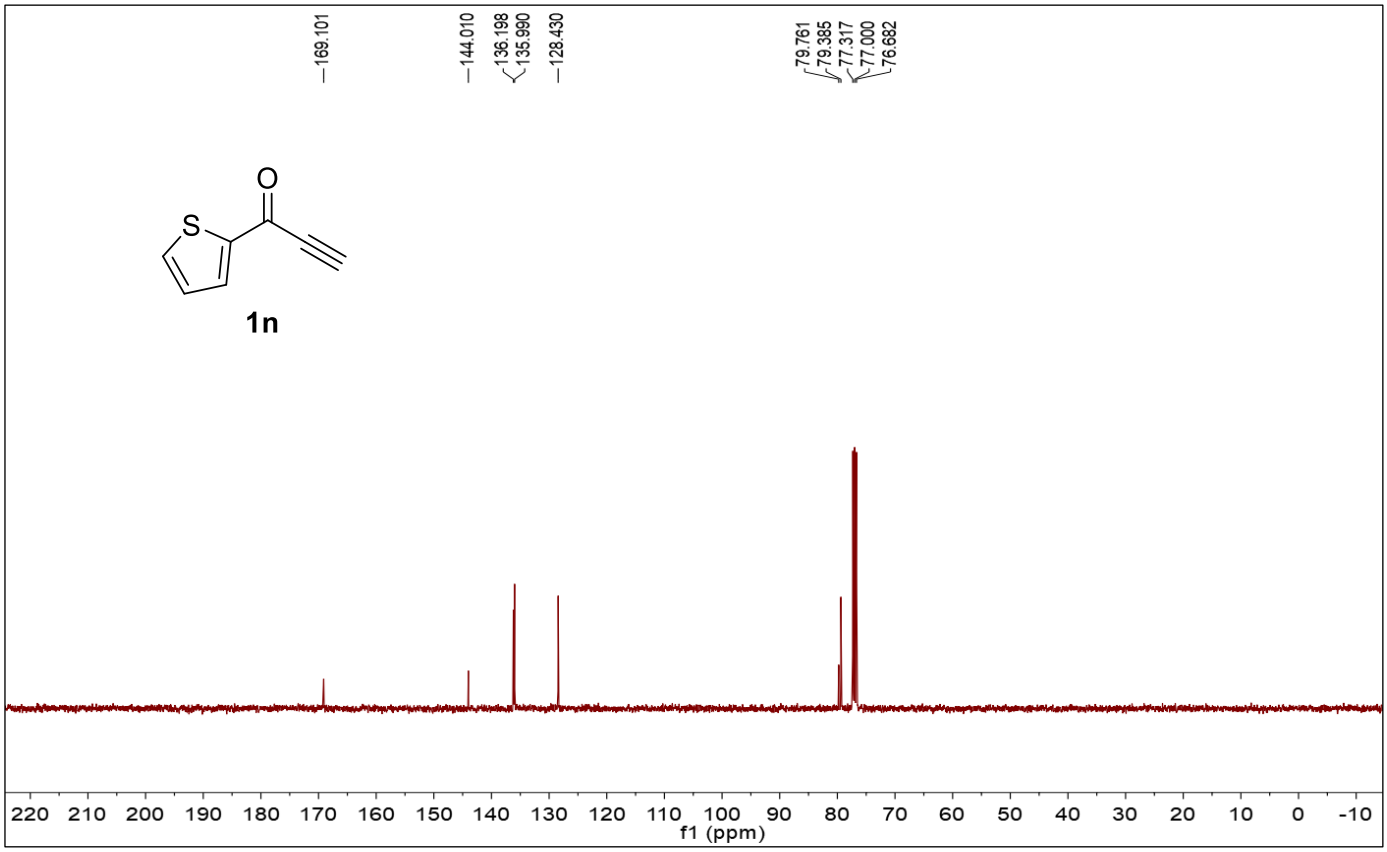

175.663
138.658
132.486
129.854
117.643
117.497
82.652
79.504
77.319
77.000
76.682
O
NC
1l
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

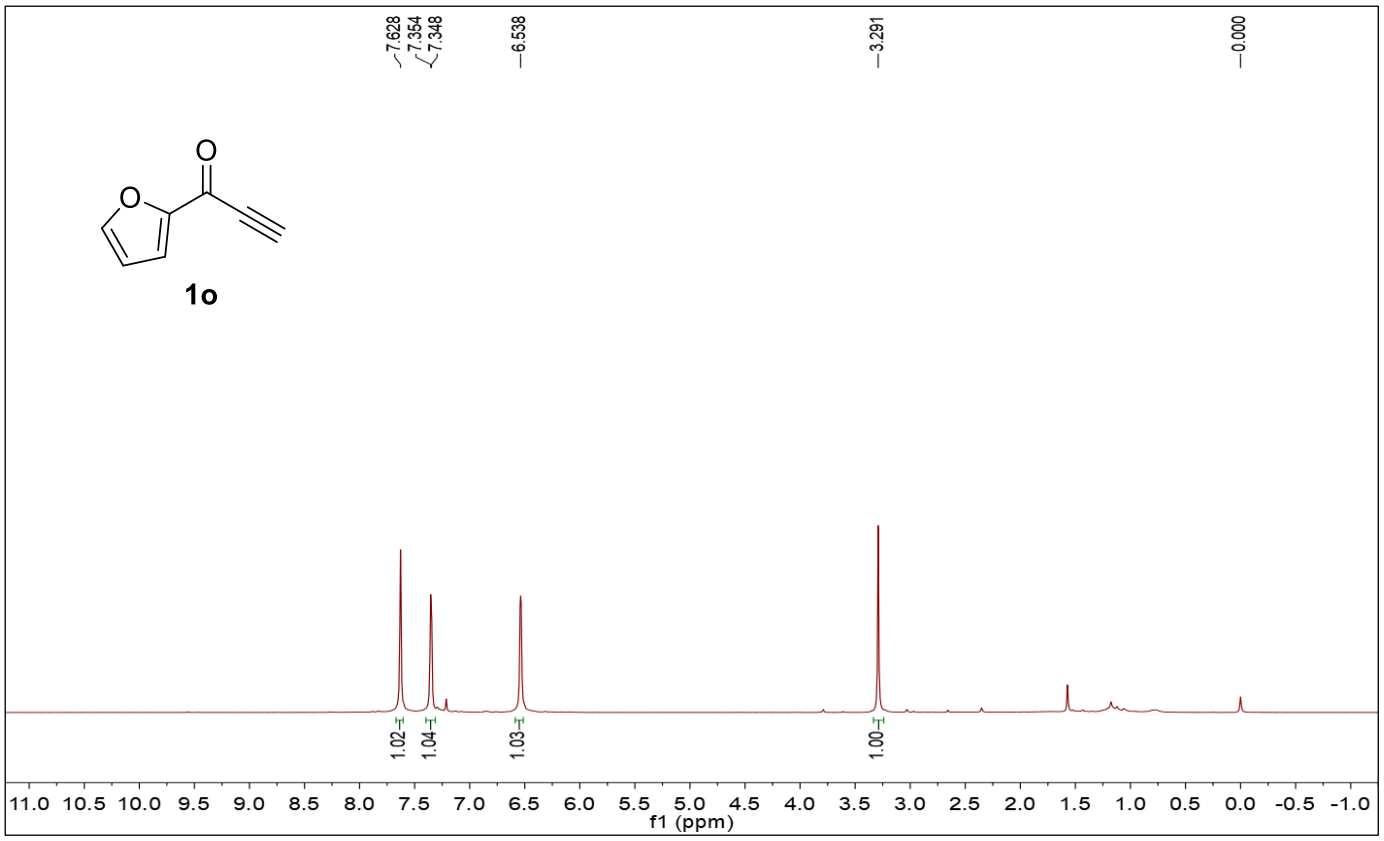

8.283
8.263
7.782
7.763
3.572
0.000
O
F3C
1m
2.02
2.09
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


176.158
138.481
135.998
135.673
135.347
135.021
129.901
127.453
125.783
125.746
125.708
125.670
124.739
122.024
119.311
82.065
79.736
77.318
77.000
76.681
O
F3C
1m
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

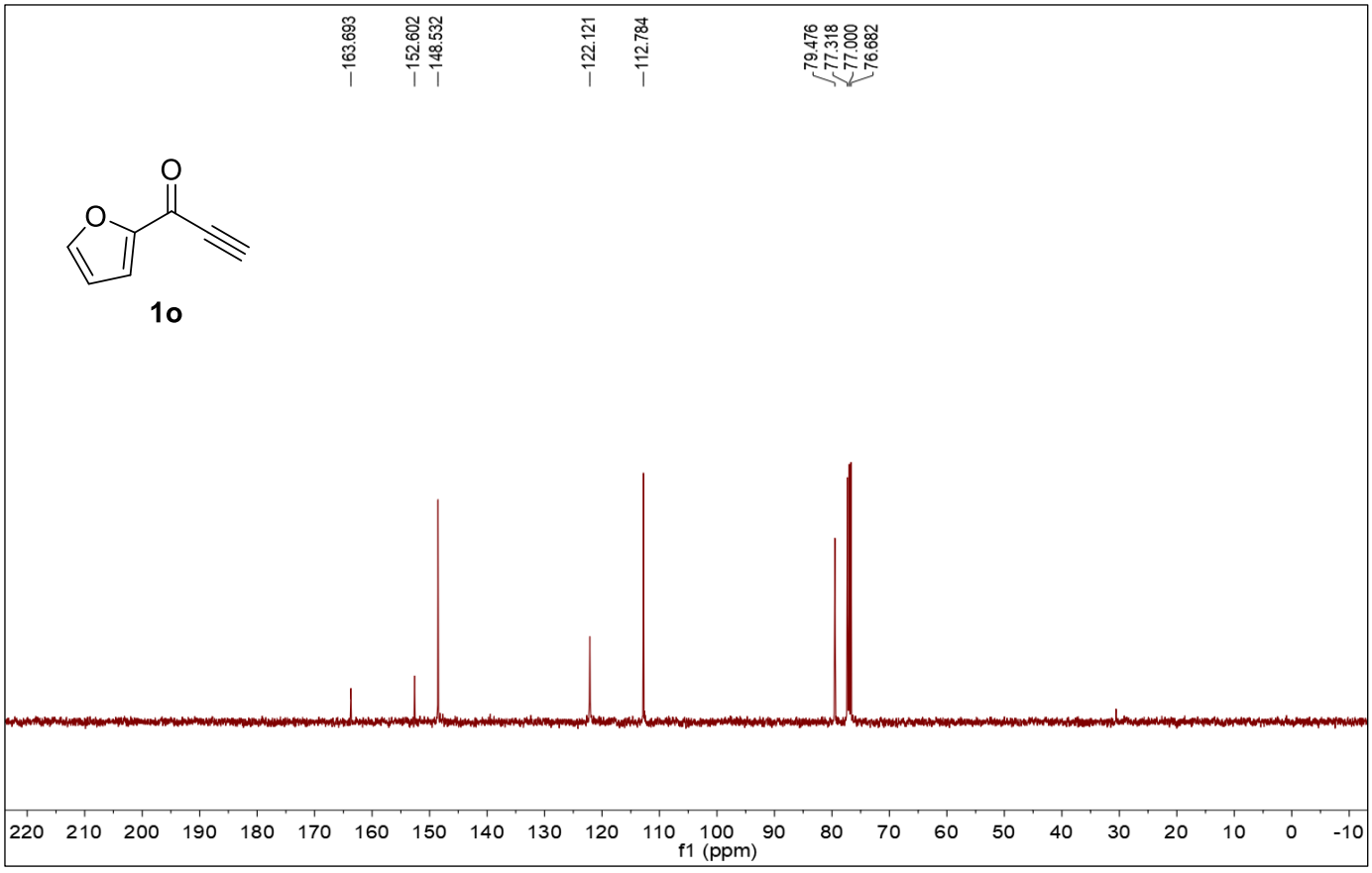

-63.152
O
F3C
1m
50
30
10
-10
-30
-50
-70
-90
-110
-130
-150
-170
-190
-210
-230
-250
f1 (ppm)


7.986
7.978
7.768
7.756
7.197
7.188
7.176
3.400
0.000
O
S
1n
1.04
1.06
1.07
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

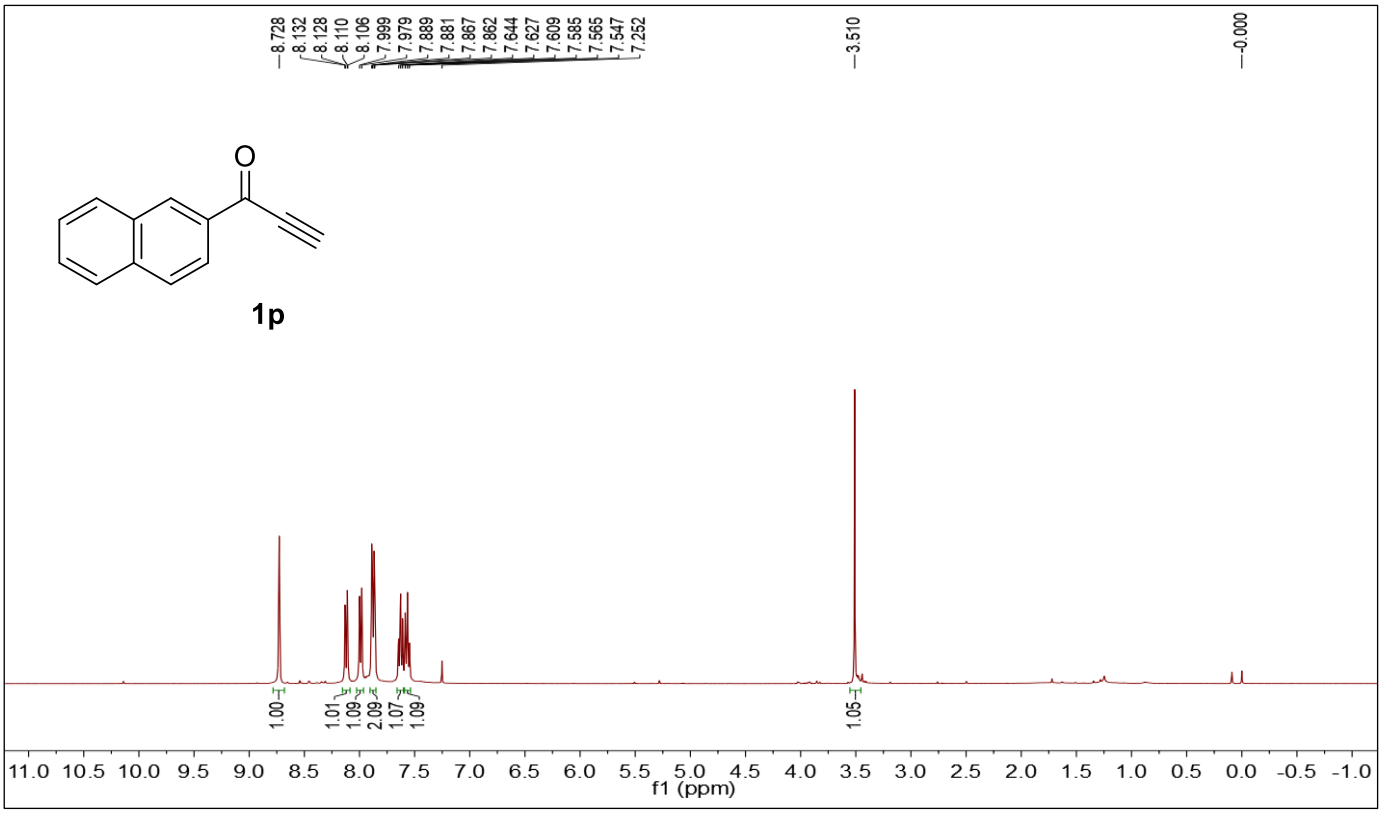

169.101
144.010
136.198
135.990
128.430
79.761
79.385
77.317
77.000
76.682
O
S
1n
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

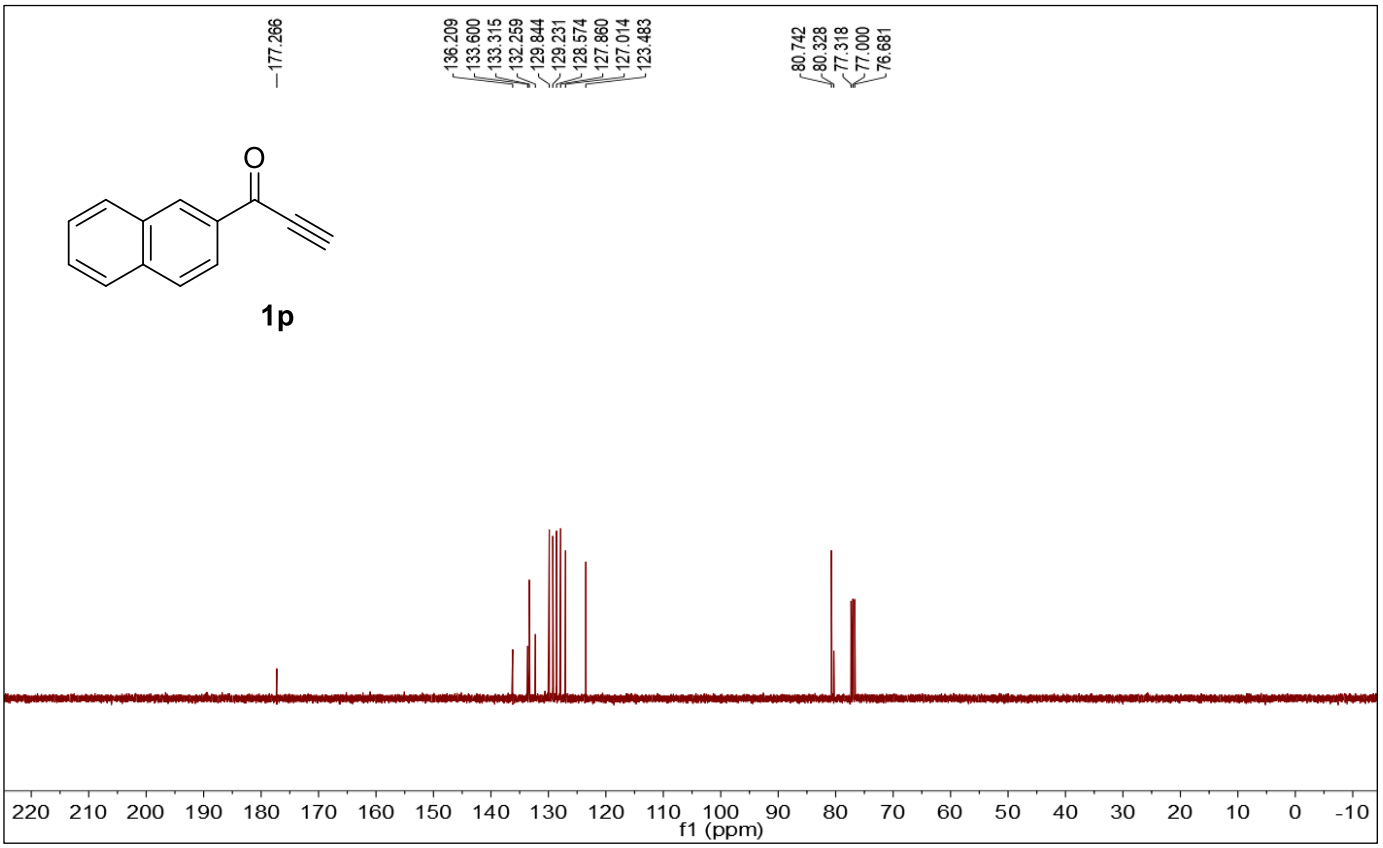

7.628
7.354
7.348
6.538
3.291
0.000
1.02
1.04
1.03
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

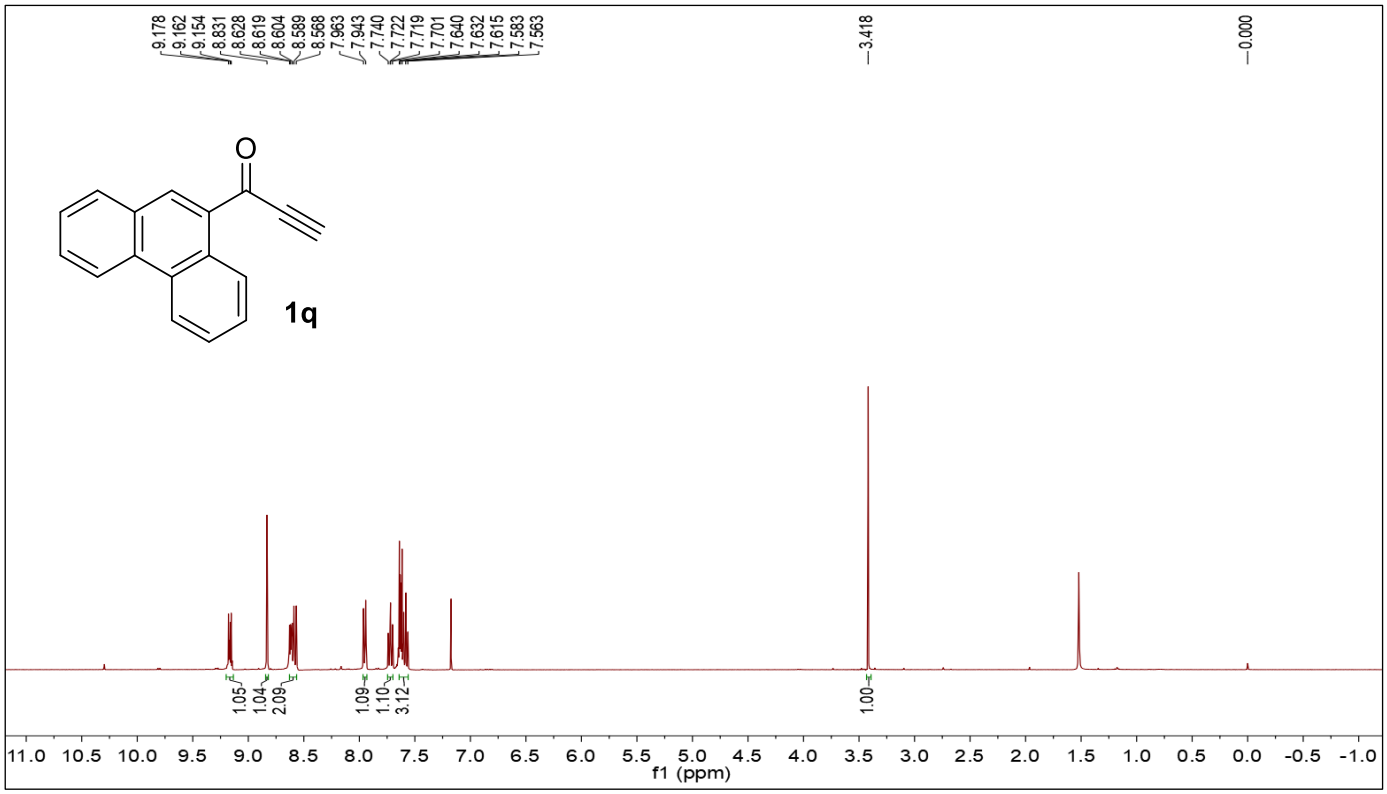
1o

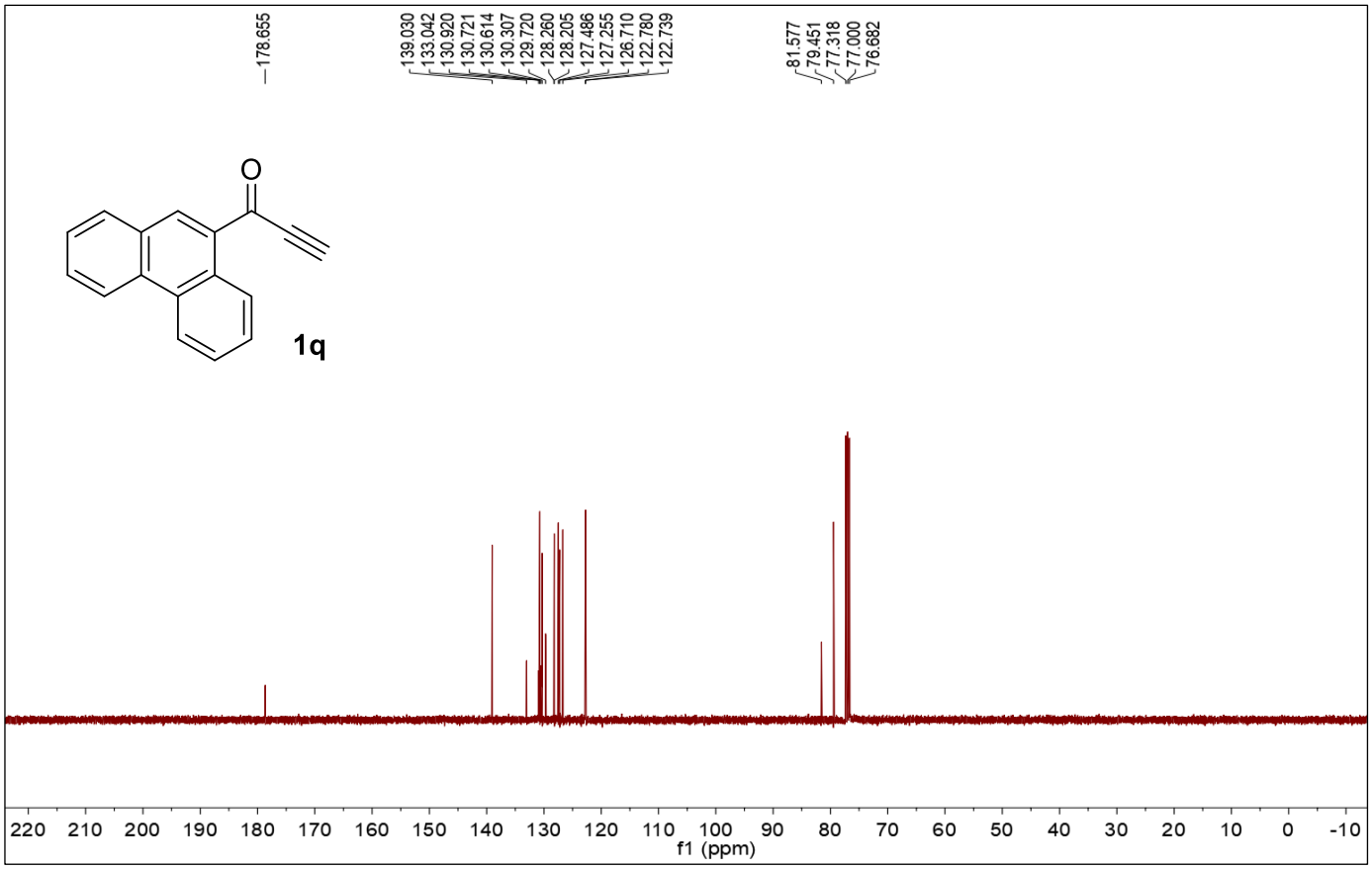

163.693
152.602
148.532
122.121
112.784
79.476
77.318
77.000
76.682
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

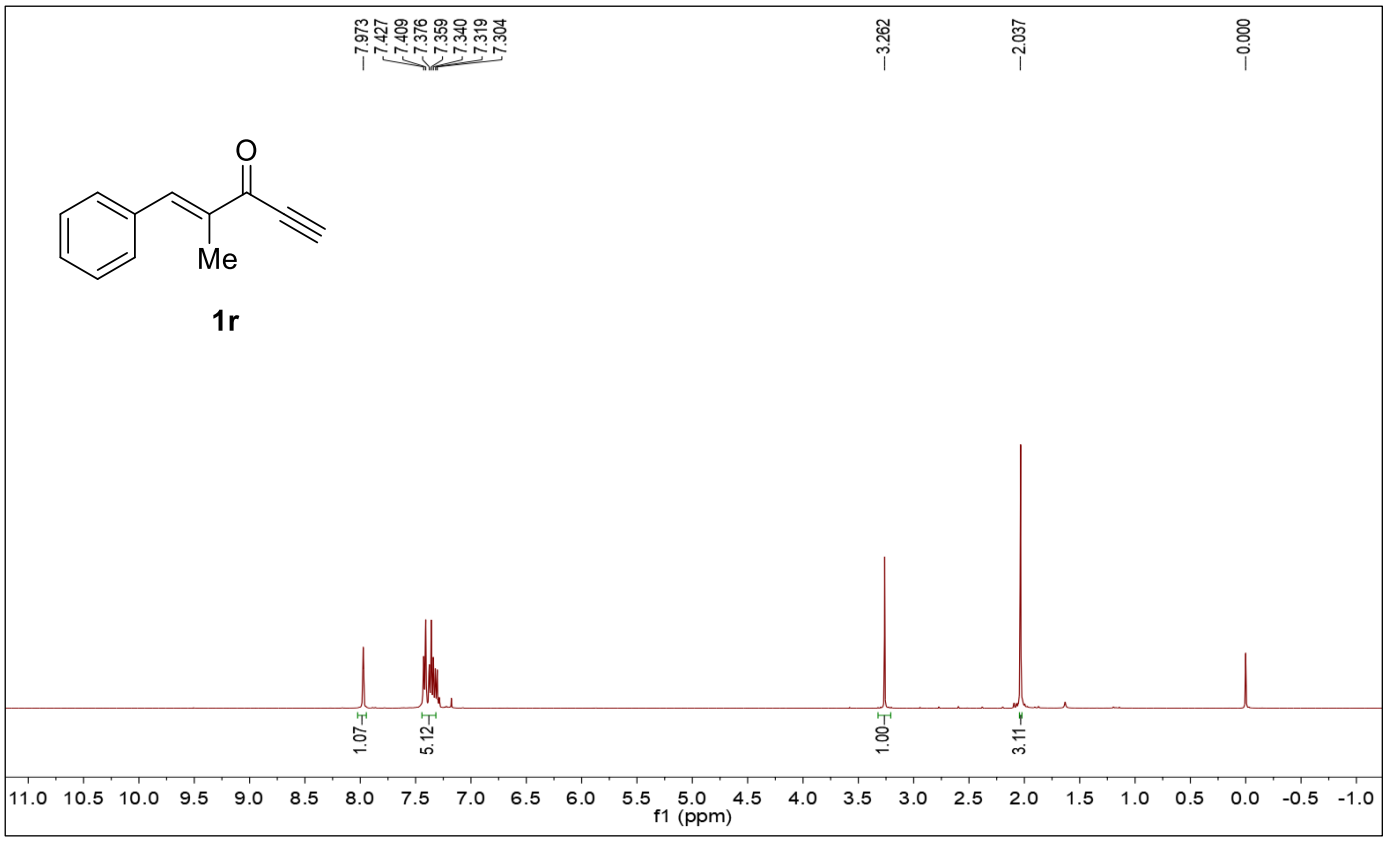
1o

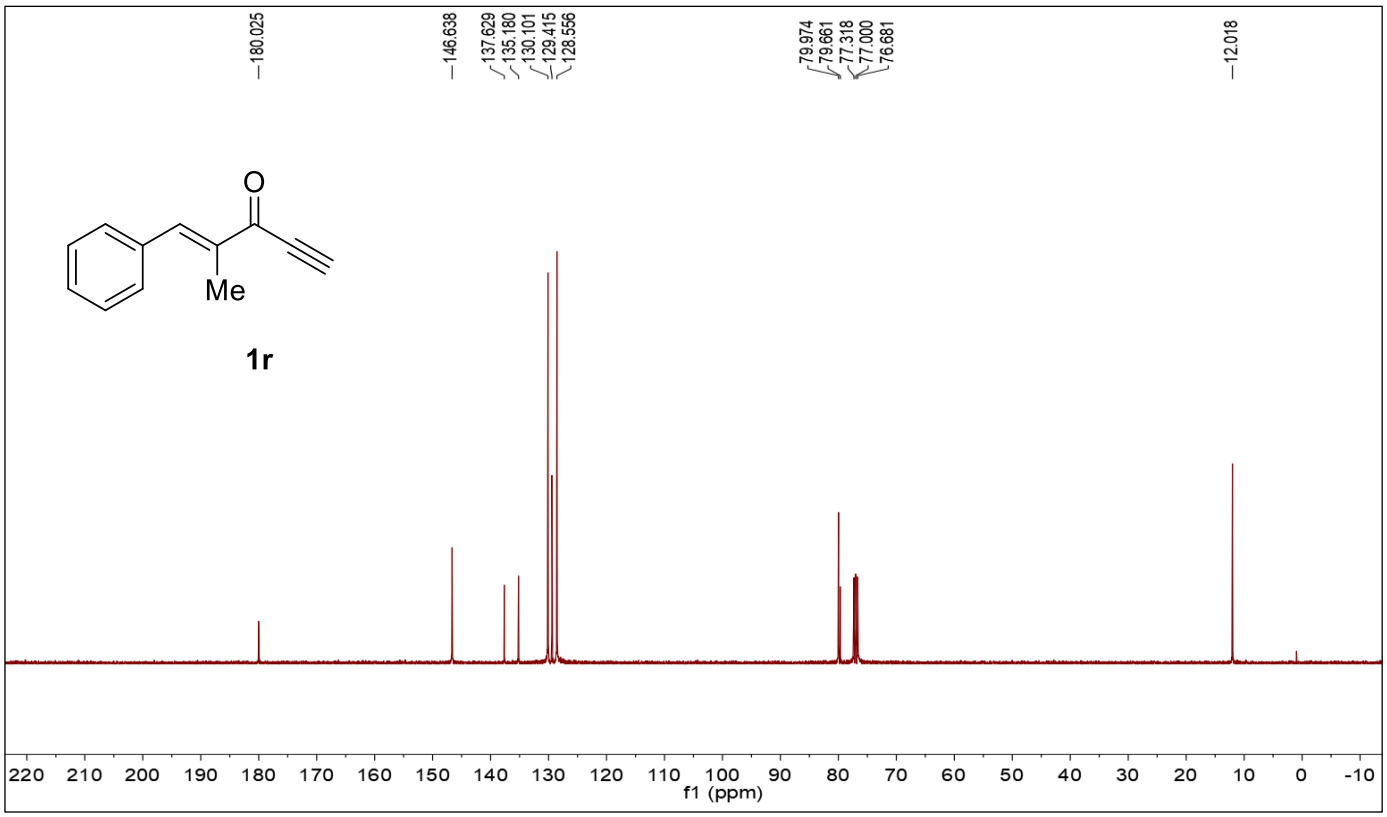

8.728
8.132
8.128
8.110
8.106
7.999
7.979
7.889
7.881
7.867
7.862
7.644
7.627
7.609
7.585
7.565
7.547
7.252
3.510
-0.000
O
1p
1.00
1.01
1.09
2.09
1.07
1.09
1.05
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

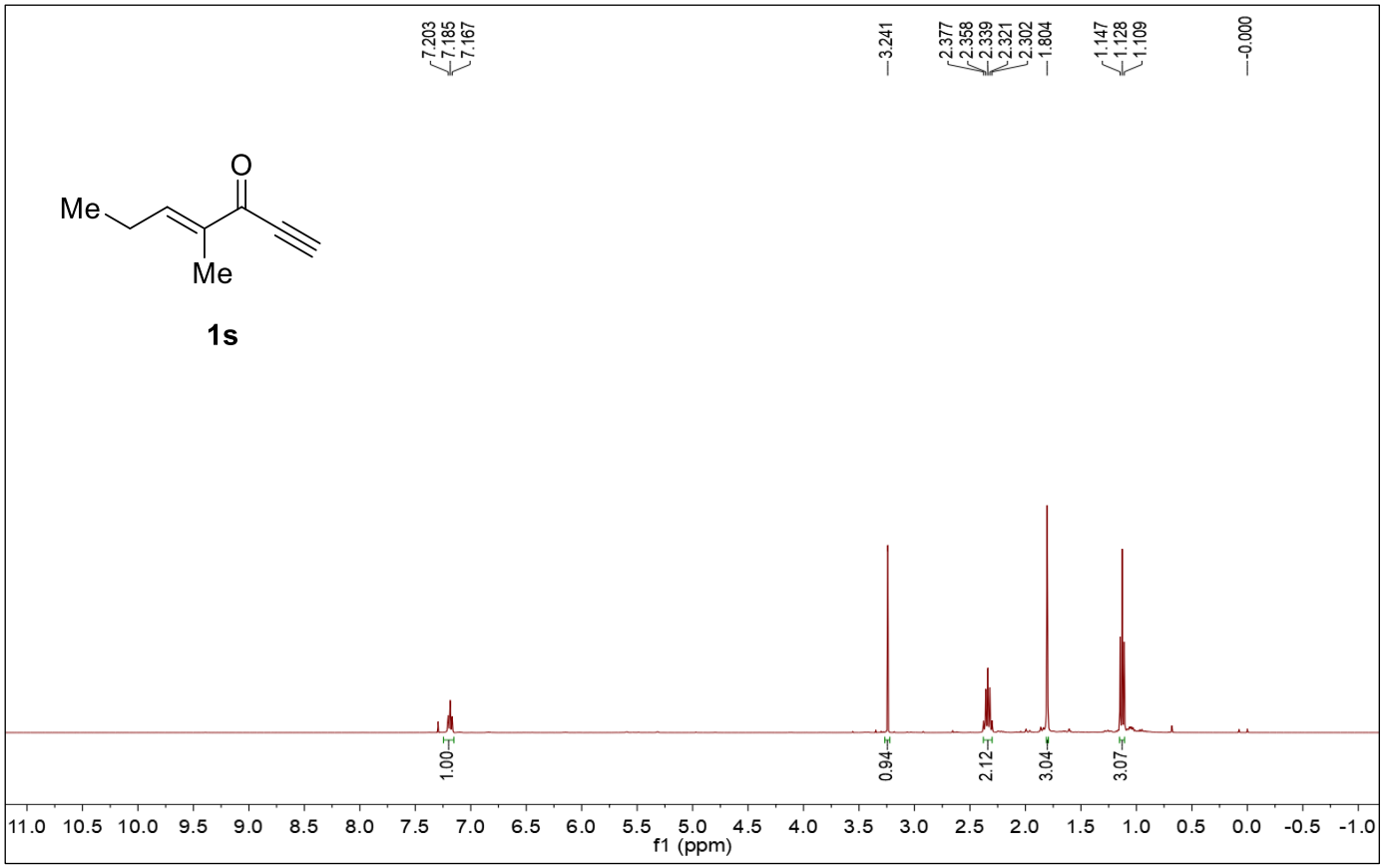

177.266
136.209
133.600
133.315
132.259
129.844
129.231
128.574
127.860
127.014
123.483
80.742
80.328
77.318
77.000
76.681
O
1p
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

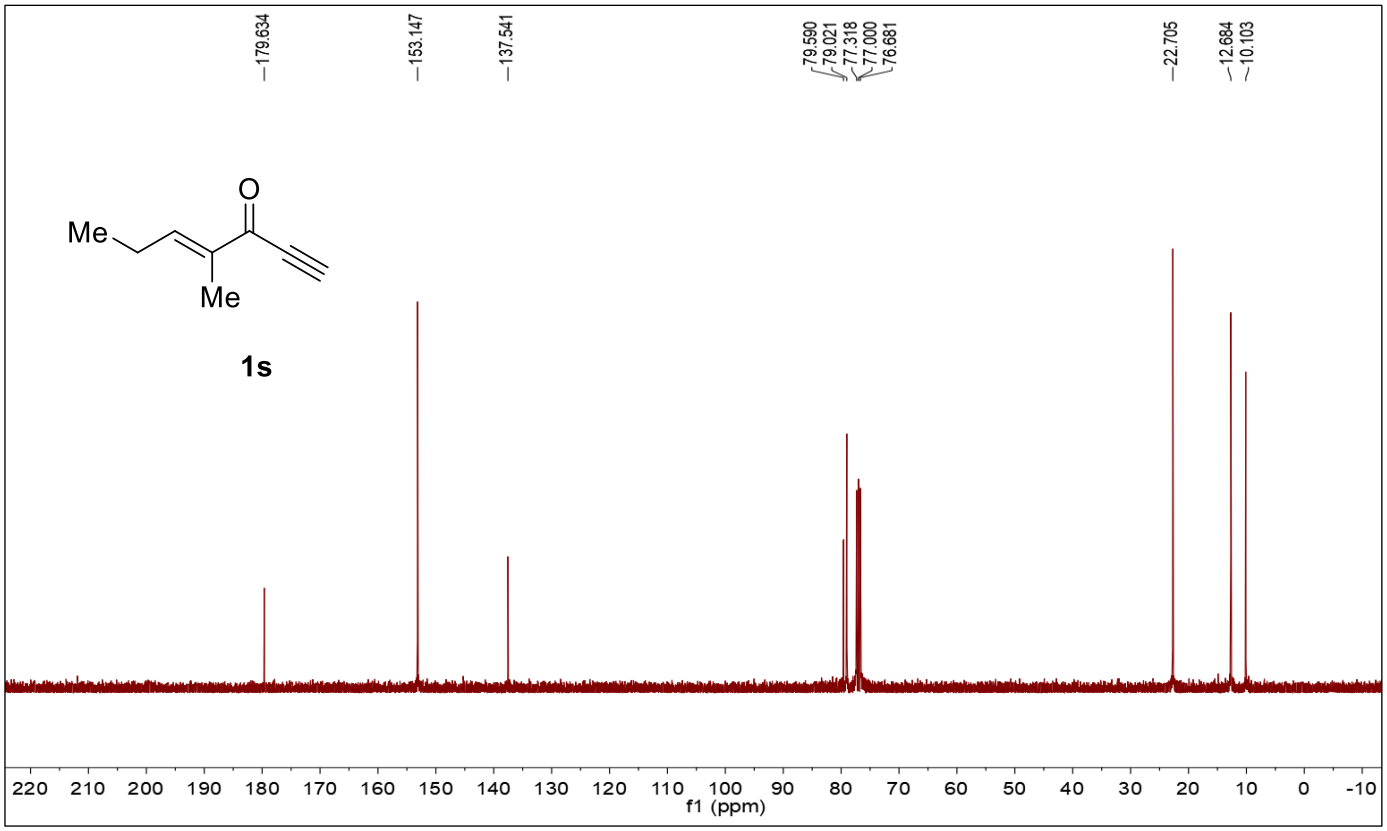

8.178
9.162
9.154
8.831
8.628
8.619
8.604
8.589
8.568
7.963
7.943
7.740
7.722
7.719
7.701
7.640
7.632
7.615
7.583
7.563
3.418
0.000
O
1q
1.05
1.04
2.09
1.09
1.10
3.12
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

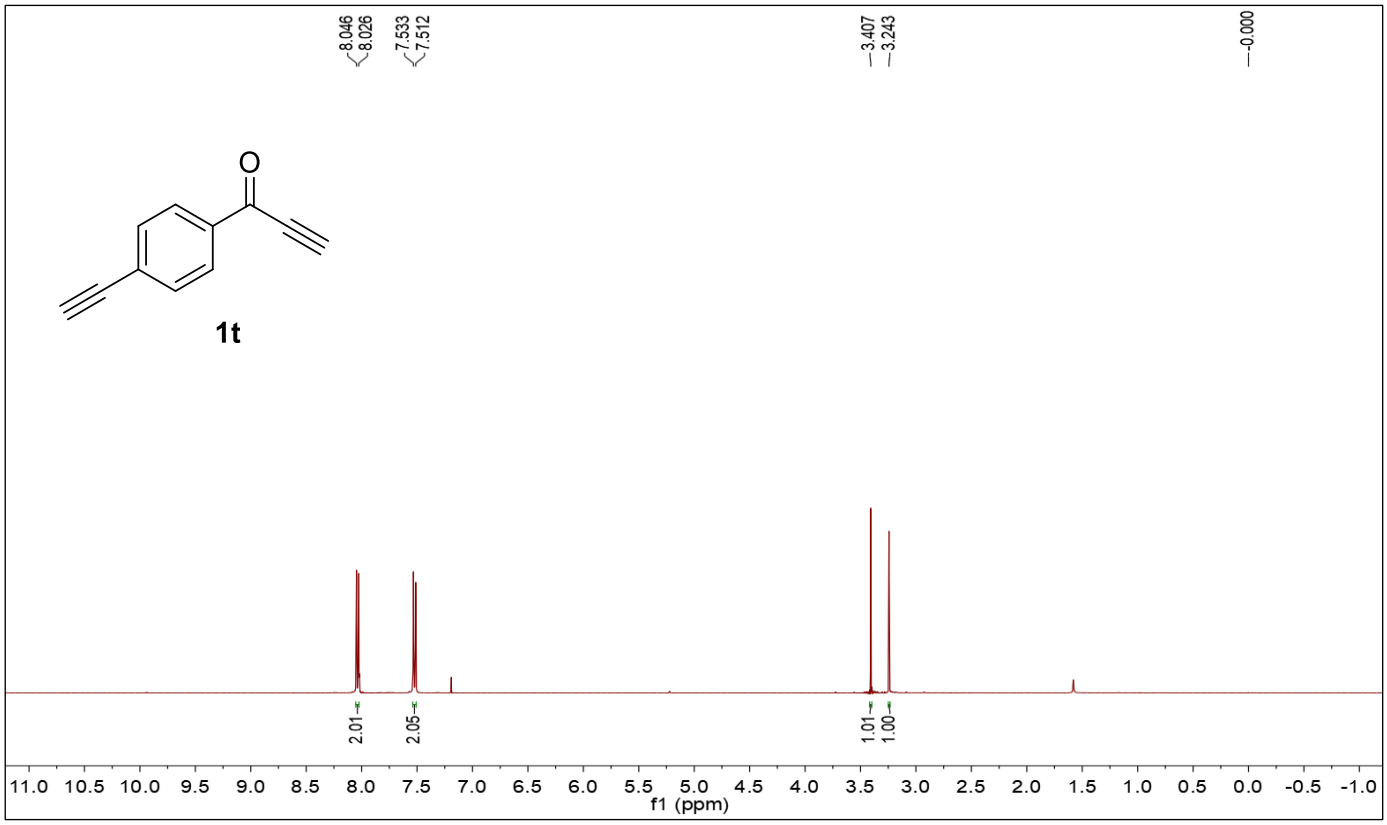

178.655
139.030
133.042
130.920
130.721
130.614
130.307
129.720
128.260
128.205
127.486
127.255
126.710
122.780
122.739
81.577
79.451
77.318
77.000
76.682
O
1q
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

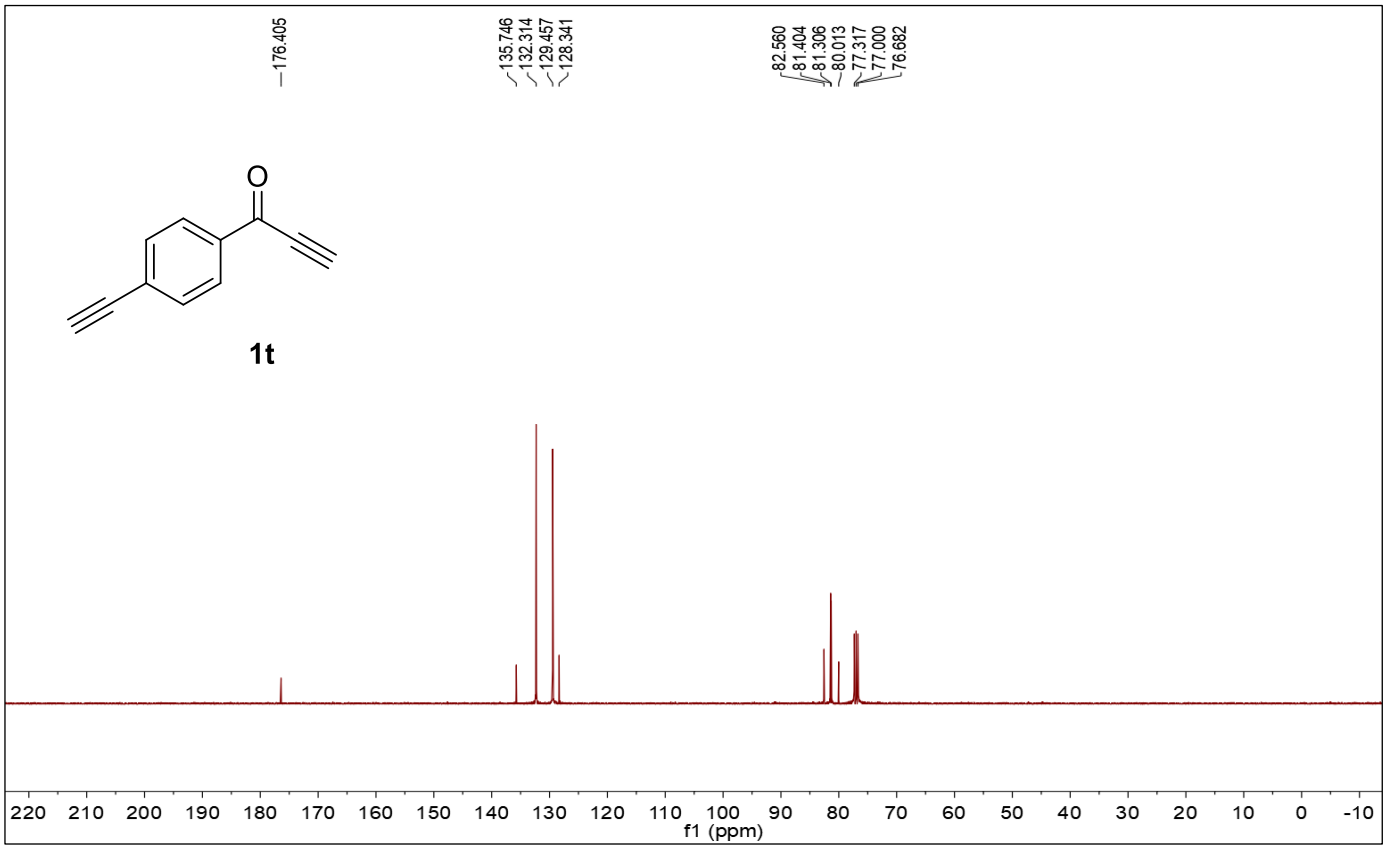

7.973
7.427
7.409
7.376
7.359
7.340
7.319
7.304
3.262
2.037
0.000
O
Me
1r
1.07
5.12
1.00
3.11
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

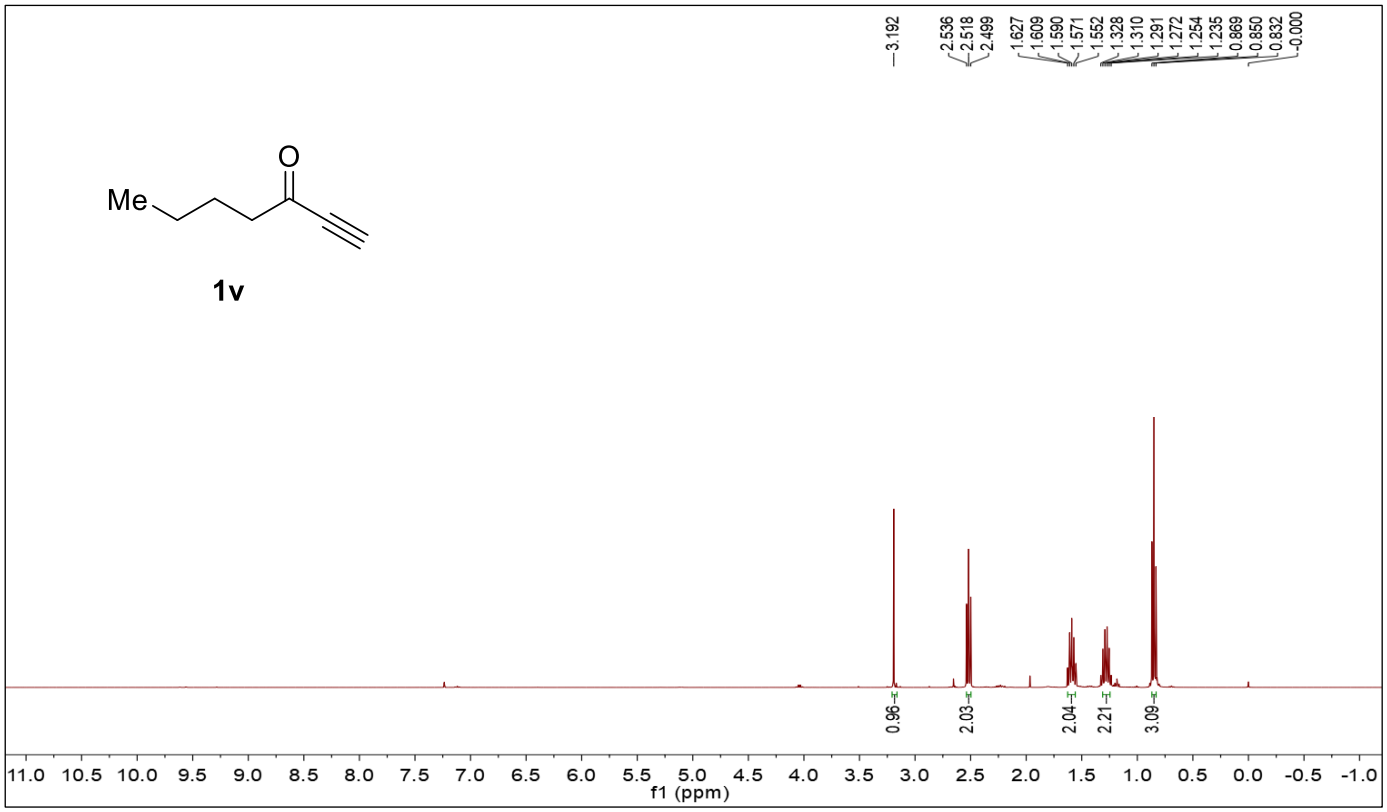

180.025
146.638
137.629
135.180
130.101
129.415
128.556
79.974
79.661
77.318
77.000
76.681
12.018
O
Me
1r
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

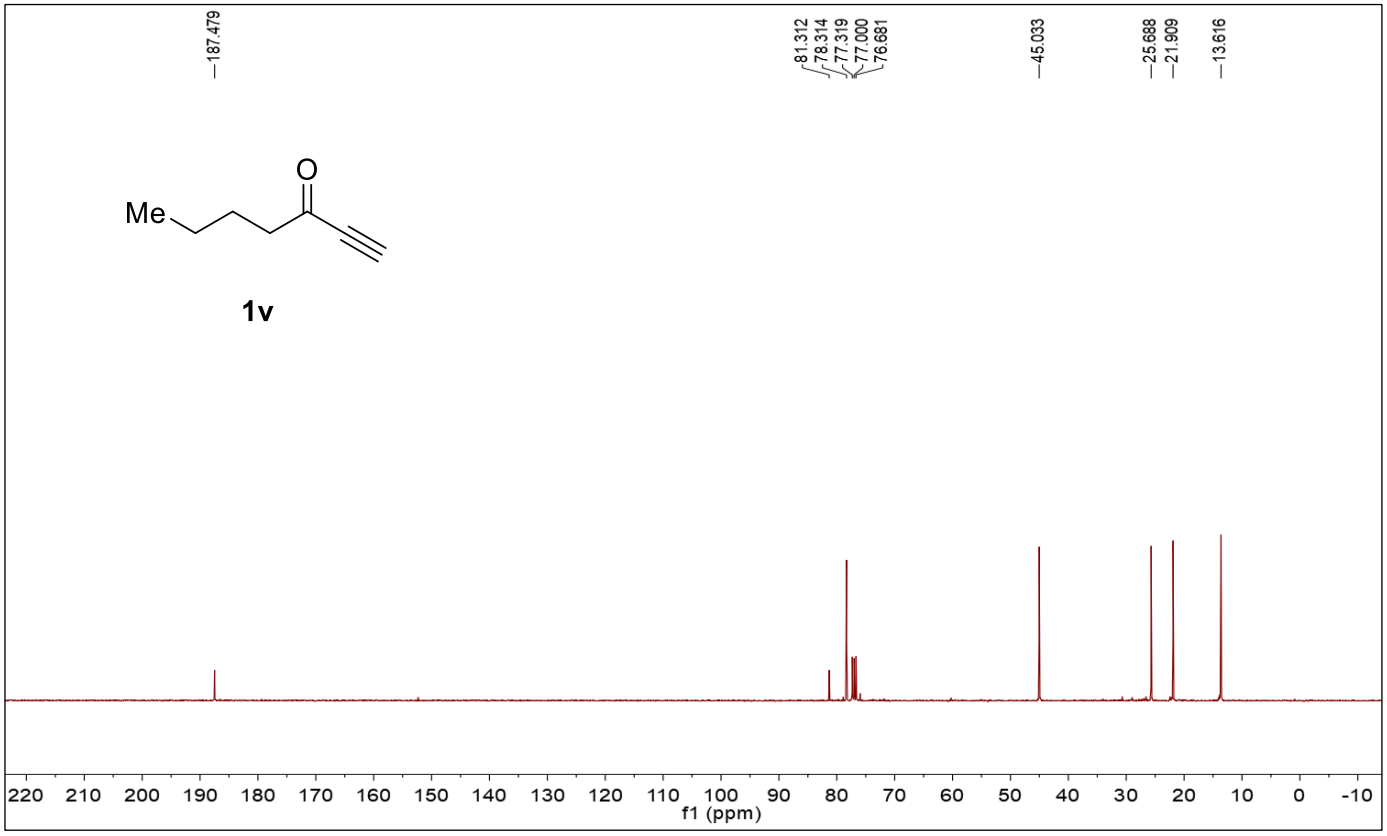

7.203
7.185
7.167
3.241
2.377
2.358
2.339
2.321
2.302
1.804
1.147
1.128
1.109
-0.000
O
Me
Me
1s
1.00
0.94
2.12
3.04
3.07
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

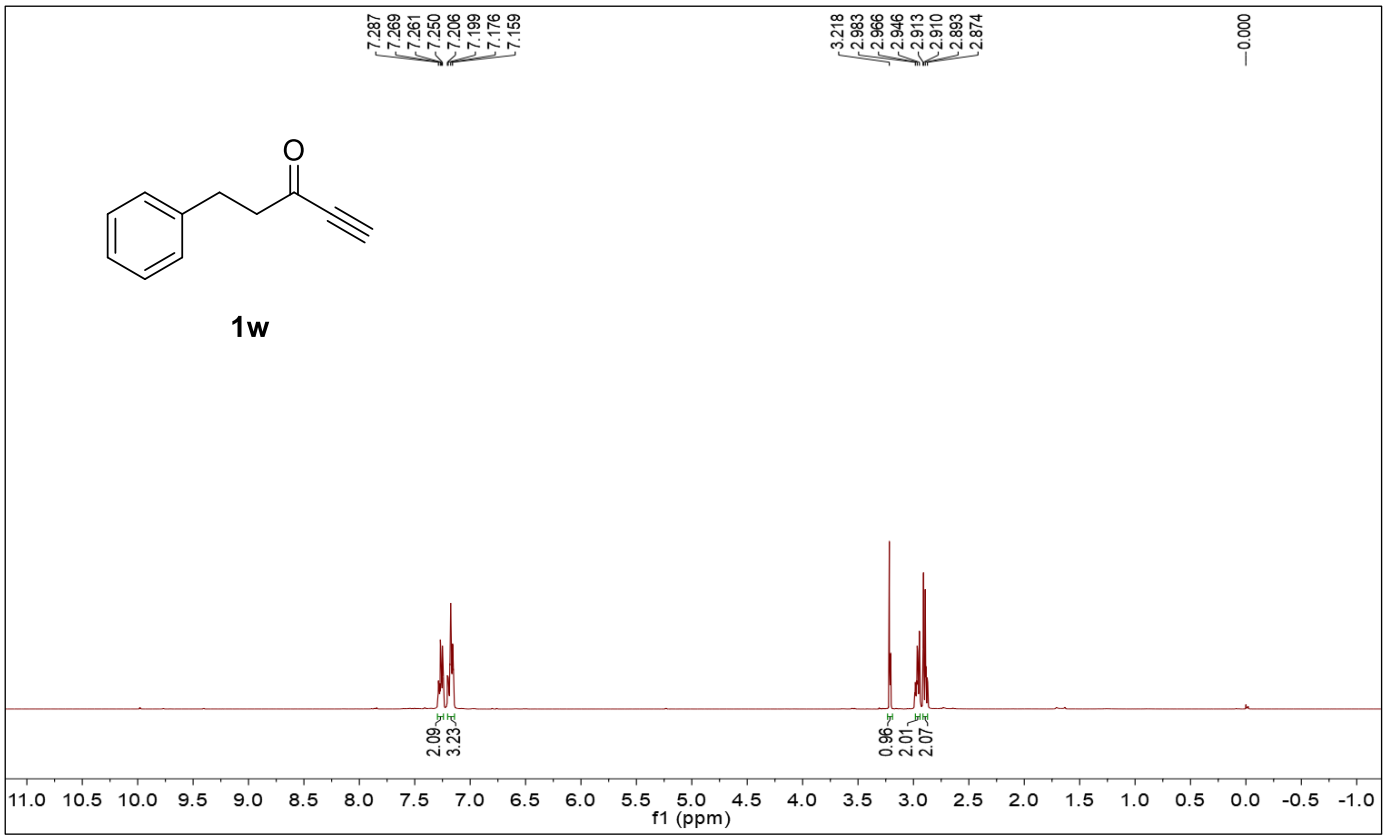

179.634
153.147
137.541
79.590
79.021
77.318
77.000
76.681
22.705
12.684
10.103
O
Me
Me
1s
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

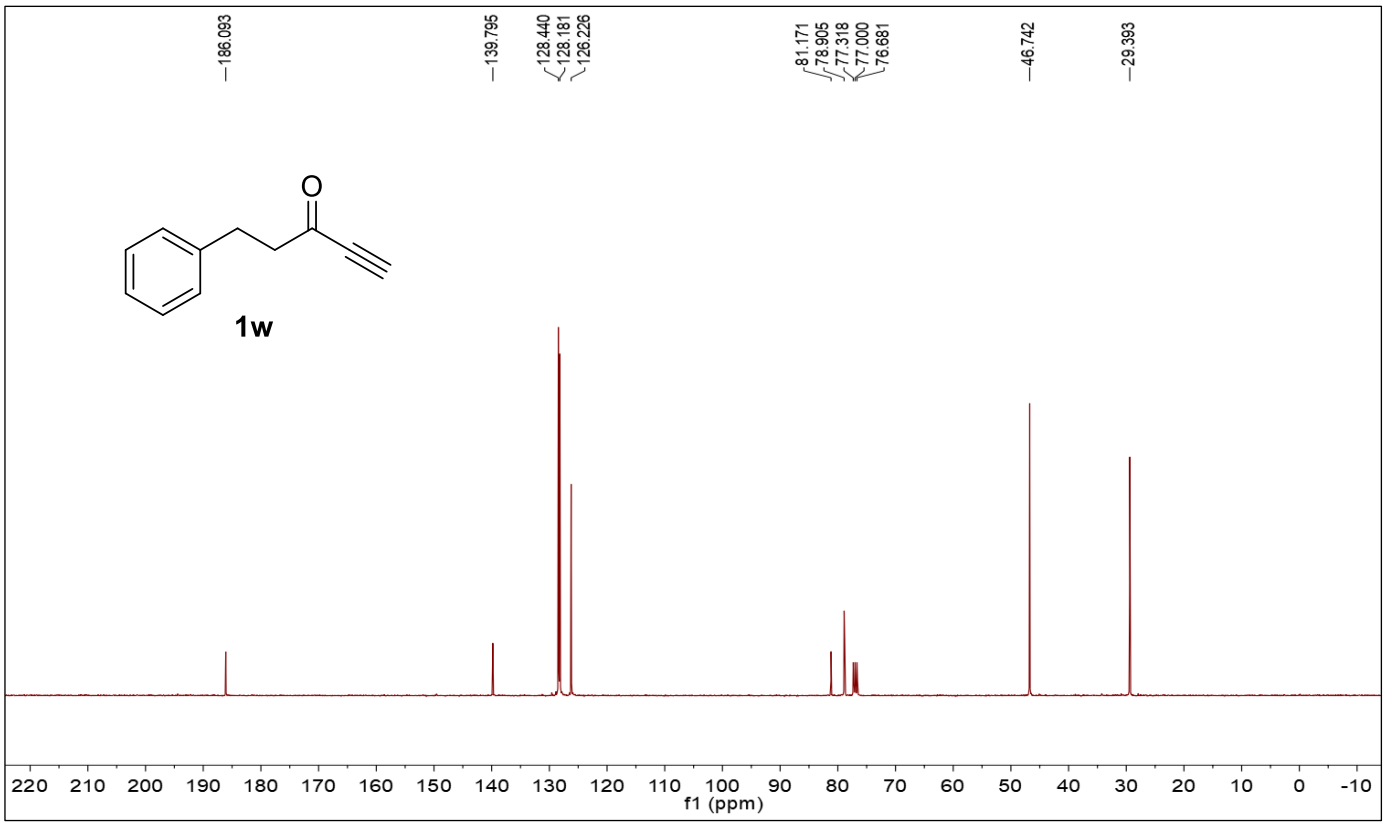

8.046
8.026
7.533
7.512
3.407
3.243
0.000
O
1t
2.01
2.05
1.01
1.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

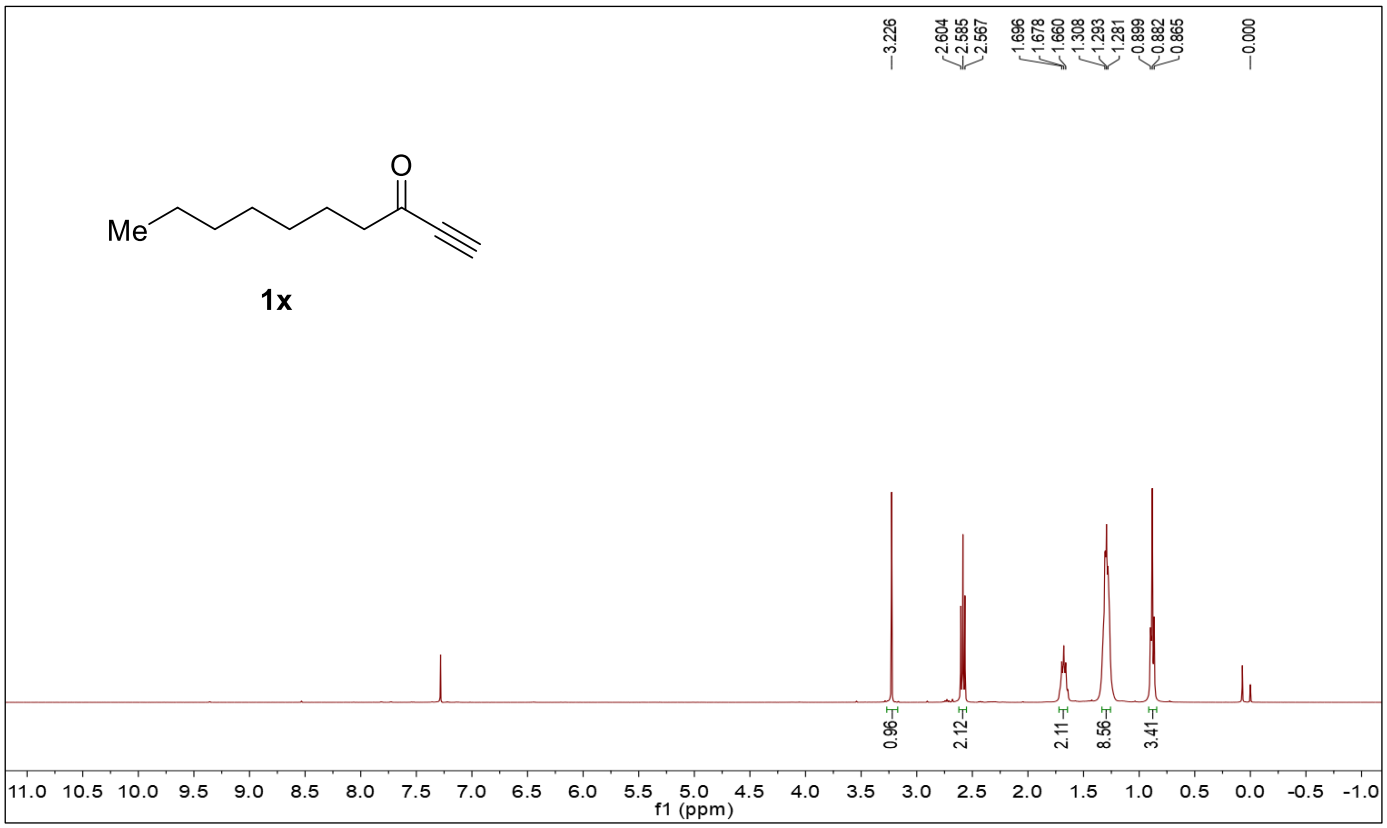

176.405
135.746
132.314
129.457
128.341
82.560
81.404
81.306
80.013
77.317
77.000
76.682
O
1t
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

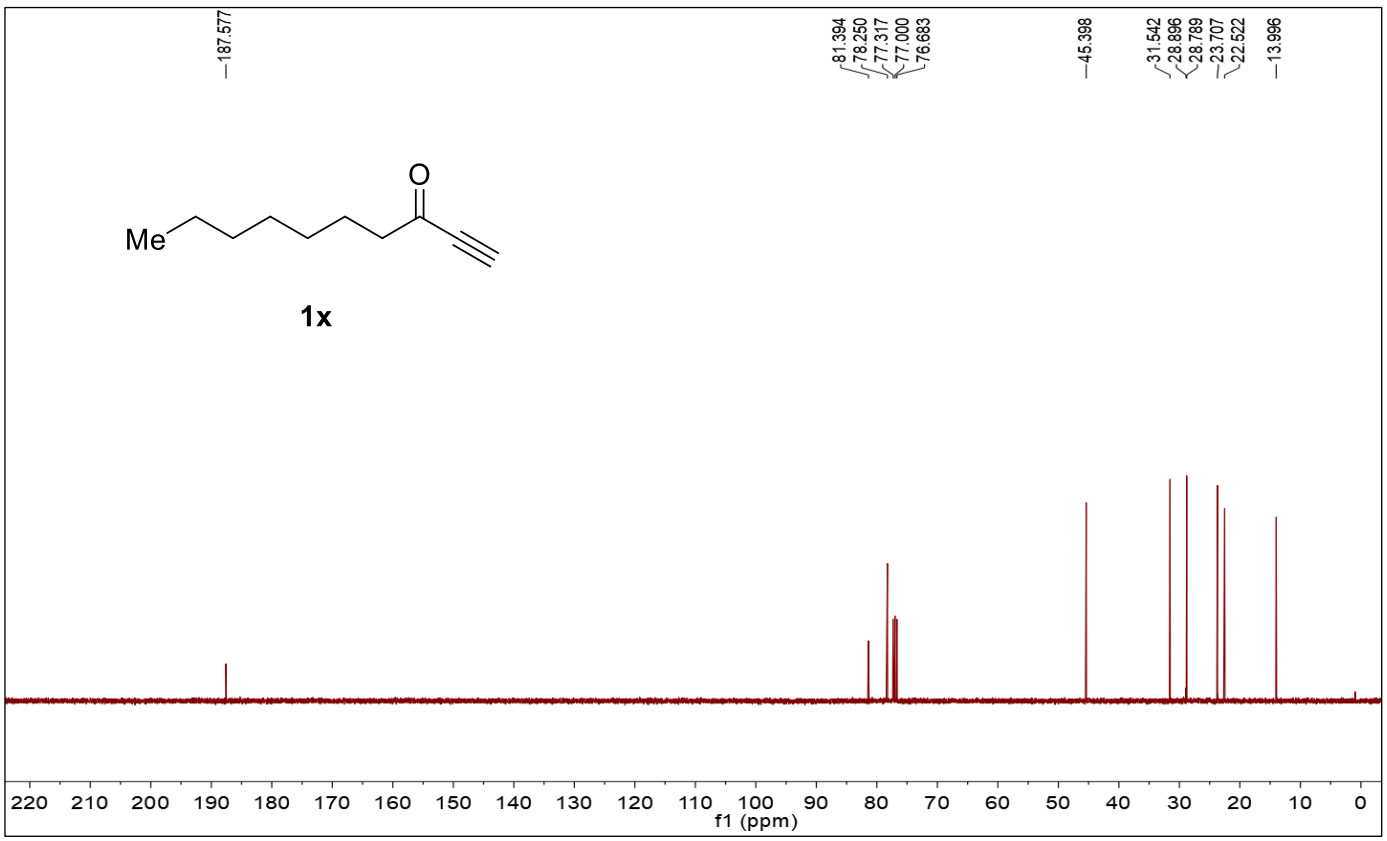

3.192
2.536
2.518
2.499
1.627
1.609
1.590
1.571
1.552
1.328
1.310
1.291
1.272
1.254
1.235
0.869
0.850
0.832
-0.000
O
Me
1v
0.96
2.03
2.04
2.21
3.09
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

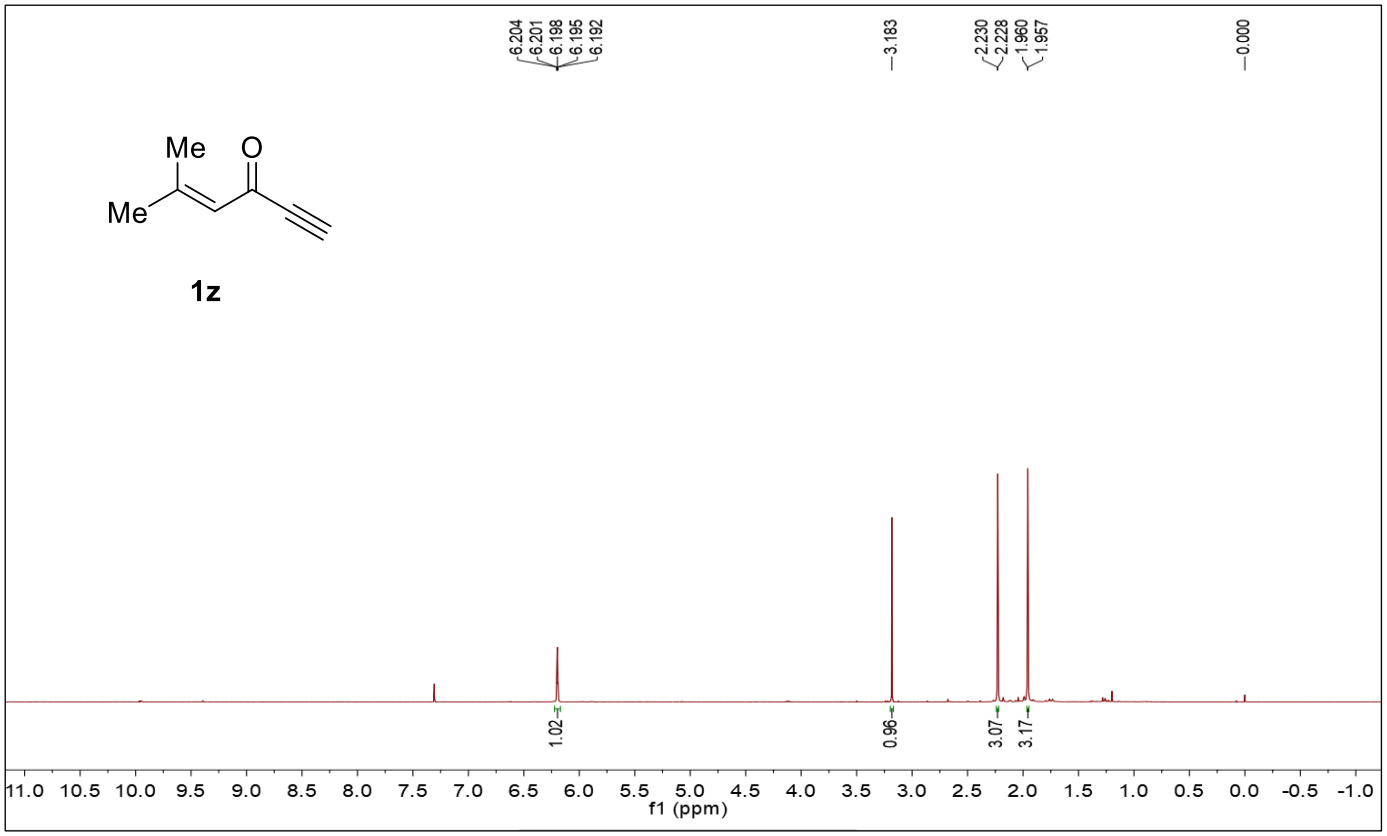

187.479
81.312
78.314
77.319
77.000
76.681
45.033
25.688
21.909
13.616
O
Me
1v
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

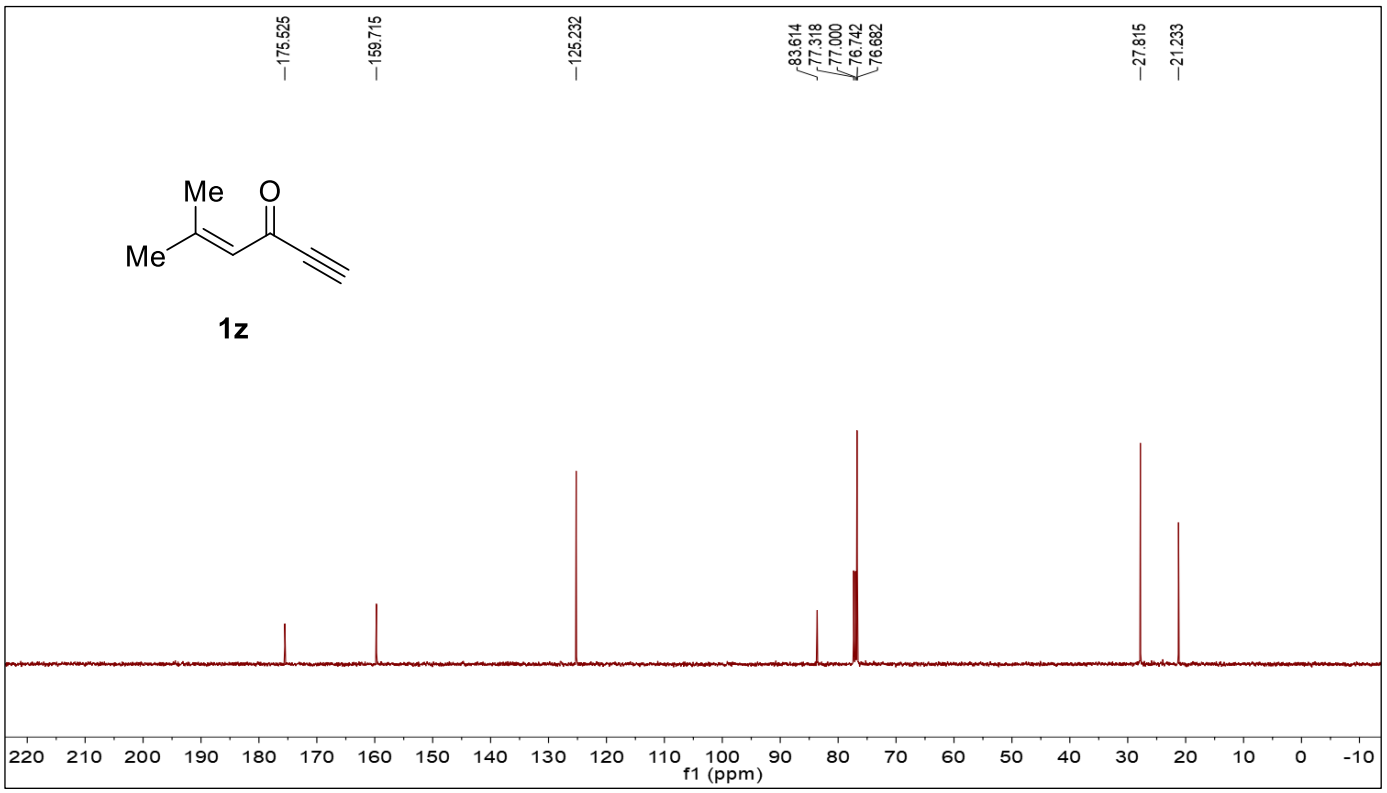

7.287
7.269
7.261
7.250
7.206
7.199
7.176
7.159
3.218
2.983
2.966
2.946
2.913
2.910
2.893
2.874
0.000
O
1w
2.09
3.23
0.96
2.01
2.07
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


186.093
139.795
128.440
128.181
126.226
81.171
78.905
77.318
77.000
76.681
46.742
29.393
O
1w
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

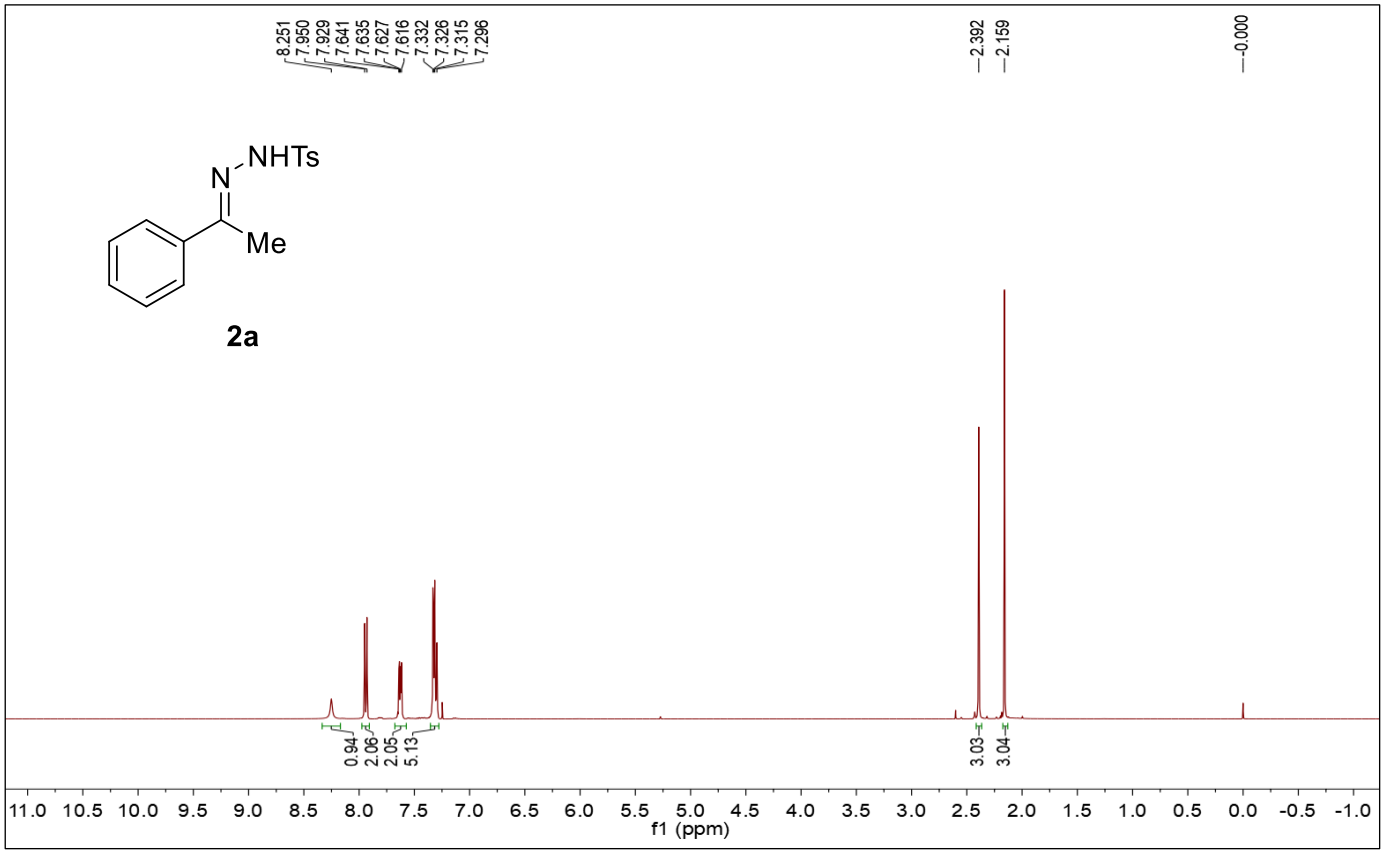

3.226
2.604
2.585
2.567
1.696
1.678
1.660
1.308
1.293
1.281
0.899
0.882
0.865
0.000
O
Me
1x
0.96
2.12
2.11
8.56
3.41
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

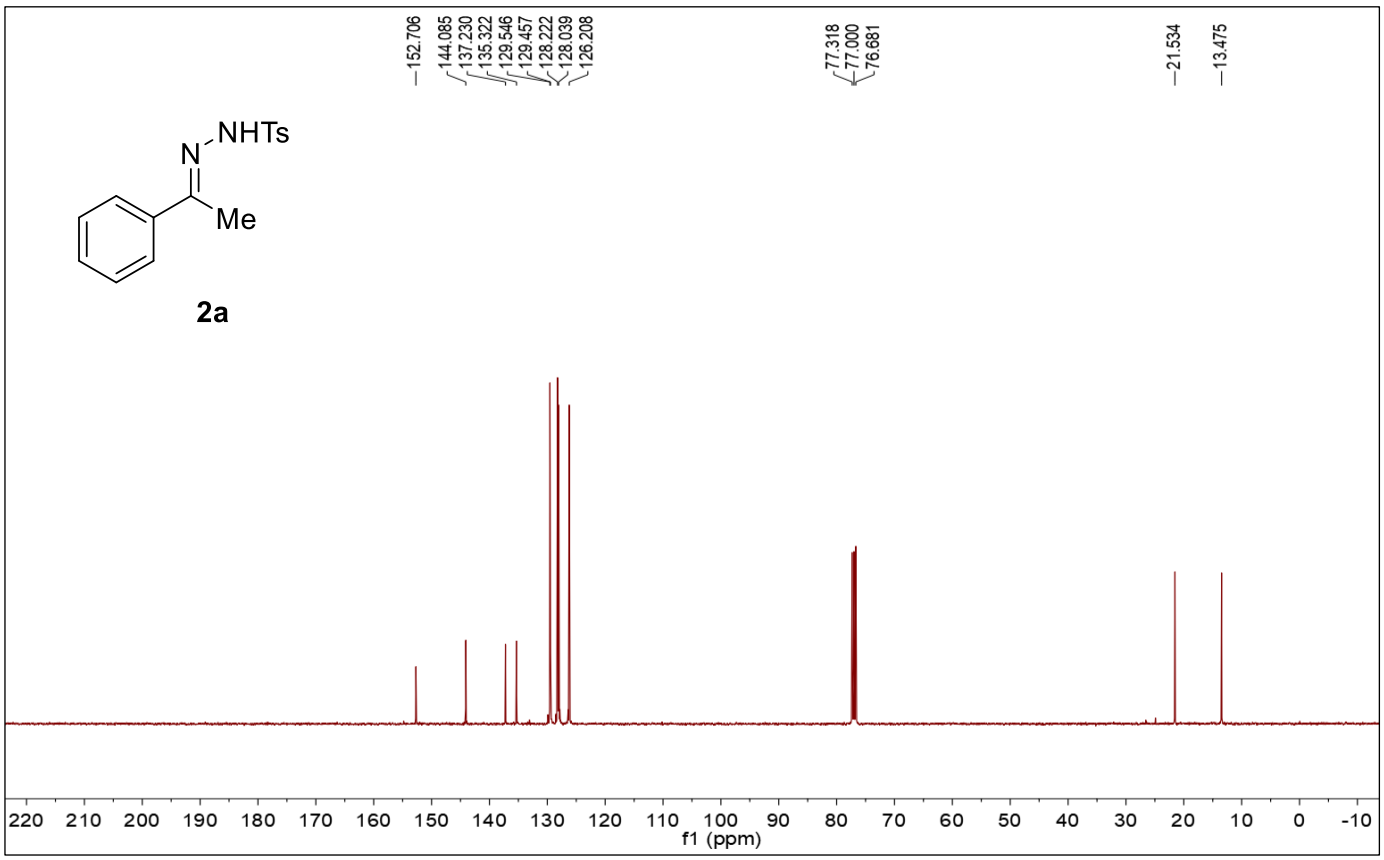

187.577
81.394
78.250
77.317
77.000
76.683
45.398
31.542
28.896
28.789
23.707
22.522
13.996
O
Me
1x
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

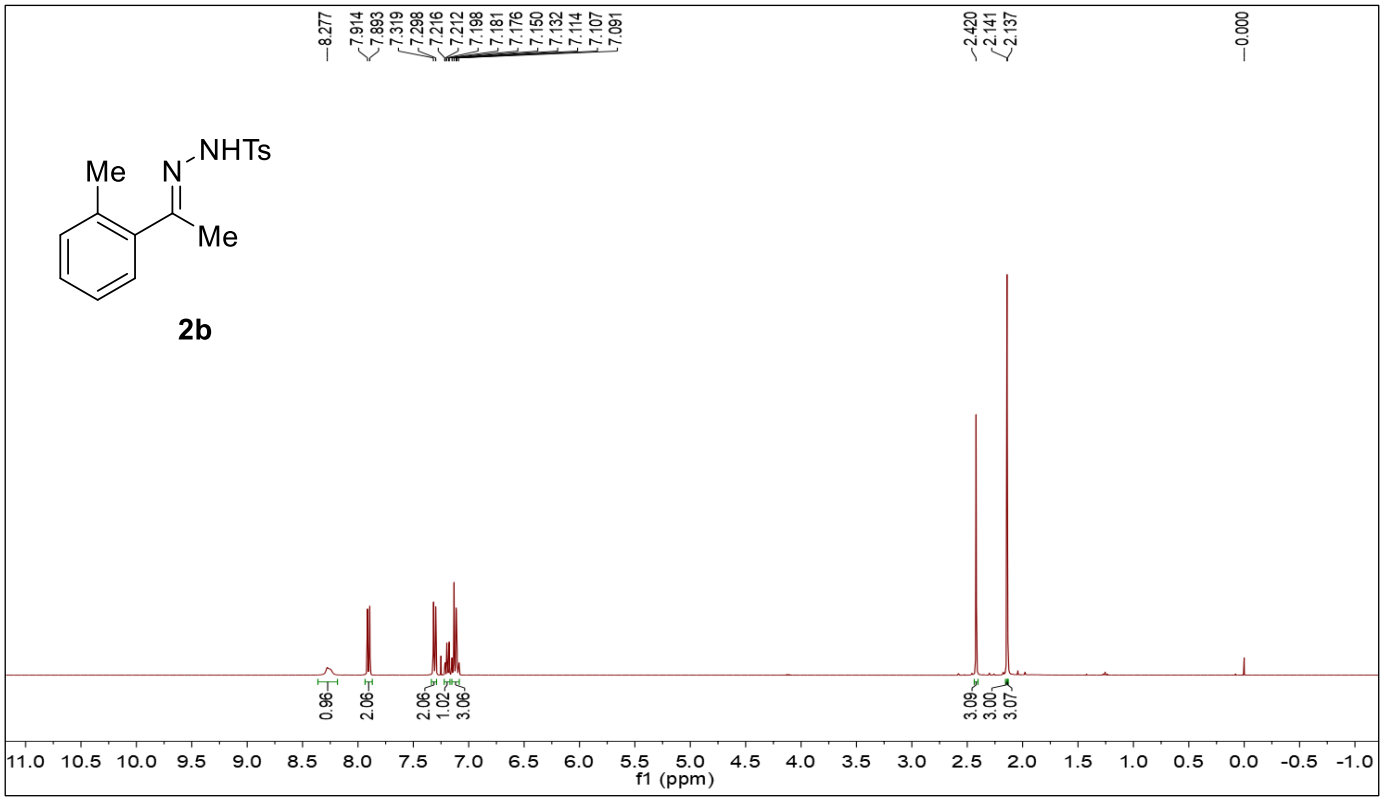

6.204
6.201
6.198
6.195
6.192
3.183
2.230
2.228
1.960
1.957
0.000
Me
O
Me
1z
1.02
0.96
3.07
3.17
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


175.525
159.715
125.232
83.614
77.318
77.000
76.742
76.682
27.815
21.233
Me
O
Me
1z
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

^{1}H and ^{13}C NMR spectra of *N*-tosylhydrazones



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7.216
7.212
7.198
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7.176
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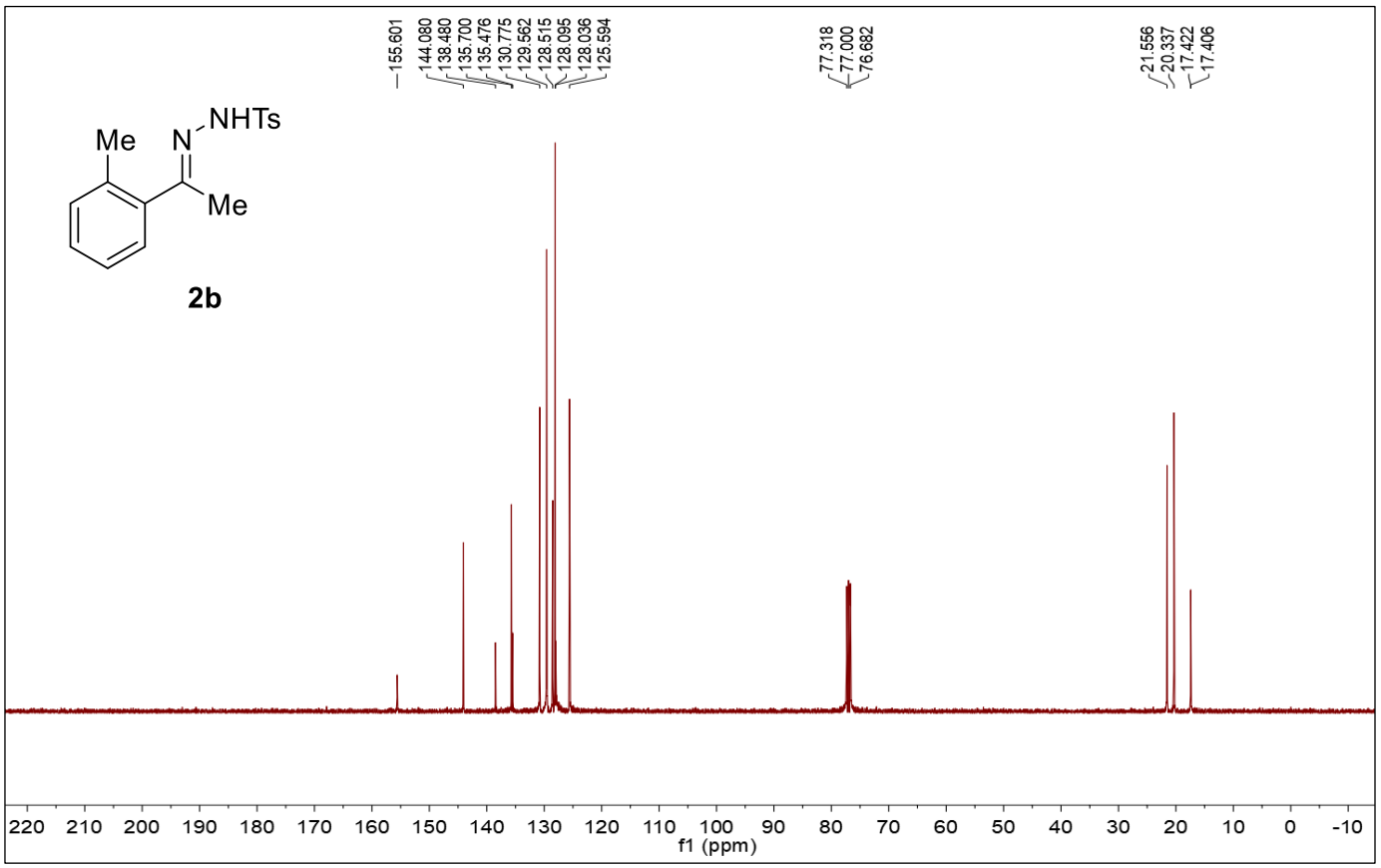
7.132
7.114
7.107
7.091
2.420
2.141
2.137
0.000
Me
N
NHTs
Me
2b
0.96
2.06
2.06
1.02
3.06
3.09
3.00
3.07
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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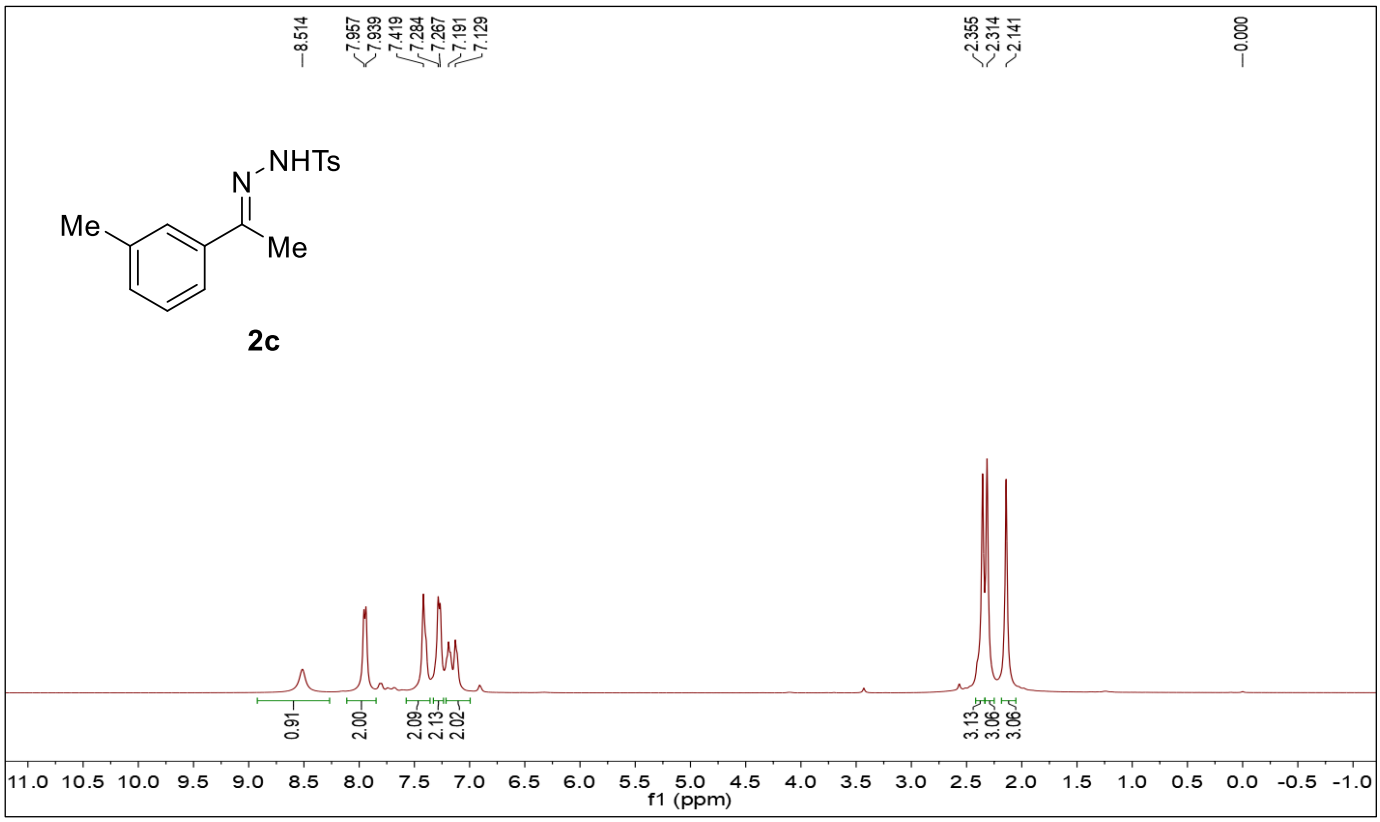
138.480
135.700
135.476
130.775
129.562
128.515
128.095
128.036
125.594
77.318
77.000
76.682
21.556
20.337
17.422
17.406
Me
N
NHTs
Me
2b
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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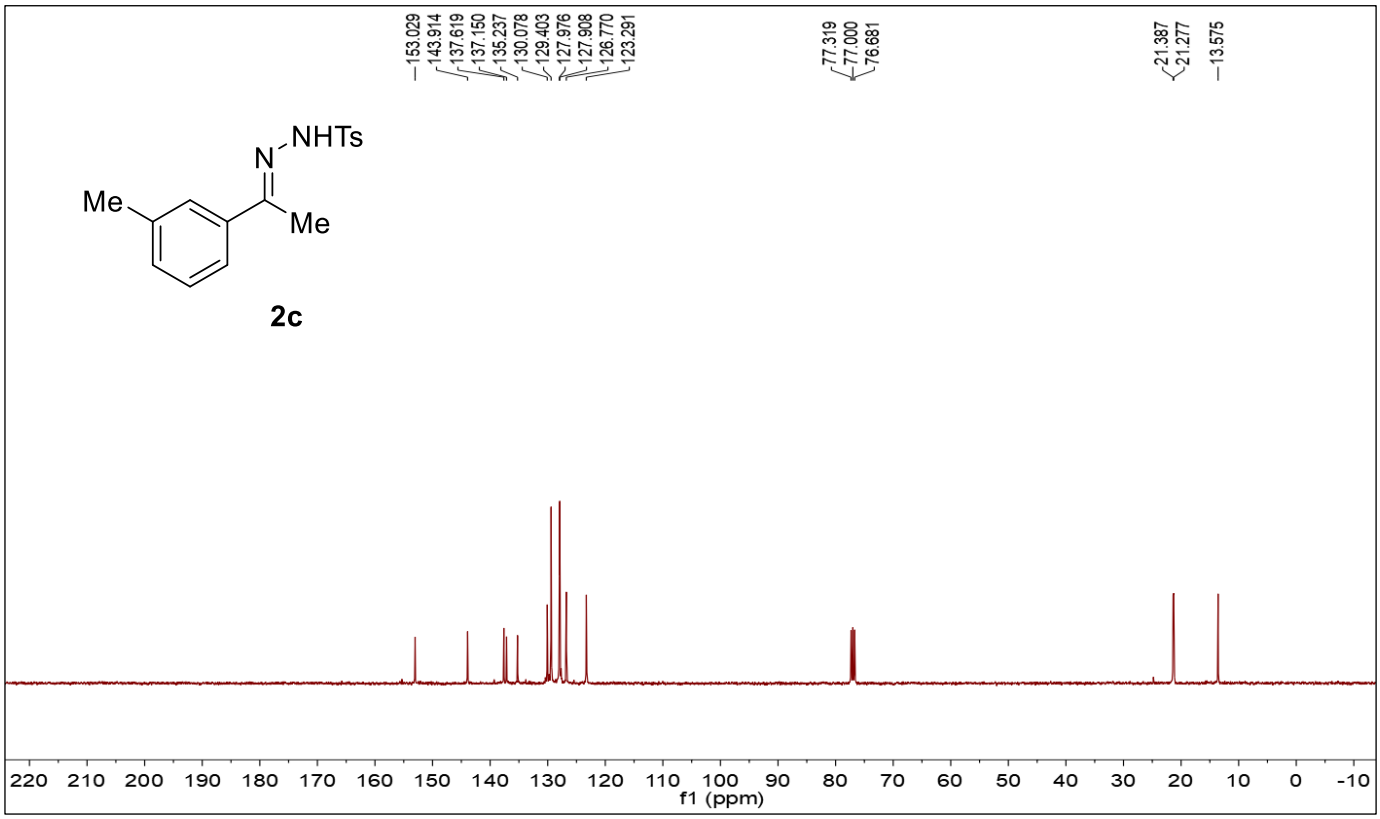
7.957
7.939
7.419
7.284
7.267
7.191
7.129
2.355
2.314
2.141
0.000
NHTs
N
Me
Me
2c
0.91
2.00
2.09
2.13
2.02
3.13
3.06
3.06
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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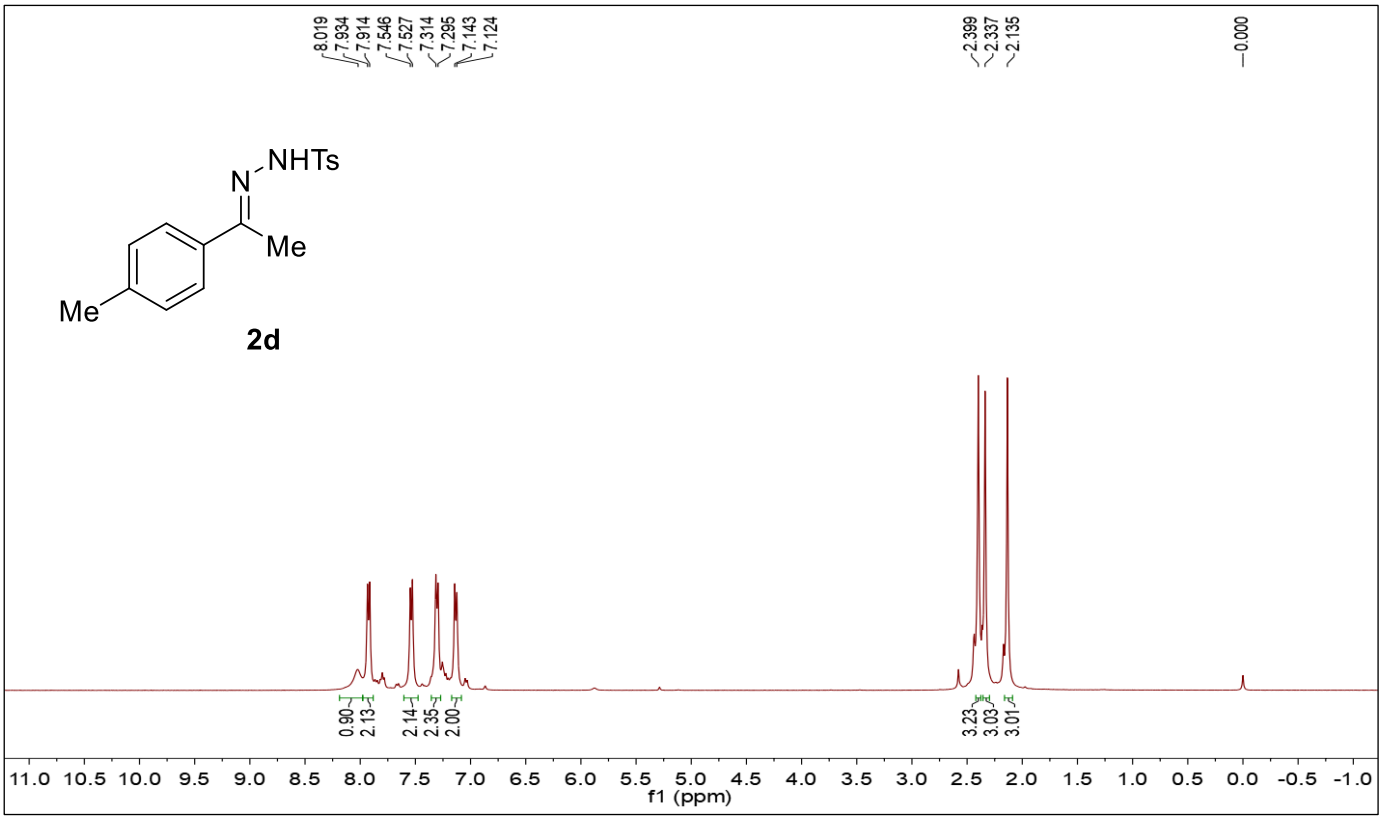
137.619
137.150
135.237
130.078
129.403
127.976
127.908
126.770
123.291
77.319
77.000
76.681
21.387
21.277
13.575
NHTs
N
Me
Me
2c
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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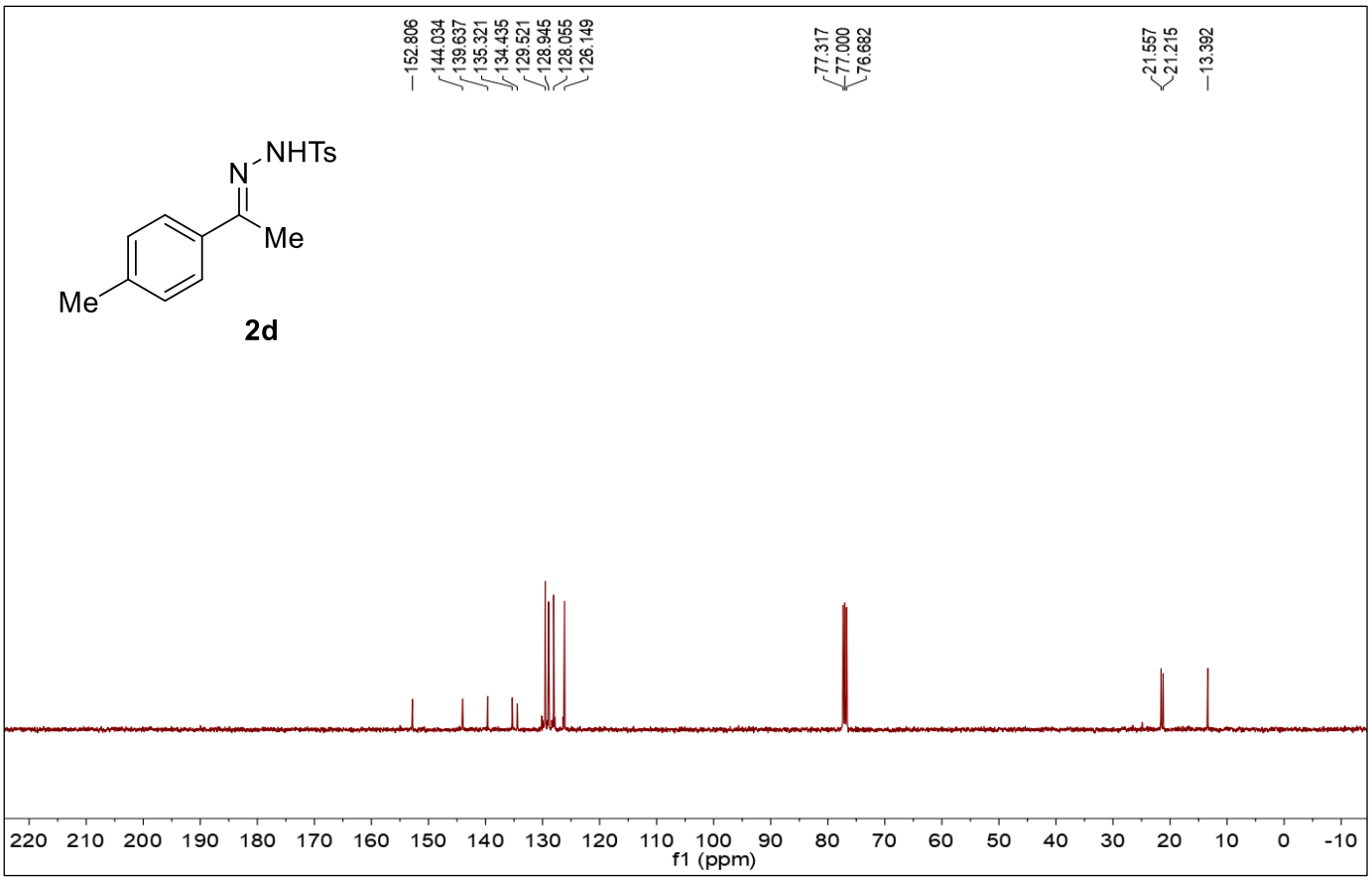
7.934
7.914
7.546
7.527
7.314
7.295
7.143
7.124
2.399
2.337
2.135
0.000
NHTs
N
Me
Me
2d
0.90
2.13
2.14
2.35
2.00
3.23
3.03
3.01
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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144.034
139.637
135.321
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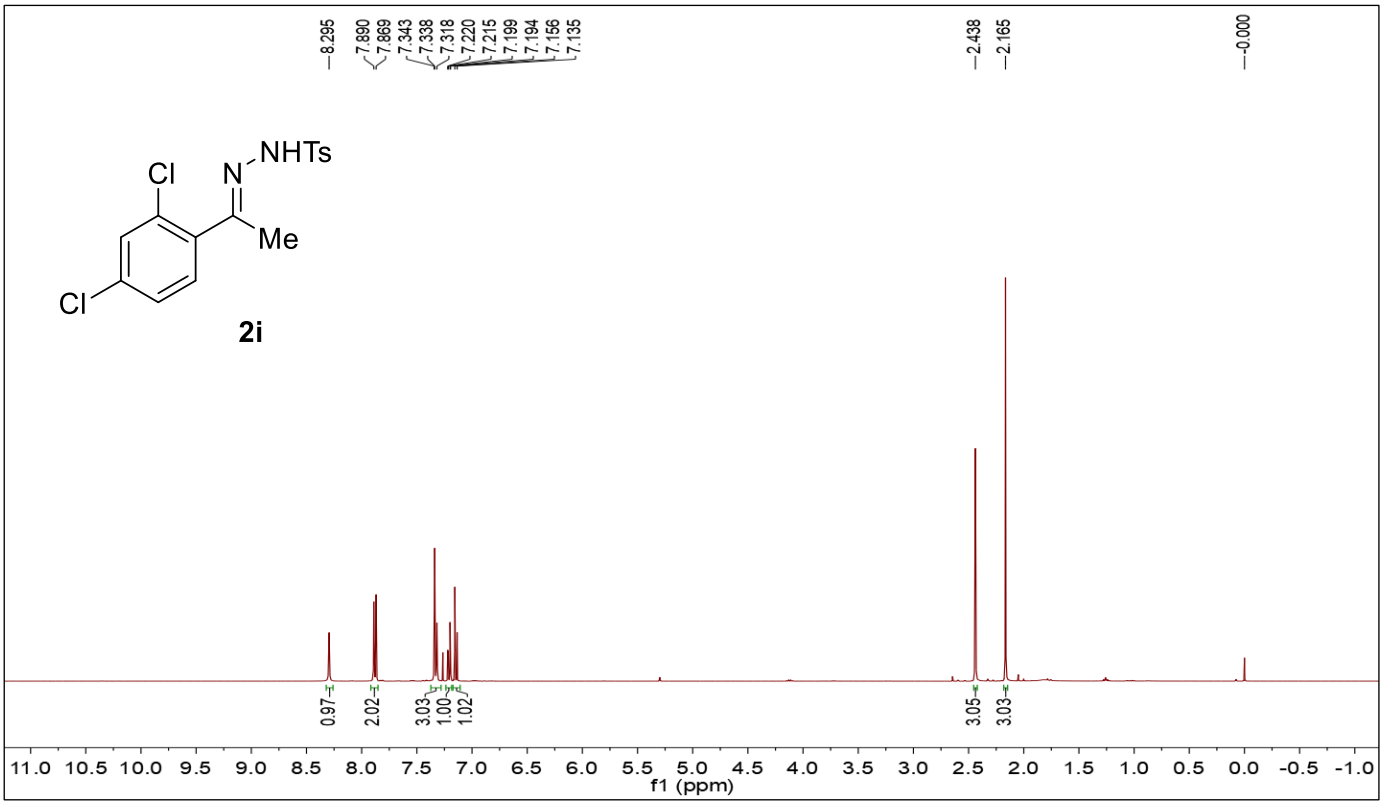
129.521
128.945
128.055
126.149
77.317
77.000
76.682
21.557
21.215
13.392
NHTs
N
Me
Me
2d
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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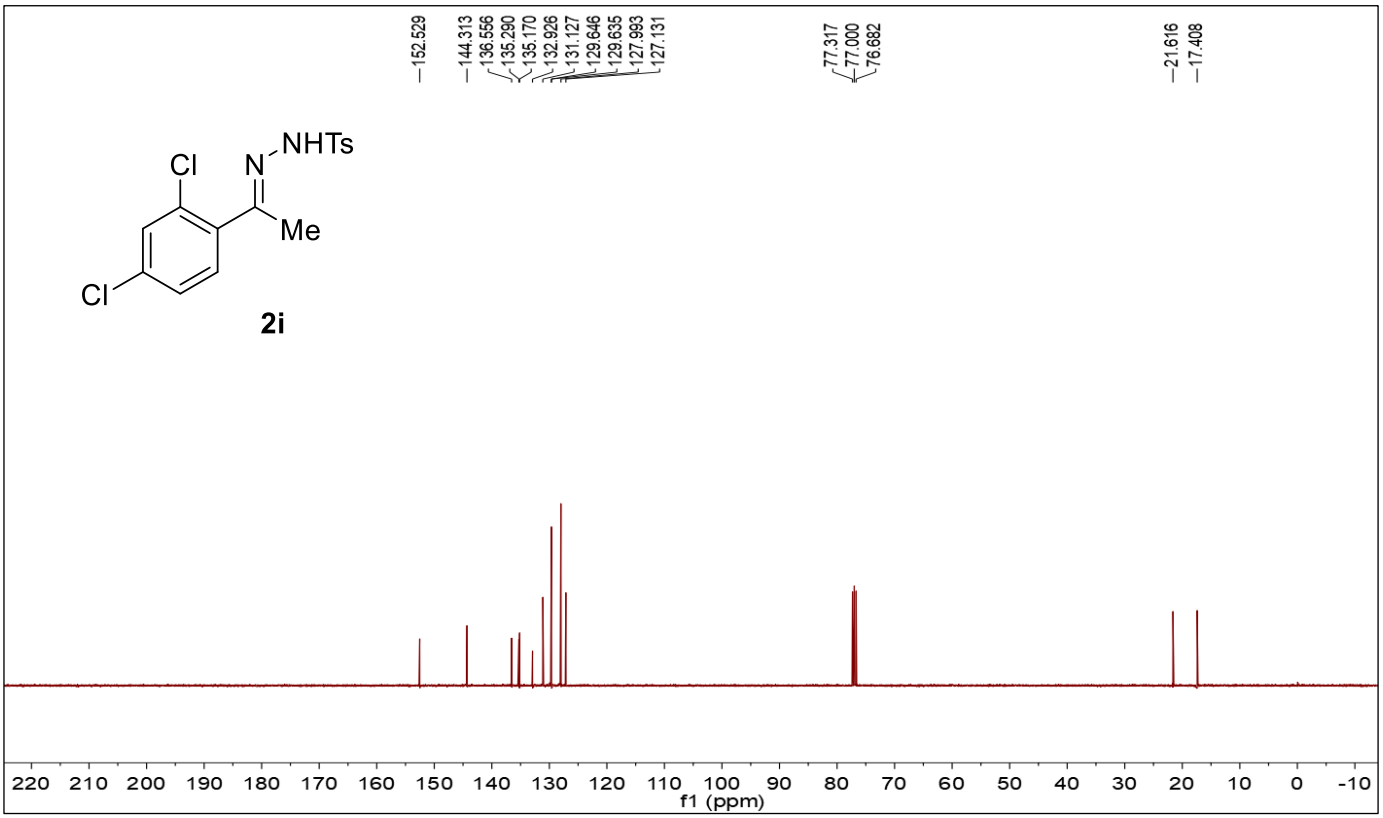
7.890
7.869
7.343
7.338
7.318
7.220
7.215
7.199
7.194
7.156
7.135
2.438
2.165
0.000
Cl
N
NHTs
Me
Cl
2i
0.97
2.02
3.03
1.00
1.02
3.05
3.03
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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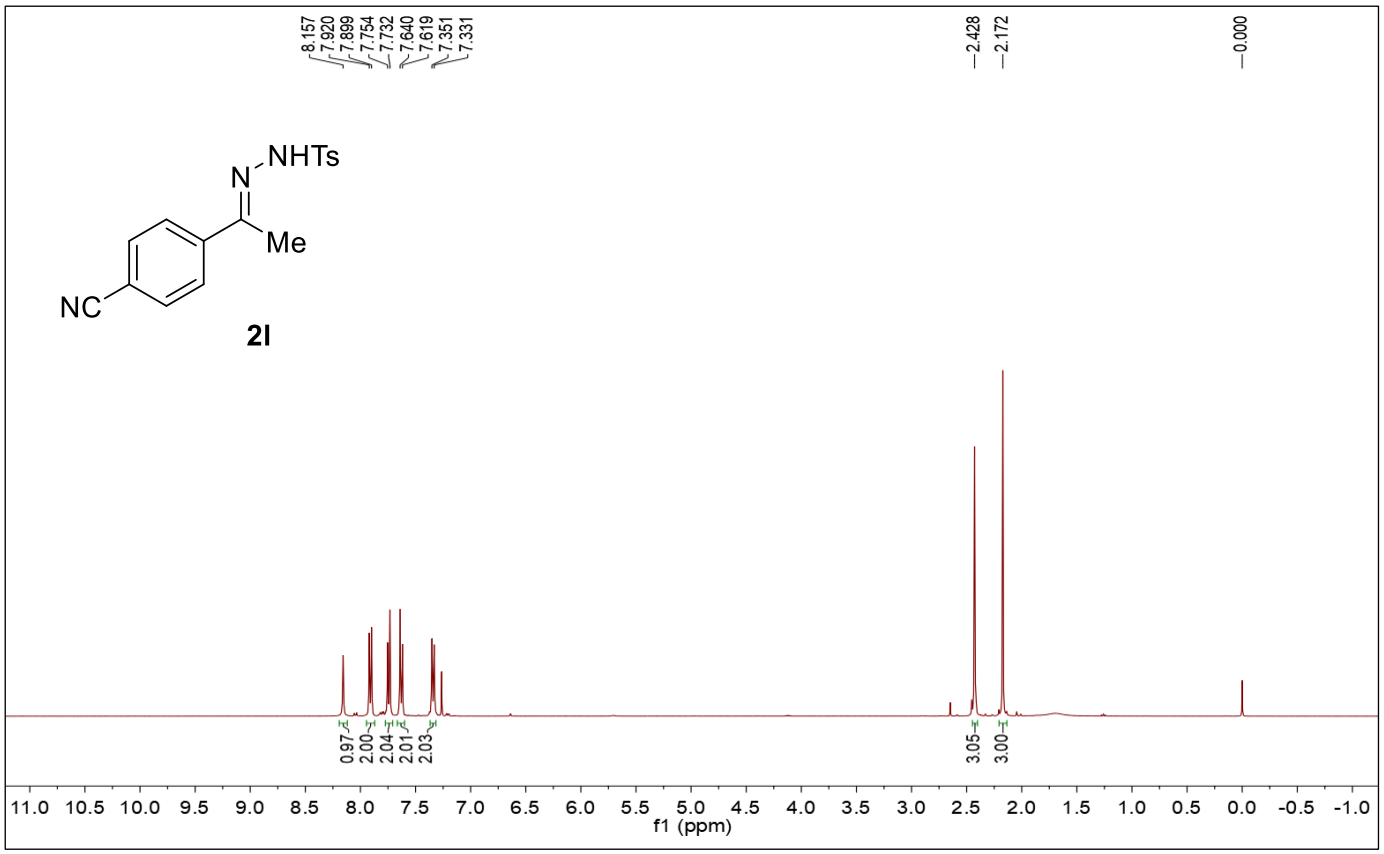
144.313
136.556
135.290
135.170
132.926
131.127
129.646
129.635
127.993
127.131
77.317
77.000
76.682
21.616
17.408
Cl
N
NHTs
Me
Cl
2i
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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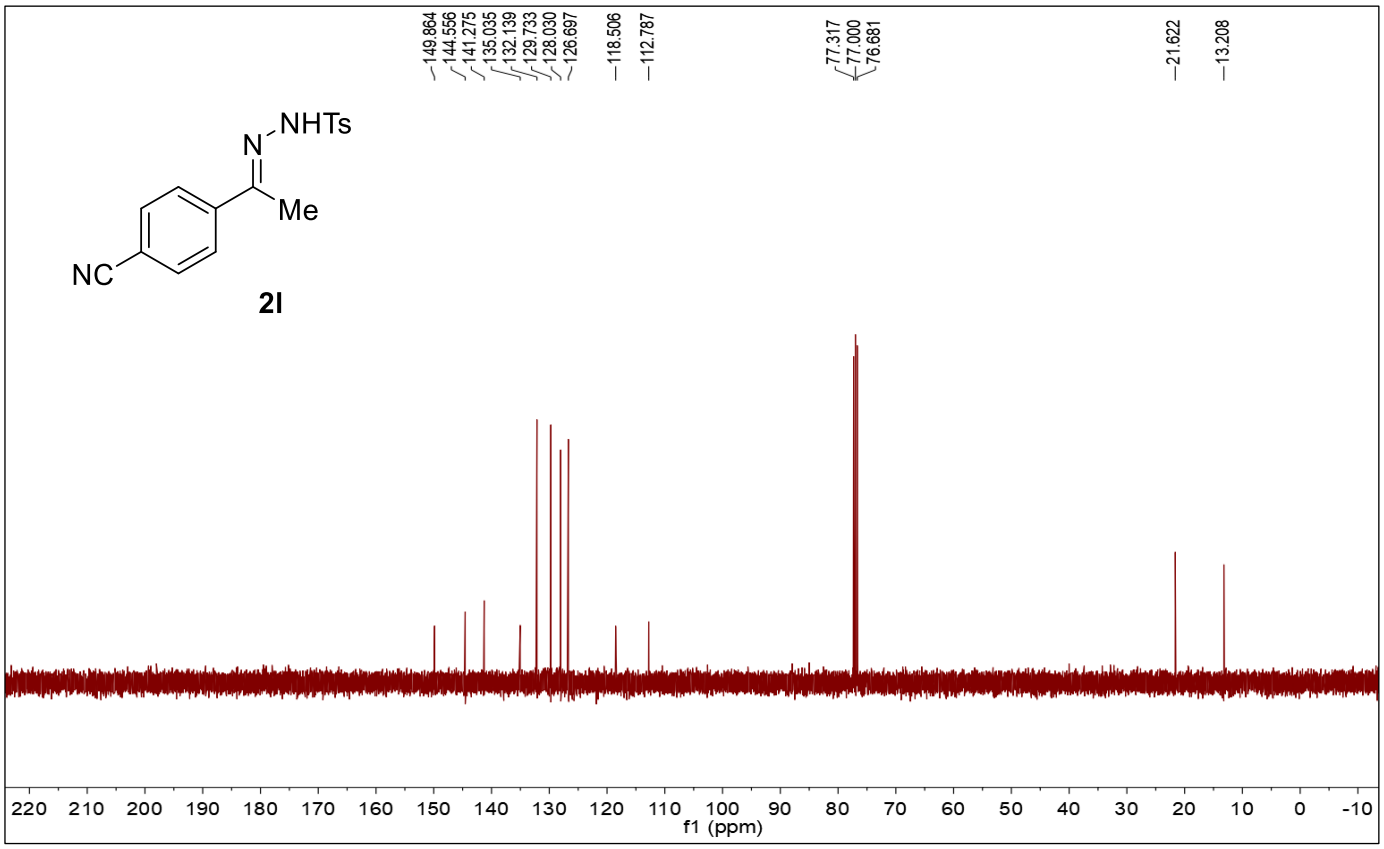
7.899
7.754
7.732
7.640
7.619
7.351
7.331
2.428
2.172
0.000
NHTs
N
Me
NC
2I
0.97
2.00
2.04
2.01
2.03
3.05
3.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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144.556
141.275
135.035
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129.733
128.030
126.697
118.506
112.787
77.317
77.000
76.681
21.622
13.208
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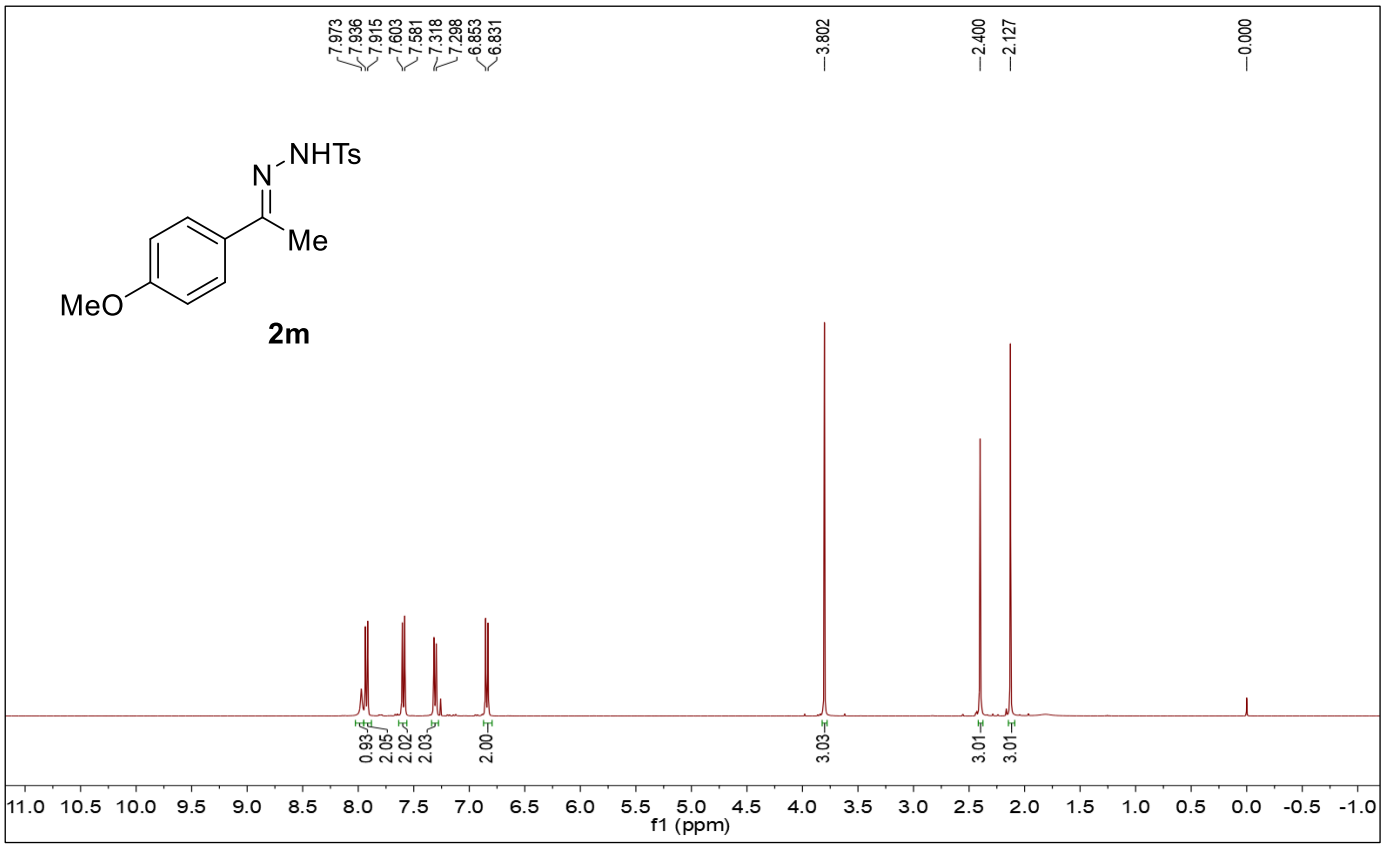
N
Me
NC
2I
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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7.603
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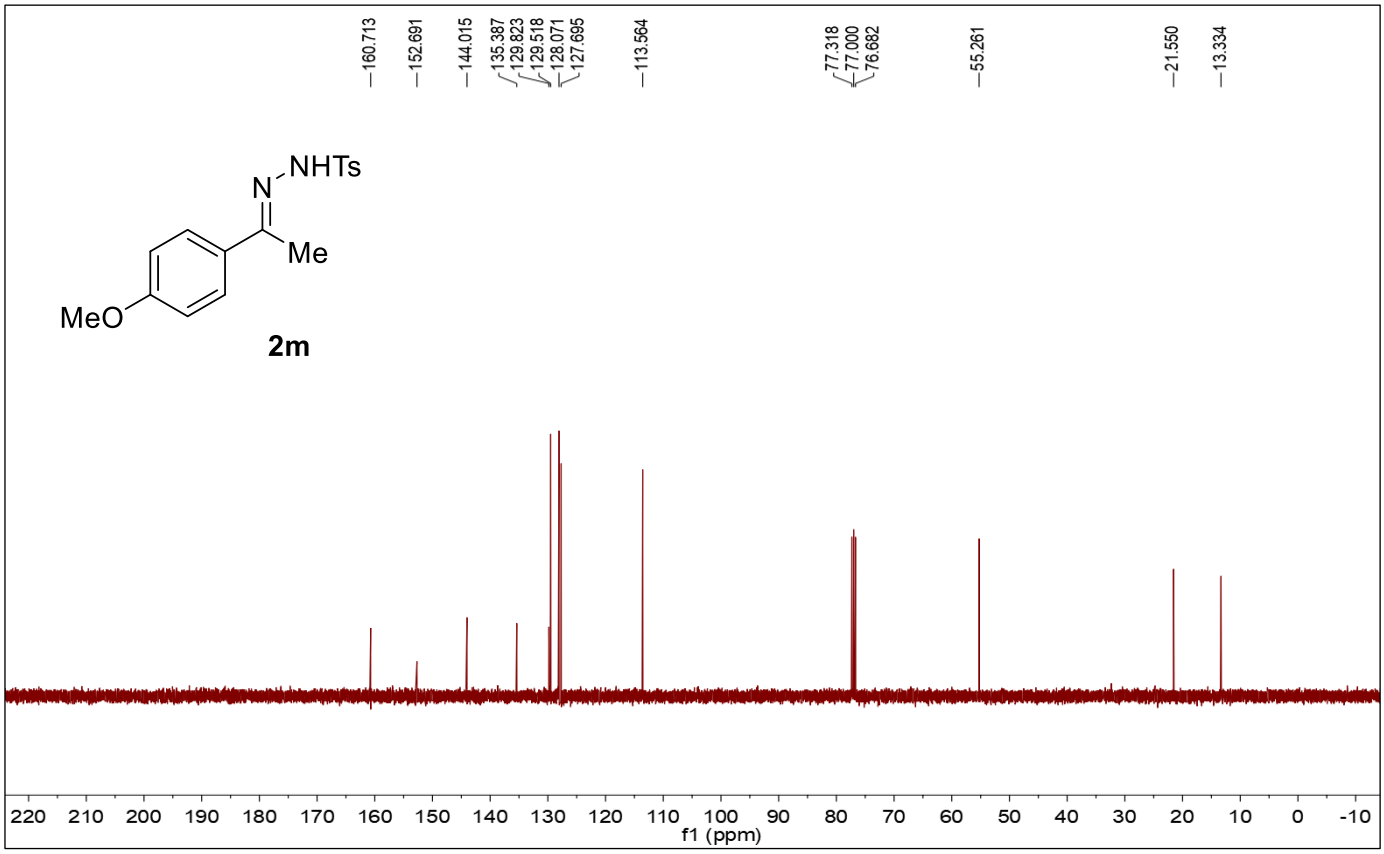
7.298
6.853
6.831
3.802
2.400
2.127
0.000
NHTs
N
Me
MeO
2m
0.93
2.05
2.02
2.03
2.00
3.03
3.01
3.01
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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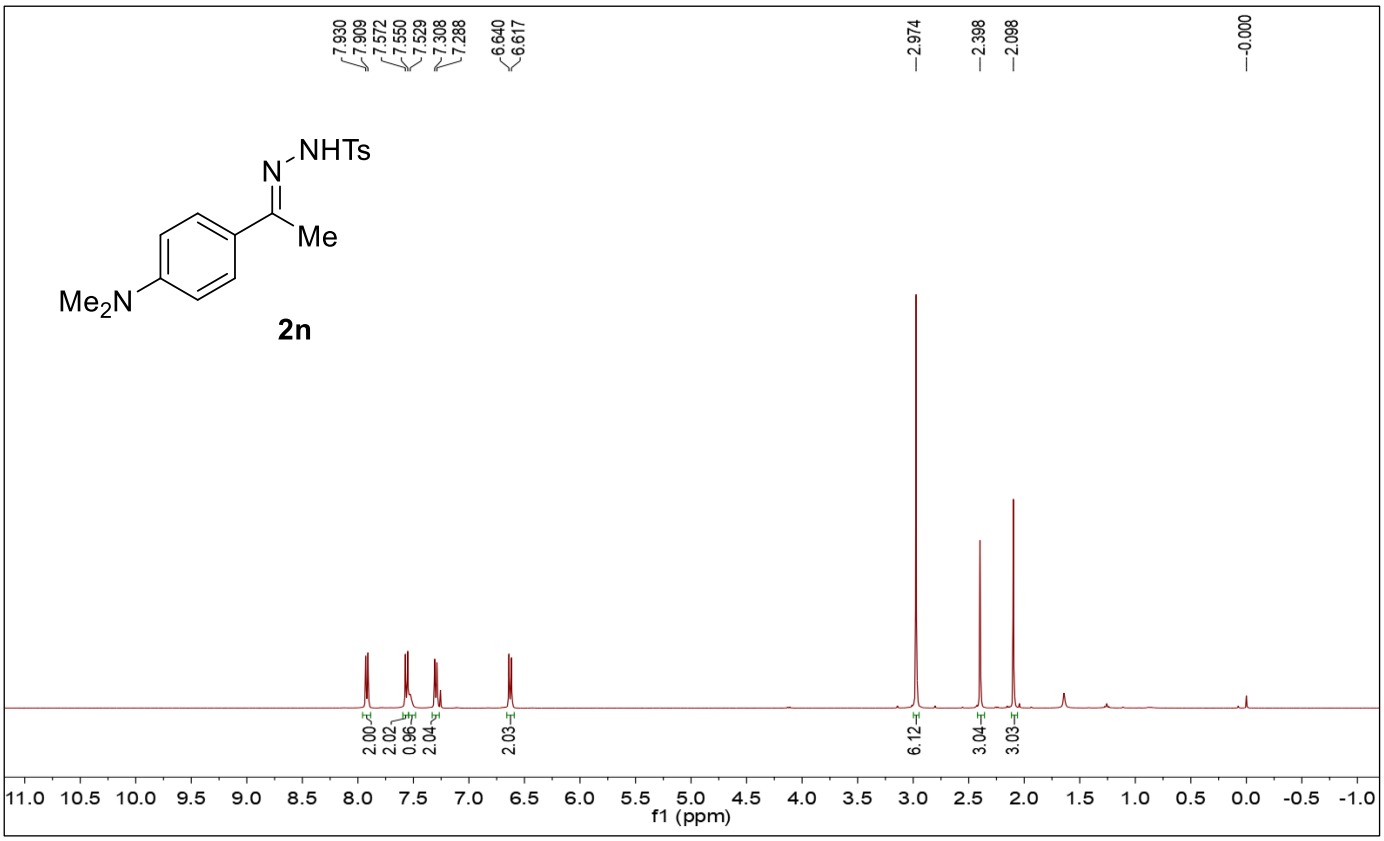
129.518
128.071
127.695
113.564
77.318
77.000
76.682
55.261
21.550
13.334
NHTs
N
Me
MeO
2m
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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7.909
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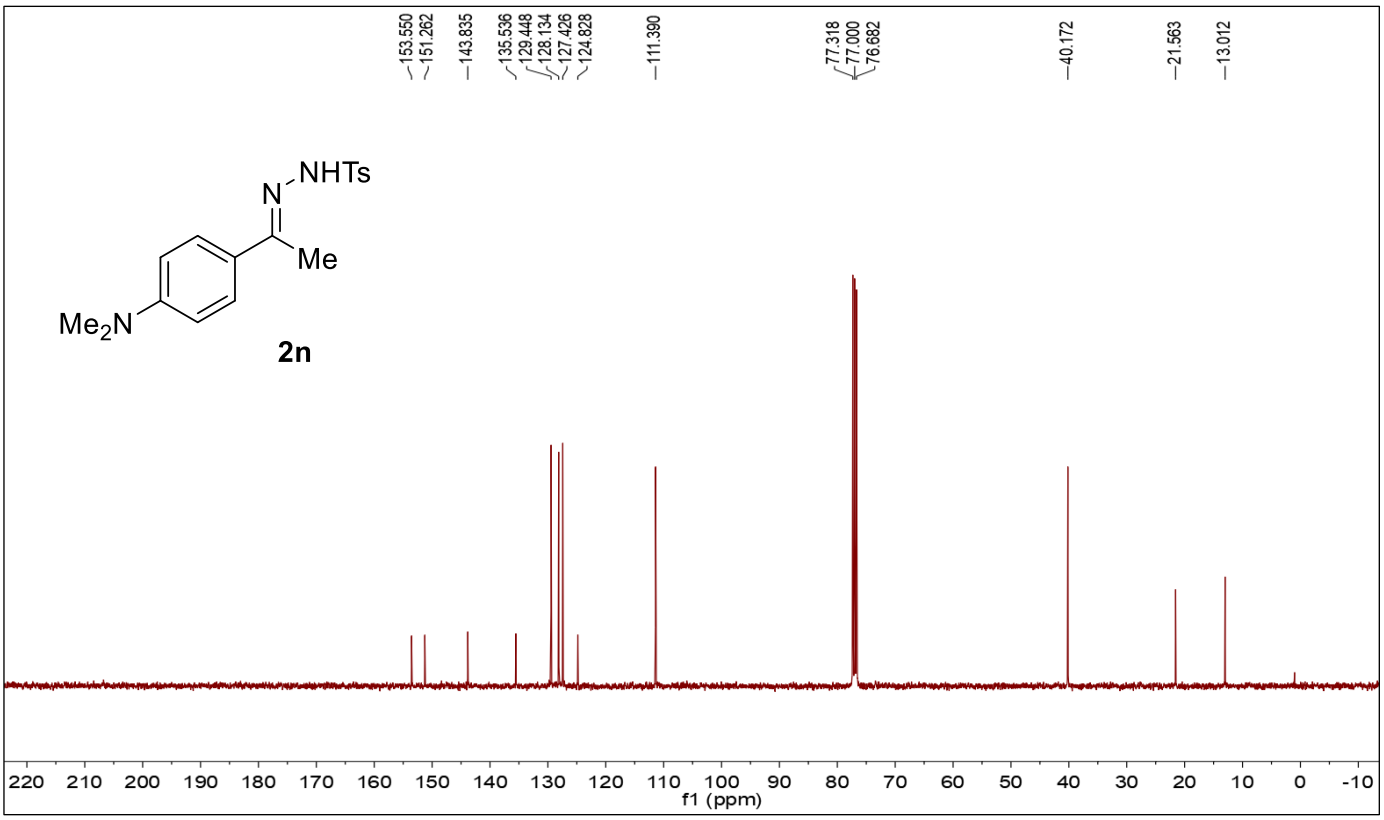
7.550
7.529
7.308
7.288
6.640
6.617
2.974
2.398
2.098
0.000
NHTs
N
Me
Me2N
2n
2.00
2.02
0.96
2.04
2.03
6.12
3.04
3.03
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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128.134
127.426
124.828
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77.318
77.000
76.682
40.172
21.563
13.012
NHTs
N
Me
Me2N
2n
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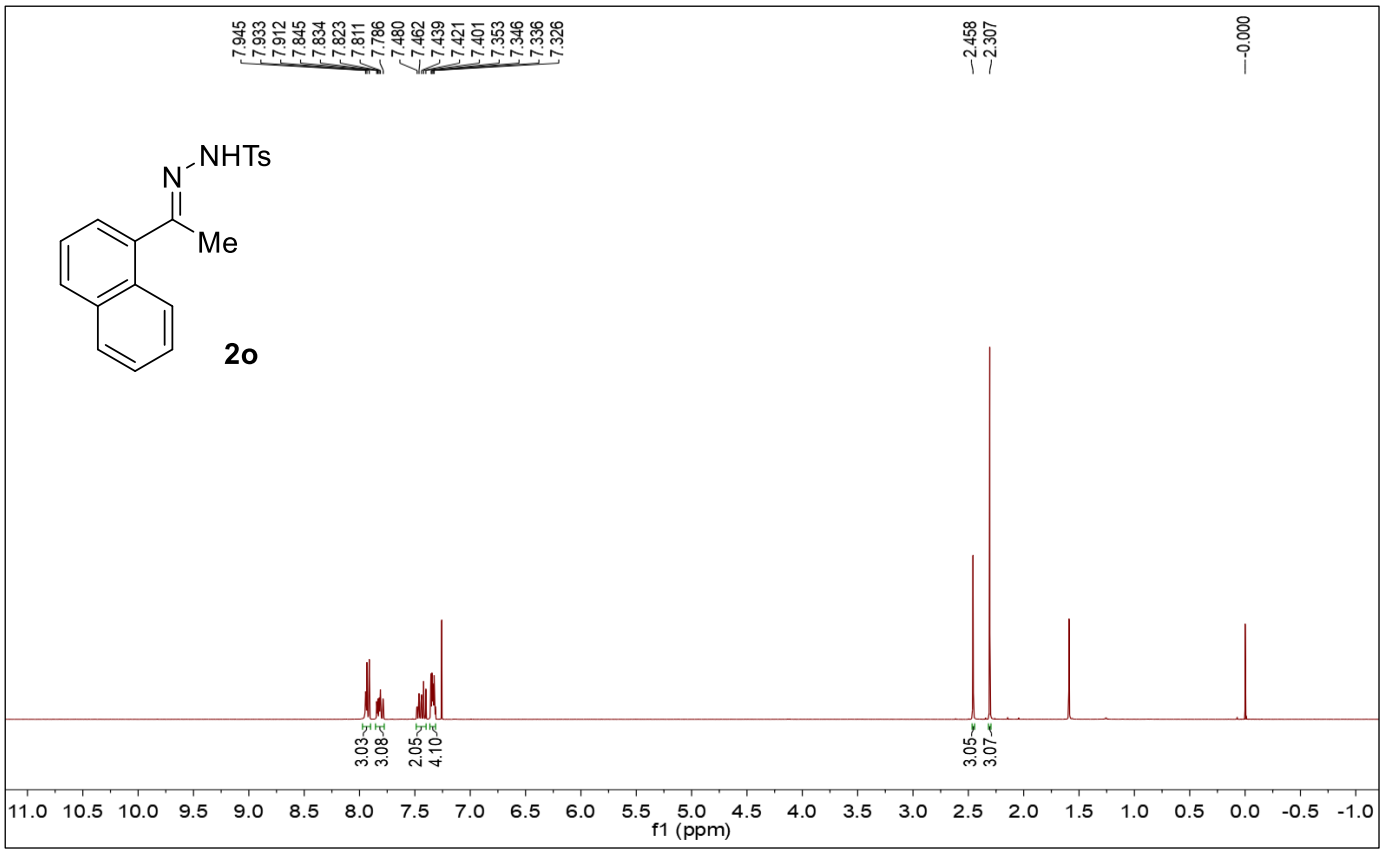
f1 (ppm)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7.845
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7.811
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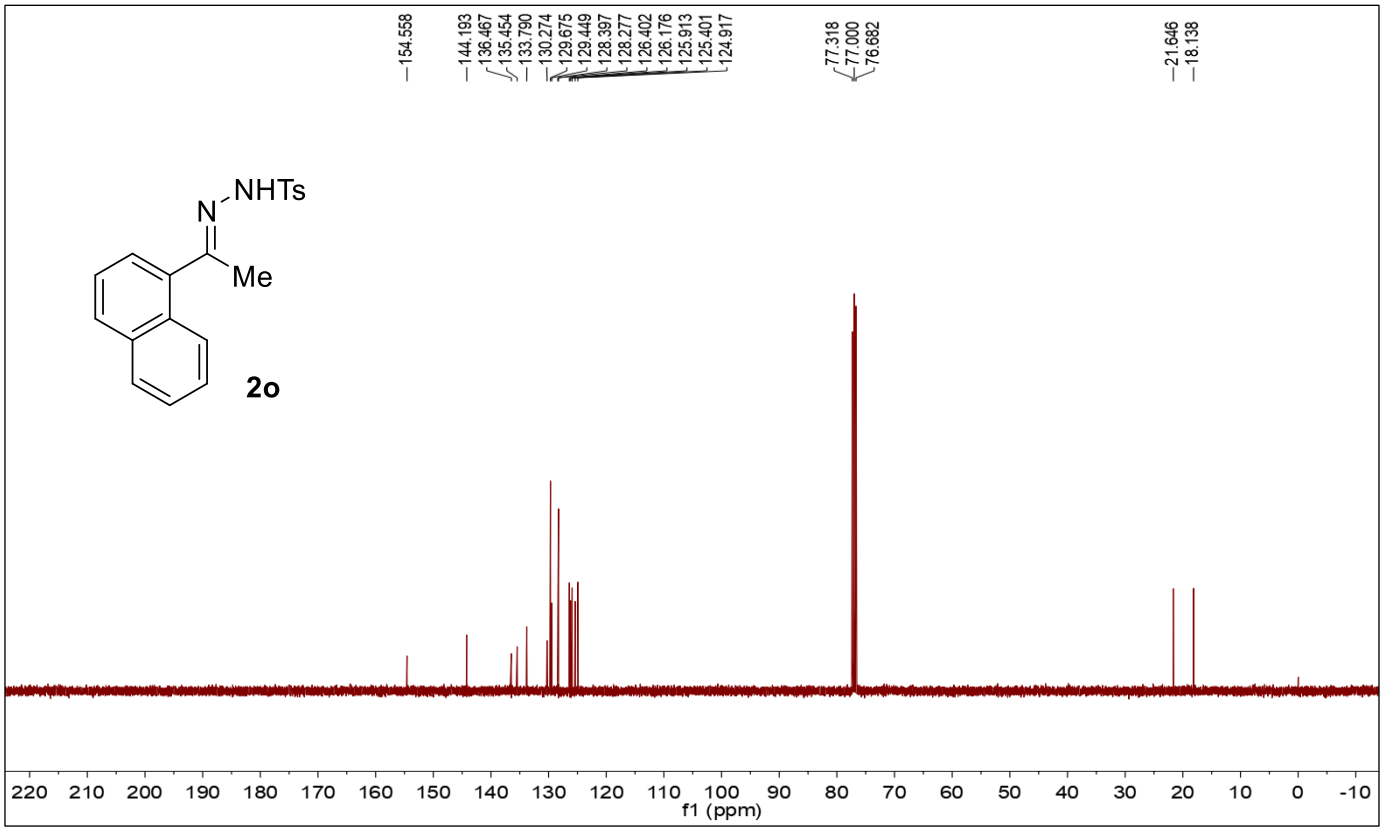
7.480
7.462
7.439
7.421
7.401
7.353
7.346
7.336
7.326
2.458
2.307
0.000
NHTs
N
Me
2o
3.03
3.08
2.05
4.10
3.05
3.07
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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128.277
126.402
126.176
125.913
125.401
124.917
77.318
77.000
76.682
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NHTs
N
Me
2o
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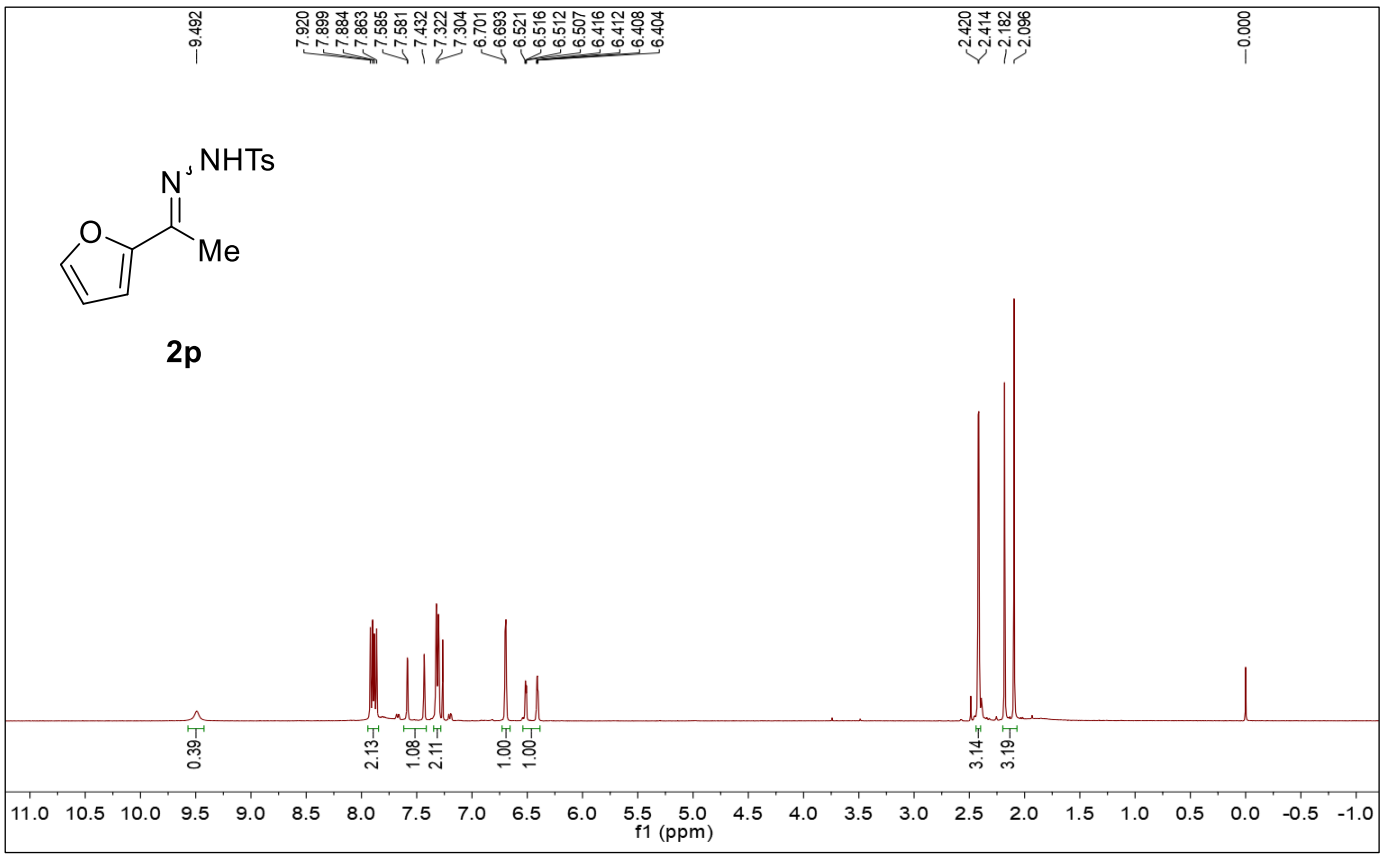
f1 (ppm)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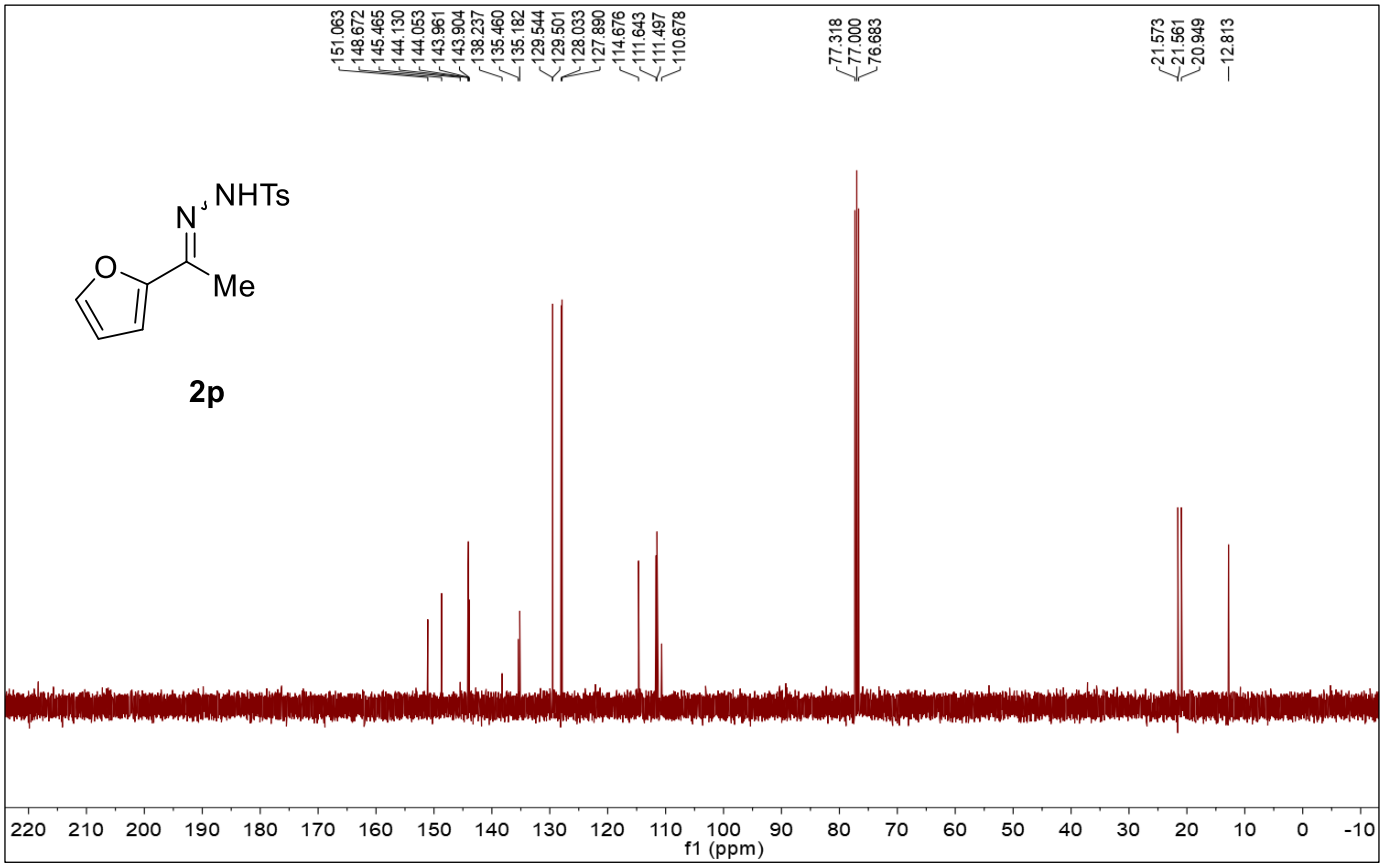
6.521
6.516
6.512
6.507
6.416
6.412
6.408
6.404
2.420
2.414
2.182
2.096
0.000
NHTs
N
O
Me
2p
0.39
2.13
1.08
2.11
1.00
1.00
3.14
3.19
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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138.237
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129.501
128.033
127.890
114.676
111.643
111.497
110.678
77.318
77.000
76.683
21.573
21.561
20.949
12.813
NHTs
N
O
Me
2p
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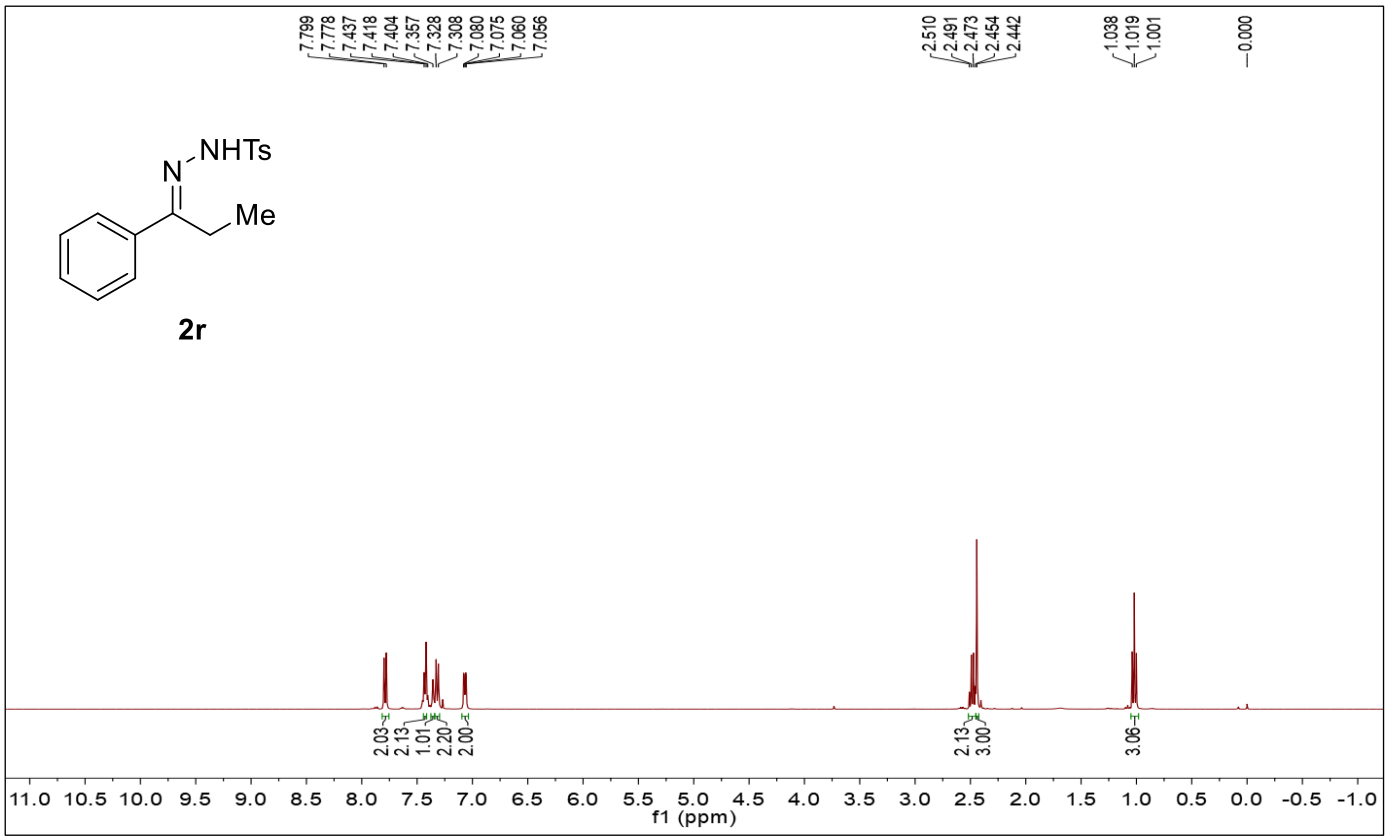
f1 (ppm)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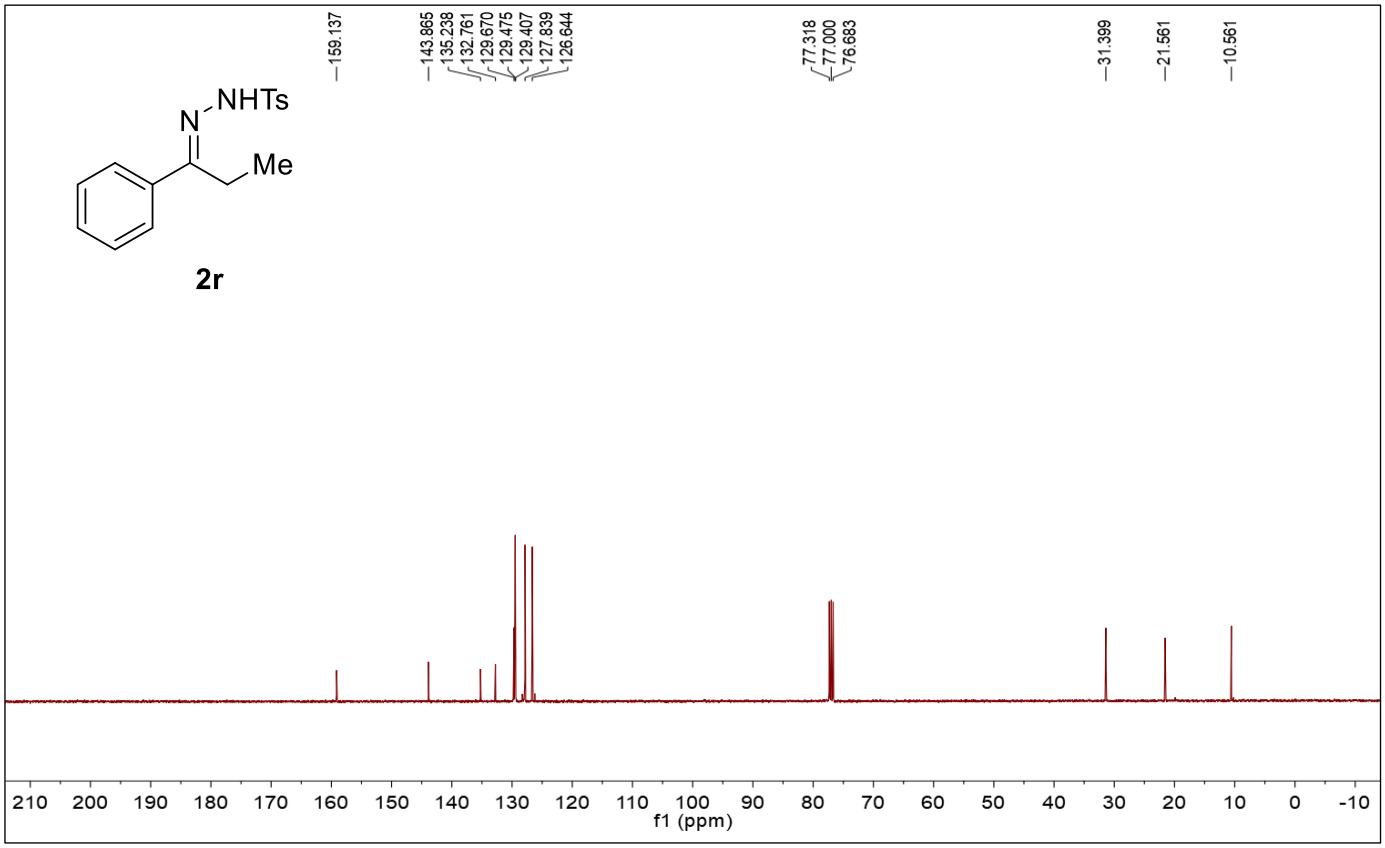
7.328
7.308
7.080
7.075
7.060
7.056
2.510
2.491
2.473
2.454
2.442
1.038
1.019
1.001
0.000
NHTs
N
Me
2r
2.03
2.13
1.01
2.20
2.00
2.13
3.00
3.06
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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127.839
126.644
77.318
77.000
76.683
31.399
21.561
10.561
NHTs
N
Me
2r
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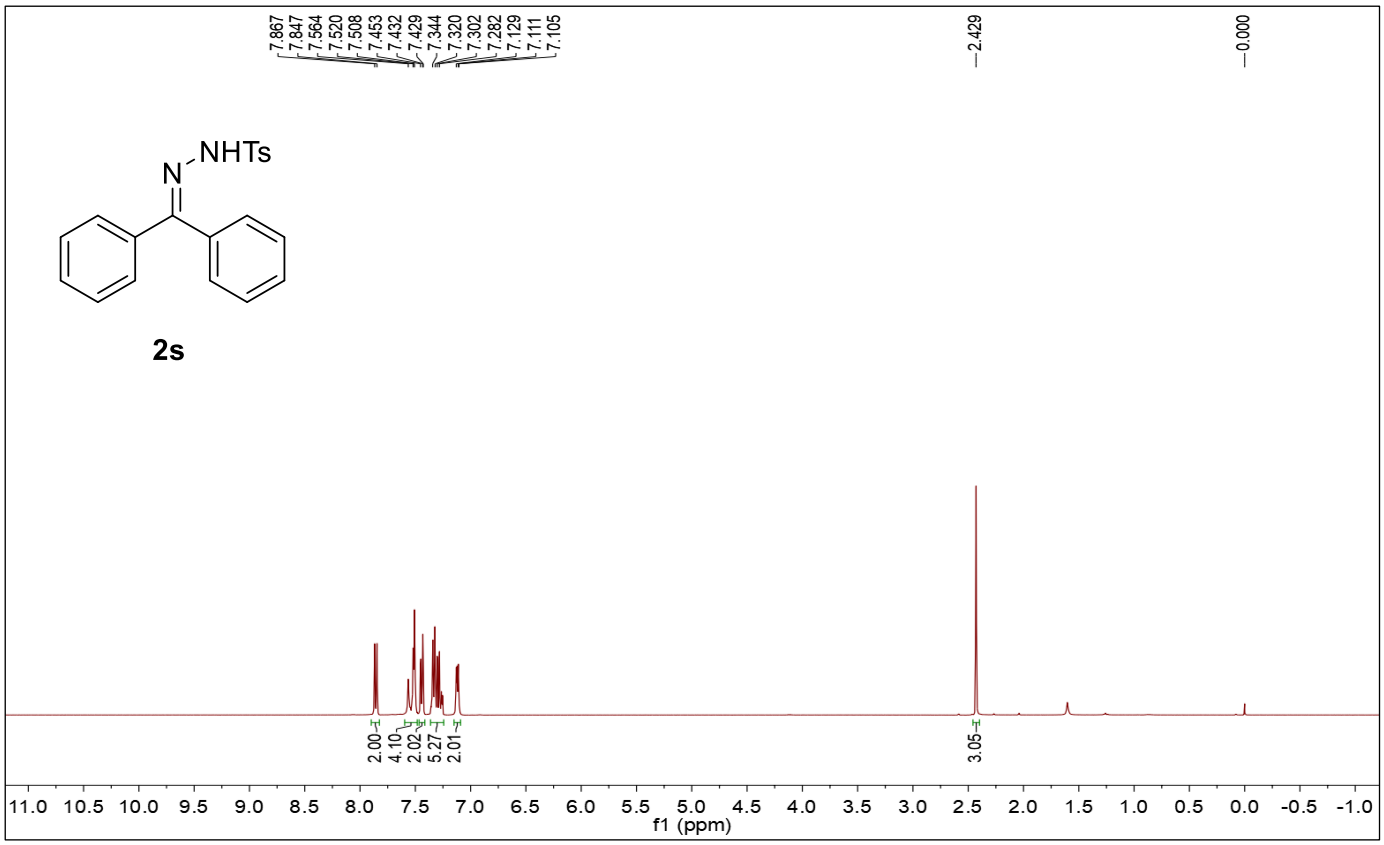
f1 (ppm)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7.429
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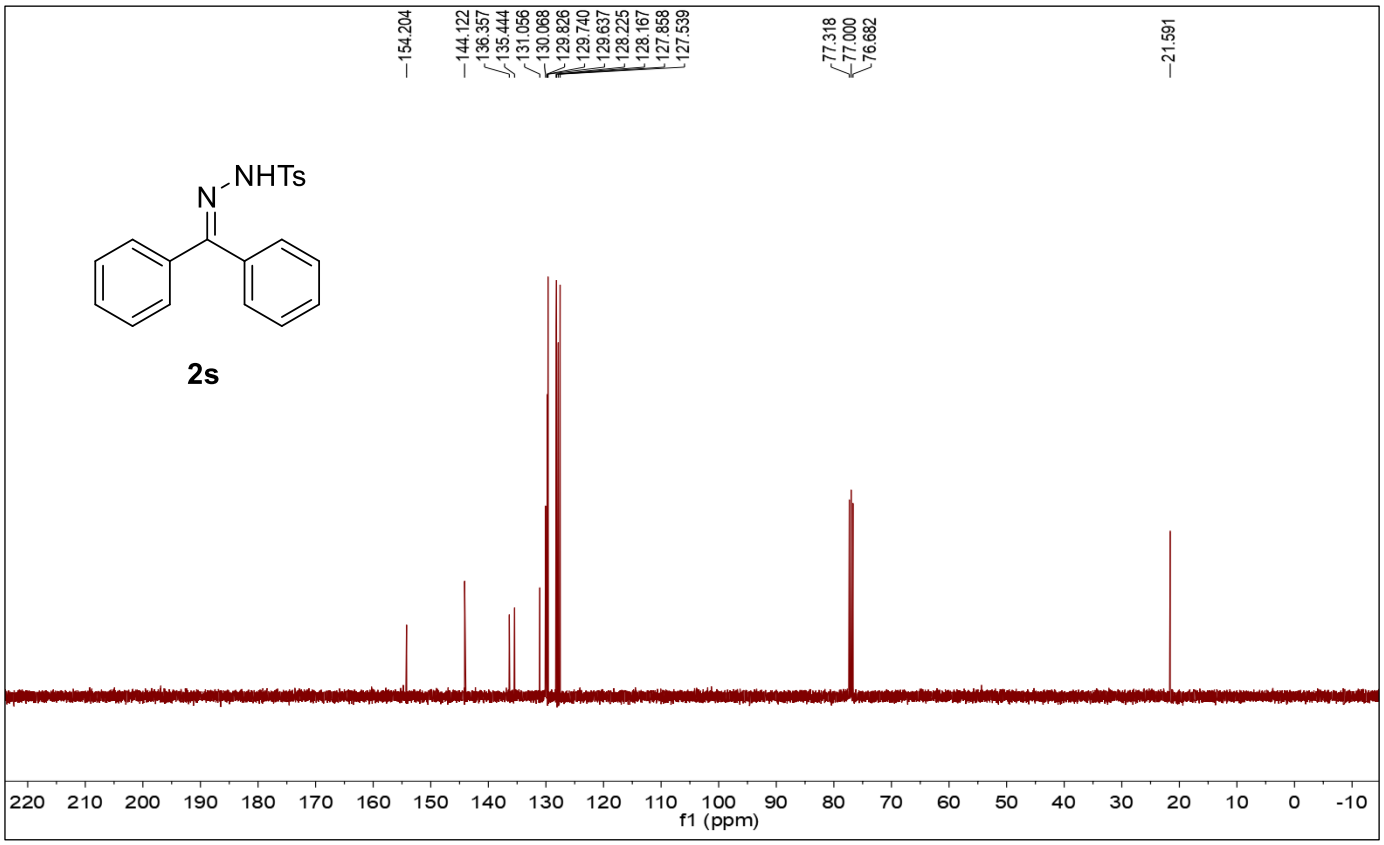
7.320
7.302
7.282
7.129
7.111
7.105
2.429
0.000
NHTs
N
2s
2.00
4.10
2.02
5.27
2.01
3.05
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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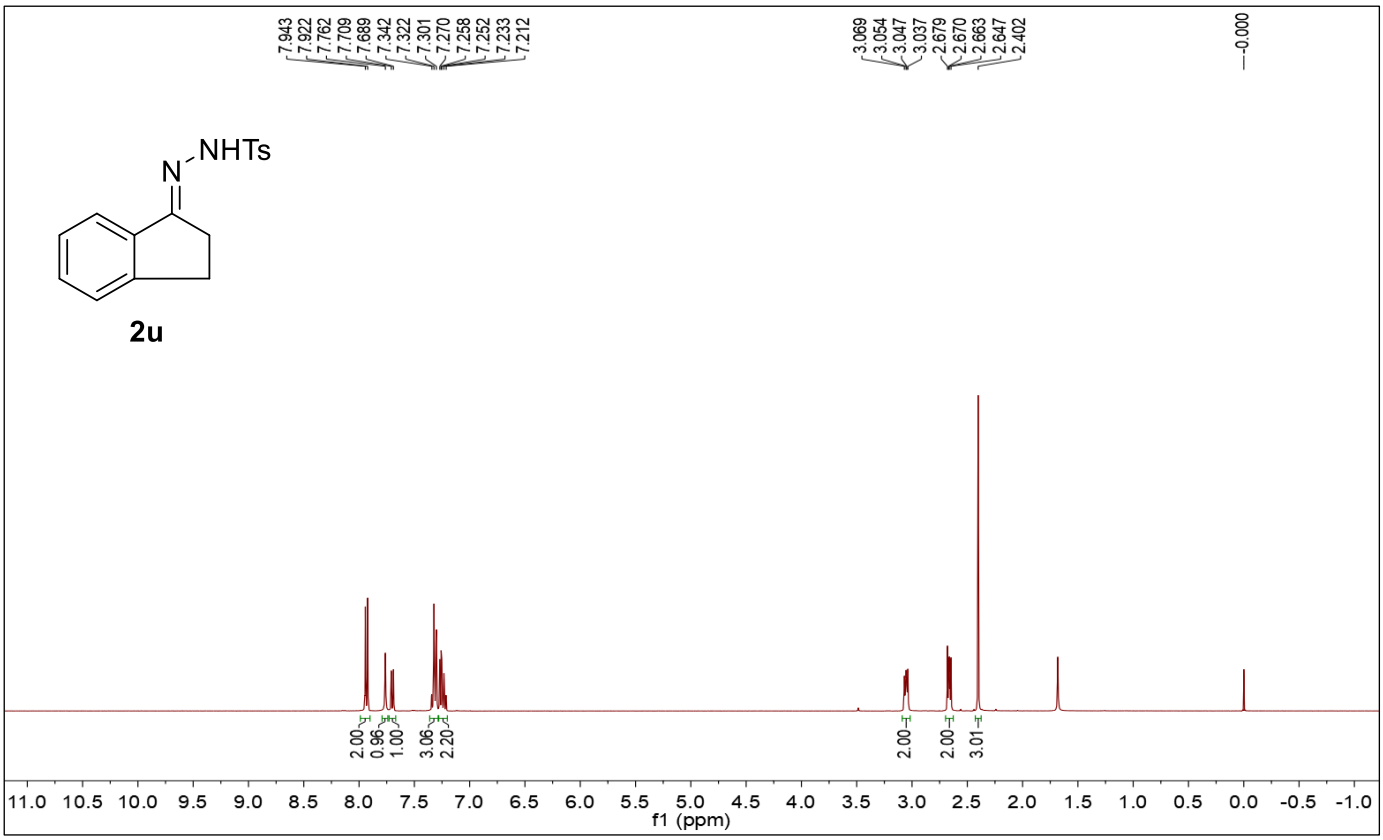
128.225
128.167
127.858
127.539
77.318
77.000
76.682
21.591
NHTs
N
2s
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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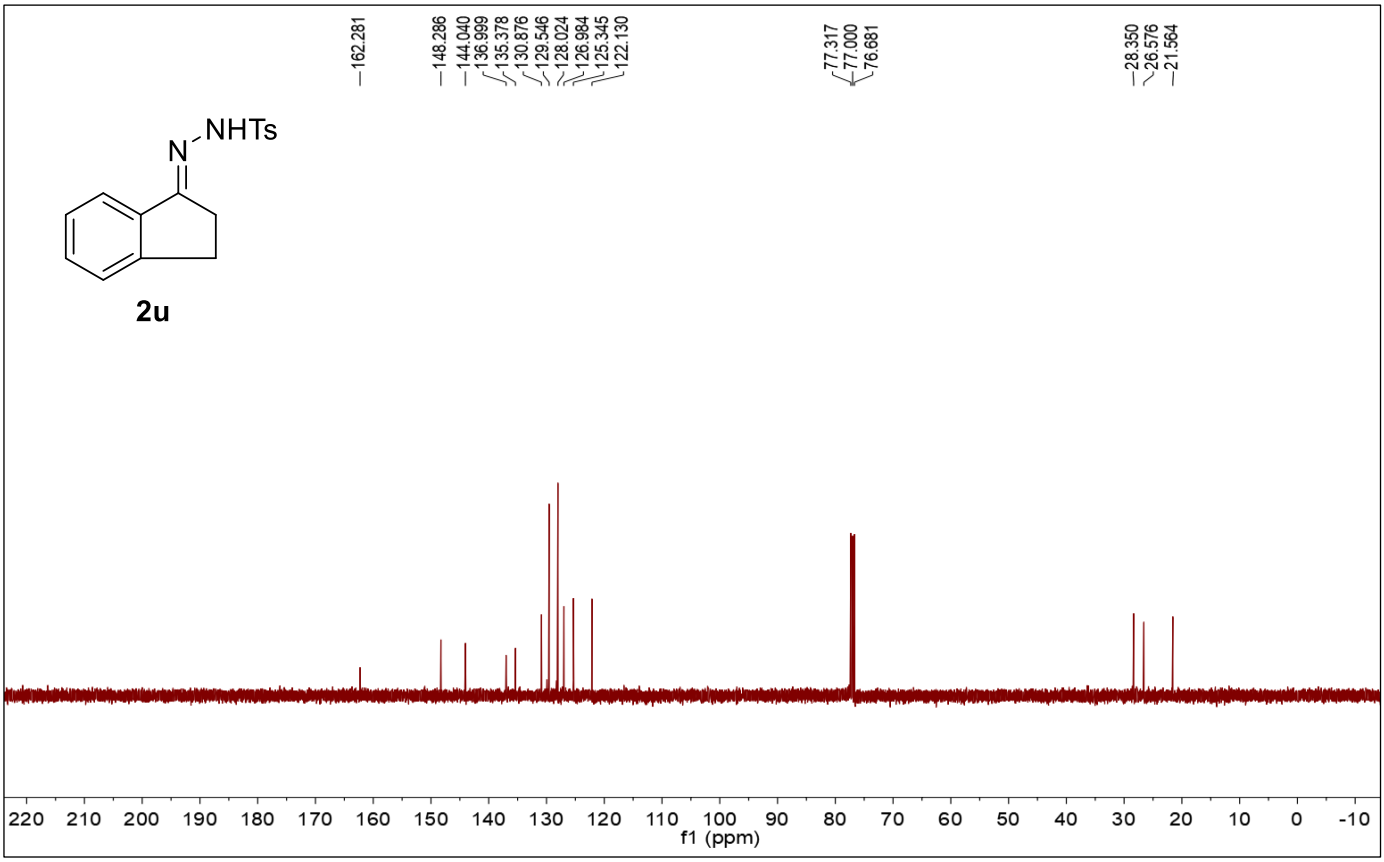
7.252
7.233
7.212
3.069
3.054
3.047
3.037
2.679
2.670
2.663
2.647
2.402
0.000
NHTs
N
2u
2.00
0.96
1.00
3.06
2.20
2.00
2.00
3.01
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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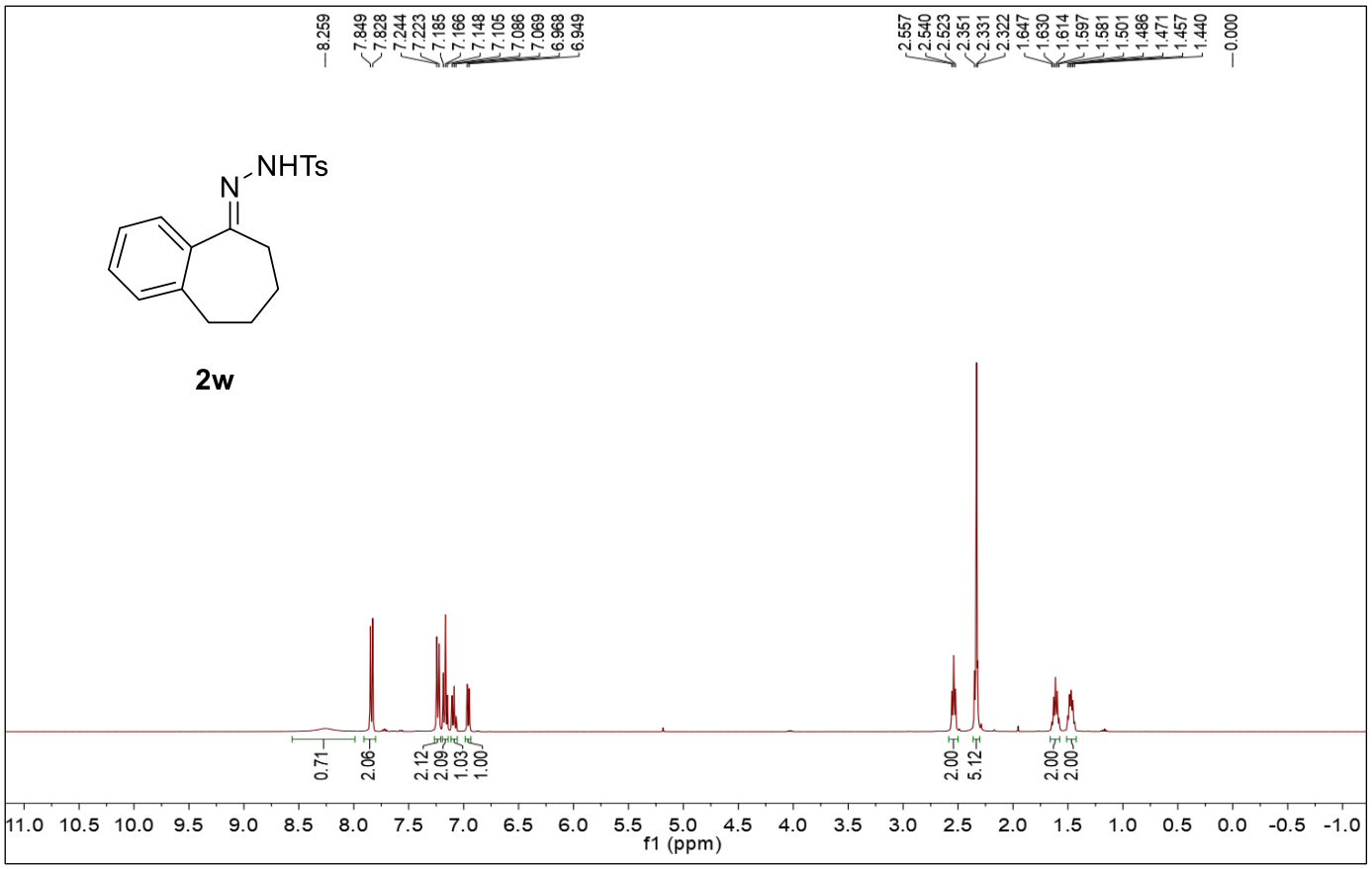
135.378
130.876
129.546
128.024
126.984
125.345
122.130
77.317
77.000
76.681
28.350
26.576
21.564
NHTs
N
2u
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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7.148
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7.069
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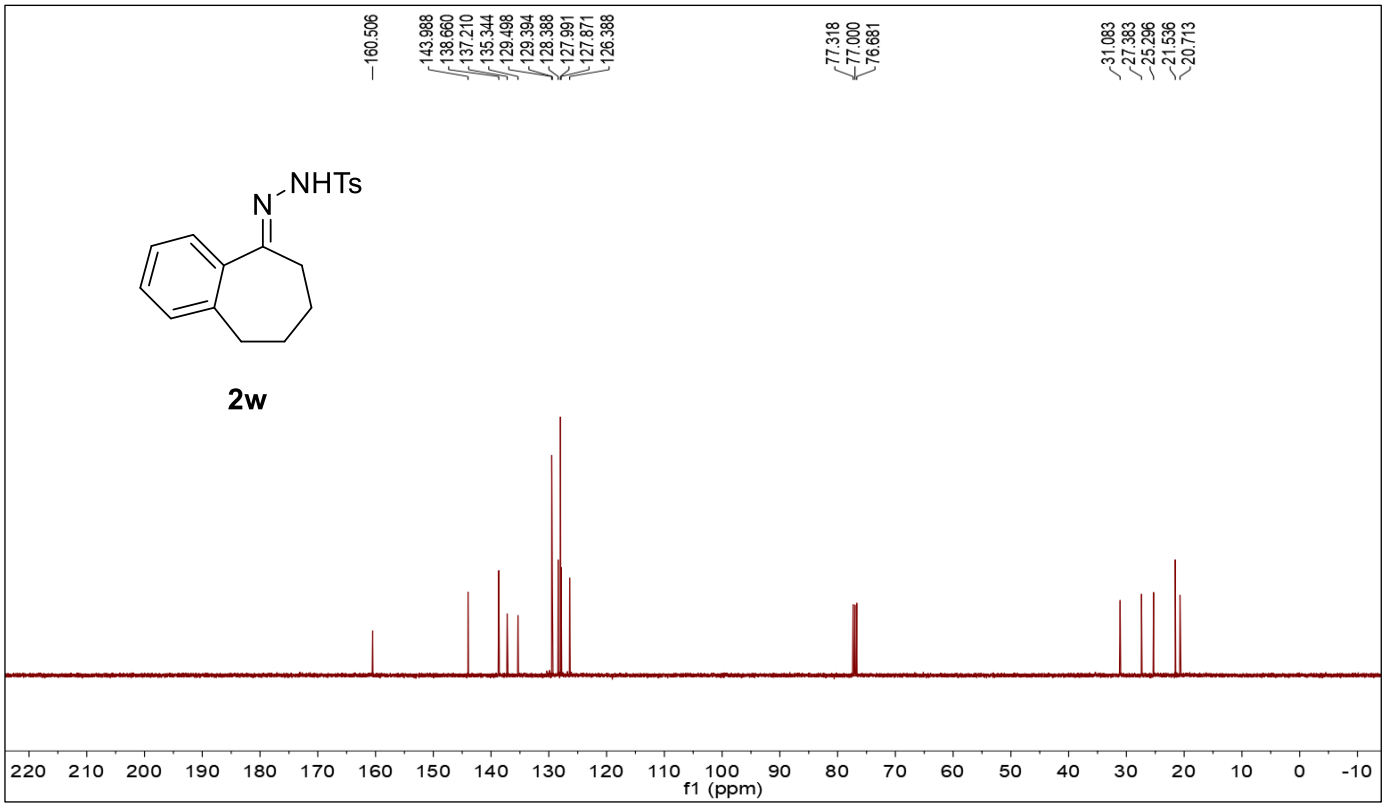
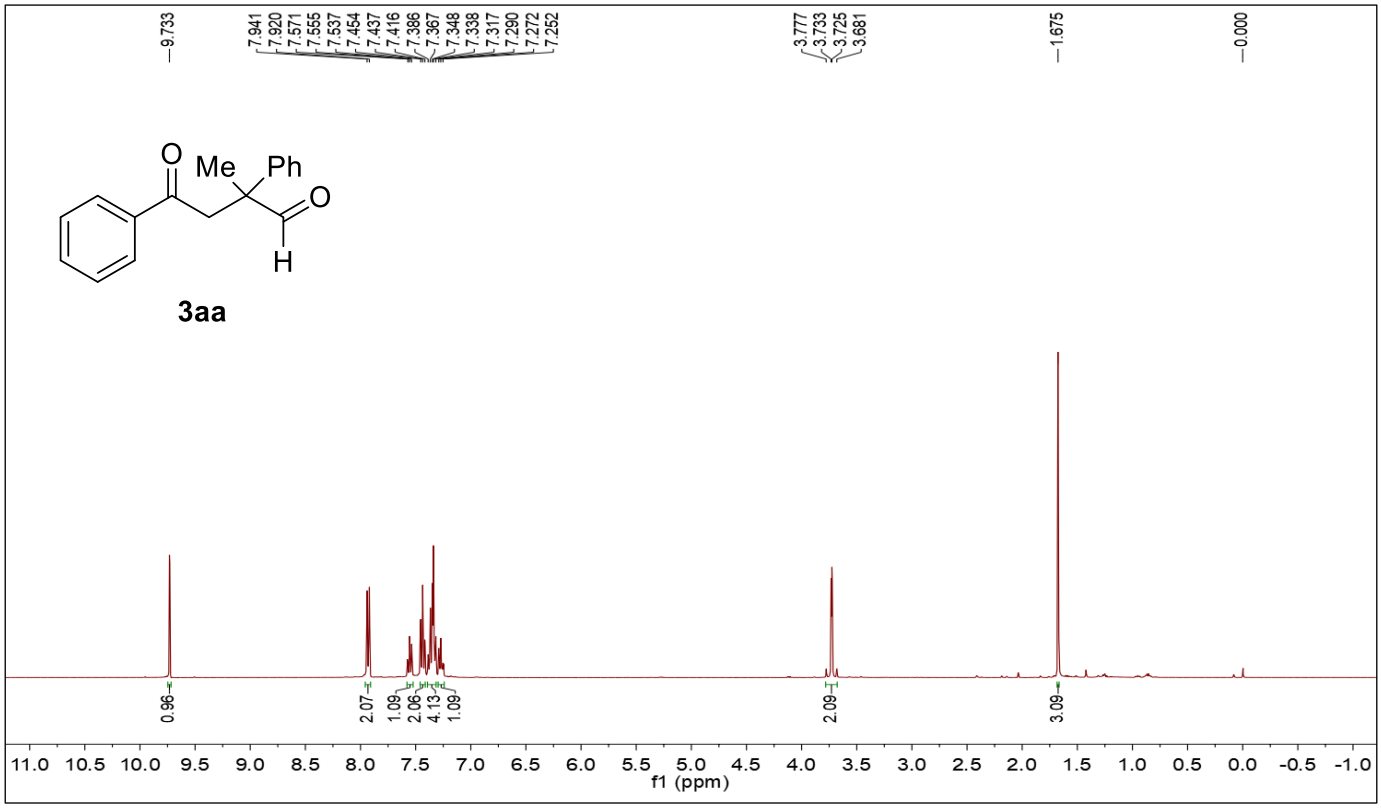
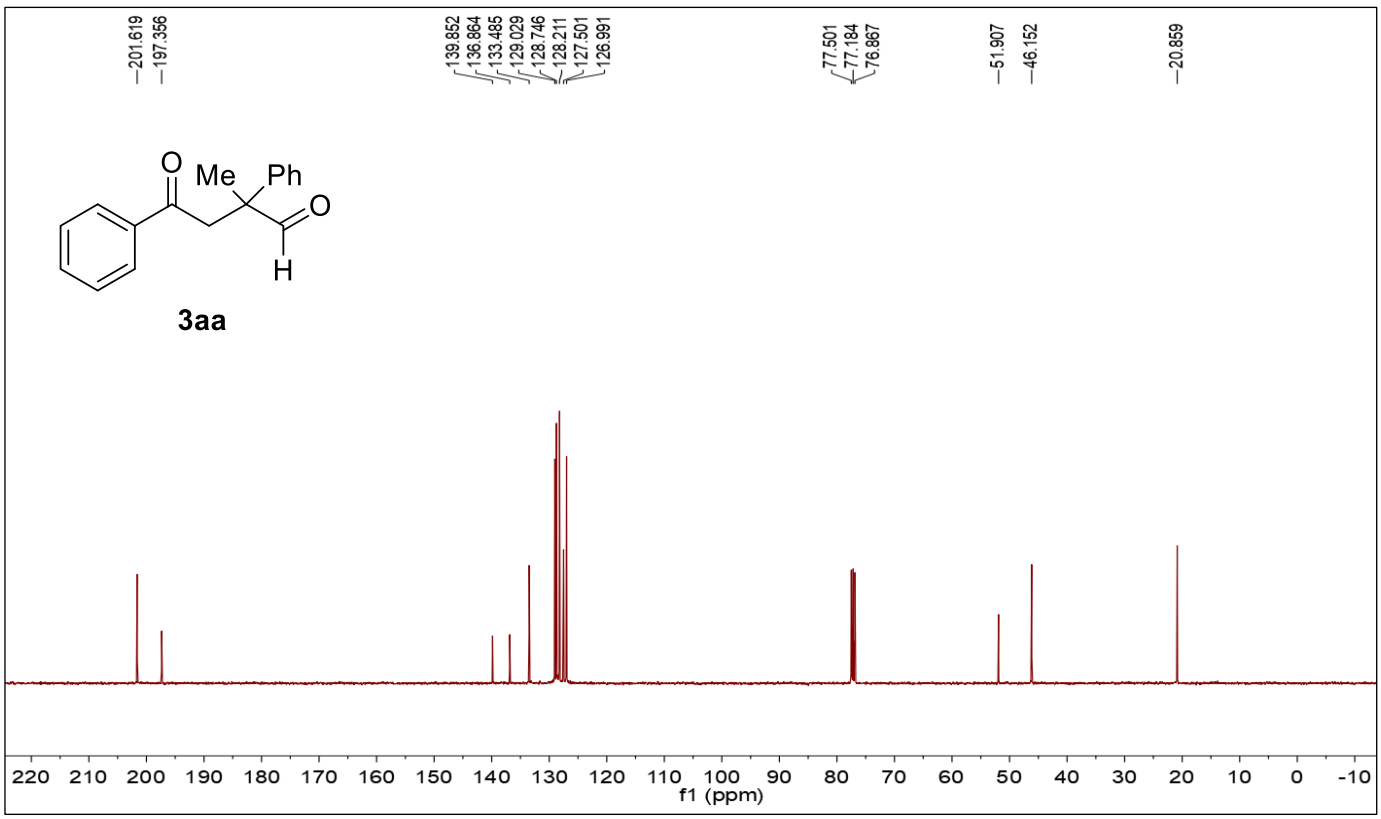
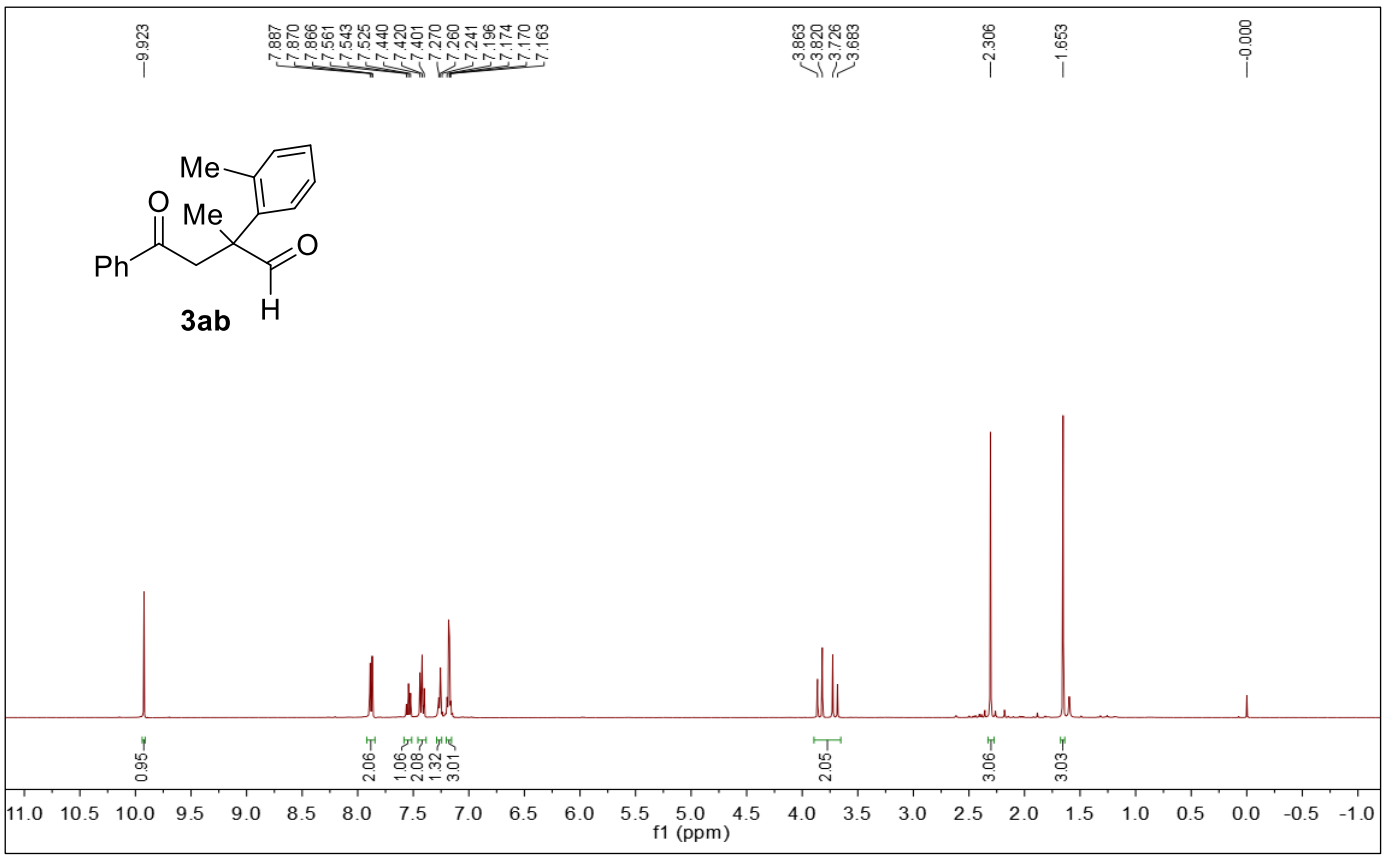
2.331
2.322
1.647
1.630
1.614
1.597
1.581
1.501
1.486
1.471
1.457
1.440
0.000
NHTs
N
2w
0.71
2.06
2.12
2.09
1.03
1.00
2.00
5.12
2.00
2.00
f1 (ppm)

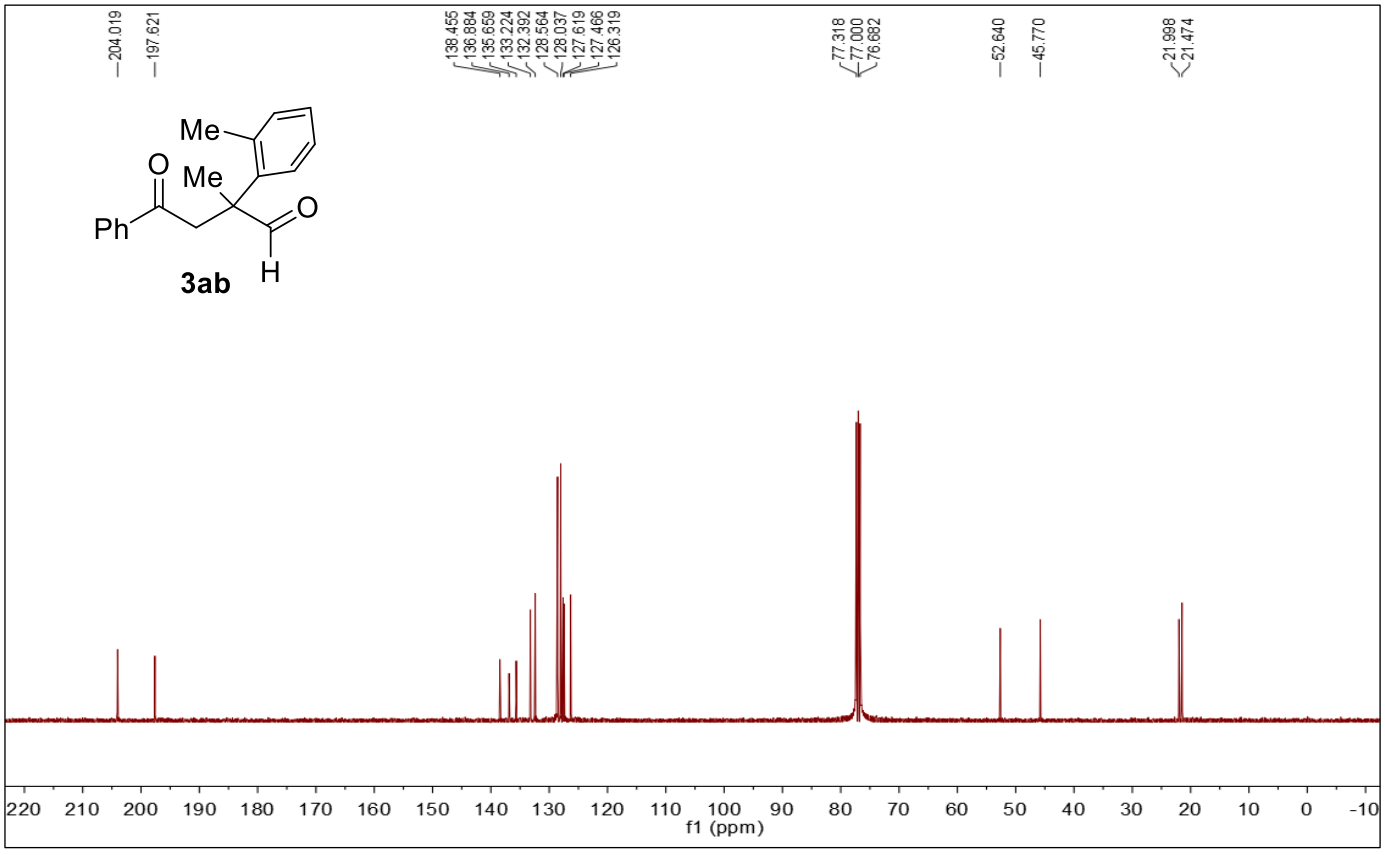

160.506
143.988
138.660
137.210
135.344
129.498
129.394
128.388
127.991
127.871
126.388
77.318
77.000
76.681
31.083
27.383
25.296
21.536
20.713
NHTs
N
2w
f1 (ppm)

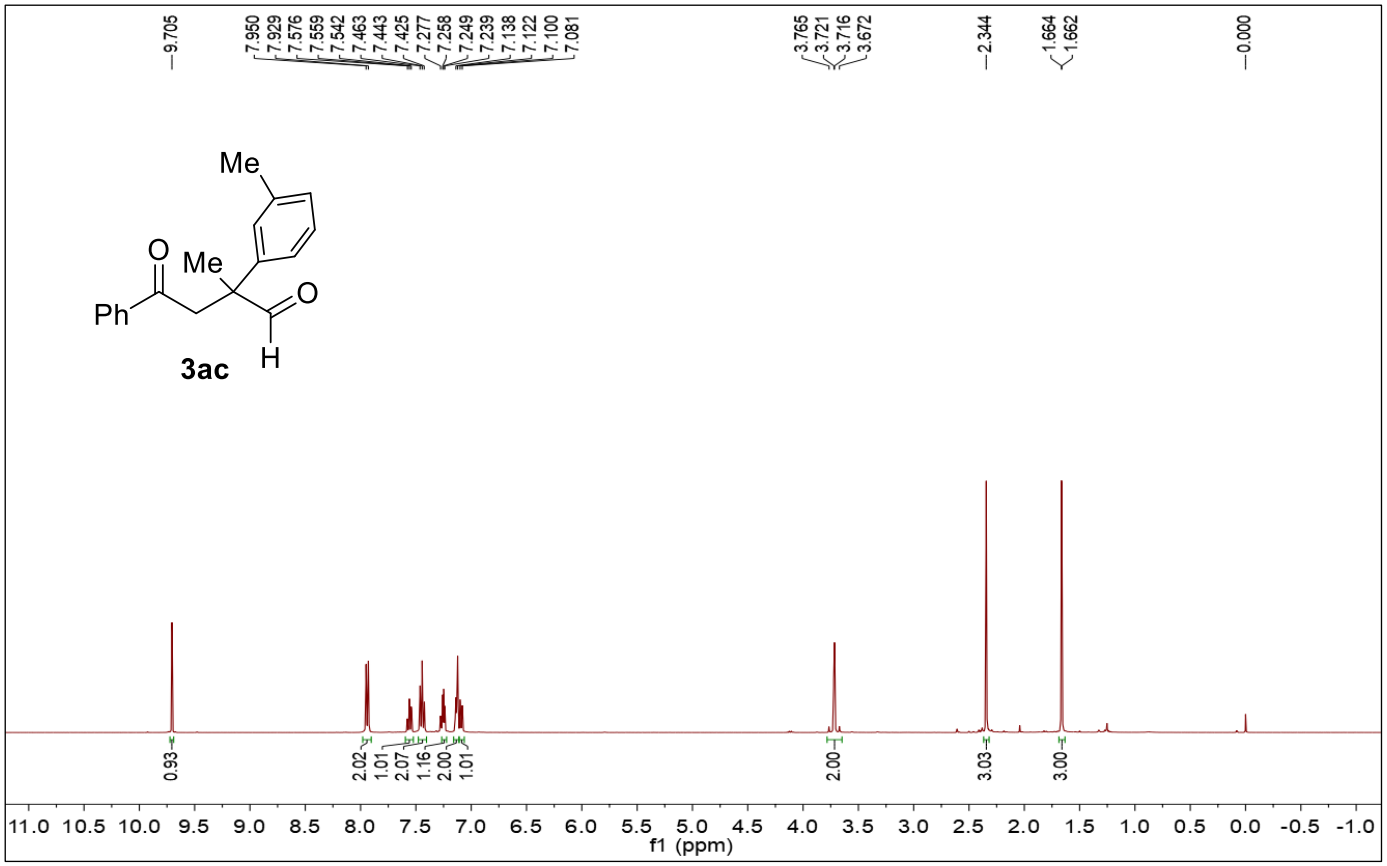
^{1}H and ^{13}C NMR spectra of 1,4-ketoaldehydes:

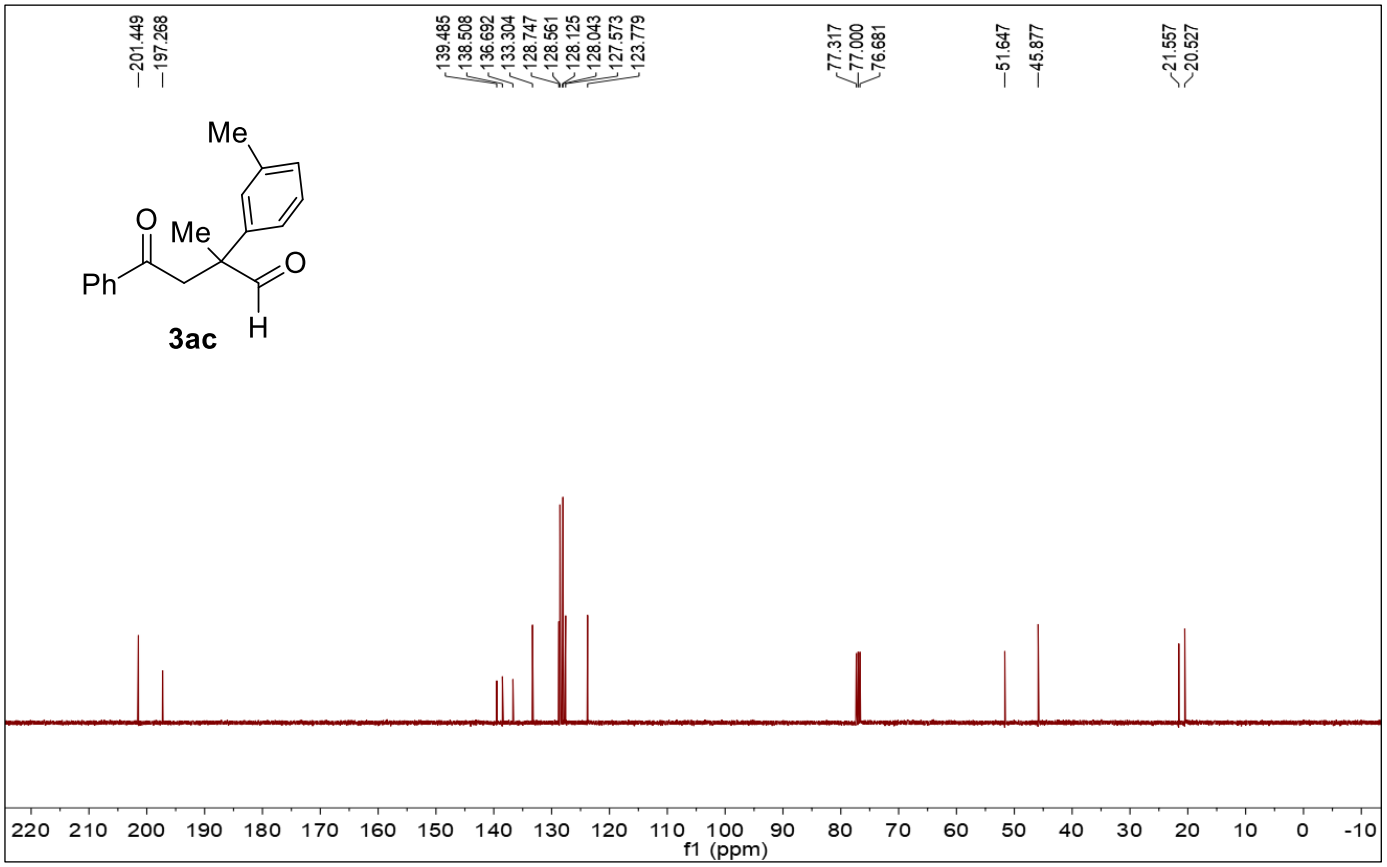





9.923
7.887
7.870
7.866
7.561
7.543
7.525
7.440
7.420
7.401
7.270
7.260
7.241
7.196
7.174
7.170
7.163
3.863
3.820
3.726
3.683
2.306
1.653
0.000
Me
O
Me
Ph
O
H
3ab
0.95
2.06
1.06
2.08
1.32
3.01
2.05
3.06
3.03
f1 (ppm)


204.019
197.621
138.455
136.884
135.659
133.224
132.392
128.564
128.037
127.619
127.466
126.319
77.318
77.000
76.682
52.640
45.770
21.998
21.474
Me
O
Me
Ph
O
H
3ab
f1 (ppm)


9.705
7.950
7.929
7.576
7.559
7.542
7.463
7.443
7.425
7.277
7.258
7.249
7.239
7.138
7.122
7.100
7.081
3.765
3.721
3.716
3.672
2.344
1.664
1.662
0.000
Me
O
Me
Ph
O
H
3ac
0.93
2.02
1.01
2.07
1.16
2.00
1.01
2.00
3.03
3.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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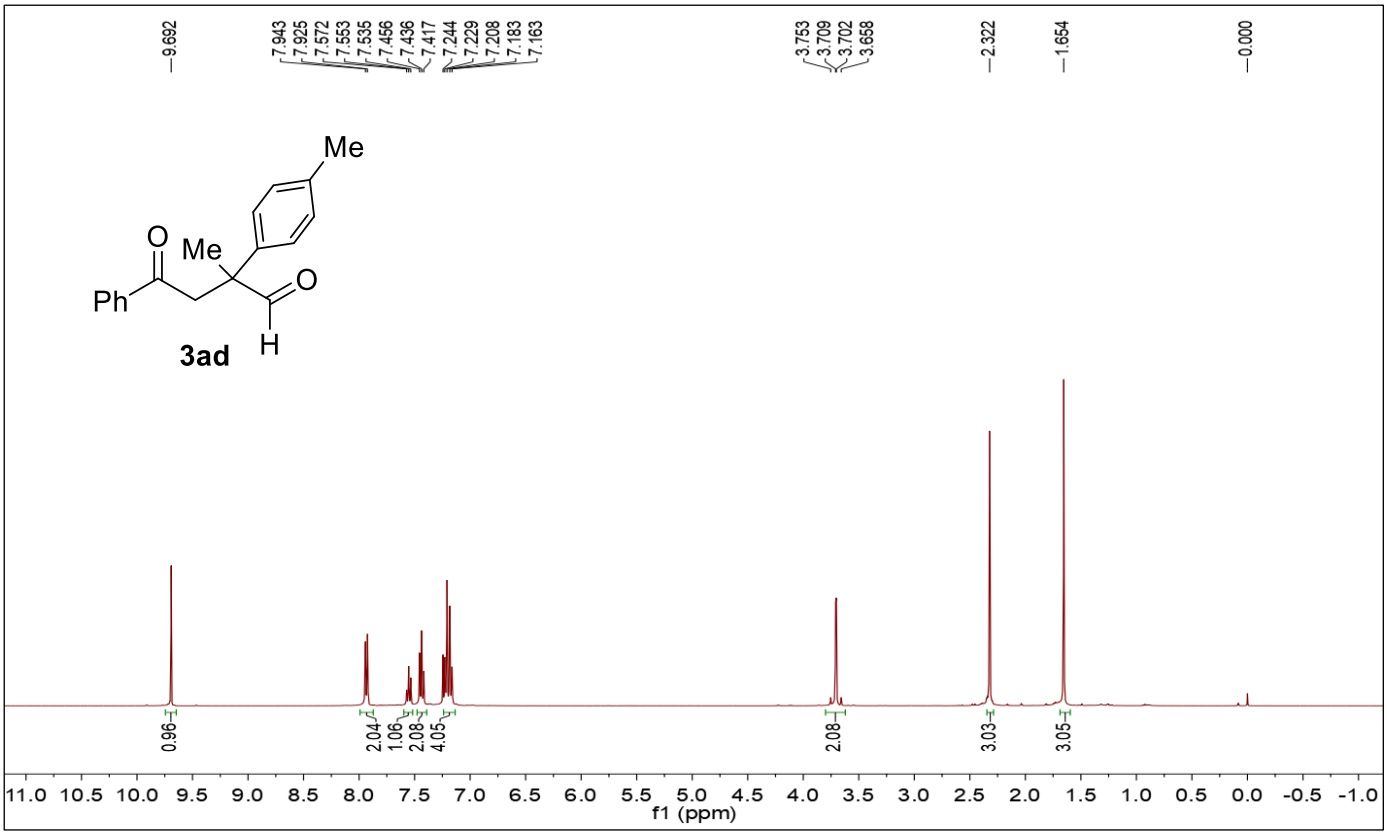
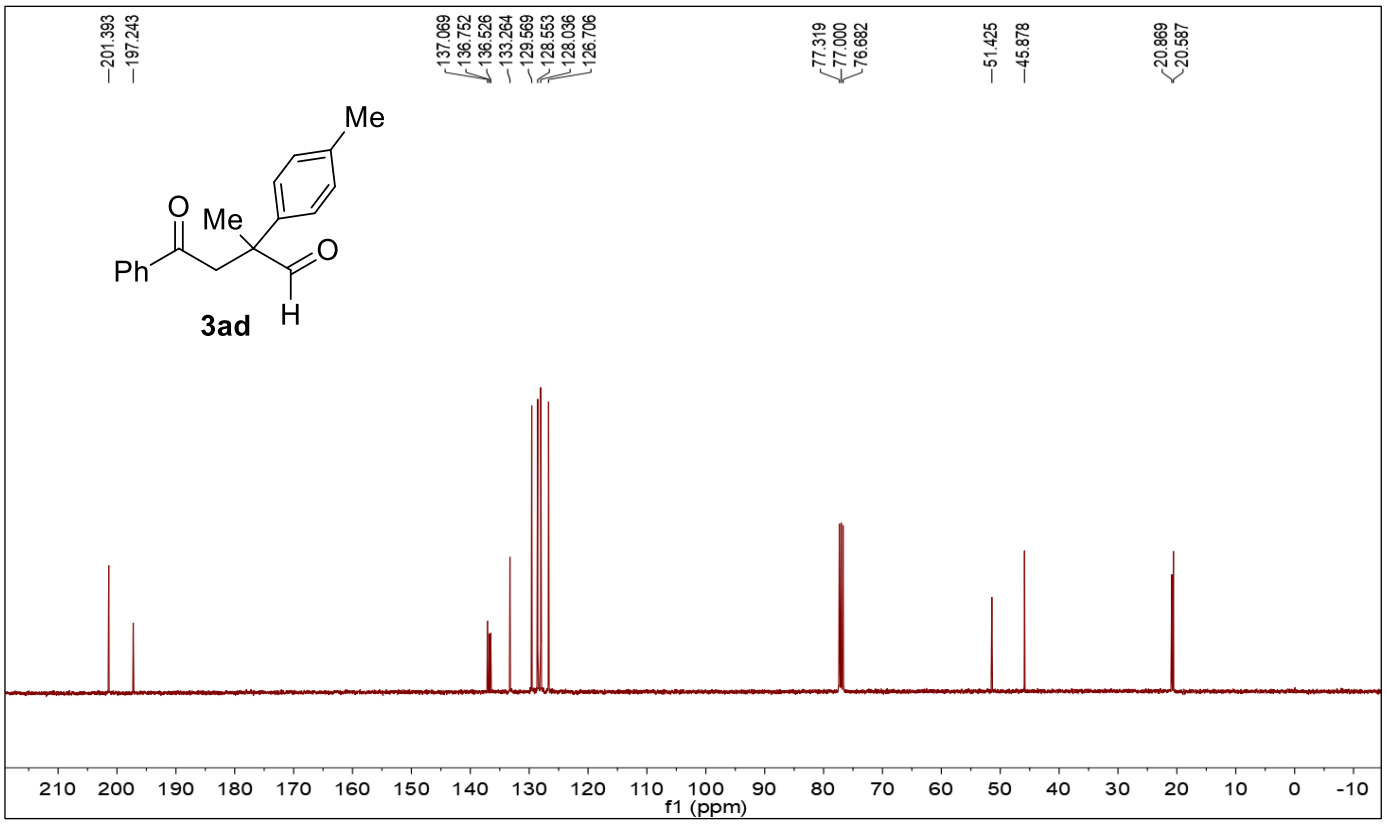
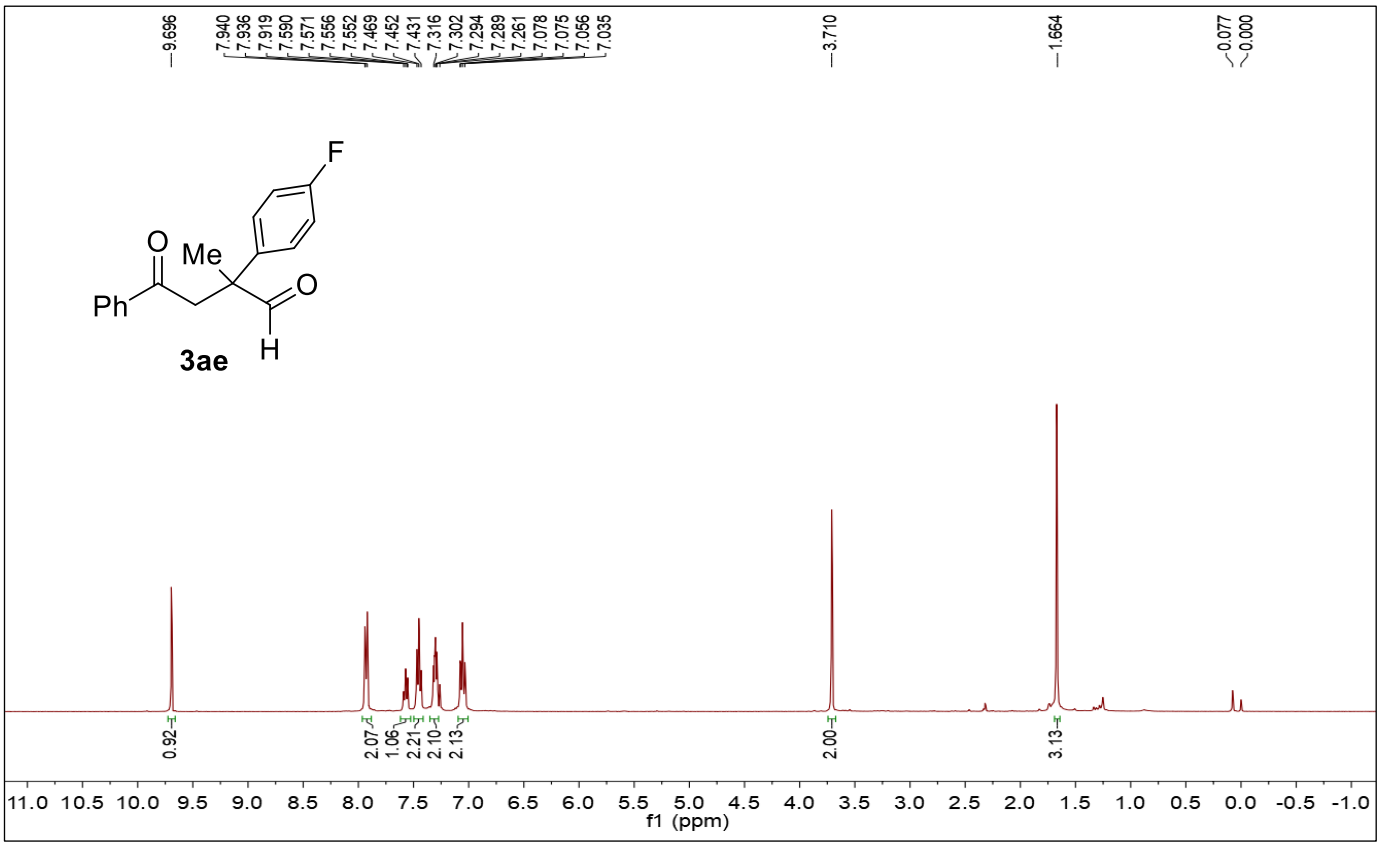
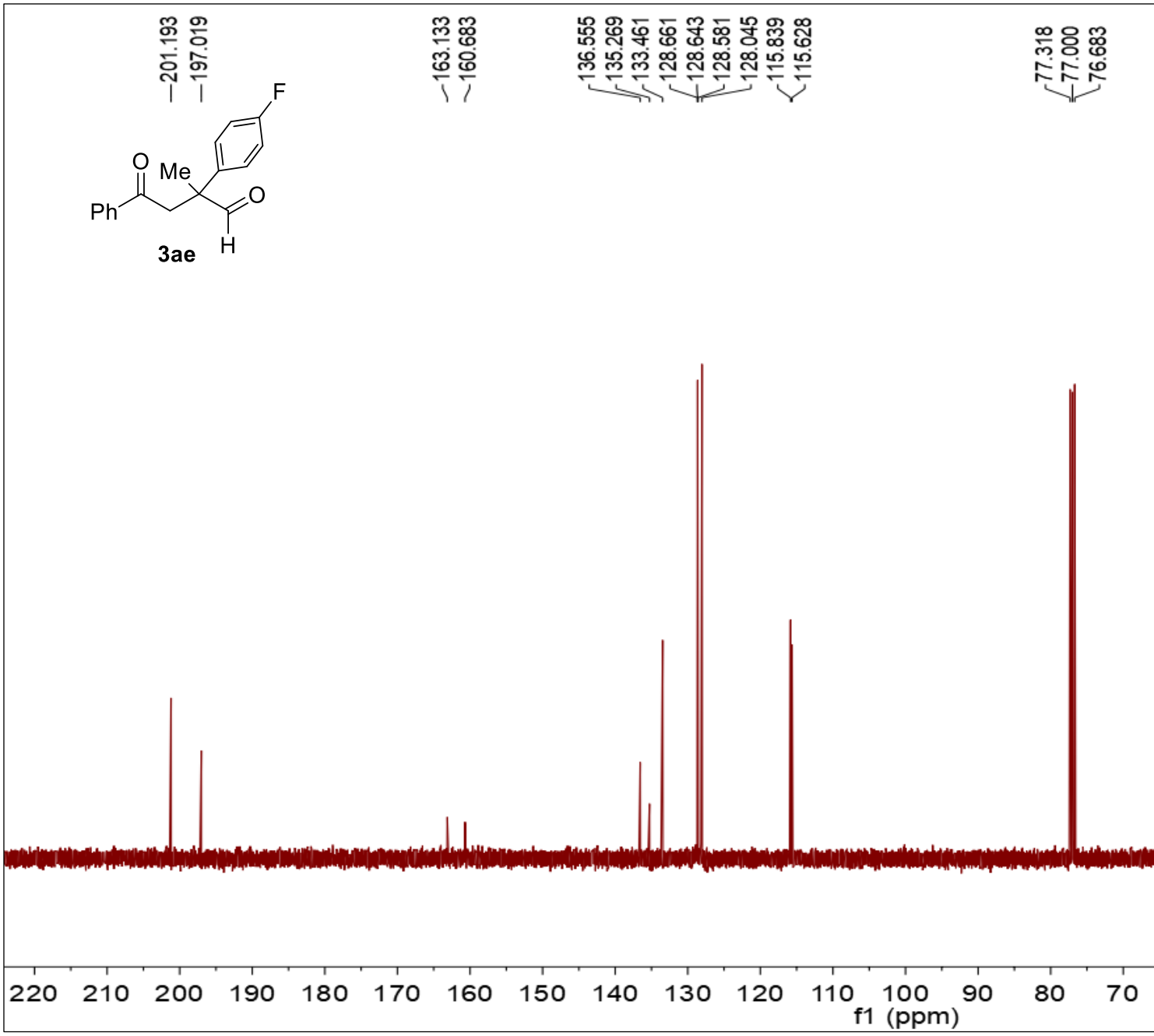
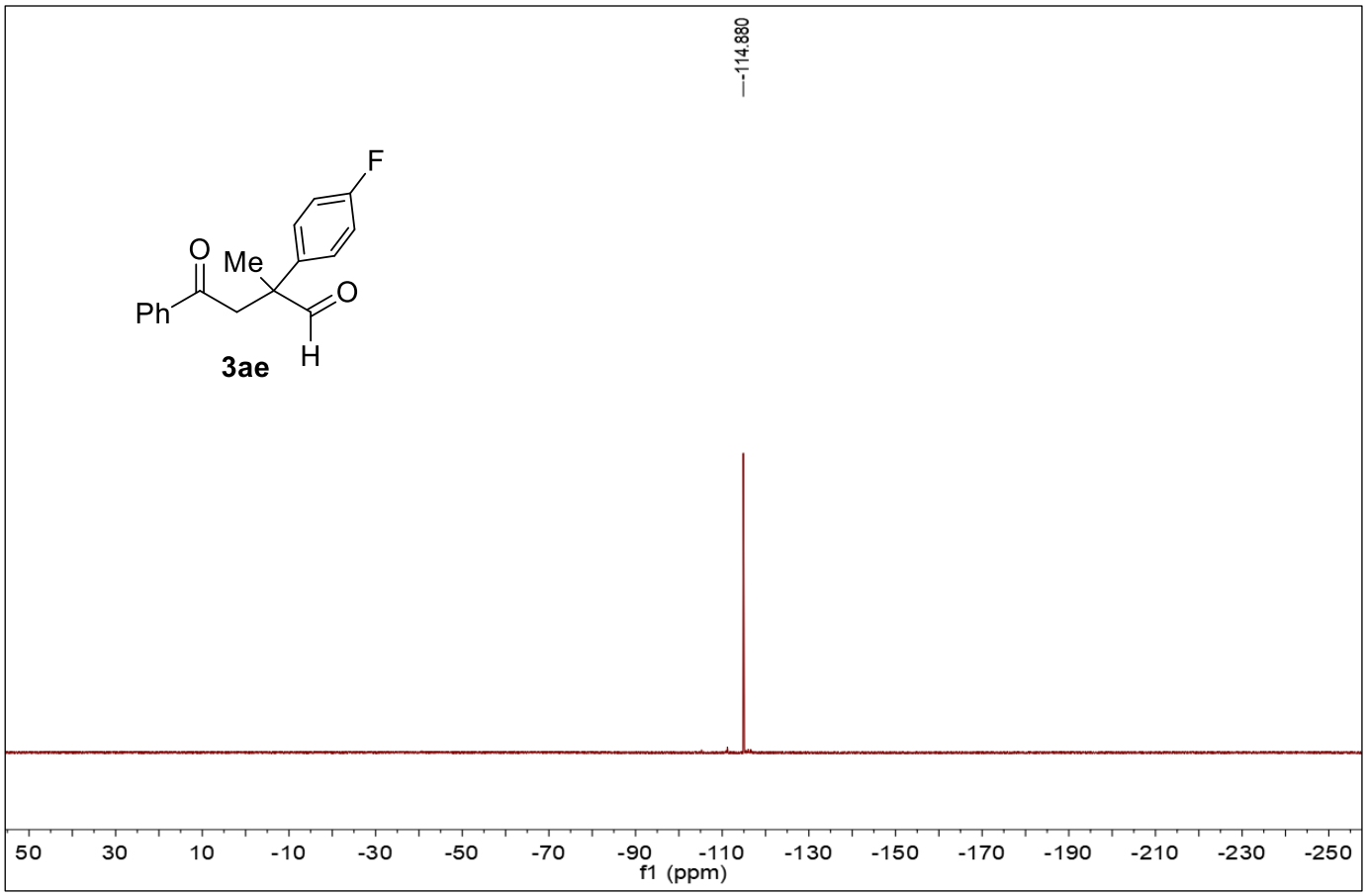
197.268
139.485
138.508
136.692
133.304
128.747
128.561
128.125
128.043
127.573
123.779
77.317
77.000
76.681
51.647
45.877
21.557
20.527
Me
O
Me
Ph
O
H
3ac
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

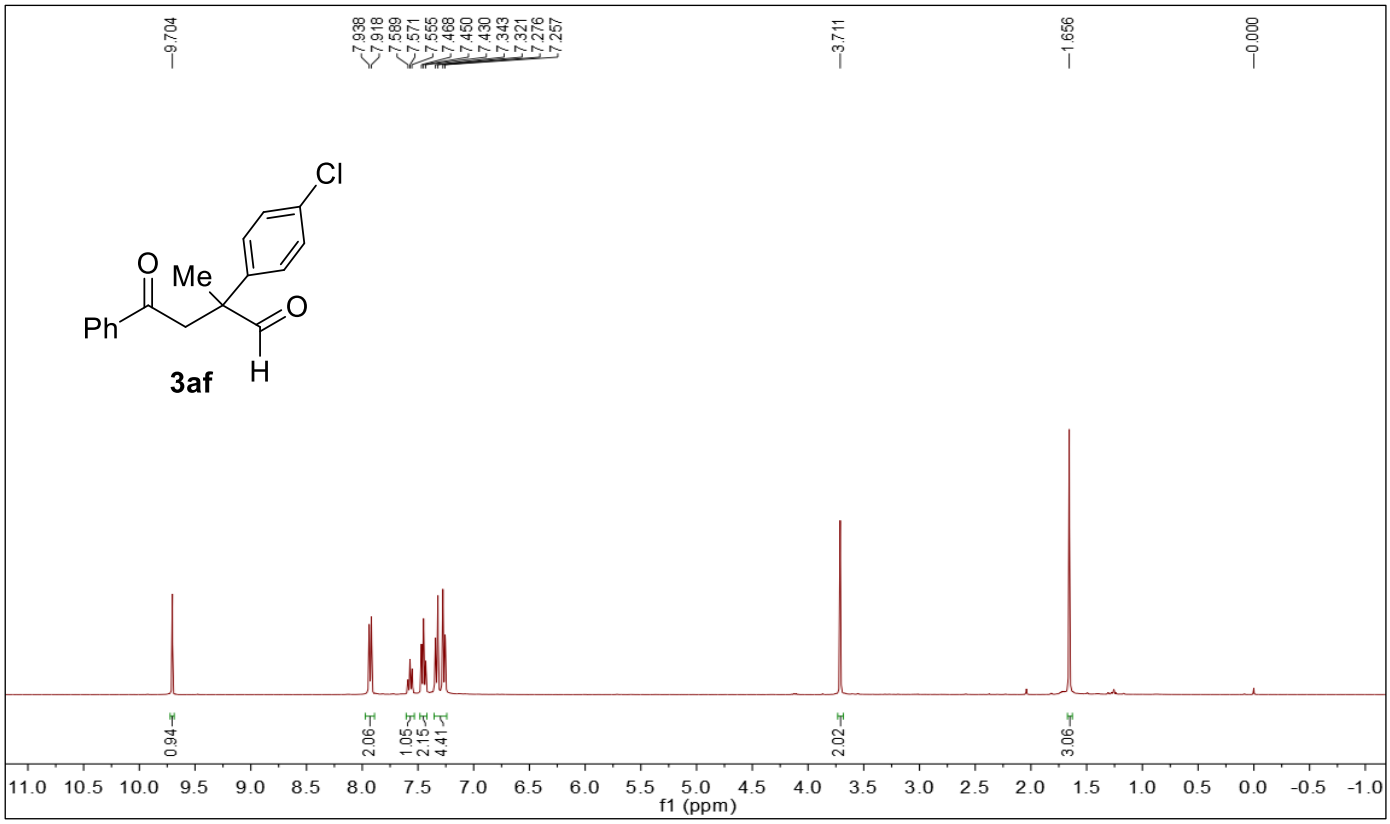

9.692
7.943
7.925
7.572
7.553
7.535
7.456
7.436
7.417
7.244
7.229
7.208
7.183
7.163
3.753
3.709
3.702
3.658
2.322
1.654
0.000
Me
O
Me
Ph
O
H
3ad
0.96
2.04
1.06
2.08
4.05
2.08
3.03
3.05
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

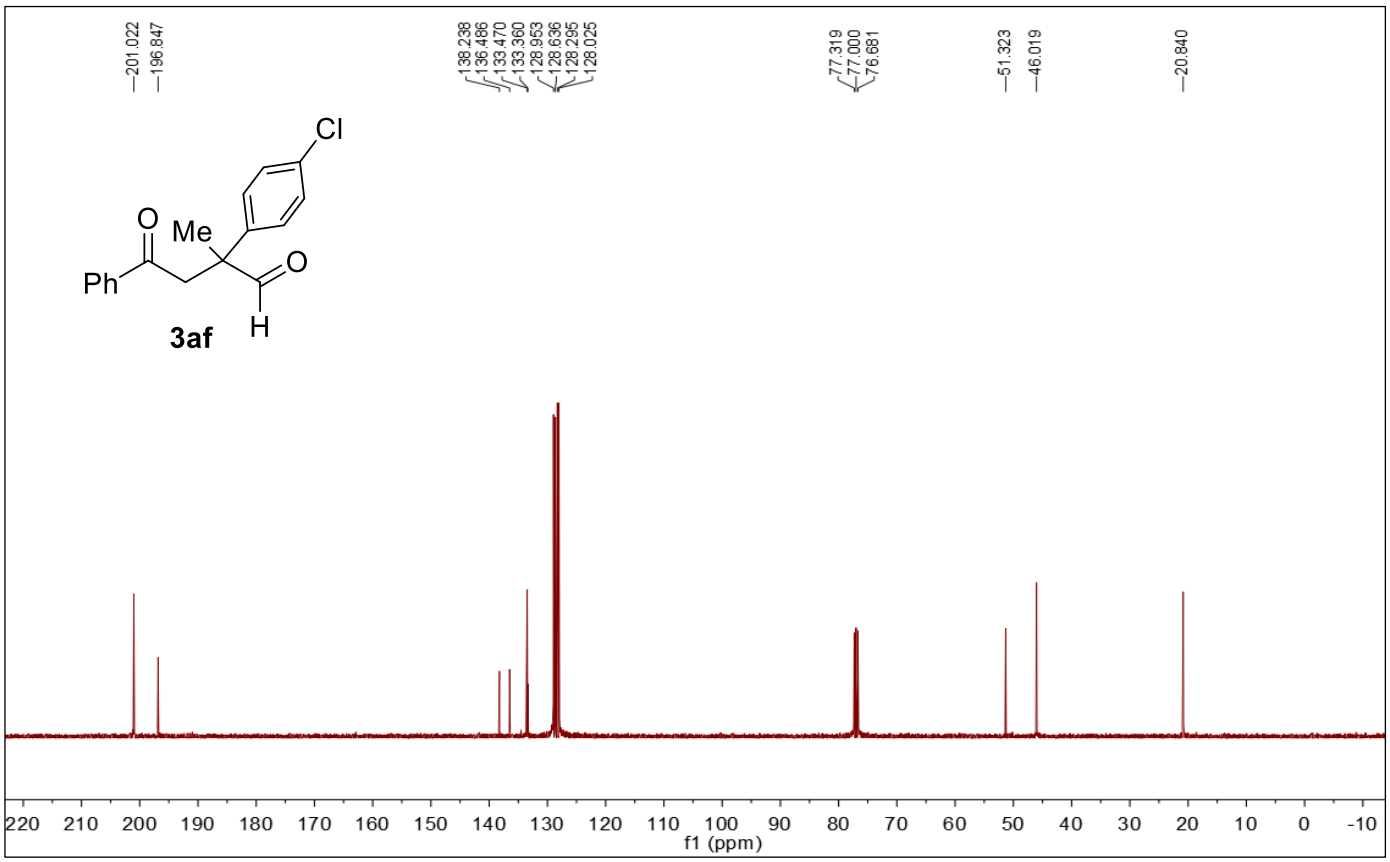

201.393
197.243
137.069
136.752
136.526
133.264
129.569
128.553
128.036
126.706
77.319
77.000
76.682
51.425
45.878
20.869
20.587
Me
O
Me
Ph
O
H
3ad
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

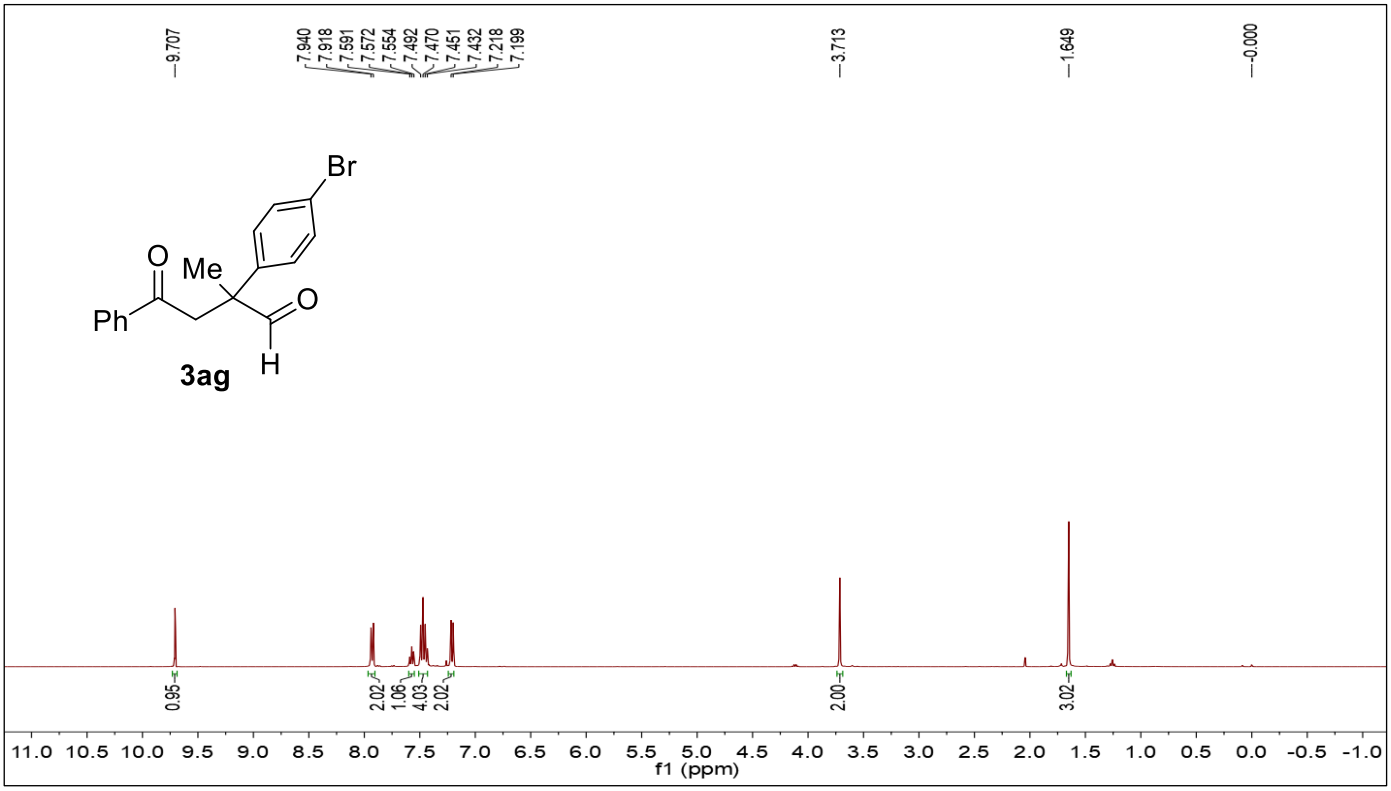

9.696
7.940
7.936
7.919
7.590
7.571
7.556
7.552
7.469
7.452
7.431
7.316
7.302
7.294
7.289
7.261
7.078
7.075
7.056
7.035
3.710
1.664
0.077
0.000
F
O
Me
Ph
O
H
3ae
0.92
2.07
1.06
2.21
2.10
2.13
2.00
3.13
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

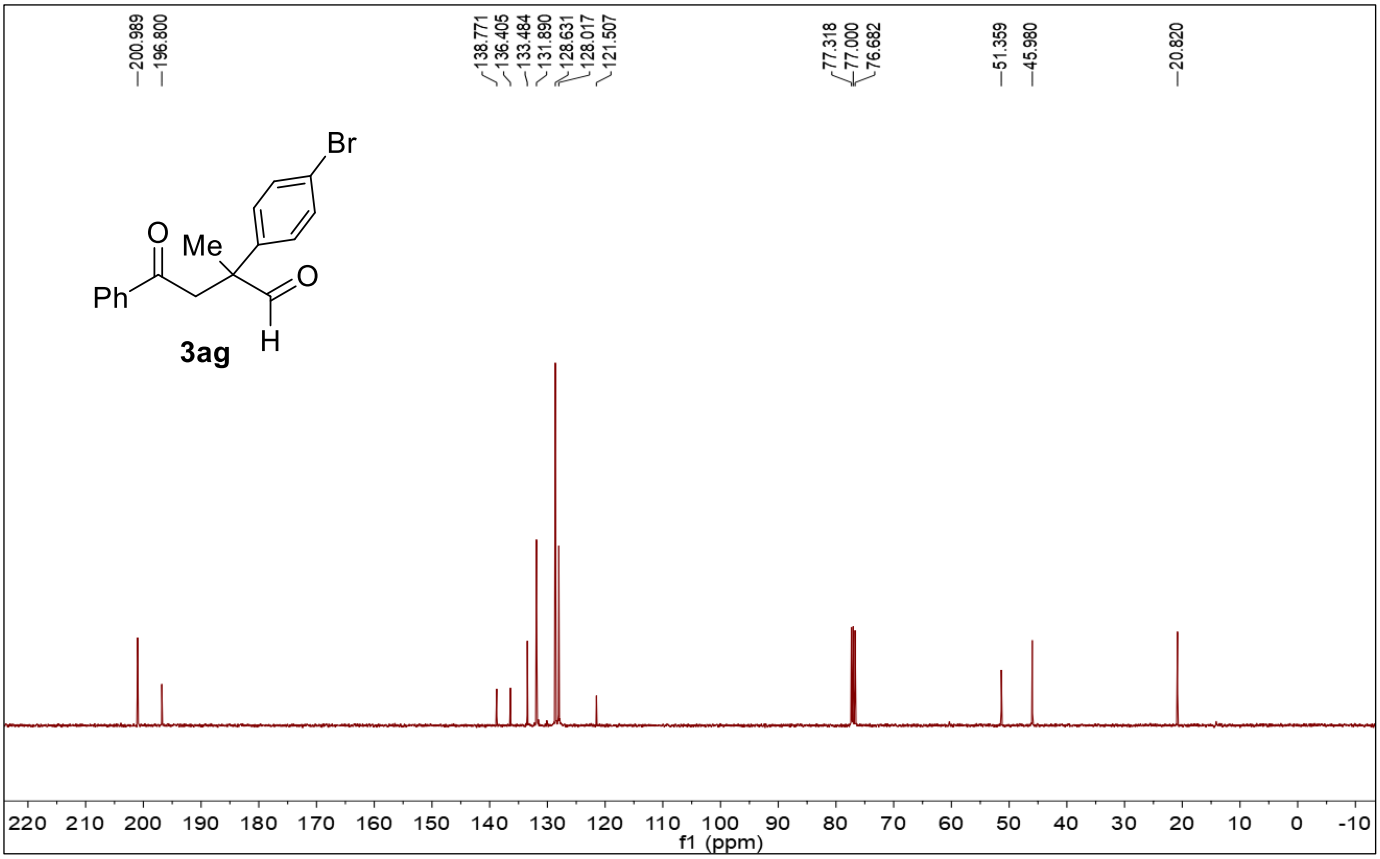

201.193
197.019
163.133
160.683
136.555
135.269
133.461
128.661
128.643
128.581
128.045
115.839
115.628
77.318
77.000
76.683
F
O
Me
Ph
O
H
3ae
220
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
f1 (ppm)

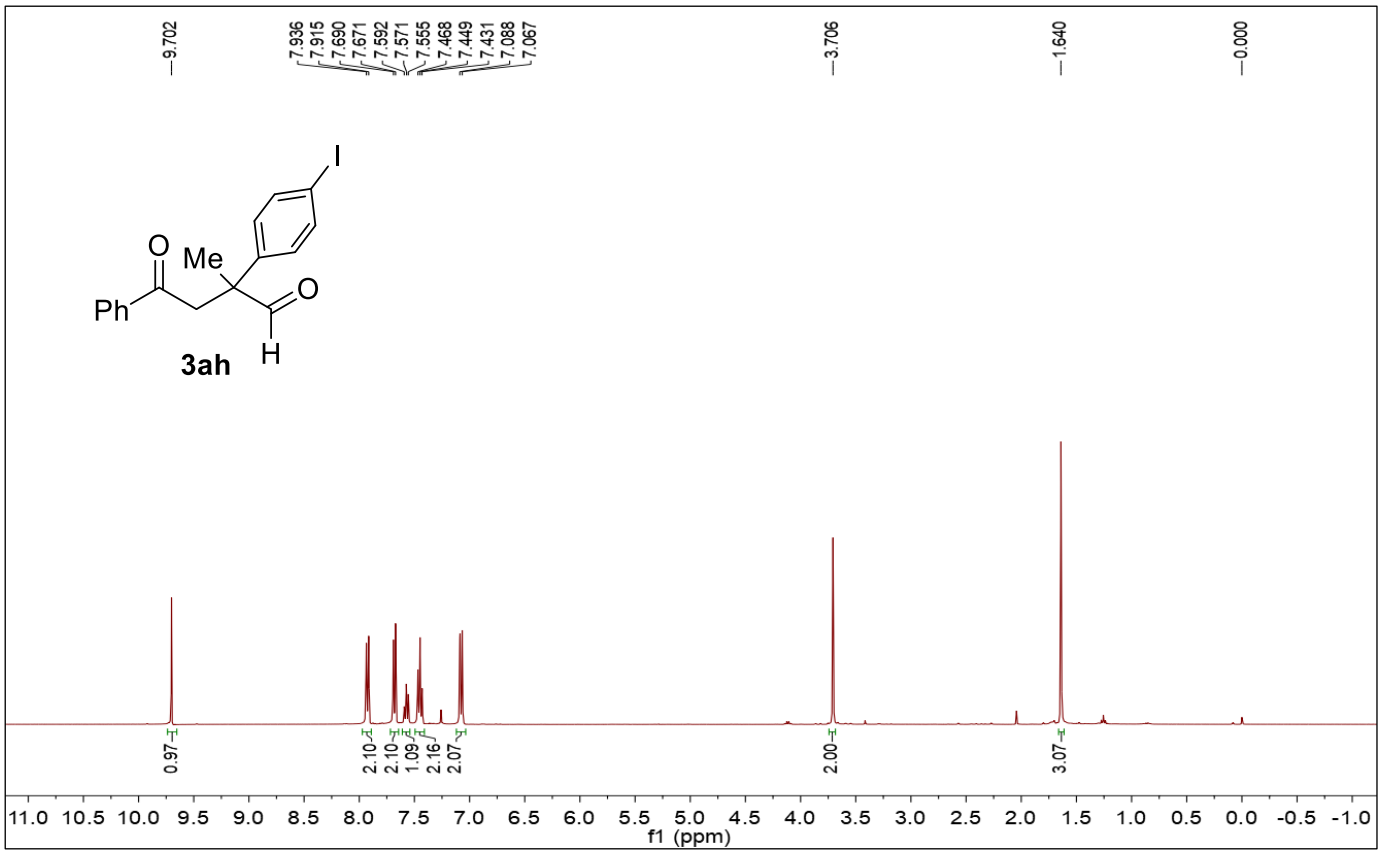

-114.880
F
O
Me
Ph
O
H
3ae
50
30
10
-10
-30
-50
-70
-90
-110
-130
-150
-170
-190
-210
-230
-250
f1 (ppm)


9.704
7.938
7.918
7.589
7.571
7.555
7.468
7.450
7.430
7.343
7.321
7.276
7.257
3.711
1.656
0.000
Cl
O
Me
Ph
O
H
3af
0.94
2.06
1.05
2.15
4.41
2.02
3.06
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


201.022
196.847
138.238
136.486
133.470
133.360
128.953
128.636
128.295
128.025
77.319
77.000
76.681
51.323
46.019
20.840
Cl
O
Me
Ph
O
H
3af
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


9.707
7.940
7.918
7.591
7.572
7.554
7.492
7.470
7.451
7.432
7.218
7.199
3.713
1.649
0.000
Br
O
Me
Ph
O
H
3ag
0.95
2.02
1.06
4.03
2.02
2.00
3.02
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


200.989
196.800
138.771
136.405
133.484
131.890
128.631
128.017
121.507
77.318
77.000
76.682
51.359
45.980
20.820
Br
O
Me
Ph
O
H
3ag
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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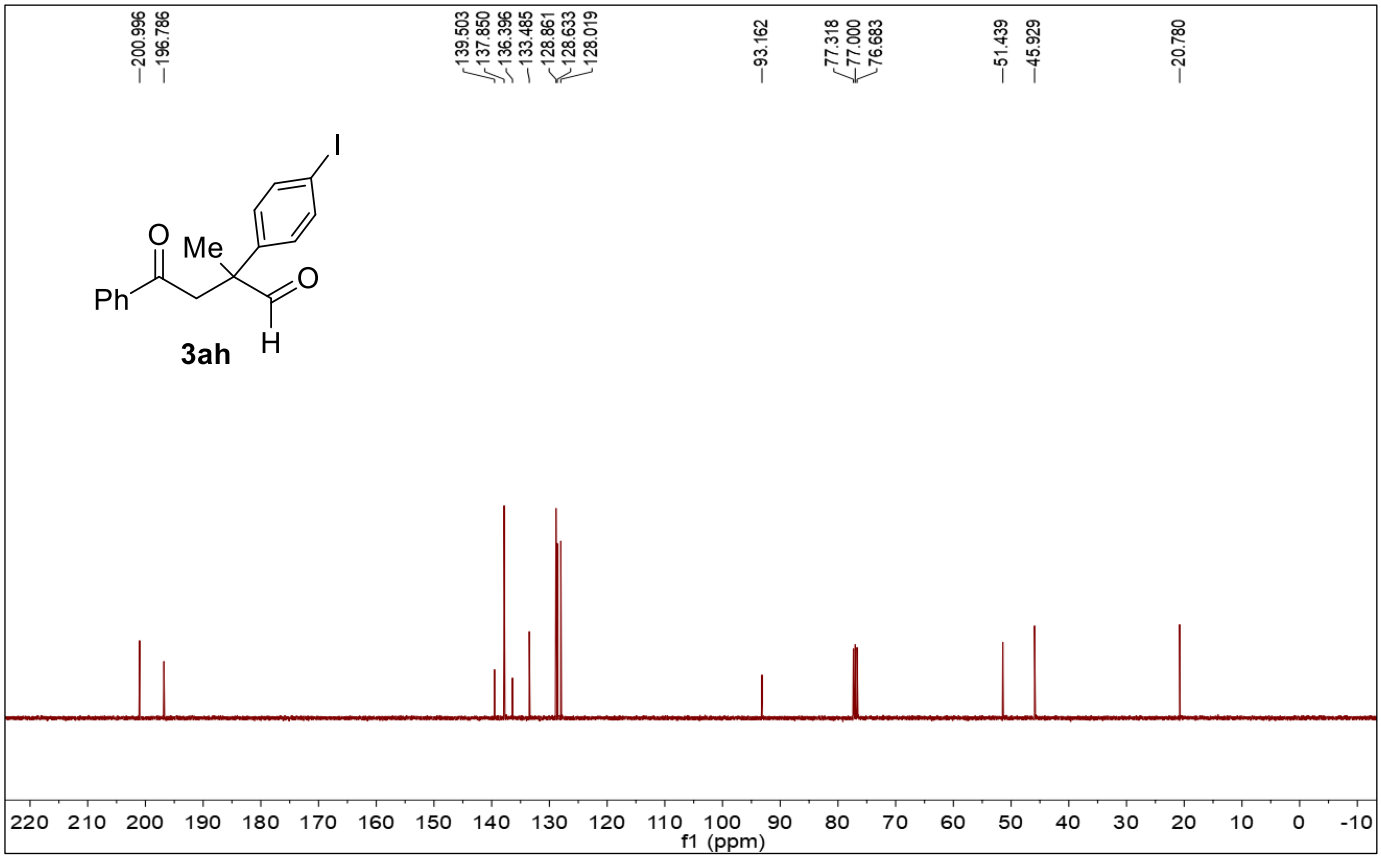
7.915
7.690
7.671
7.592
7.571
7.555
7.468
7.449
7.431
7.088
7.067
3.706
1.640
0.000
I
O
Me
Ph
O
H
3ah
0.97
2.10
2.10
1.09
2.16
2.07
2.00
3.07
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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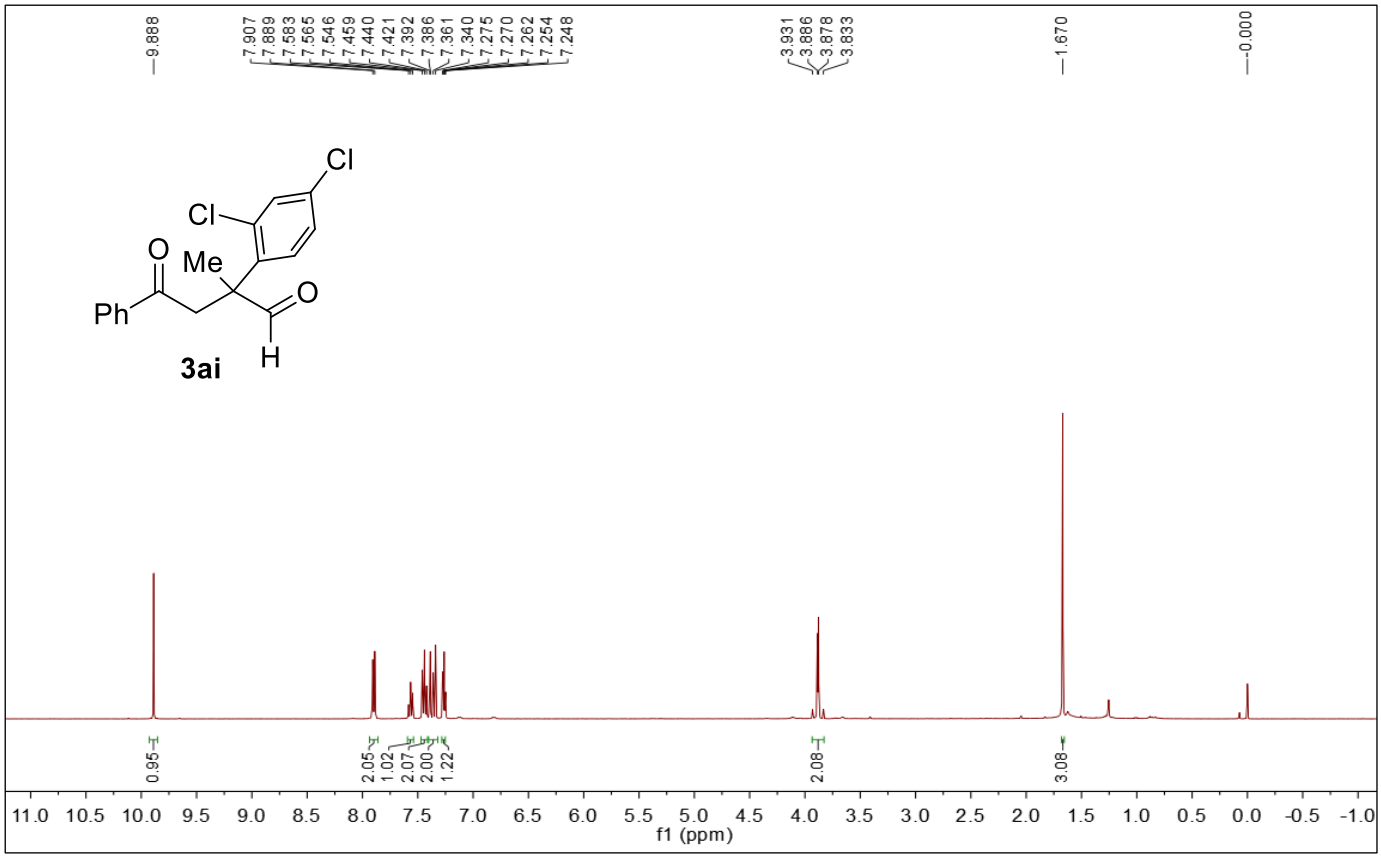
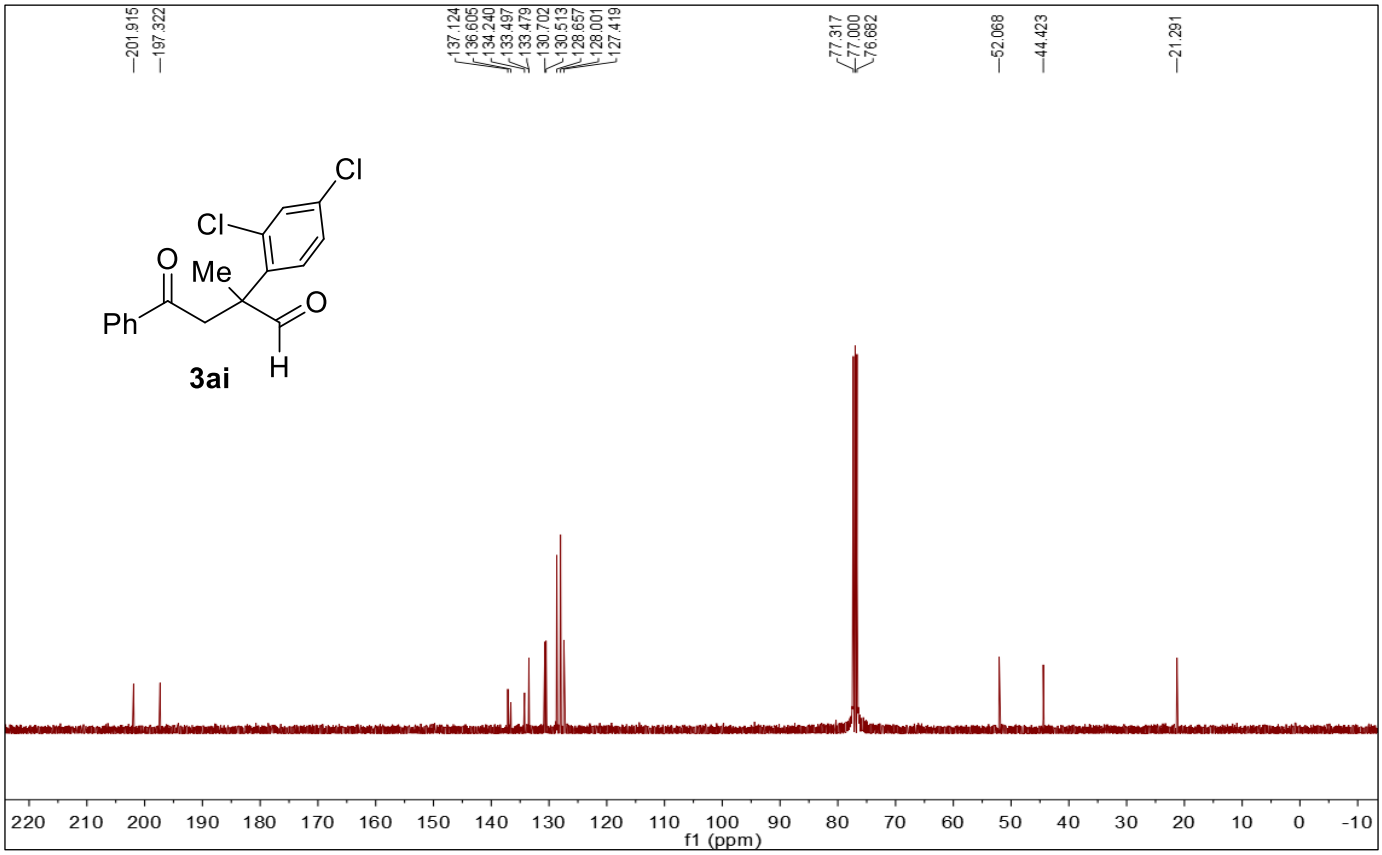
137.850
136.396
133.485
128.861
128.633
128.019
93.162
77.318
77.000
76.683
51.439
45.929
20.780
I
O
Me
Ph
O
H
3ah
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


9.888
7.907
7.889
7.583
7.565
7.546
7.459
7.440
7.421
7.392
7.386
7.361
7.340
7.275
7.270
7.262
7.254
7.248
3.931
3.886
3.878
3.833
1.670
0.000
Cl
Cl
O
Me
Ph
O
H
3ai
0.95
2.05
1.02
2.07
2.00
1.22
2.08
3.08
f1 (ppm)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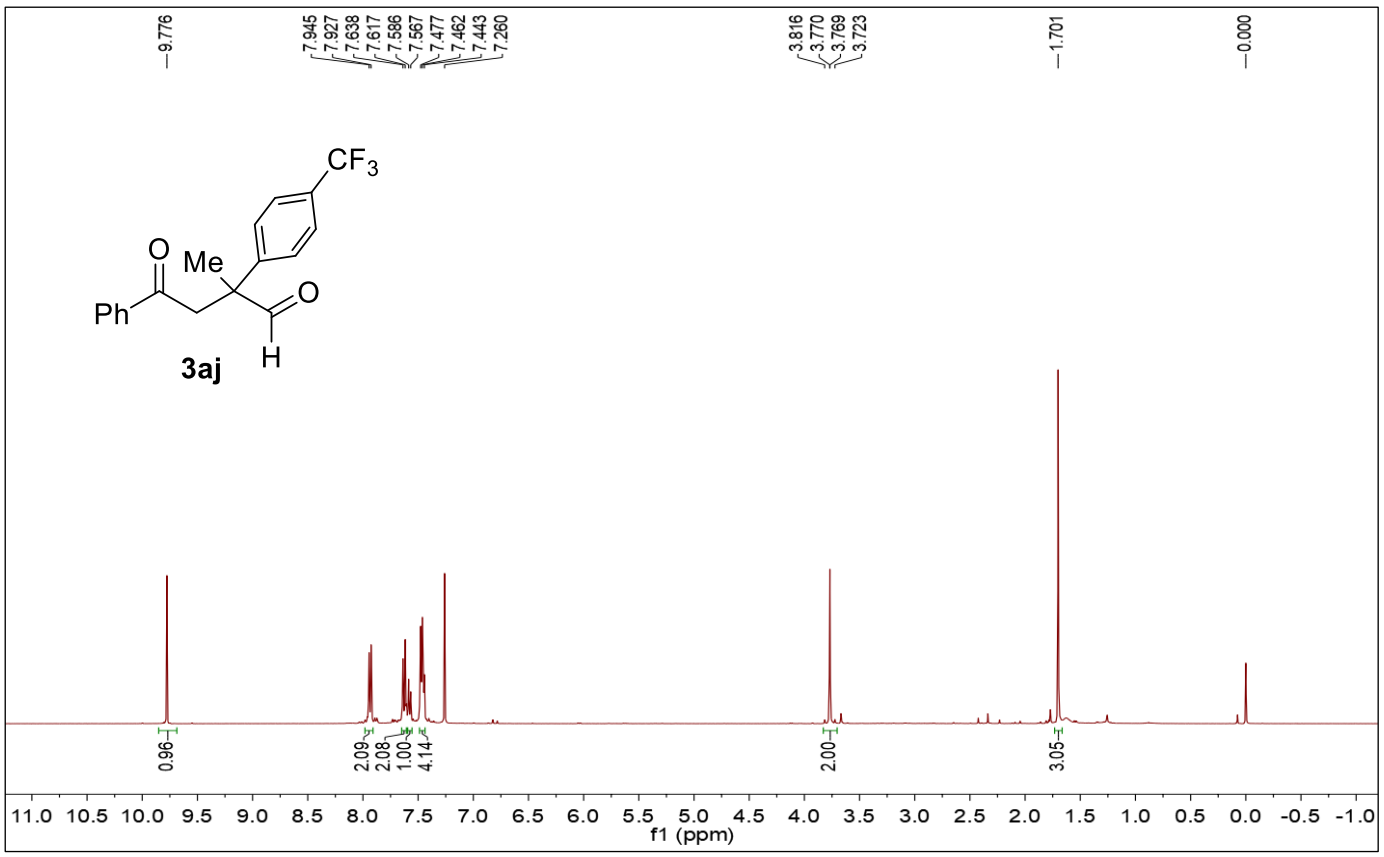
136.605
134.240
133.497
133.479
130.702
130.513
128.657
128.001
127.419
77.317
77.000
76.682
52.068
44.423
21.291
Cl
Cl
O
Me
Ph
O
H
3ai
f1 (ppm)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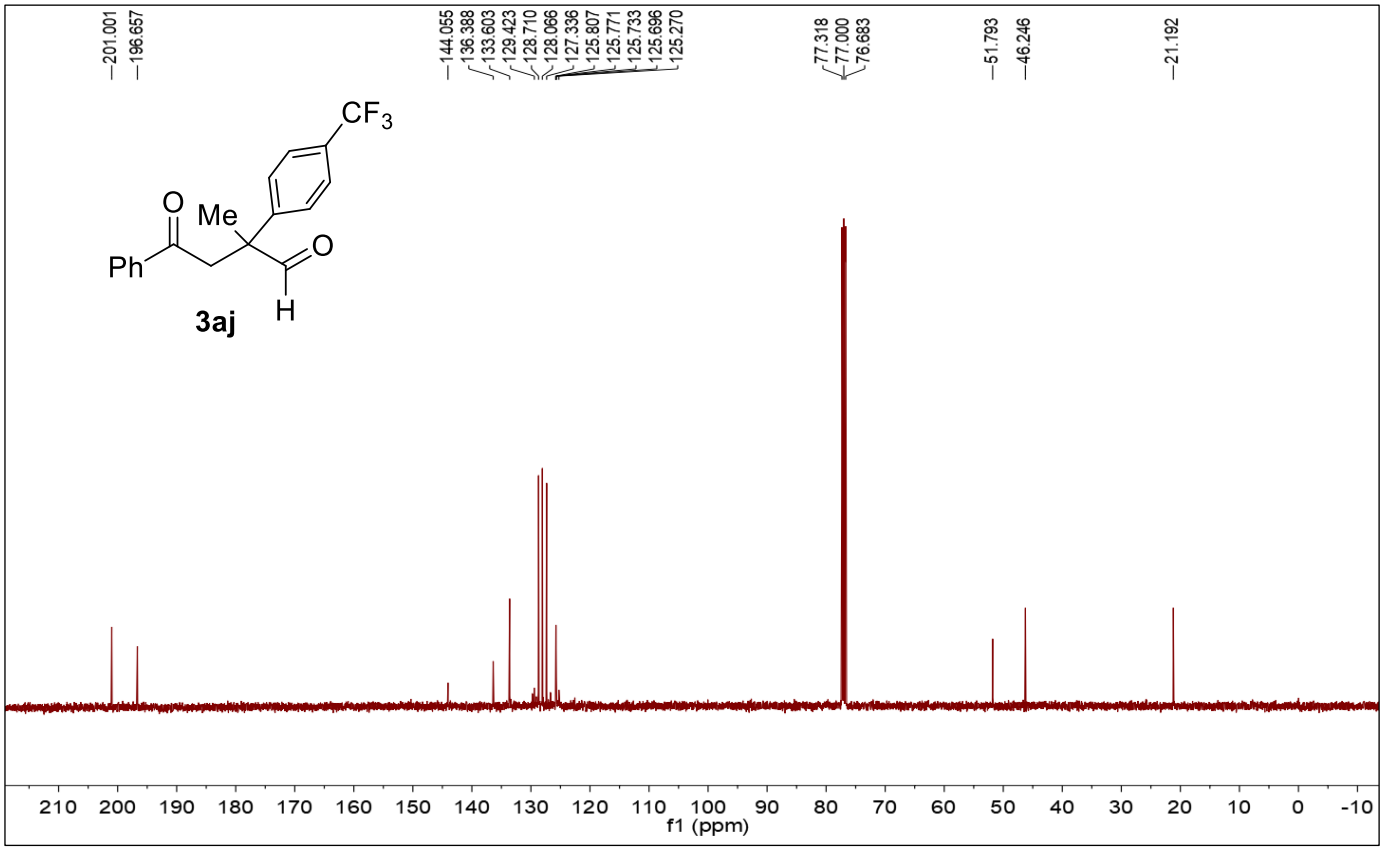
7.586
7.567
7.477
7.462
7.443
7.260
3.816
3.770
3.769
3.723
1.701
0.000
CF3
O
Me
Ph
O
H
3aj
0.96
2.09
2.08
1.00
4.14
2.00
3.05
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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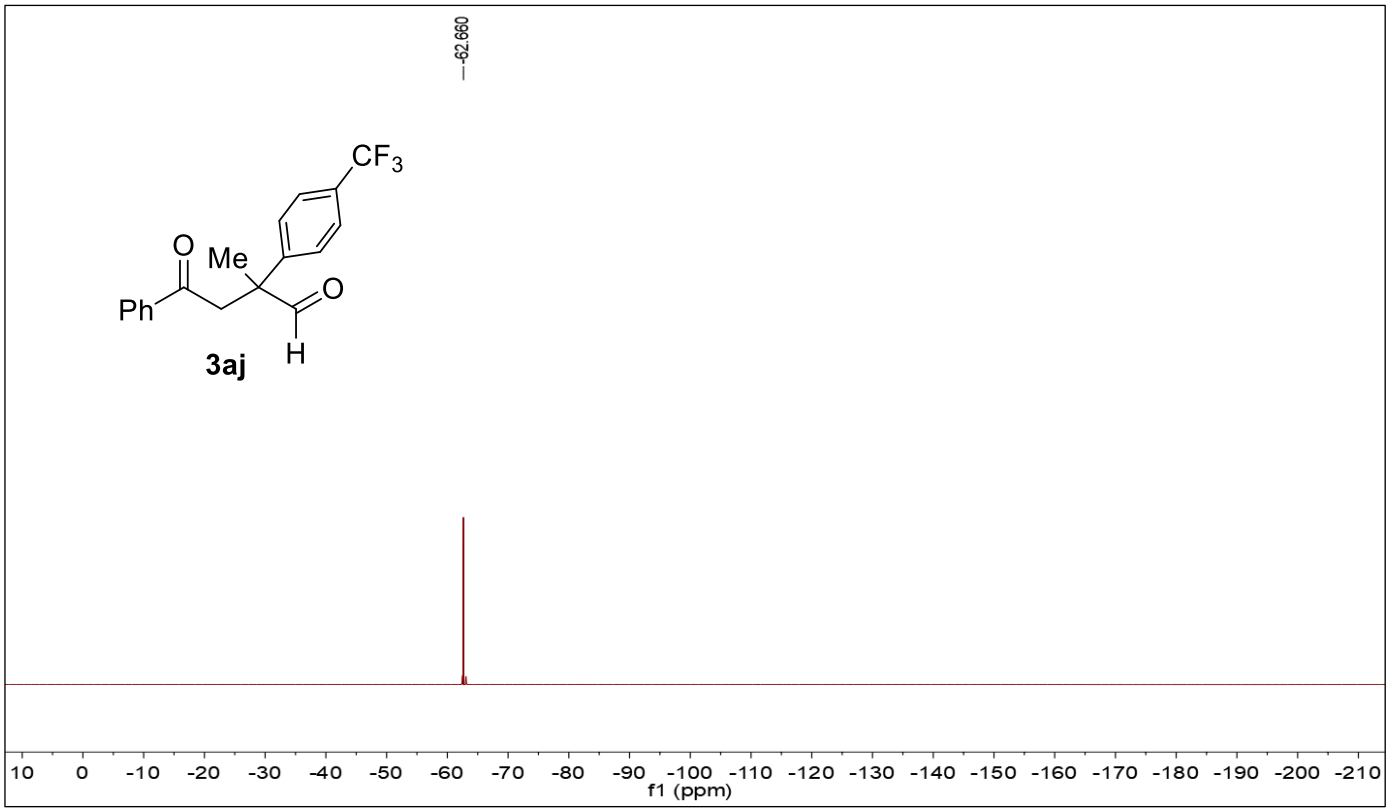
129.423
128.710
128.066
127.336
125.807
125.771
125.733
125.696
125.270
77.318
77.000
76.683
51.793
46.246
21.192
CF3
O
Me
Ph
O
H
3aj
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


-62.660
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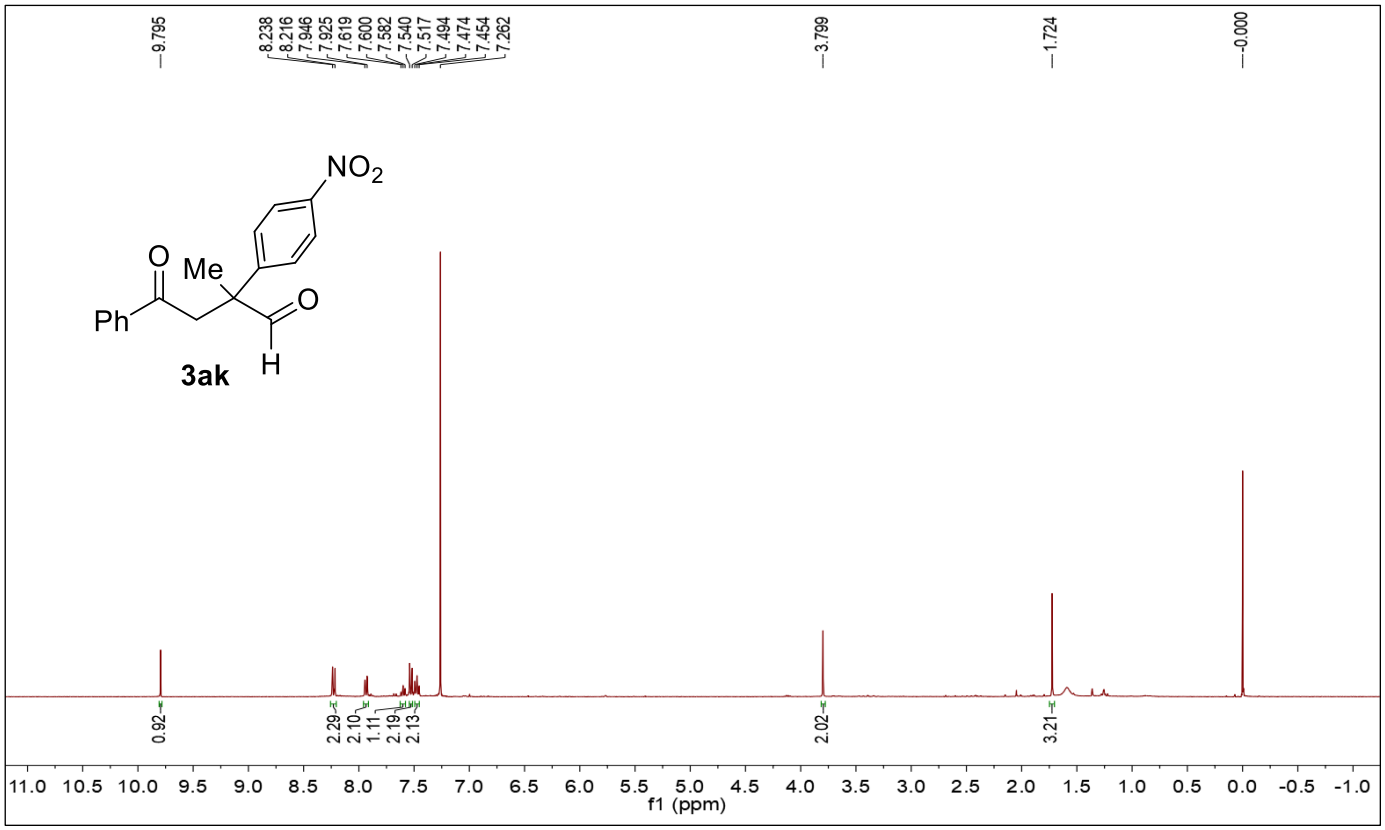
Me
Ph
O
H
3aj
10 0 -10 -20 -30 -40 -50 -60 -70 -80 -90 -100 -110 -120 -130 -140 -150 -160 -170 -180 -190 -200 -210
f1 (ppm)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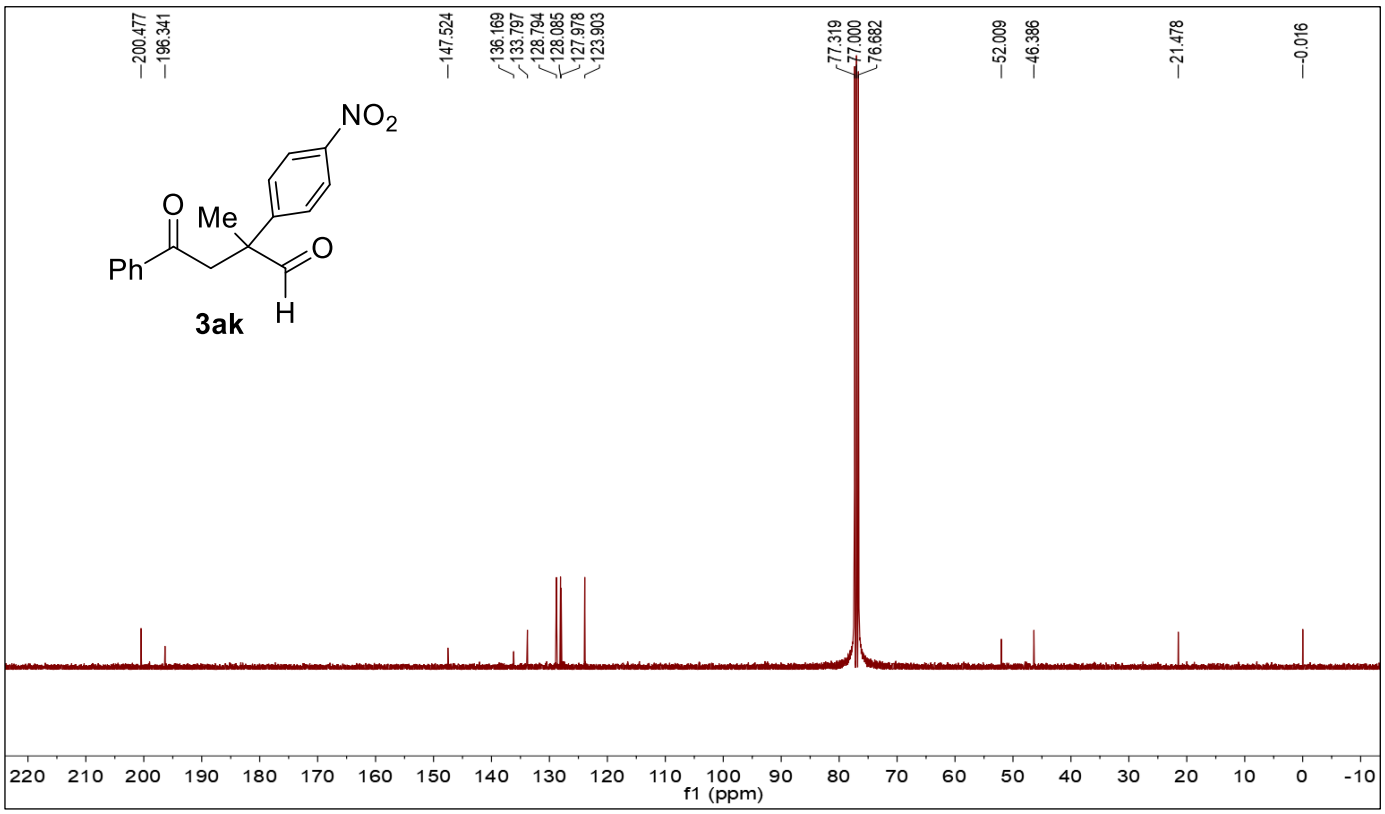
7.619
7.600
7.582
7.540
7.517
7.494
7.474
7.454
7.262
3.799
1.724
0.000
NO2
O
Me
Ph
O
H
3ak
0.92
2.29
2.10
1.11
2.19
2.13
2.02
3.21
f1 (ppm)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136.169
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128.794
128.085
127.978
123.903
77.319
77.000
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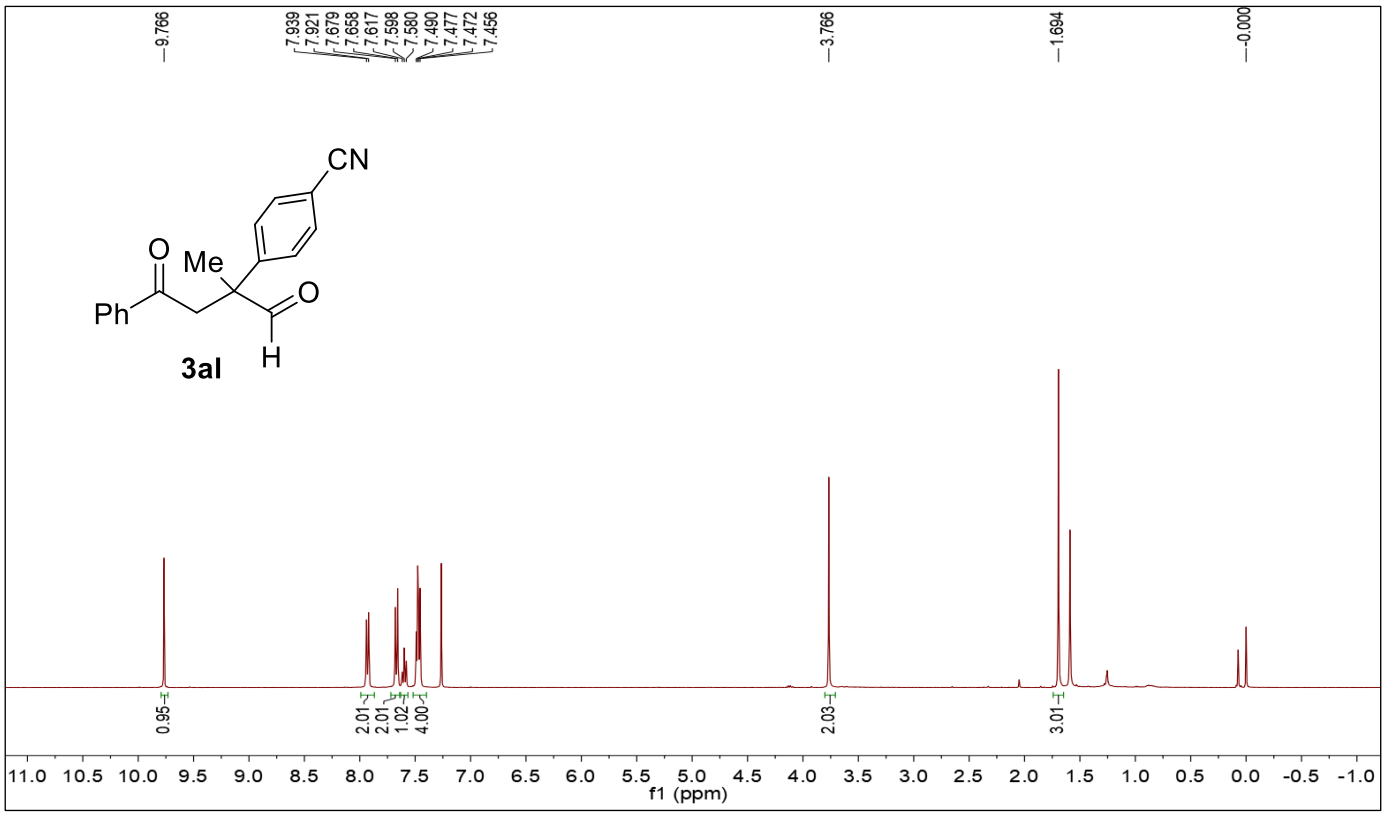
52.009
46.386
21.478
0.016
NO2
O
Me
Ph
O
H
3ak
f1 (ppm)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7.679
7.658
7.617
7.598
7.580
7.490
7.477
7.472
7.456
3.766
1.694
0.000
CN
O
Me
Ph
O
H
3al
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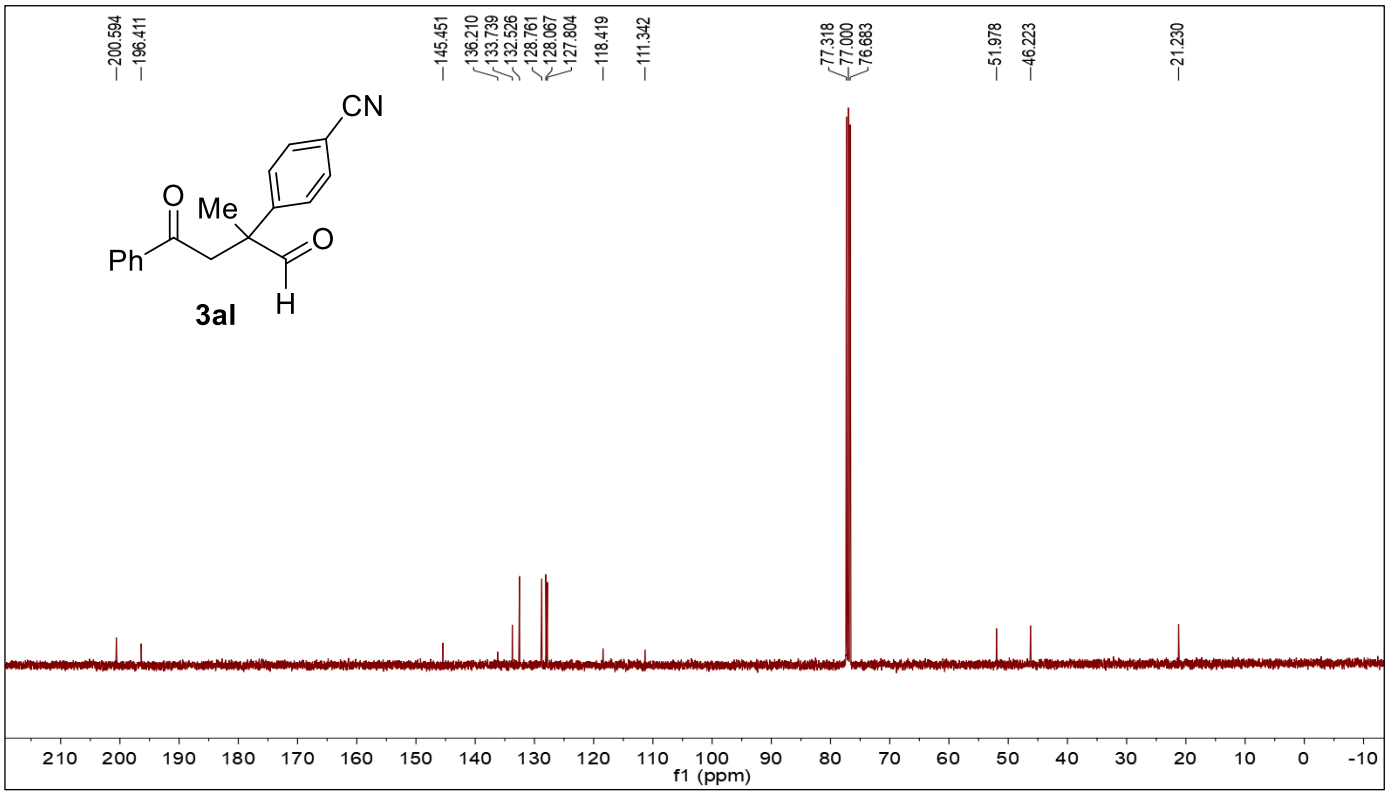
2.01
2.01
1.02
4.00
2.03
3.01
f1 (ppm)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136.210
133.739
132.526
128.761
128.067
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111.342
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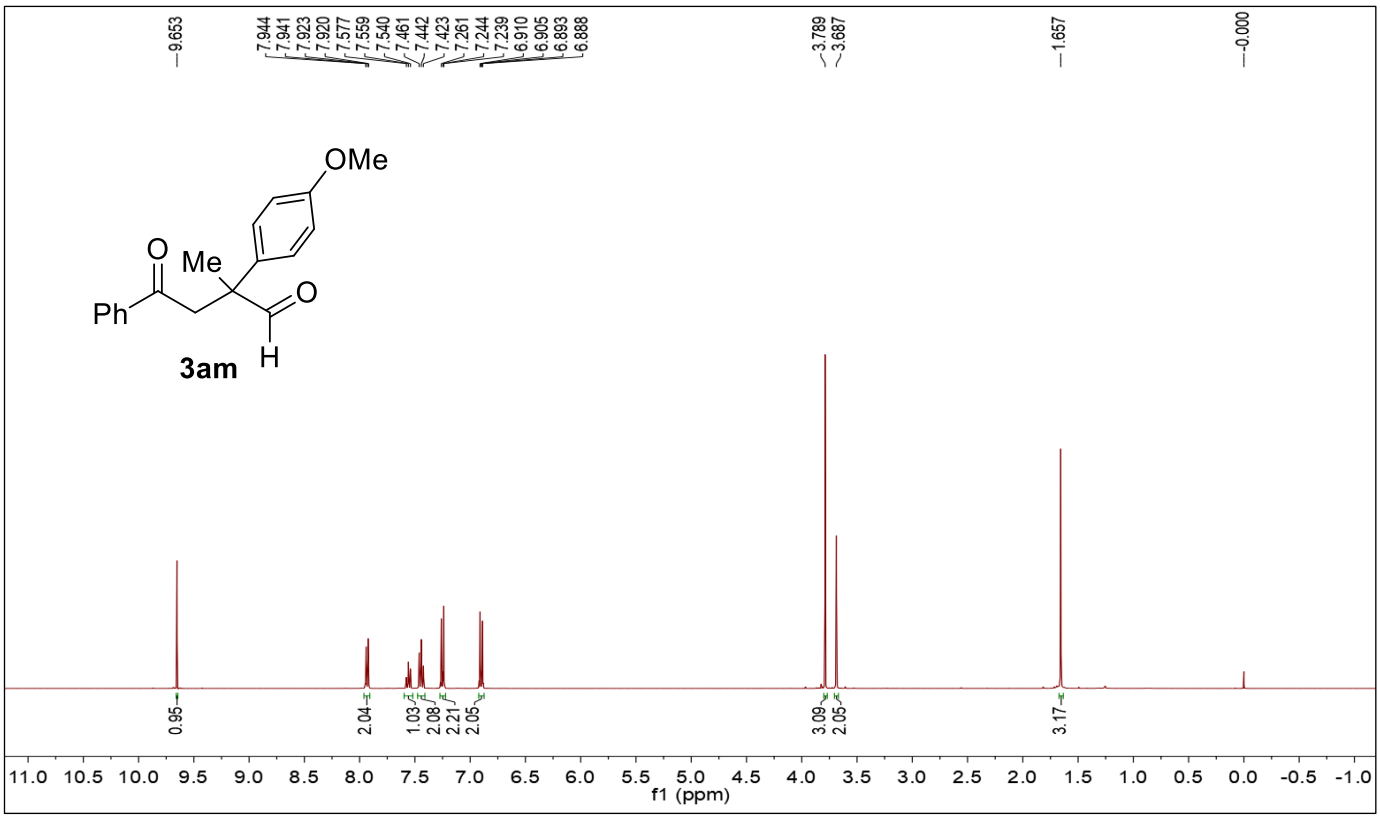
76.683
51.978
46.223
21.230
CN
O
Me
Ph
O
H
3al
f1 (ppm)


9.653
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7.920
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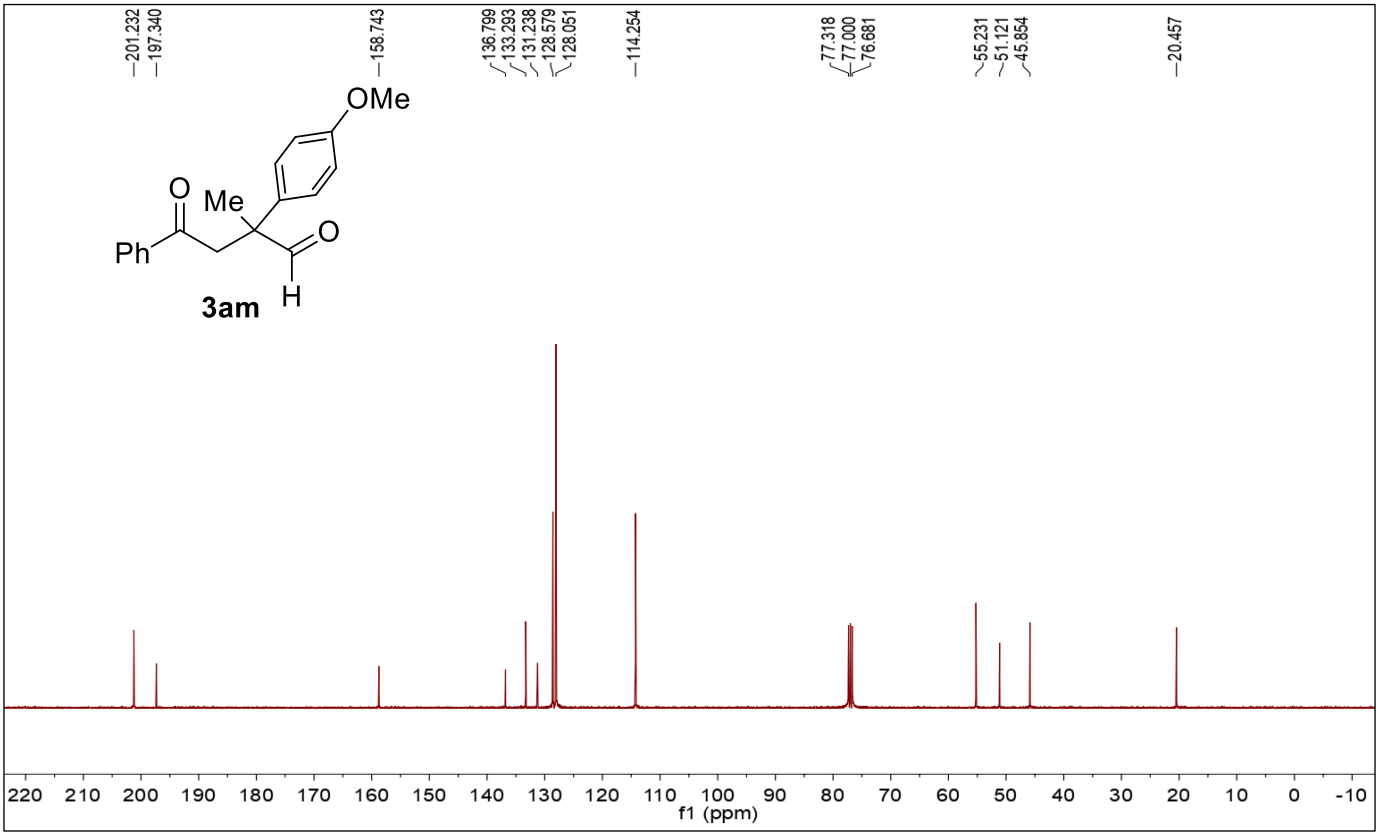
7.559
7.540
7.461
7.442
7.423
7.261
7.244
7.239
6.910
6.905
6.893
6.888
3.789
3.687
1.657
0.000
OMe
O
Me
Ph
O
H
3am
0.95
2.04
1.03
2.08
2.21
2.05
3.09
2.05
3.17
f1 (ppm)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158.743
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133.293
131.238
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128.051
114.254
77.318
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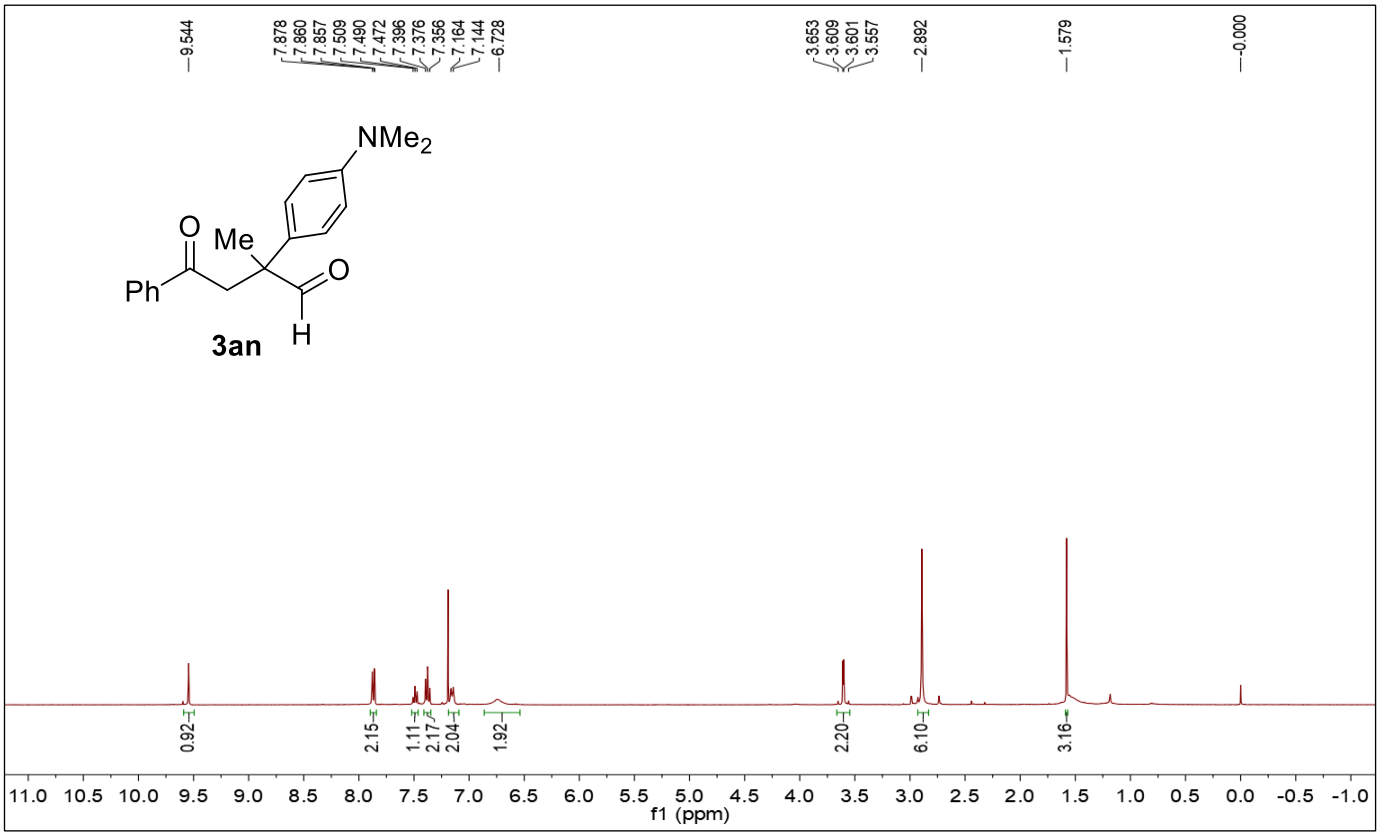
76.681
55.231
51.121
45.854
20.457
OMe
O
Me
Ph
O
H
3am
f1 (ppm)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7.860
7.857
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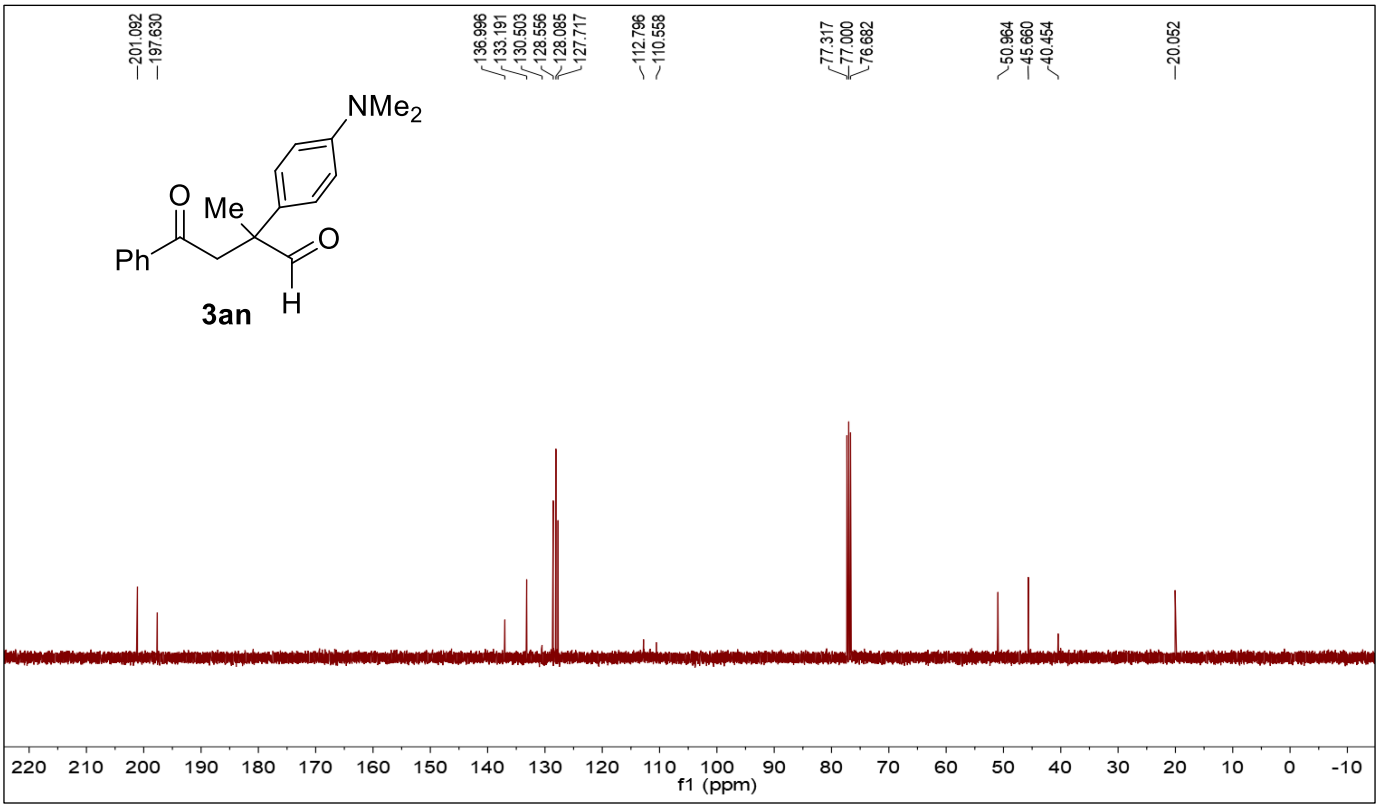
7.490
7.472
7.396
7.376
7.356
7.164
7.144
6.728
3.653
3.609
3.601
3.557
2.892
1.579
0.000
NMe2
O
Me
Ph
O
H
3an
0.92
2.15
1.11
2.17
2.04
1.92
2.20
6.10
3.16
f1 (ppm)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136.996
133.191
130.503
128.556
128.085
127.717
112.796
110.558
77.317
77.000
76.682
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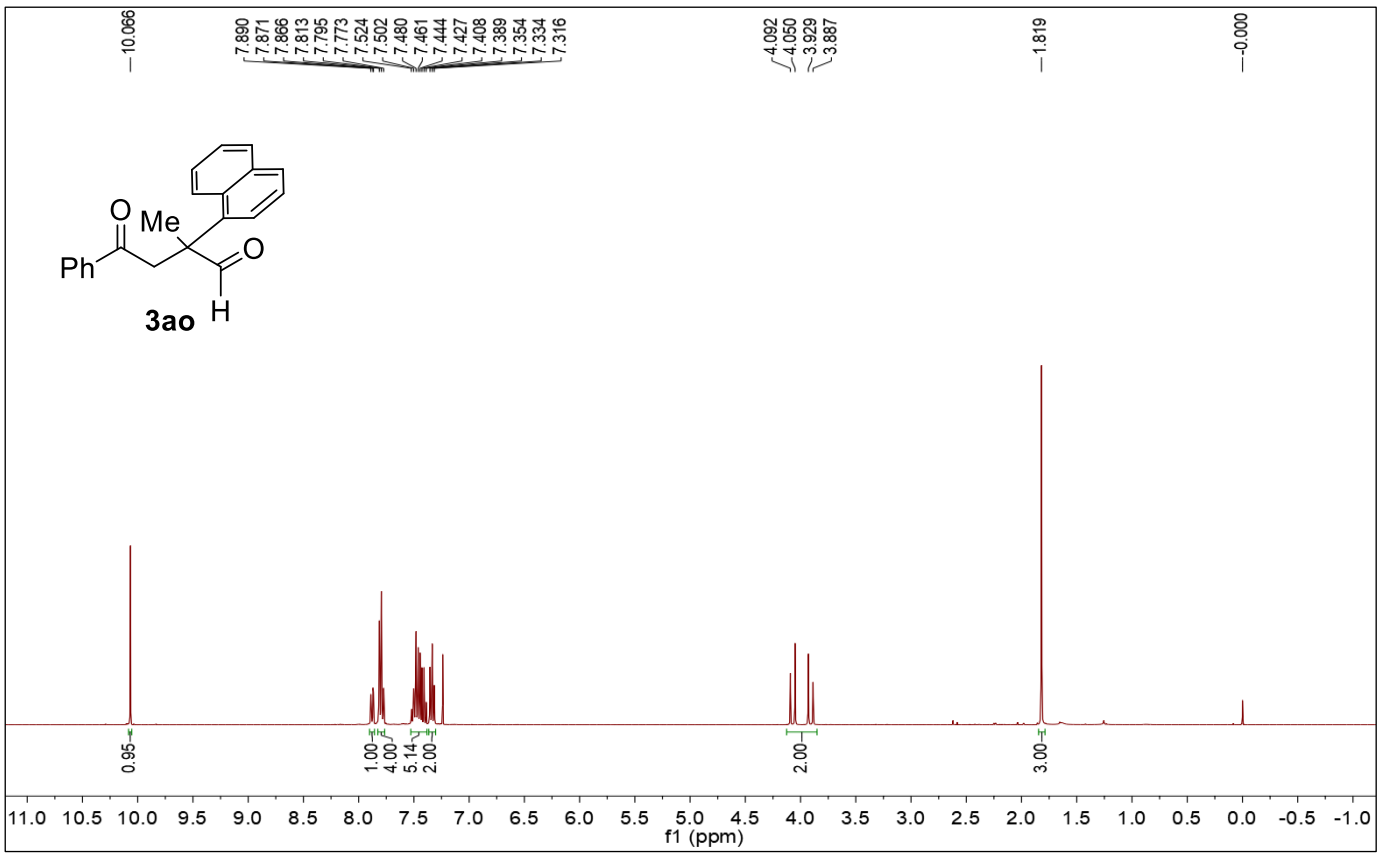
45.660
40.454
20.052
NMe2
O
Me
Ph
O
H
3an
f1 (ppm)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7.871
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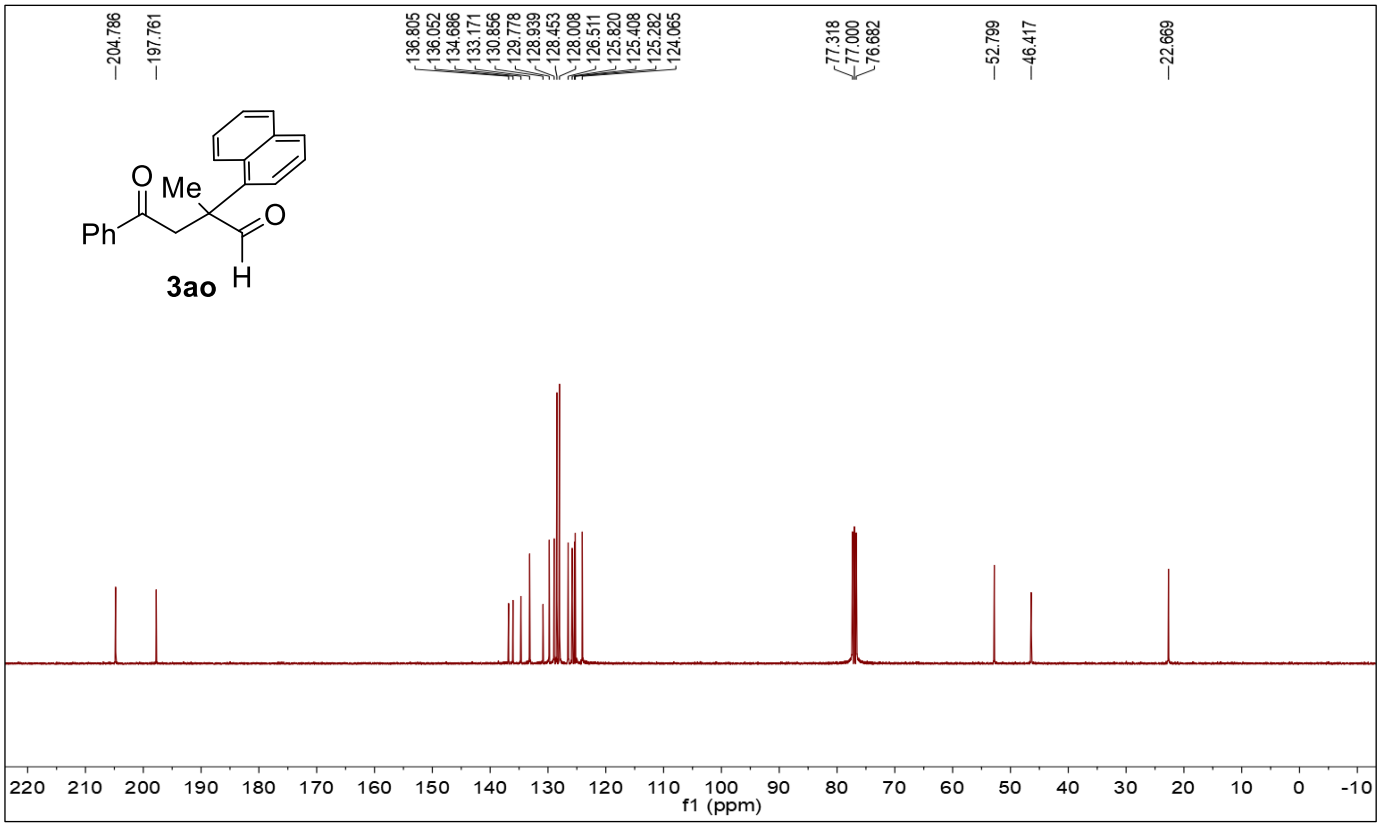
7.773
7.524
7.502
7.480
7.461
7.444
7.427
7.408
7.389
7.354
7.334
7.316
4.092
4.050
3.929
3.887
1.819
0.000
O
Me
Ph
O
H
3ao
0.95
1.00
4.00
5.14
2.00
2.00
3.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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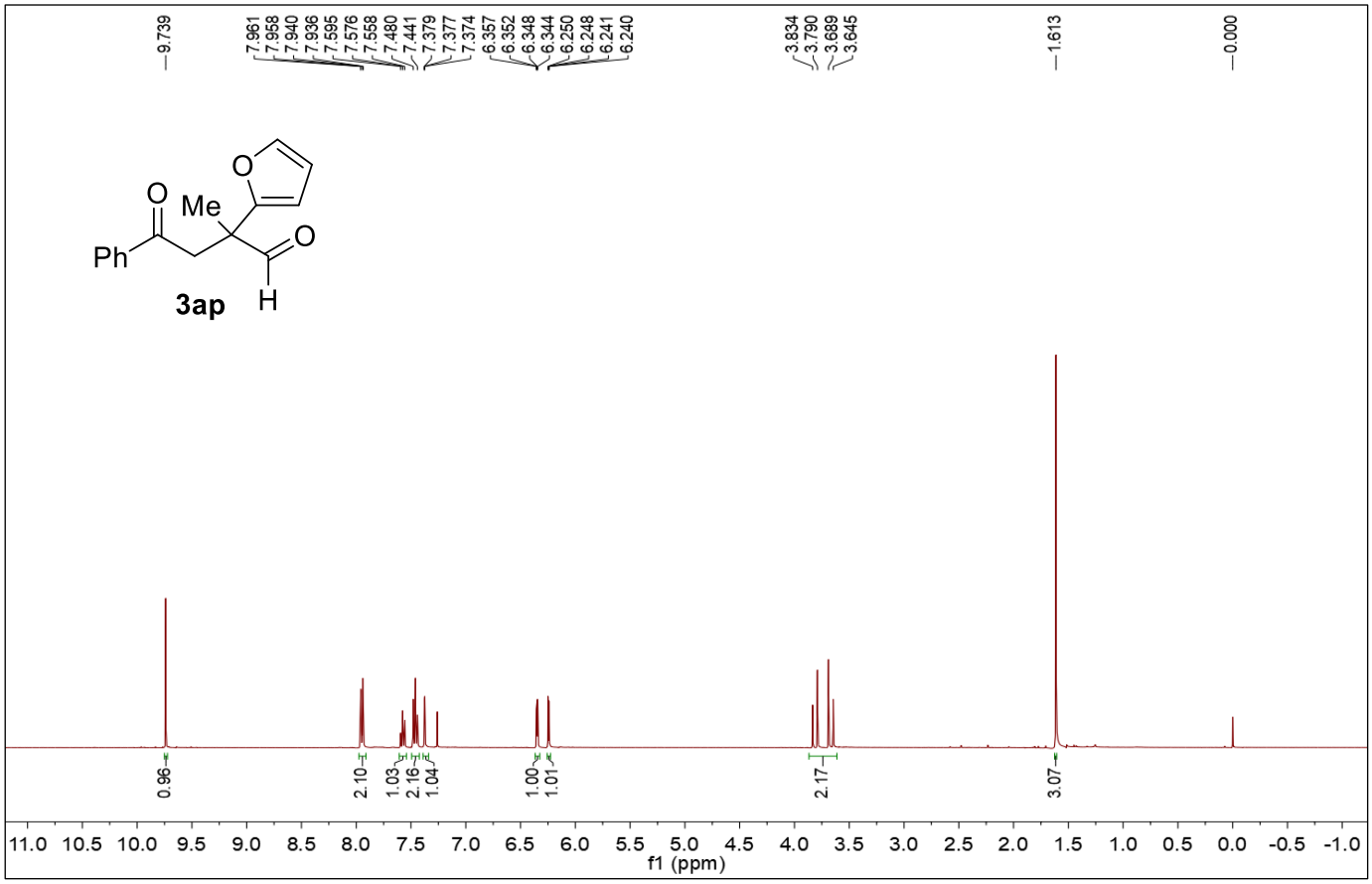
197.761
136.805
136.052
134.686
133.171
130.856
129.778
128.939
128.453
128.008
126.511
125.820
125.408
125.282
124.065
77.318
77.000
76.682
52.799
46.417
22.669
O
Me
Ph
O
H
3ao
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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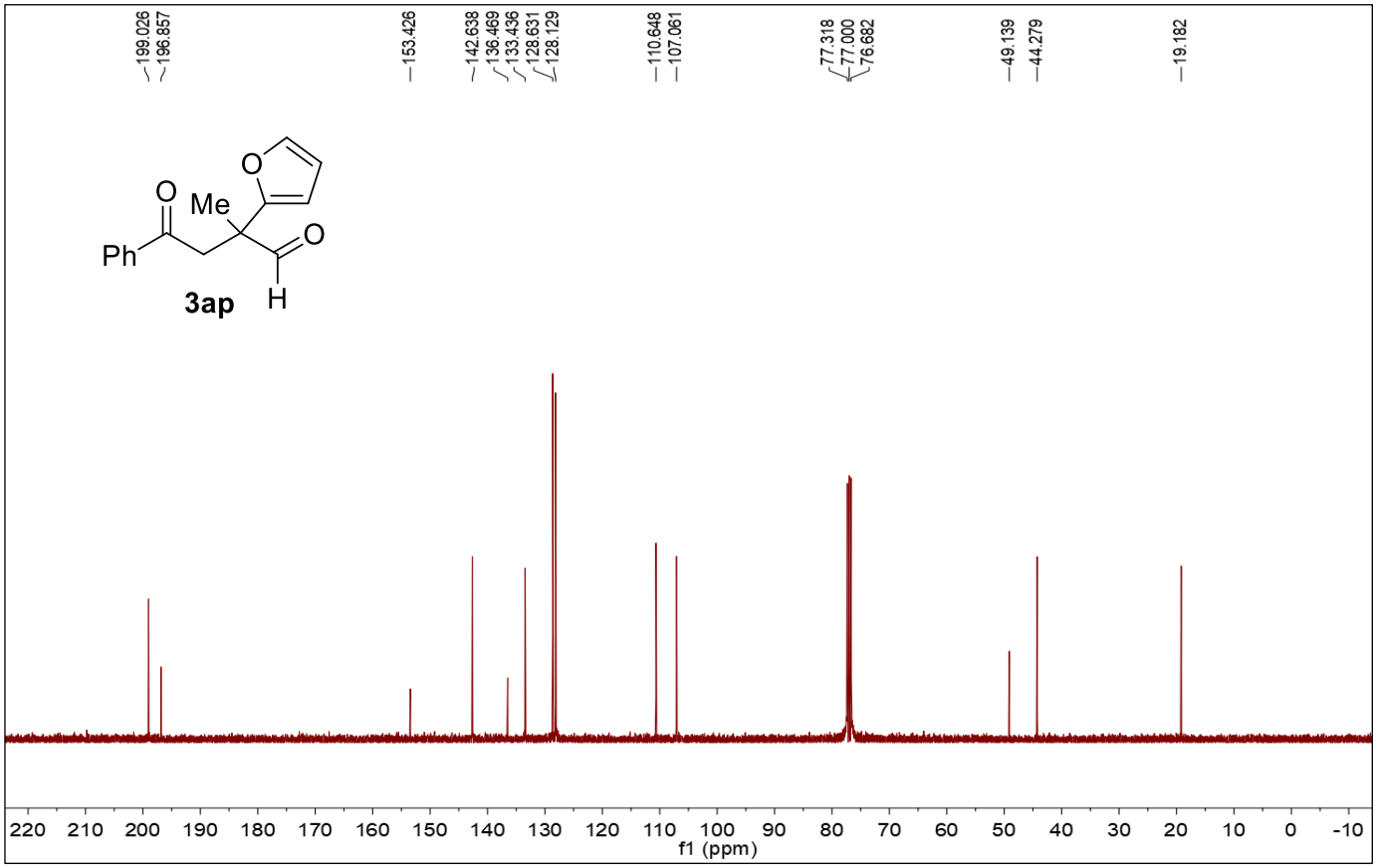
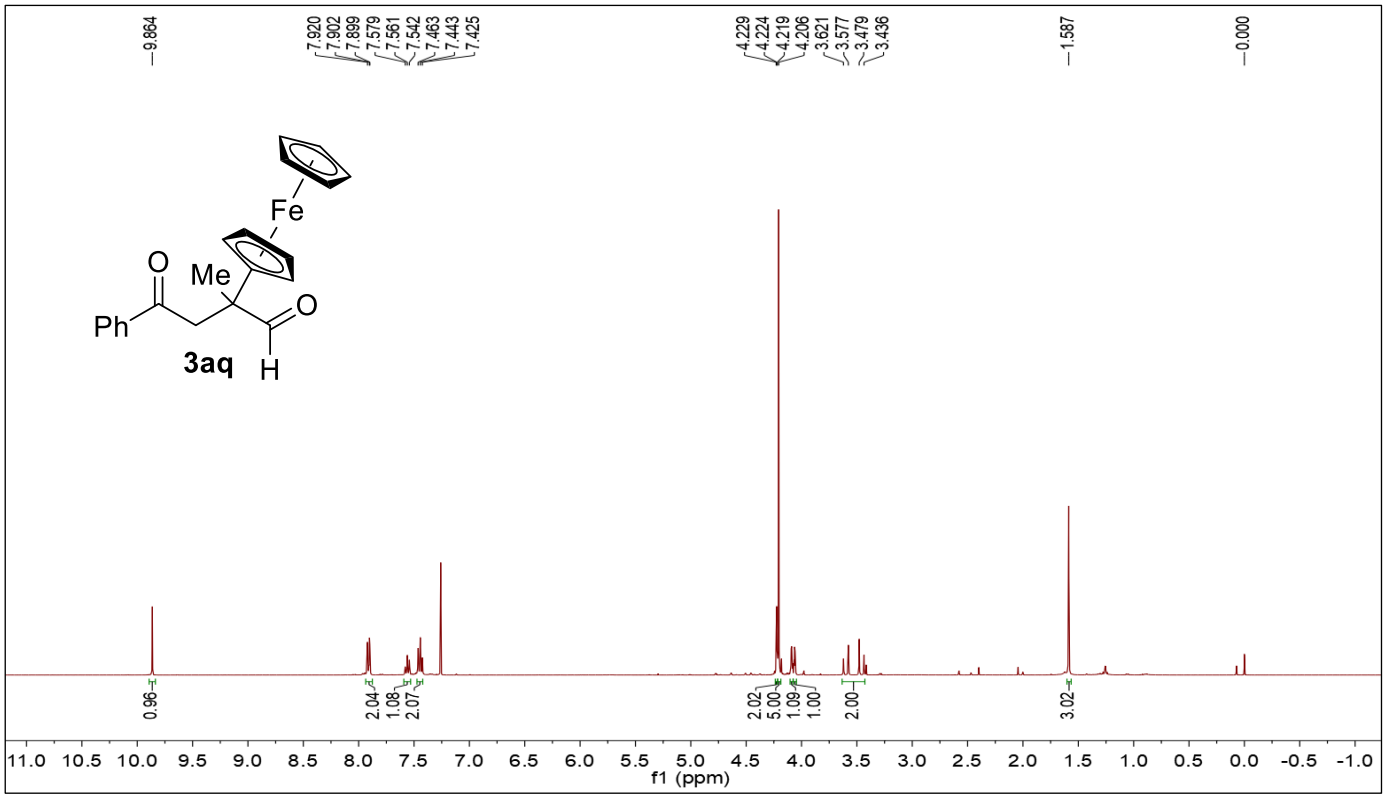
7.958
7.940
7.936
7.595
7.576
7.558
7.480
7.441
7.379
7.377
7.374
6.357
6.352
6.348
6.344
6.250
6.248
6.241
6.240
3.834
3.790
3.689
3.645
1.613
0.000
O
Me
O
Ph
O
H
3ap
0.96
2.10
1.03
2.16
1.04
1.00
1.01
2.17
3.07
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


199.026
196.857
153.426
142.638
136.469
133.436
128.631
128.129
110.648
107.061
77.318
77.000
76.682
49.139
44.279
19.182
O
Me
O
Ph
O
H
3ap
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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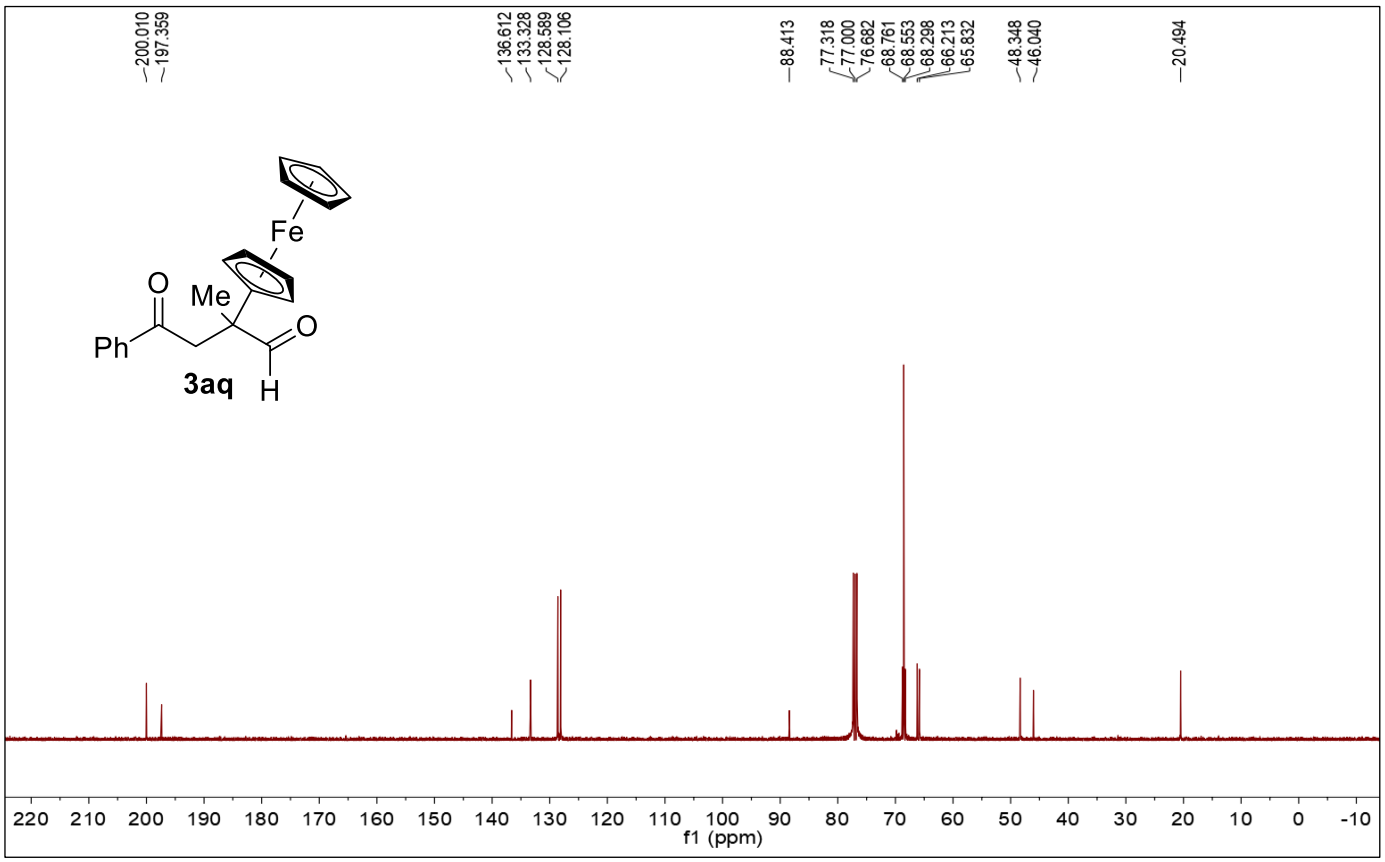
7.902
7.899
7.579
7.561
7.542
7.463
7.443
7.425
4.229
4.224
4.219
4.206
3.621
3.577
3.479
3.436
1.587
0.000
Fe
O
Me
O
Ph
3aq
H
0.96
2.04
1.08
2.07
2.02
5.00
1.09
1.00
2.00
3.02
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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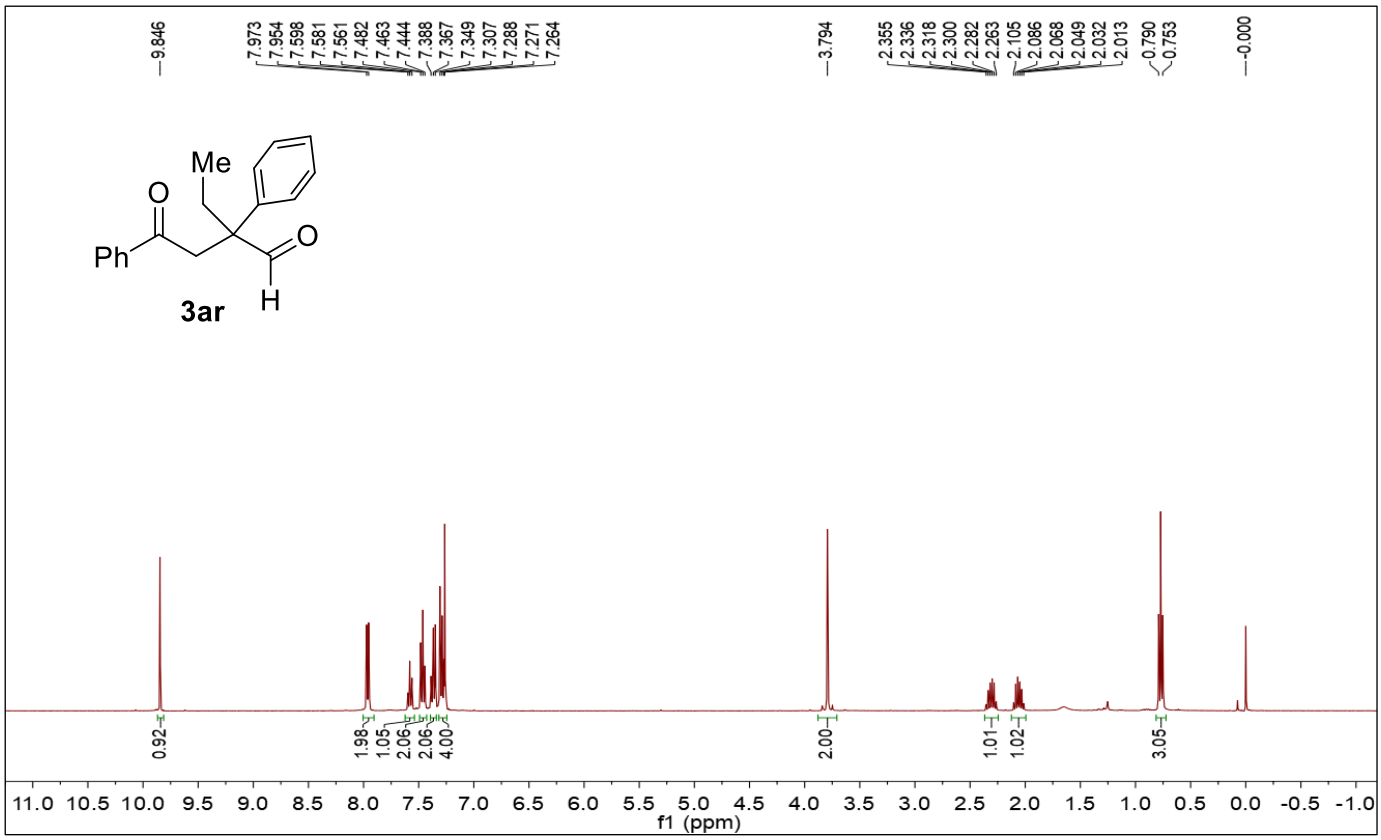
197.359
136.612
133.328
128.589
128.106
88.413
77.318
77.000
76.682
68.761
68.553
68.298
66.213
65.832
48.348
46.040
20.494
Fe
O
Me
O
Ph
3aq
H
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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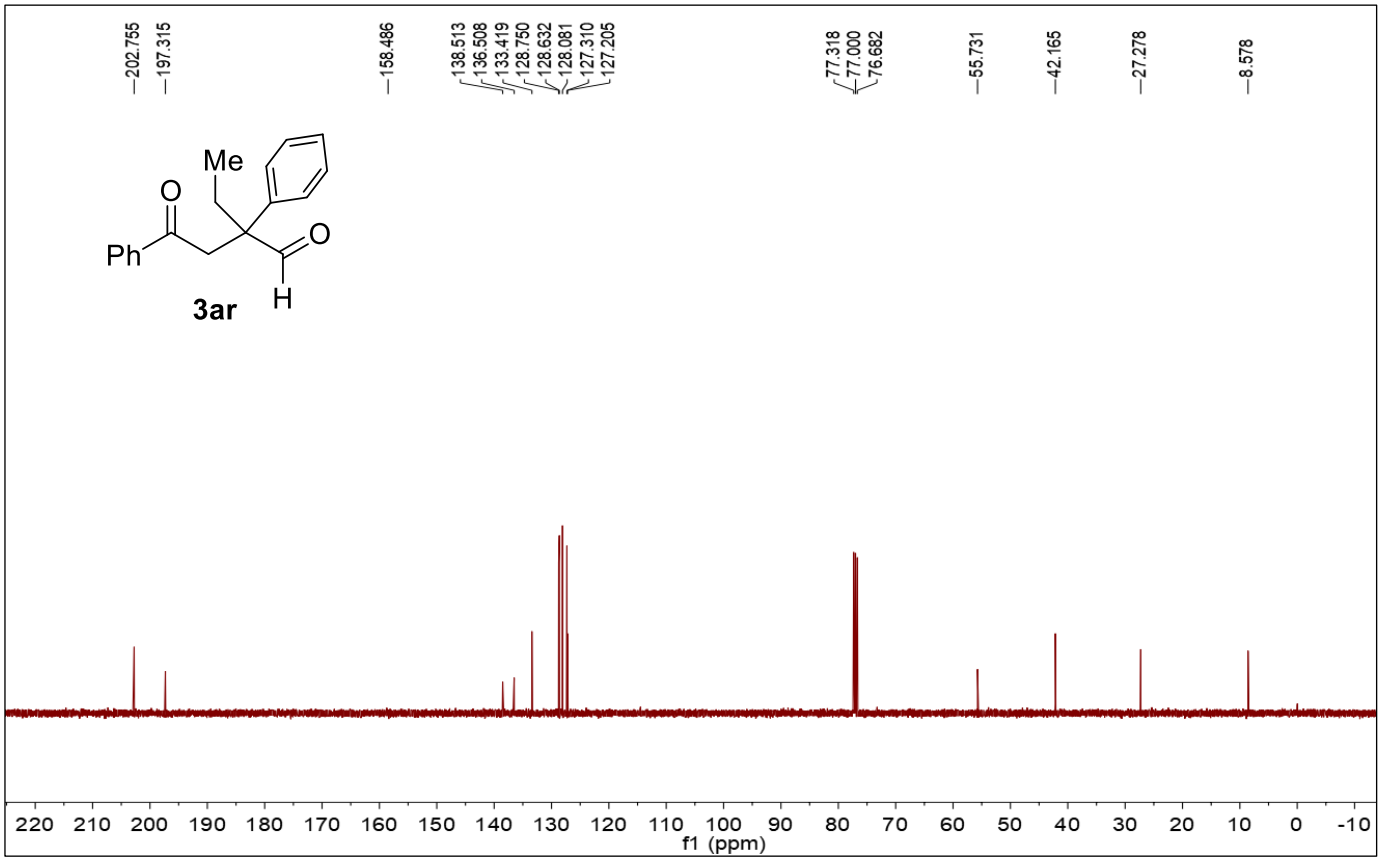
7.973
7.954
7.598
7.581
7.561
7.482
7.463
7.444
7.388
7.367
7.349
7.307
7.288
7.271
7.264
3.794
2.355
2.336
2.318
2.300
2.282
2.263
2.105
2.086
2.068
2.049
2.032
2.013
0.790
0.753
0.000
Me
O
Ph
O
H
3ar
0.92
1.98
1.05
2.06
2.06
4.00
2.00
1.01
1.02
3.05
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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136.508
133.419
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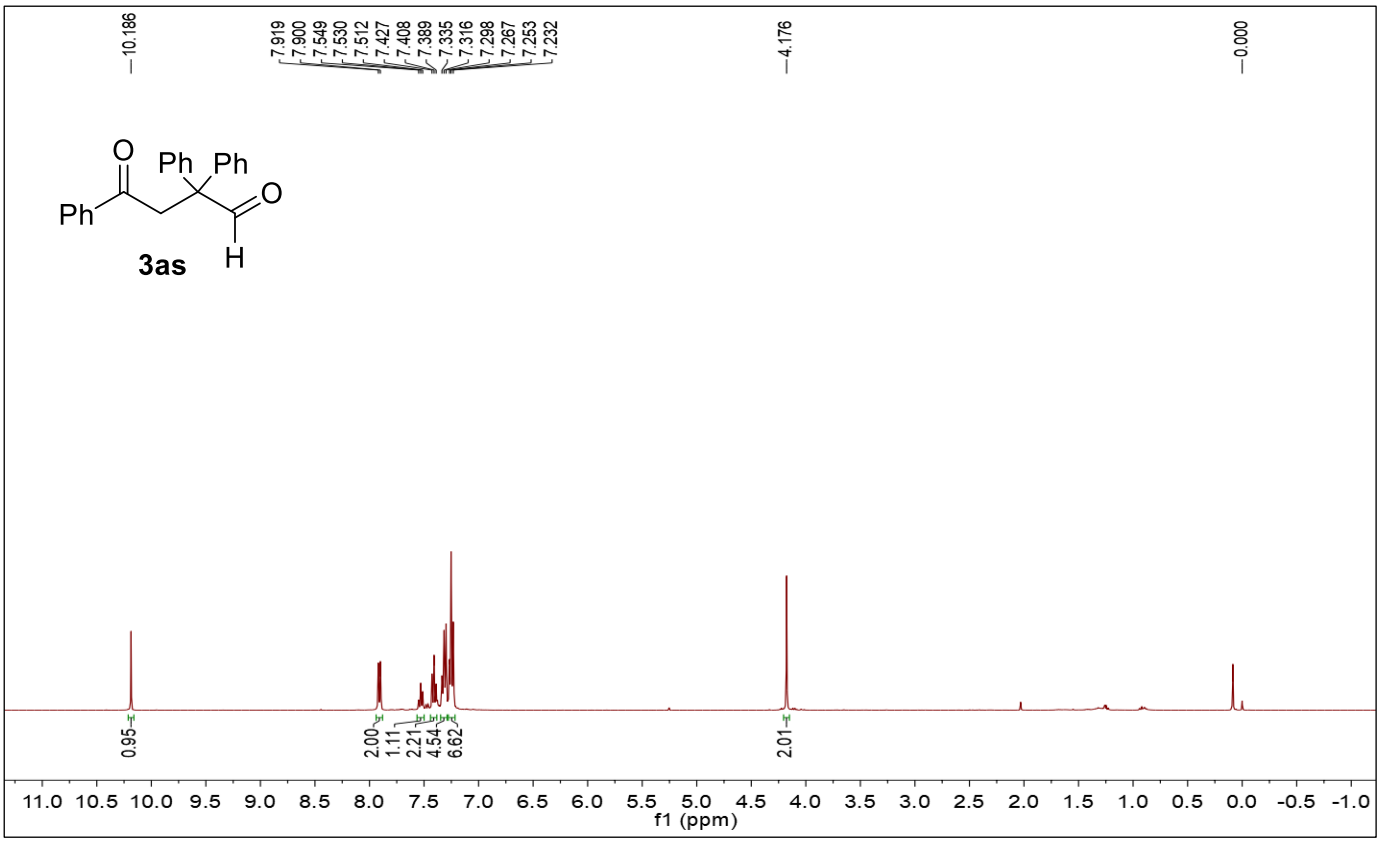
128.632
128.081
127.310
127.205
77.318
77.000
76.682
55.731
42.165
27.278
8.578
Me
O
Ph
O
H
3ar
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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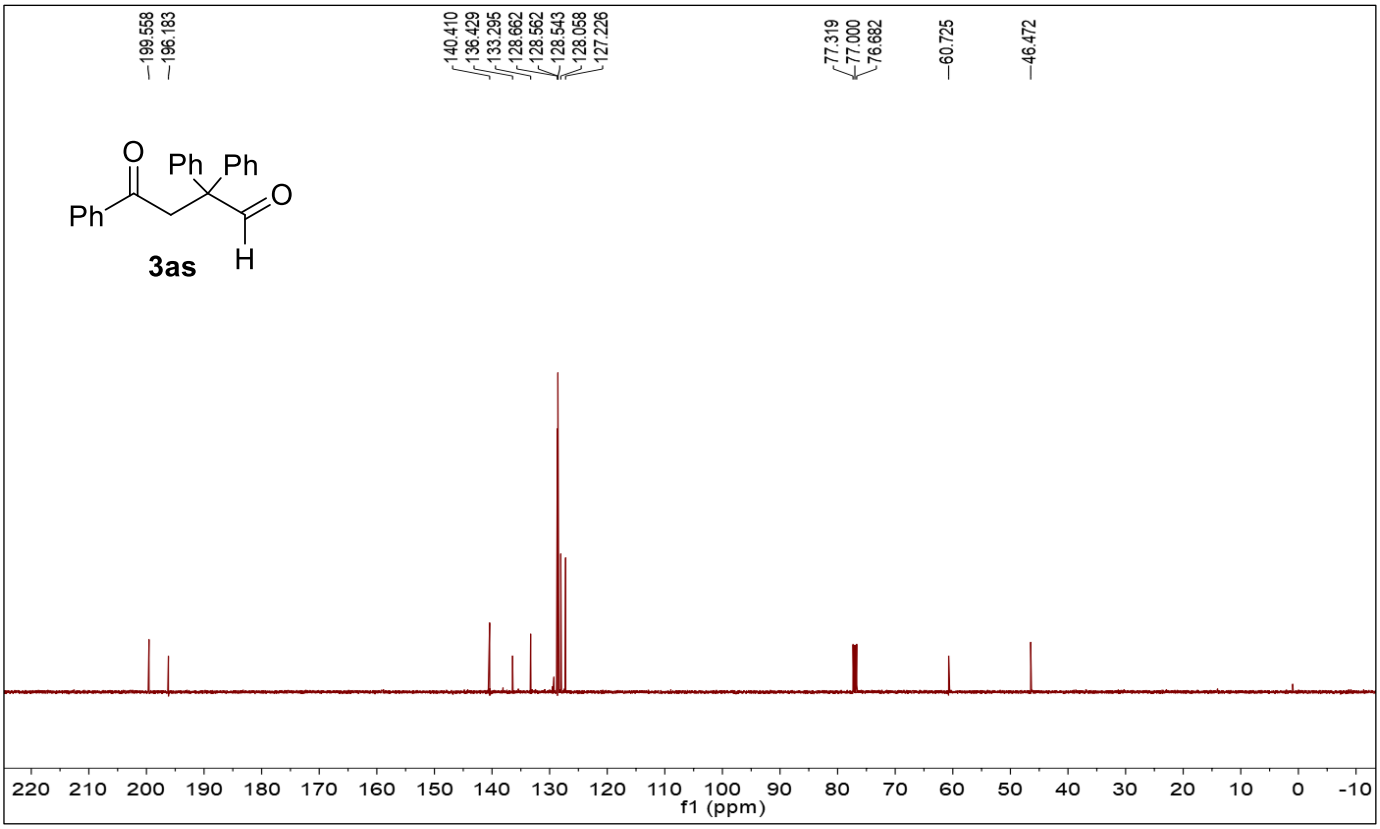
7.530
7.512
7.427
7.408
7.389
7.335
7.316
7.298
7.267
7.253
7.232
4.176
0.000
O
Ph
Ph
Ph
O
H
3as
0.95
2.00
1.11
2.21
4.54
6.62
2.01
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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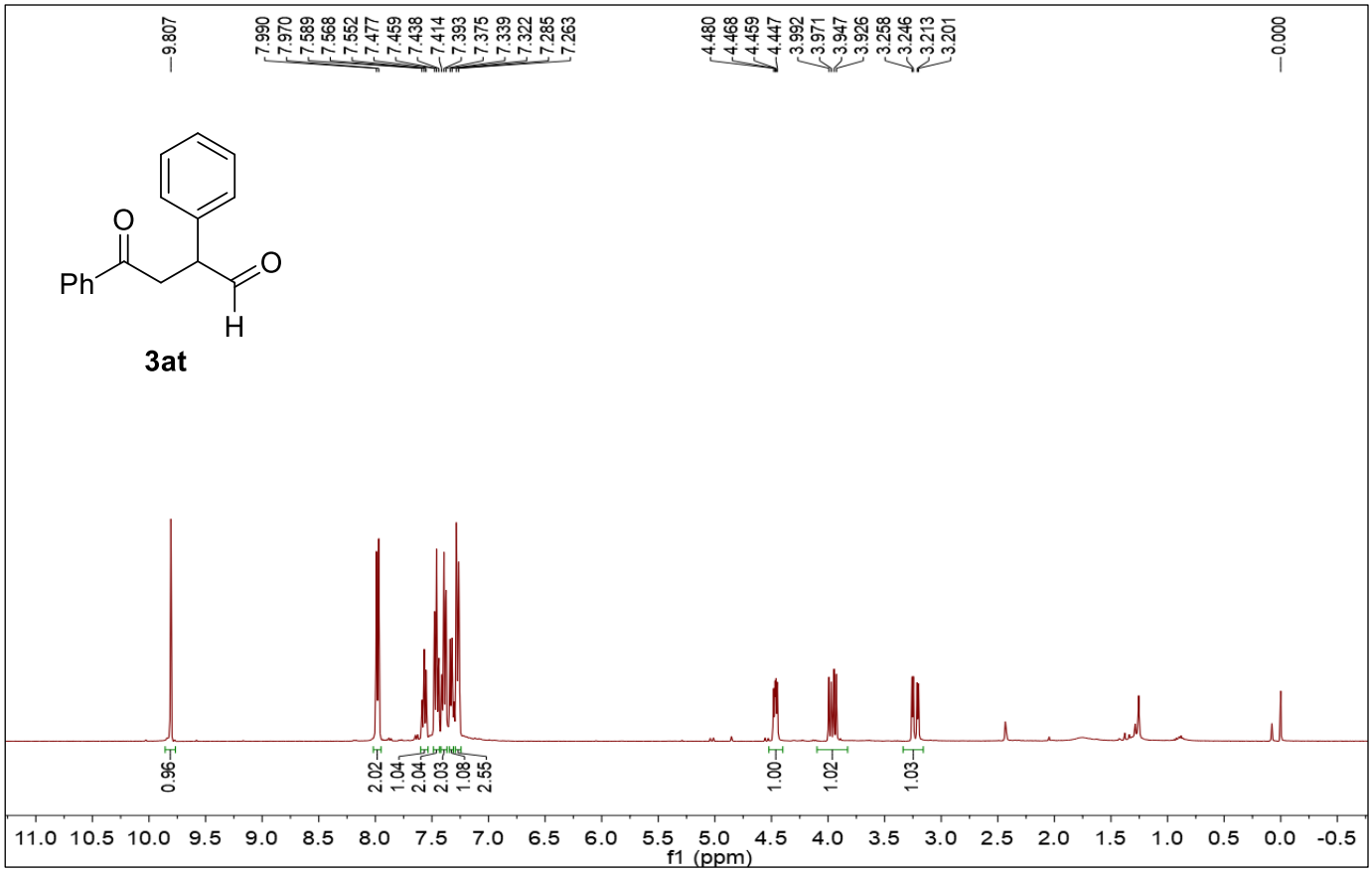
140.410
136.429
133.295
128.662
128.562
128.543
128.058
127.226
77.319
77.000
76.682
60.725
46.472
O
Ph
Ph
Ph
O
H
3as
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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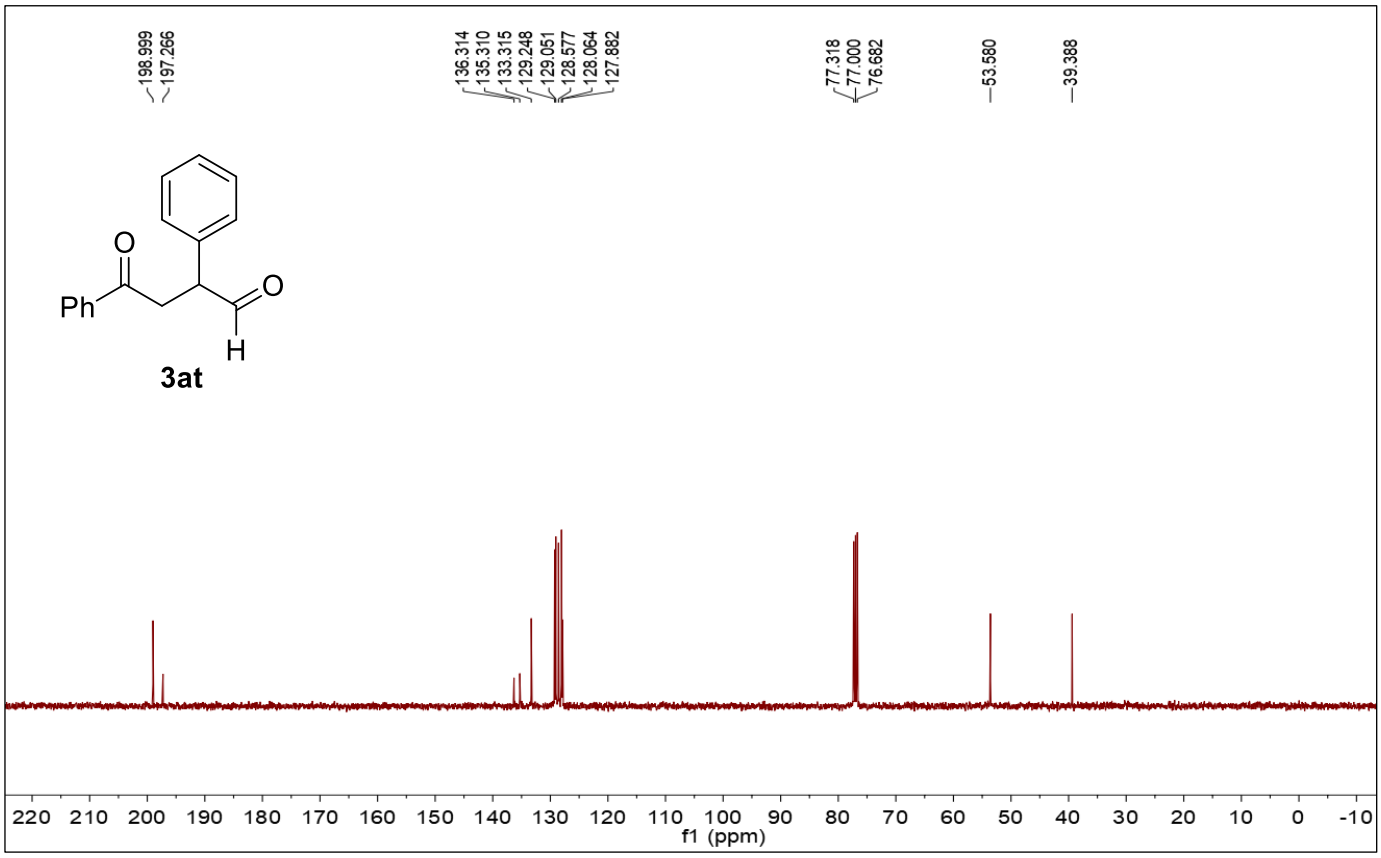
7.589
7.568
7.552
7.477
7.459
7.438
7.414
7.393
7.375
7.339
7.322
7.285
7.263
4.480
4.468
4.459
4.447
3.992
3.971
3.947
3.926
3.258
3.246
3.213
3.201
0.000
O
Ph
O
H
3at
0.96
2.02
1.04
2.04
2.03
1.08
2.55
1.00
1.02
1.03
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5
f1 (ppm)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133.315
129.248
129.051
128.577
128.064
127.882
77.318
77.000
76.682
53.580
39.388
O
Ph
O
H
3at
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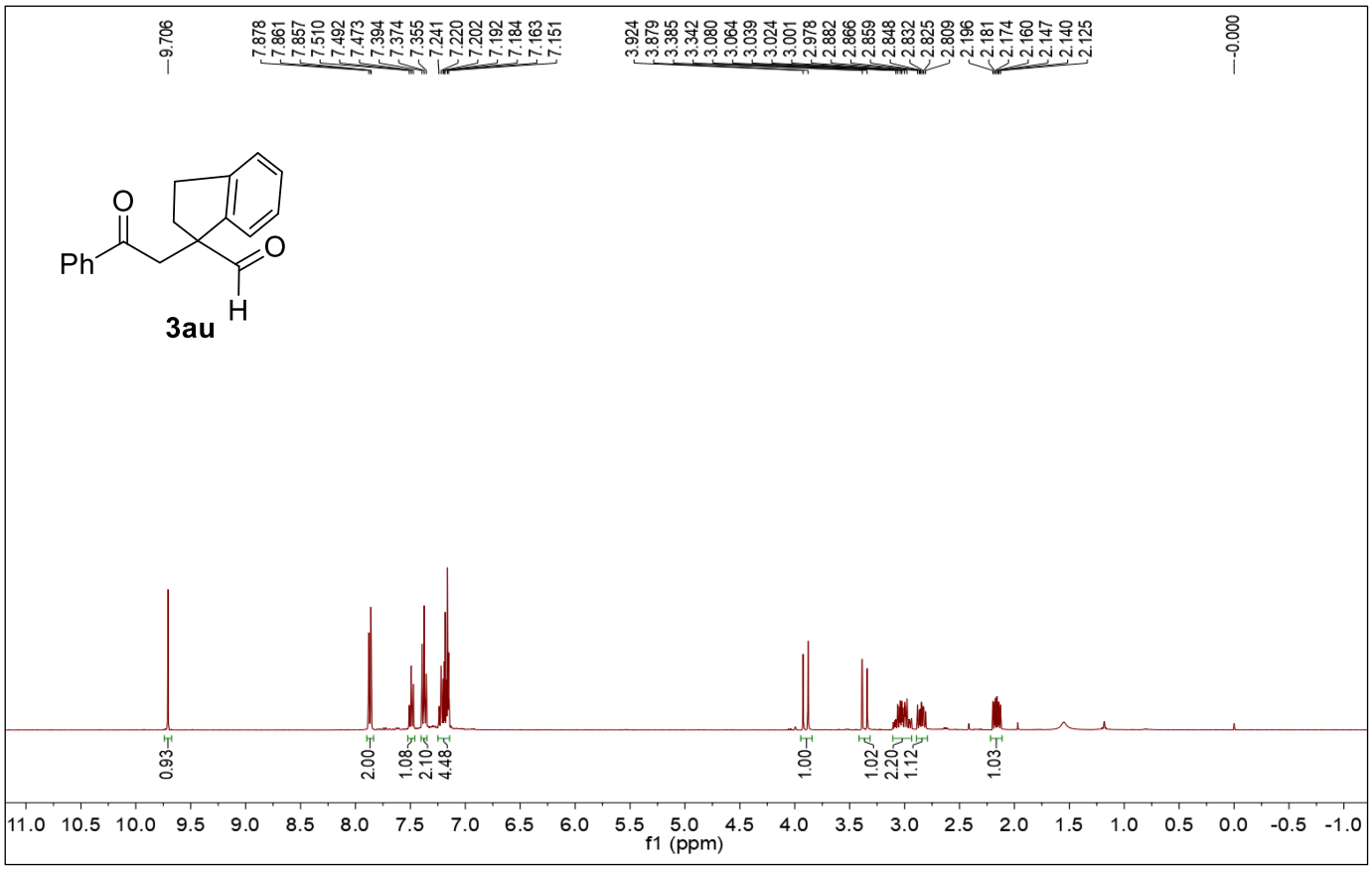
f1 (ppm)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7.355
7.241
7.220
7.202
7.192
7.184
7.163
7.151
3.924
3.879
3.385
3.342
3.080
3.064
3.039
3.024
3.001
2.978
2.882
2.866
2.859
2.848
2.832
2.825
2.809
2.196
2.181
2.174
2.160
2.147
2.140
2.125
0.000
O
Ph
O
H
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2.10
4.48
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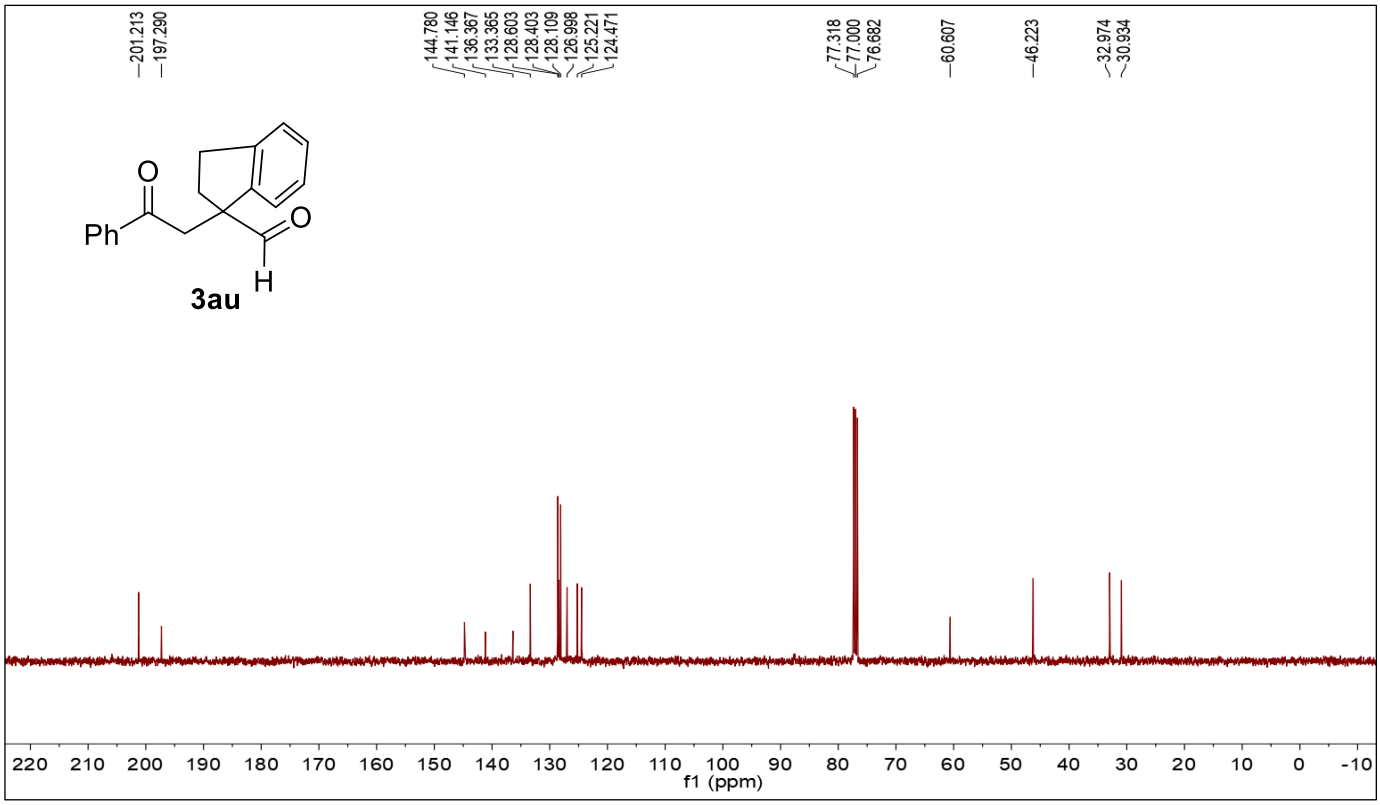
2.20
1.12
1.03
f1 (ppm)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128.403
128.109
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125.221
124.471
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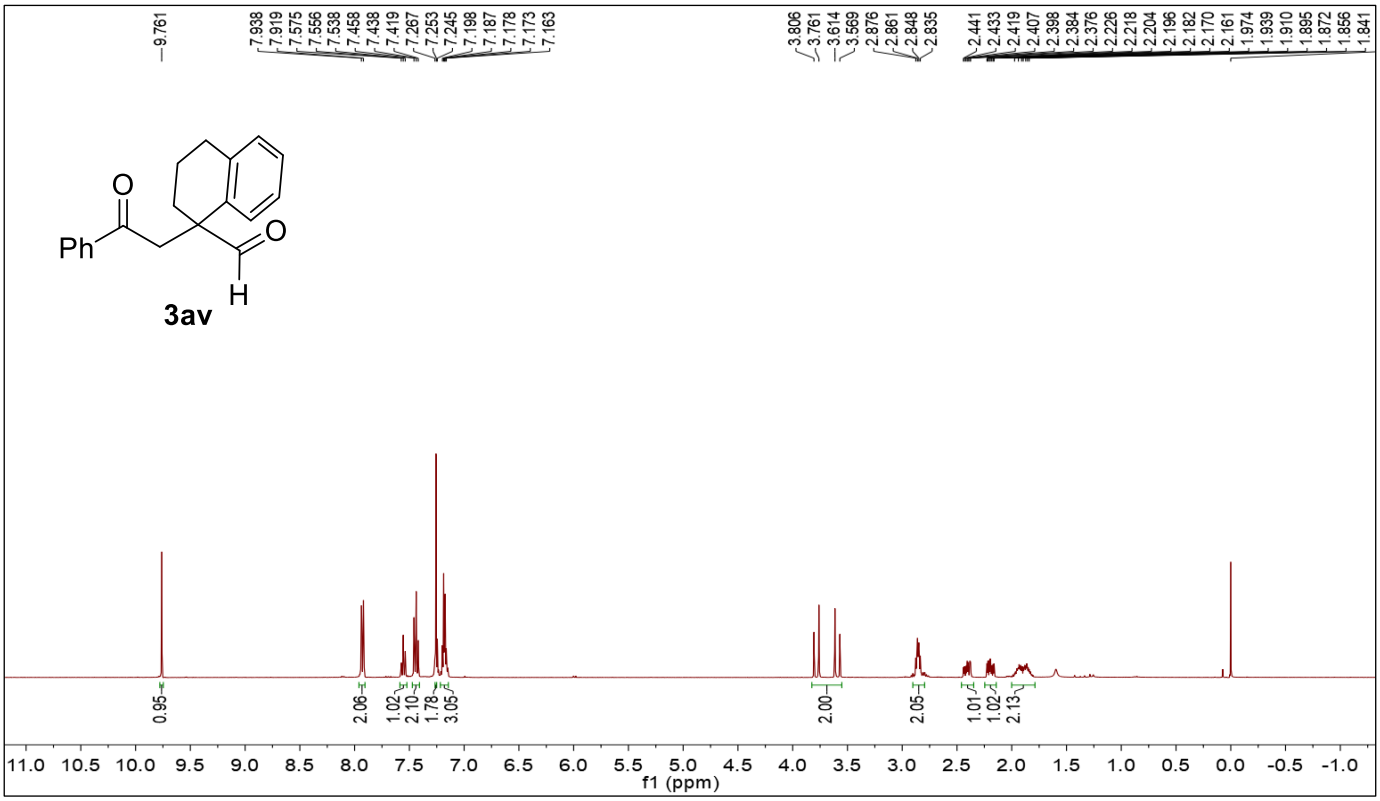
77.000
76.682
60.607
46.223
32.974
30.934
O
Ph
O
H
3au
f1 (ppm)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7.438
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7.267
7.253
7.245
7.198
7.187
7.178
7.173
7.163
3.806
3.761
3.614
3.569
2.876
2.861
2.848
2.835
2.441
2.433
2.419
2.407
2.398
2.384
2.376
2.226
2.218
2.204
2.196
2.182
2.170
2.161
1.974
1.939
1.910
1.895
1.872
1.856
1.841
O
Ph
O
H
3av
0.95
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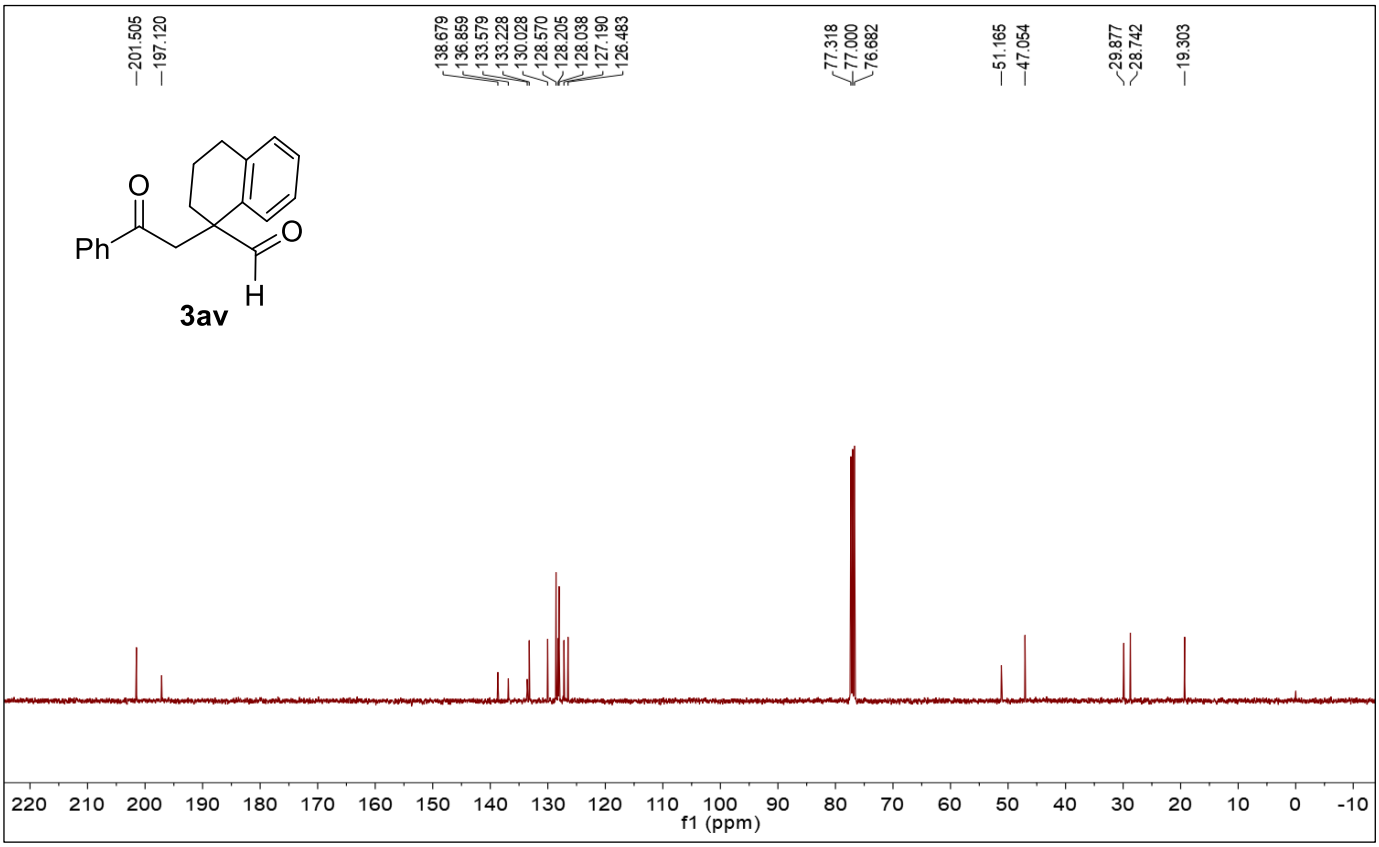
1.02
2.10
1.78
3.05
2.00
2.05
1.01
1.02
2.13
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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133.228
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128.570
128.205
128.038
127.190
126.483
77.318
77.000
76.682
51.165
47.054
29.877
28.742
19.303
O
Ph
O
H
3av
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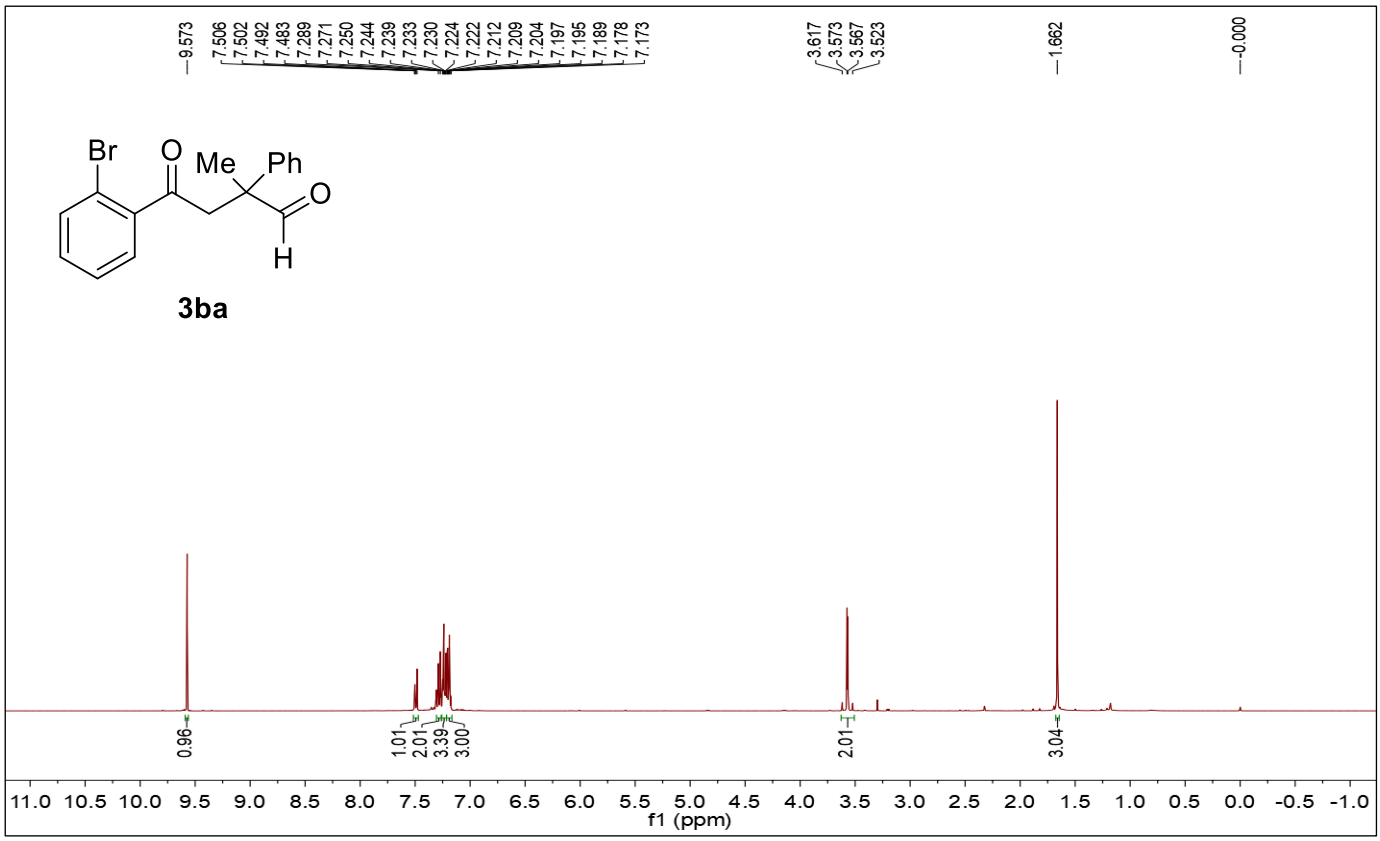
f1 (ppm)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7.271
7.250
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7.239
7.233
7.230
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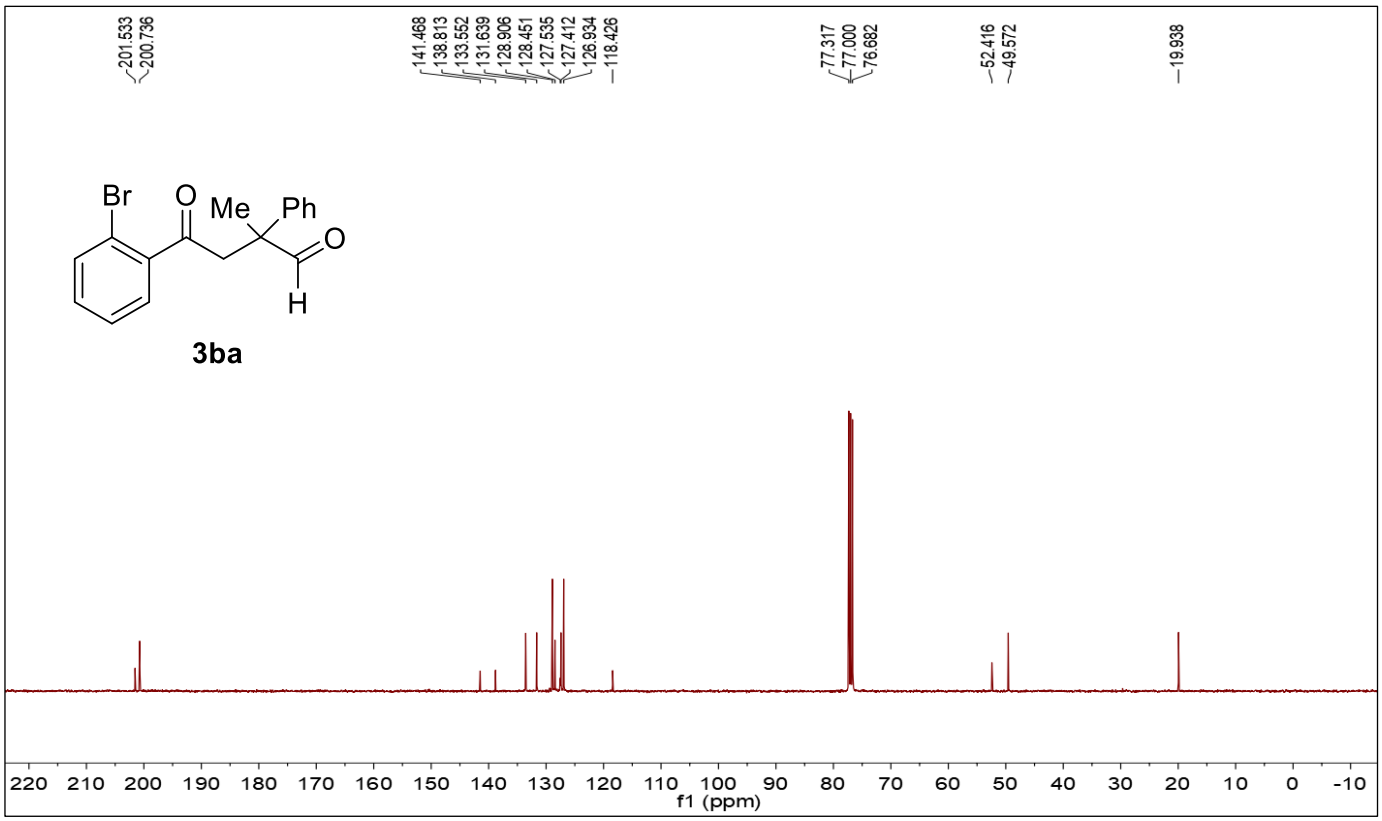
7.222
7.212
7.209
7.204
7.197
7.195
7.189
7.178
7.173
3.617
3.573
3.567
3.523
1.662
0.000
Br
O
Me
Ph
O
H
3ba
0.96
1.01
2.01
3.39
3.00
2.01
3.04
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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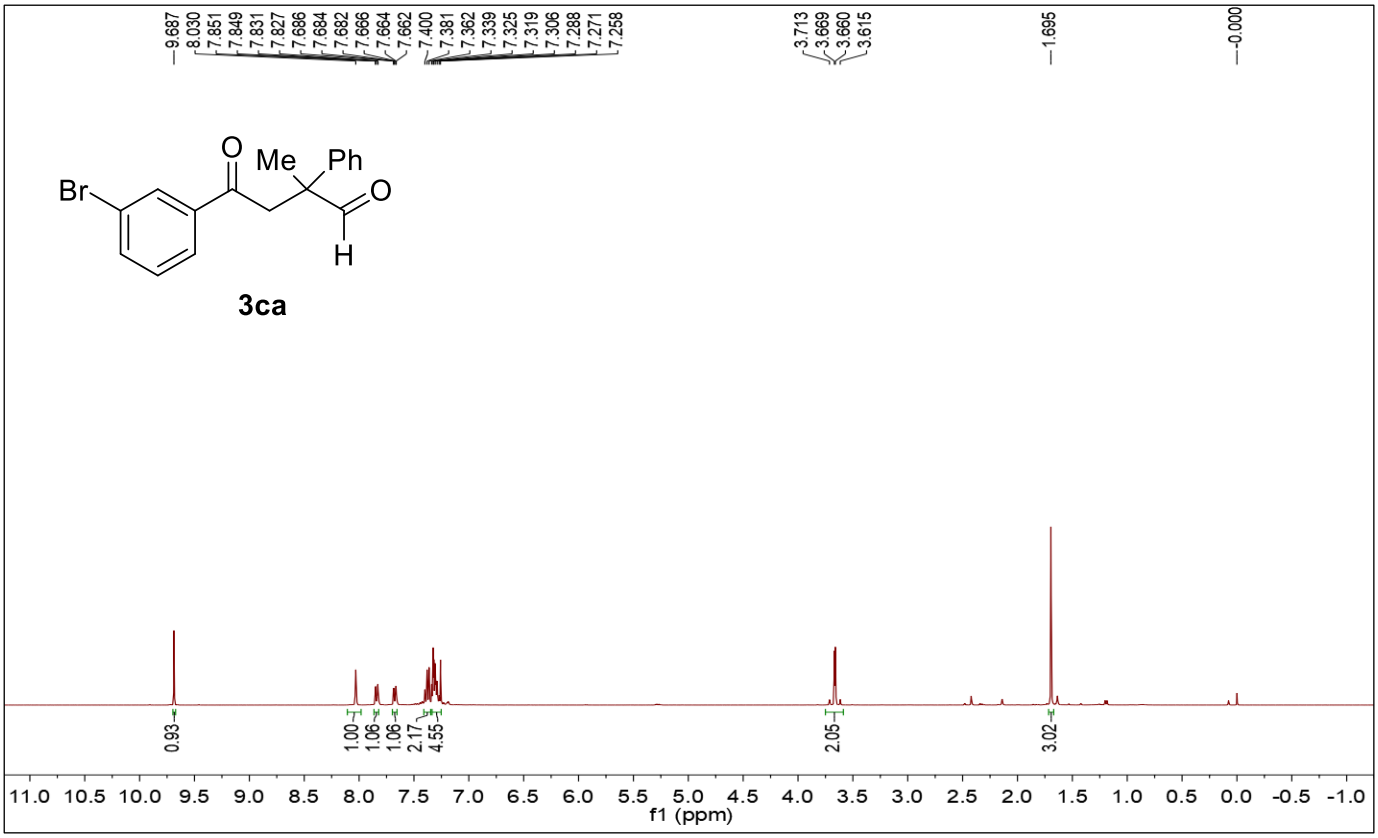
200.736
141.468
138.813
133.552
131.639
128.906
128.451
127.535
127.412
126.934
118.426
77.317
77.000
76.682
52.416
49.572
19.938
Br
O
Me
Ph
O
H
3ba
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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7.682
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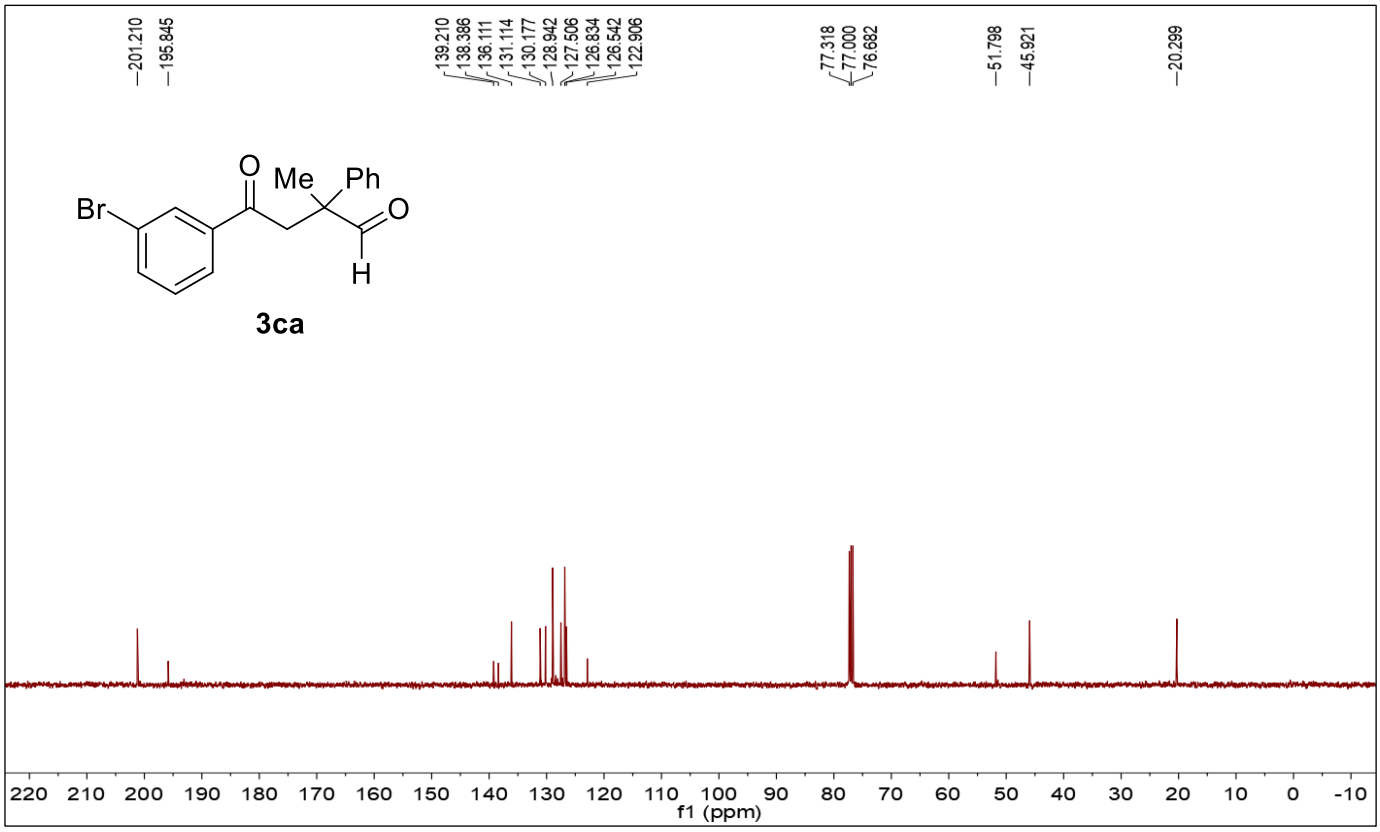
7.400
7.381
7.362
7.339
7.325
7.319
7.306
7.288
7.271
7.258
3.713
3.669
3.660
3.615
1.695
0.000
O
Me
Ph
Br
O
H
3ca
0.93
1.00
1.06
1.06
2.17
4.55
2.05
3.02
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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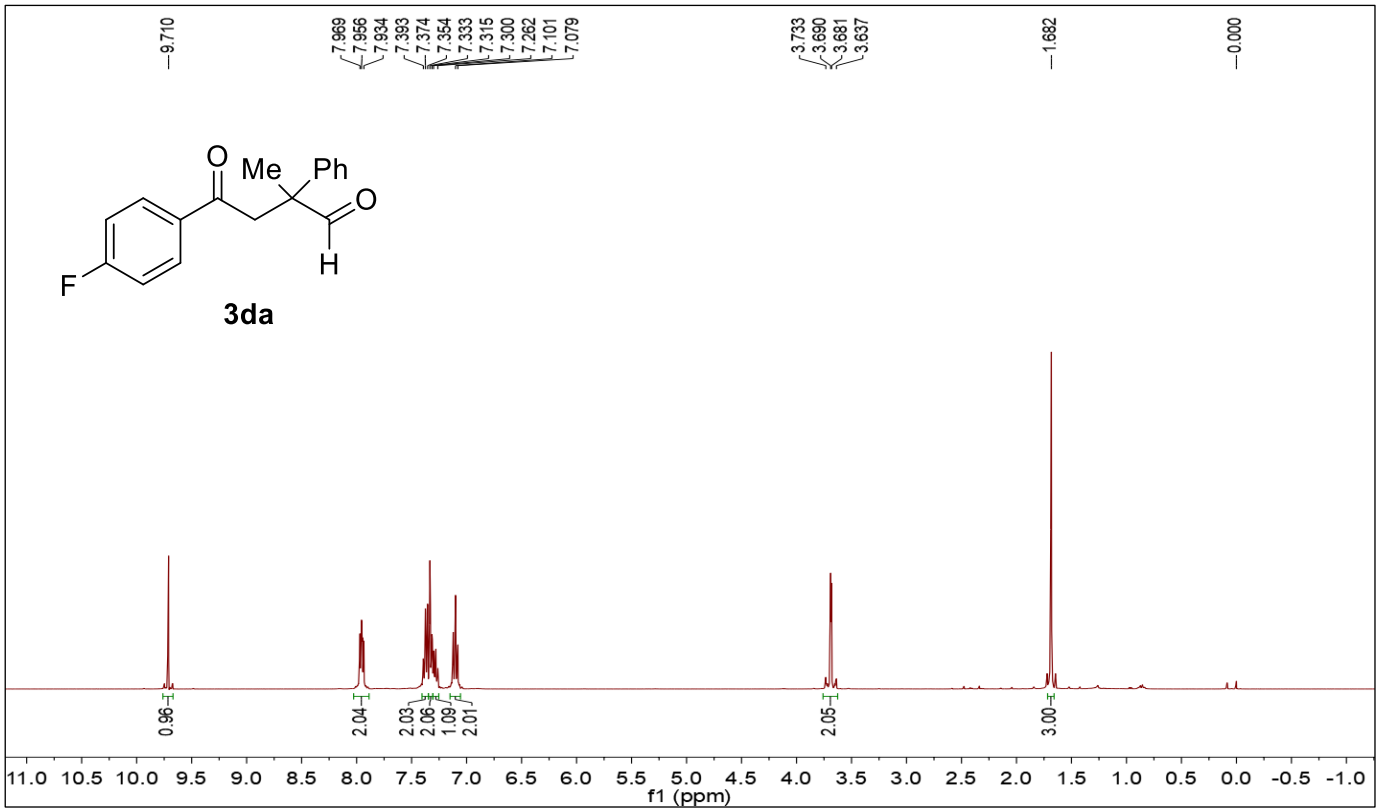
122.906
77.318
77.000
76.682
51.798
45.921
20.299
O
Me
Ph
Br
O
H
3ca
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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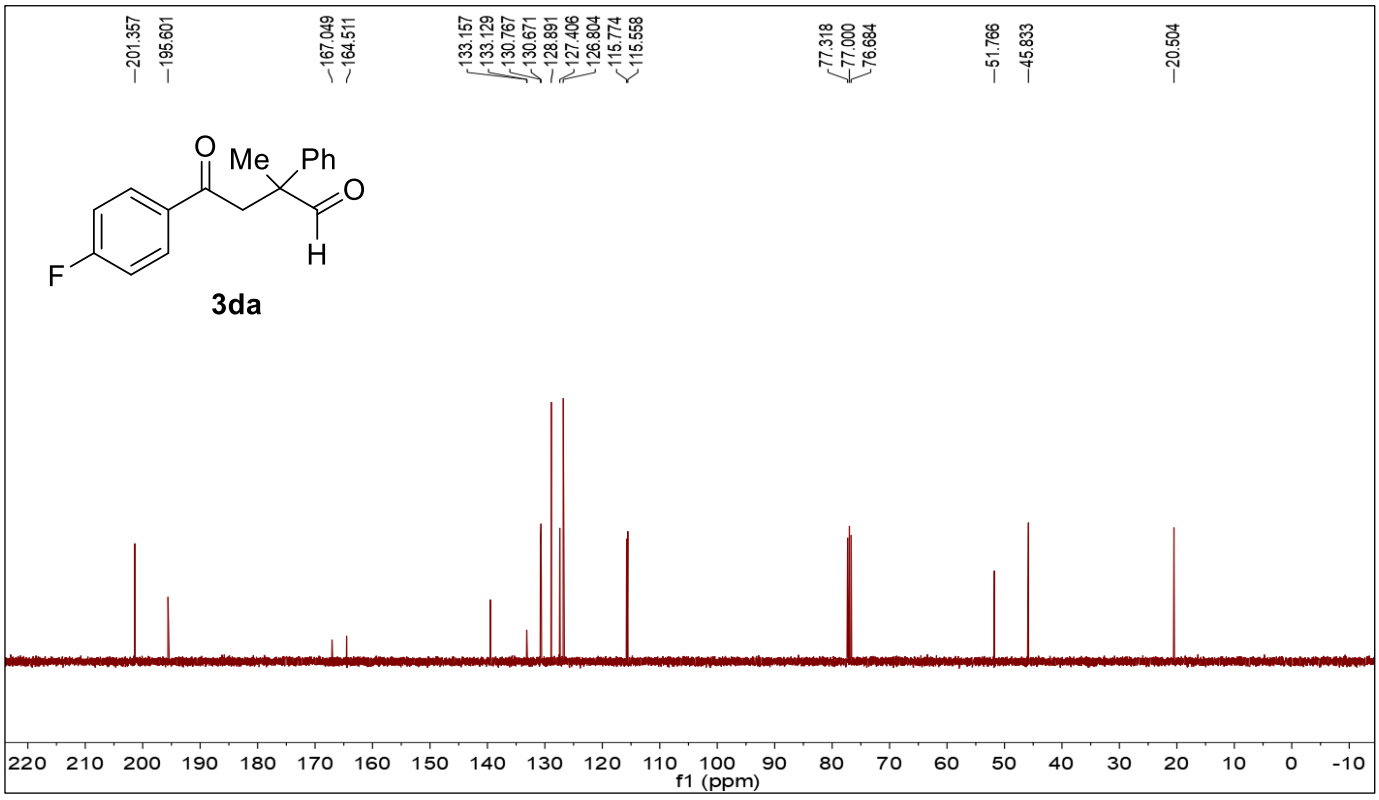
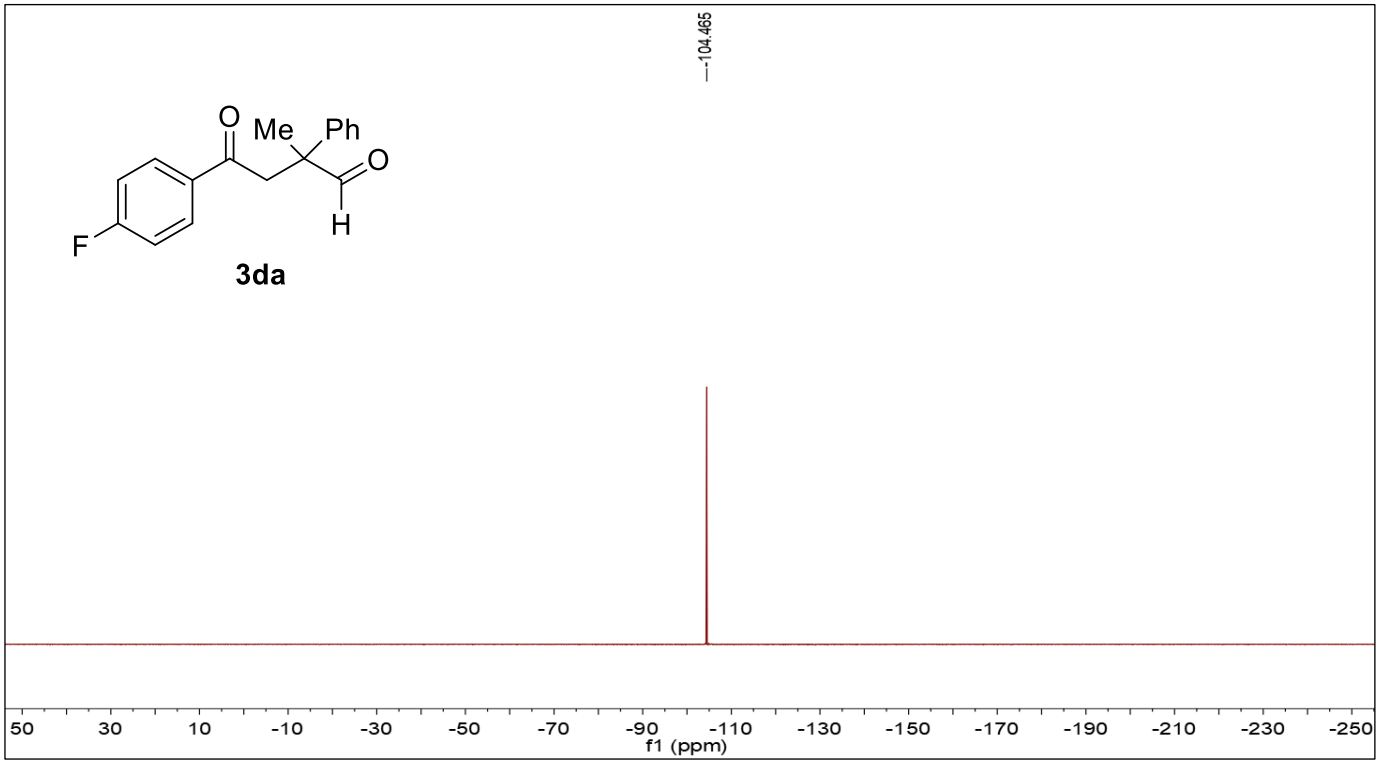
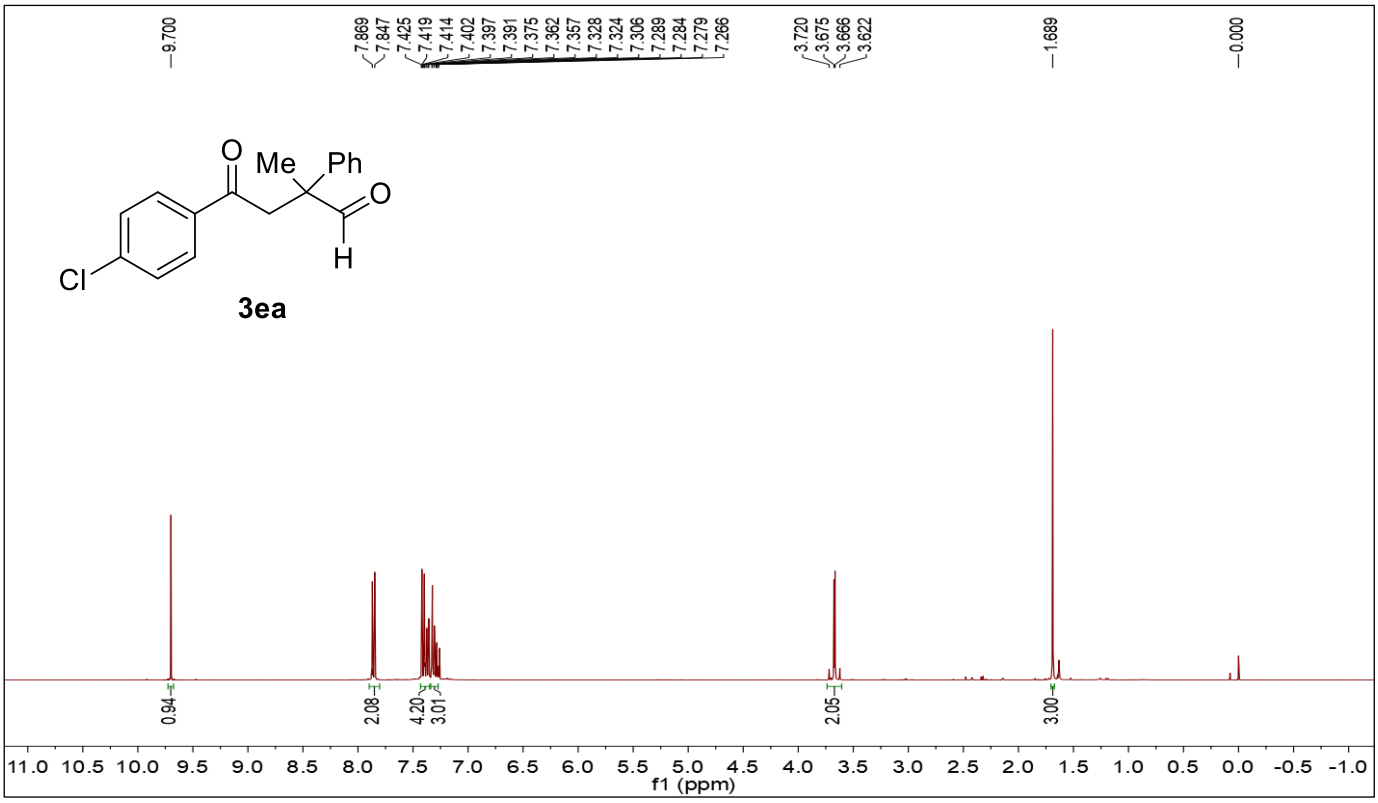
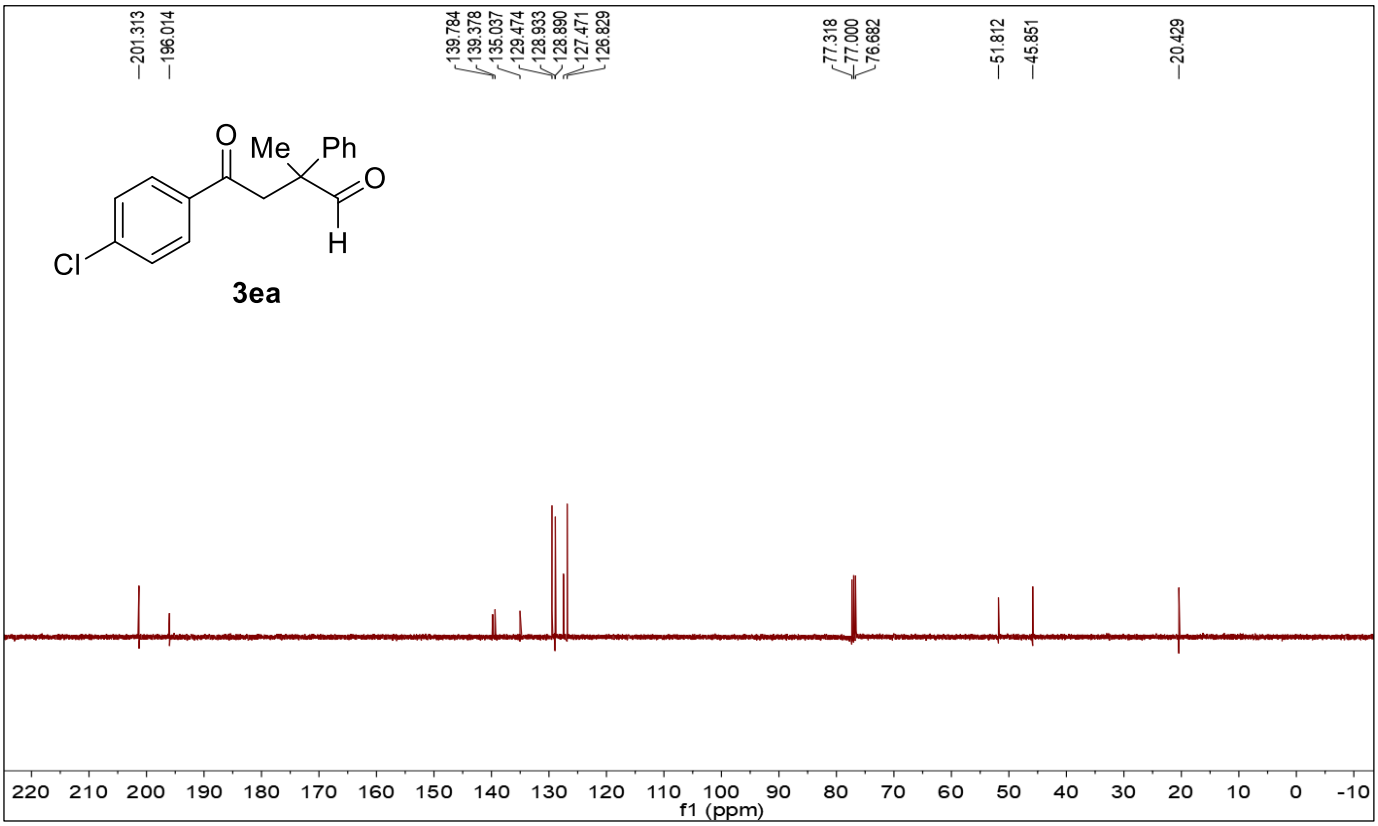
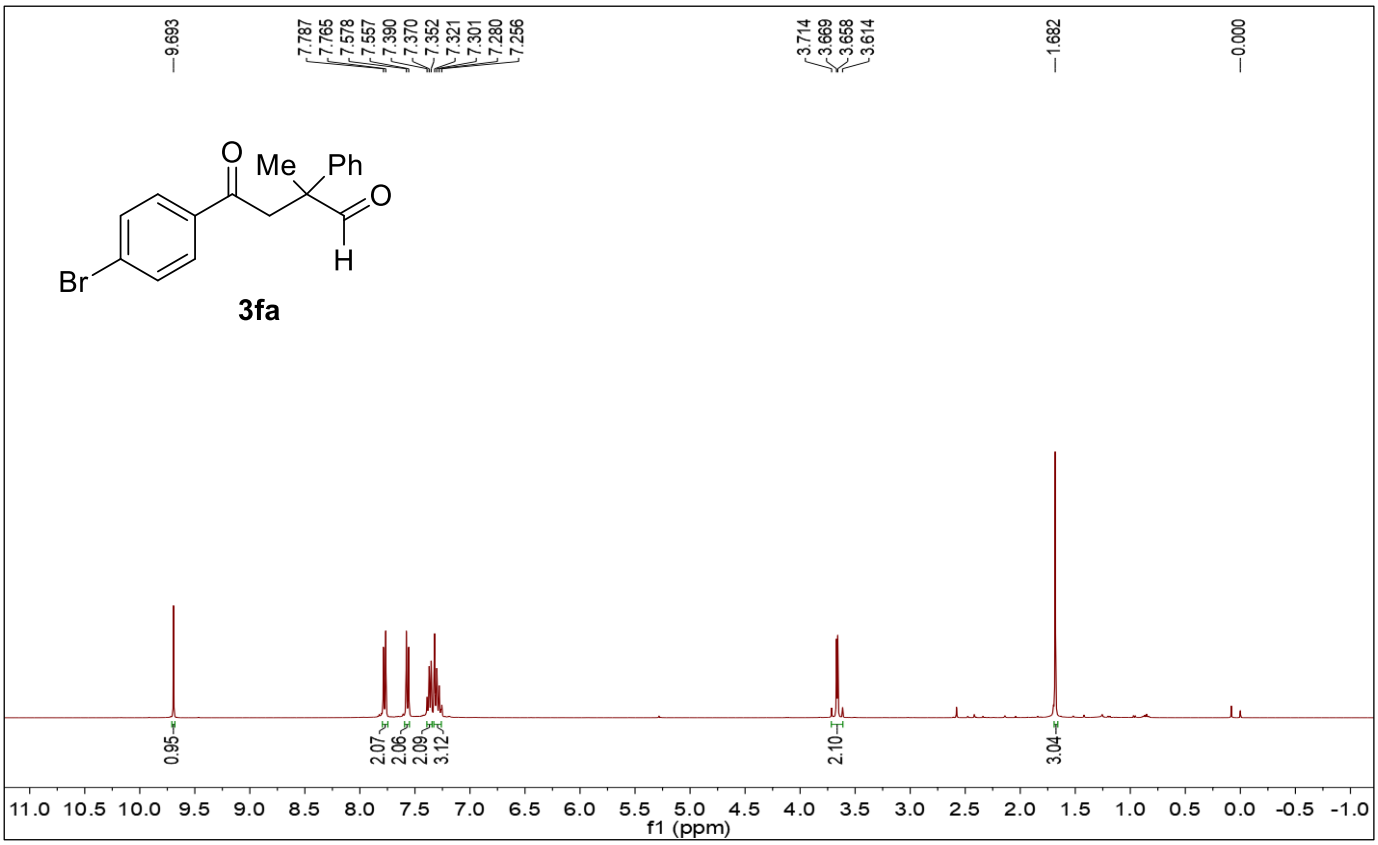
7.101
7.079
3.733
3.690
3.681
3.637
1.682
0.000
O
Me
Ph
O
H
F
3da
0.96
2.04
2.03
2.06
1.09
2.01
2.05
3.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

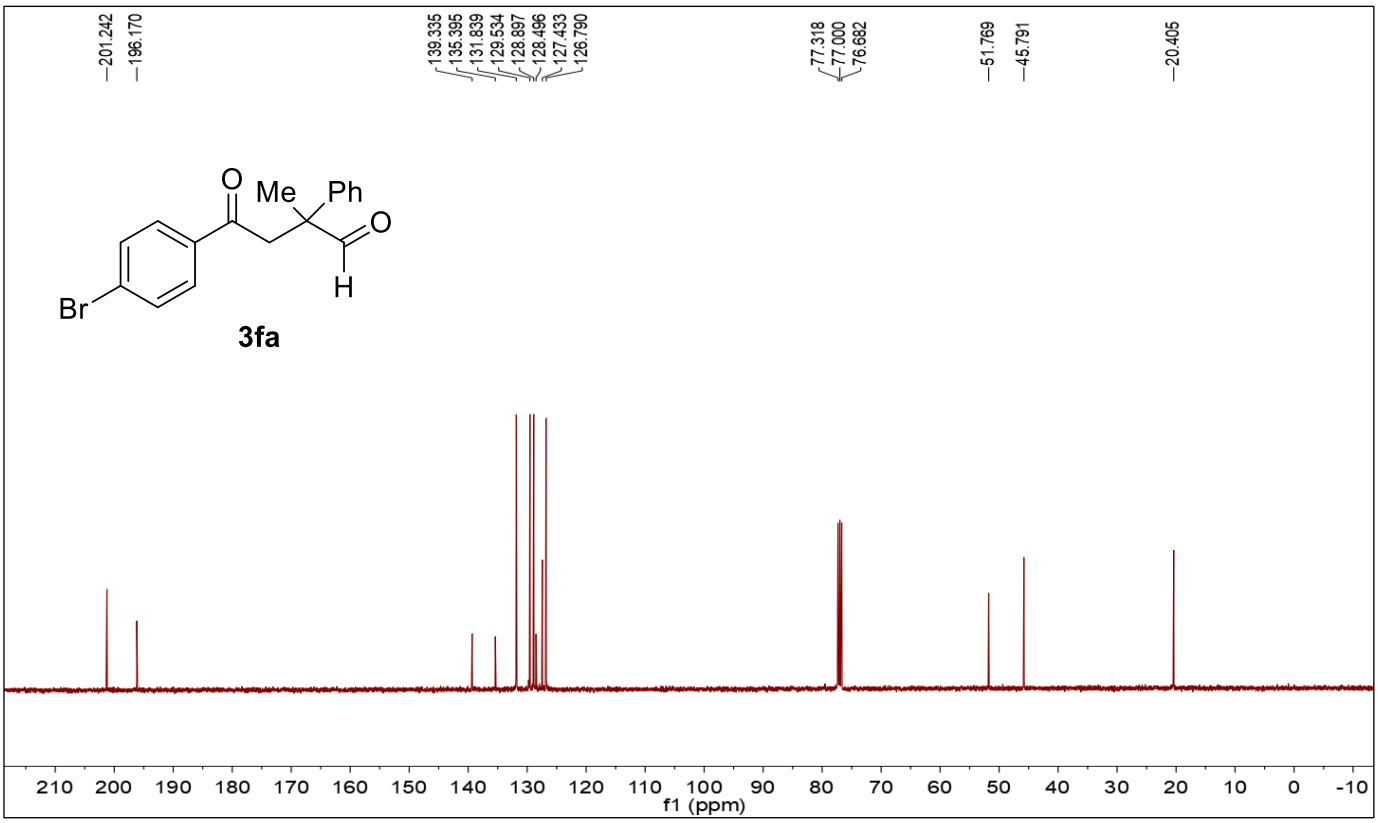

201.357
195.601
167.049
164.511
133.157
133.129
130.767
130.671
128.891
127.406
126.804
115.774
115.558
77.318
77.000
76.684
51.766
45.833
20.504
O
Me
Ph
O
H
F
3da
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

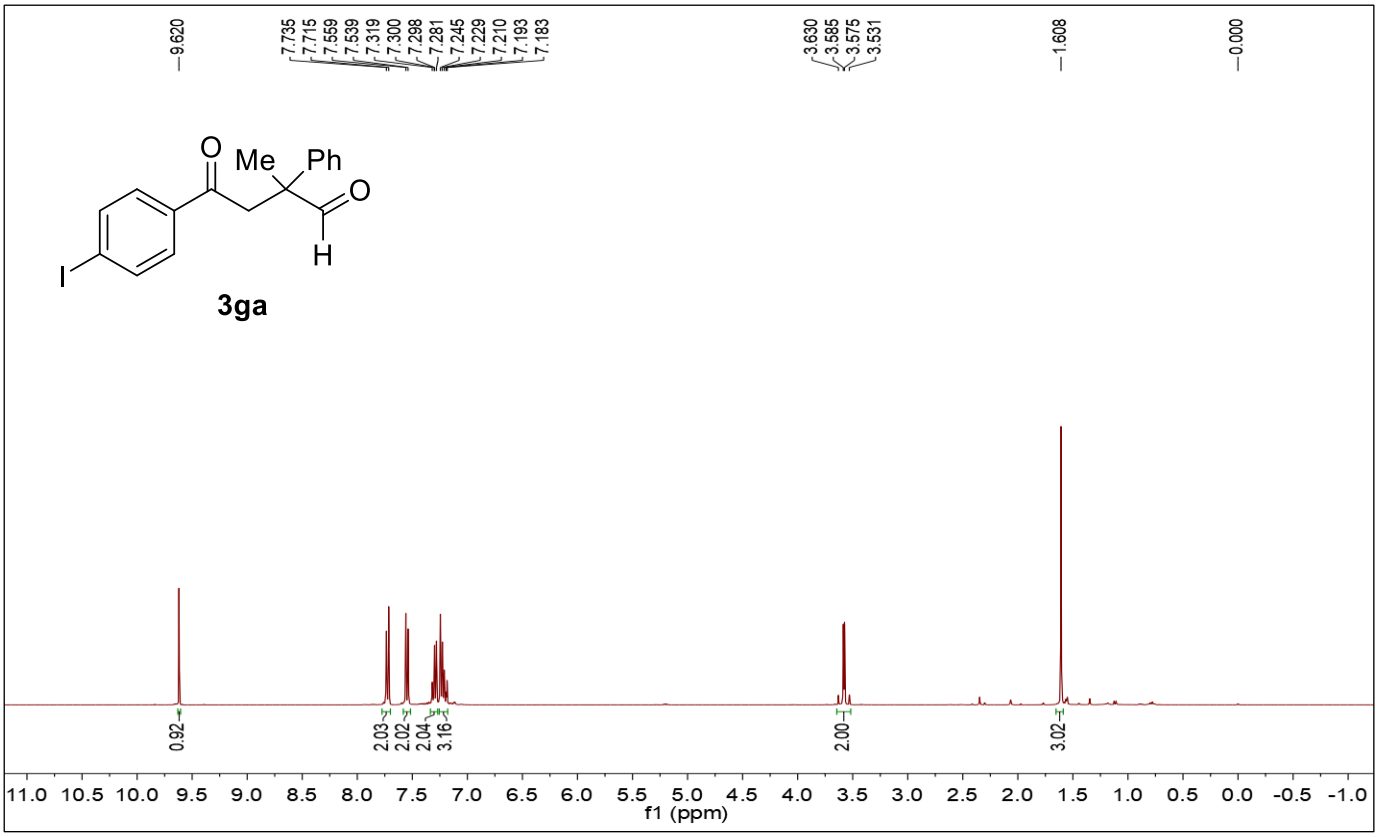

-104.465
O
Me
Ph
O
H
F
3da
50
30
10
-10
-30
-50
-70
-90
-110
-130
-150
-170
-190
-210
-230
-250
f1 (ppm)

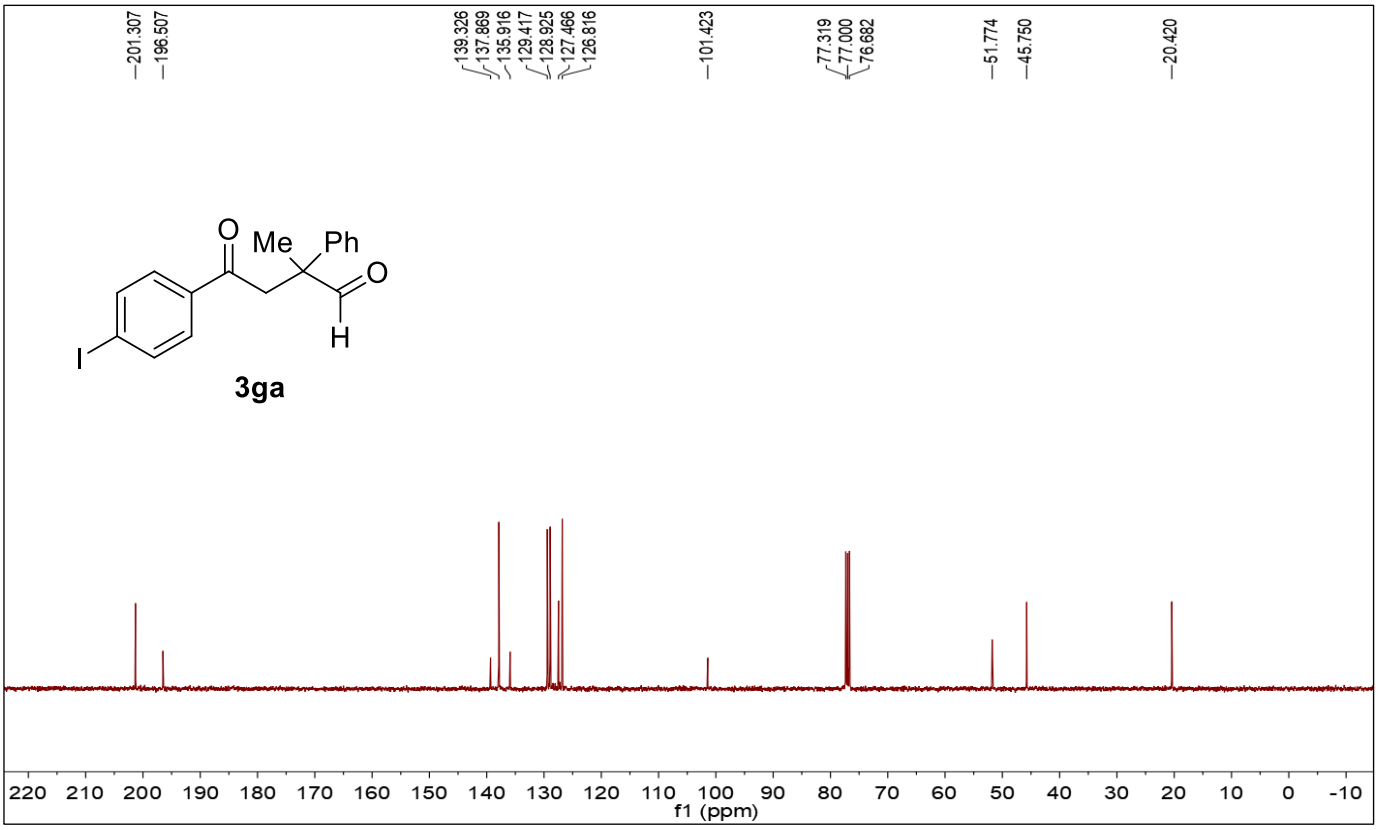

9.700
7.869
7.847
7.425
7.419
7.414
7.402
7.397
7.391
7.375
7.362
7.357
7.328
7.324
7.306
7.289
7.284
7.279
7.266
3.720
3.675
3.666
3.622
1.689
0.000
O
Me
Ph
O
H
Cl
3ea
0.94
2.08
4.20
3.01
2.05
3.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

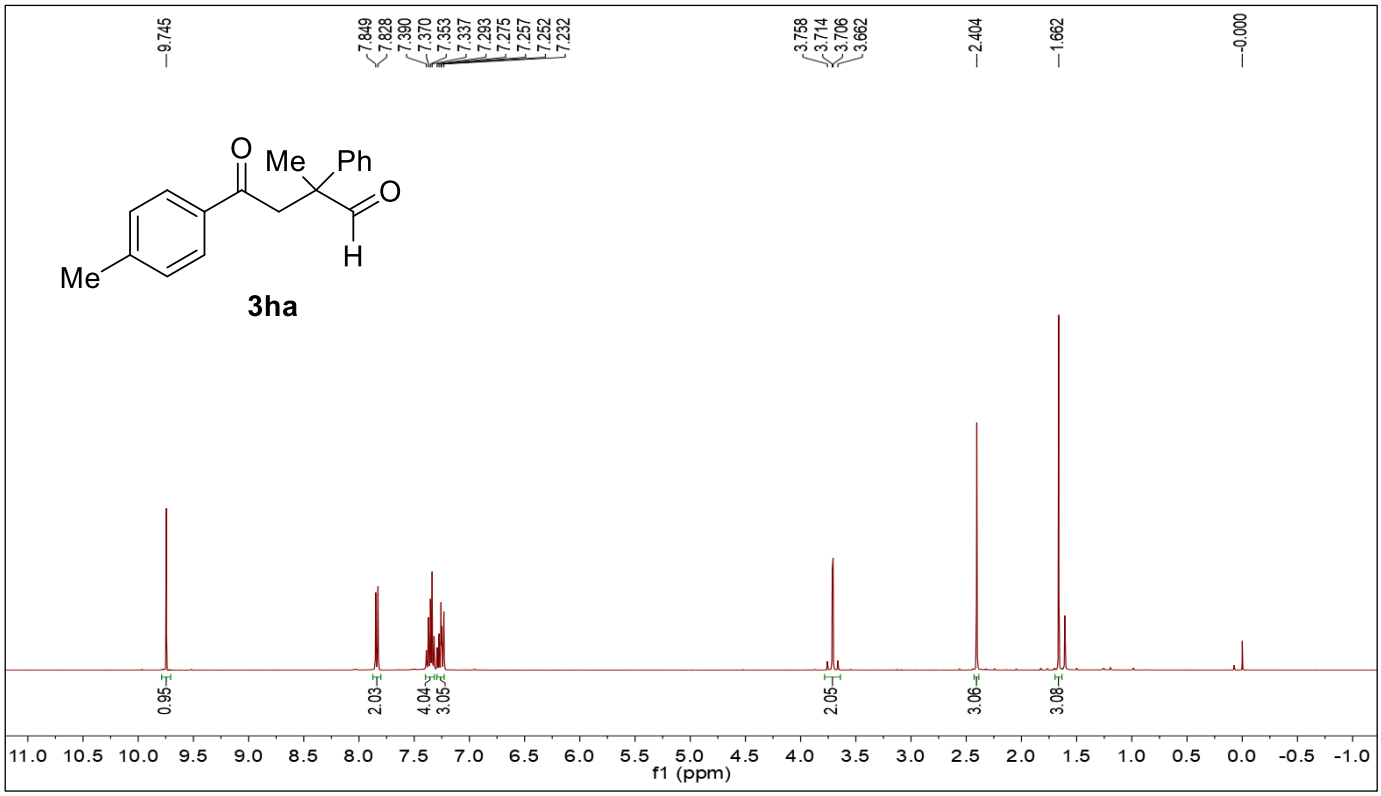

201.313
196.014
139.784
139.378
135.037
129.474
128.933
128.890
127.471
126.829
77.318
77.000
76.682
51.812
45.851
20.429
O
Me
Ph
O
H
Cl
3ea
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

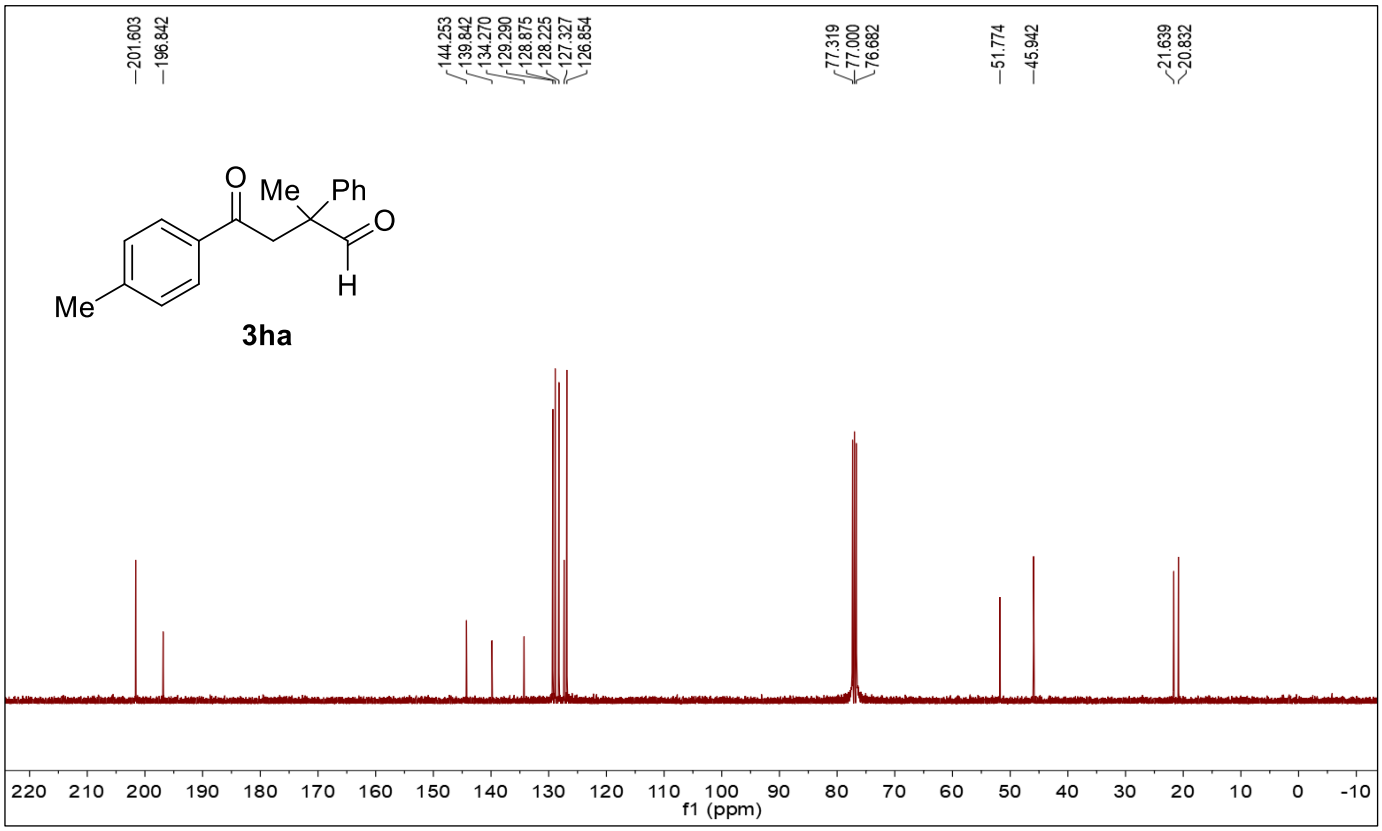

9.693
7.787
7.765
7.578
7.557
7.390
7.370
7.352
7.321
7.301
7.280
7.256
3.714
3.669
3.658
3.614
1.682
0.000
O
Me
Ph
O
H
Br
3fa
0.95
2.07
2.06
2.09
3.12
2.10
3.04
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

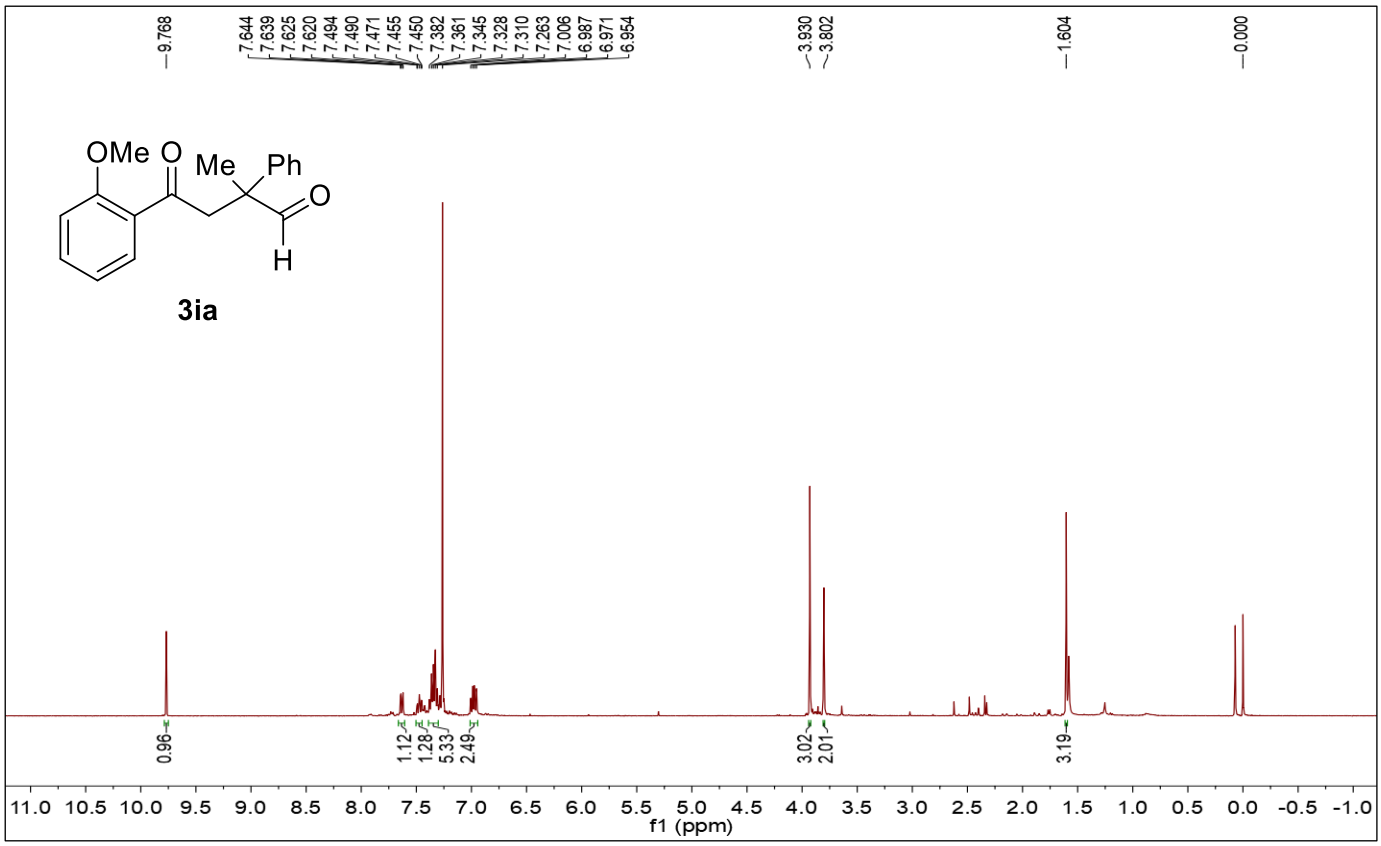

201.242
196.170
139.335
135.395
131.839
129.534
128.897
128.496
127.433
126.790
77.318
77.000
76.682
51.769
45.791
20.405
O
Me
Ph
O
H
Br
3fa
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

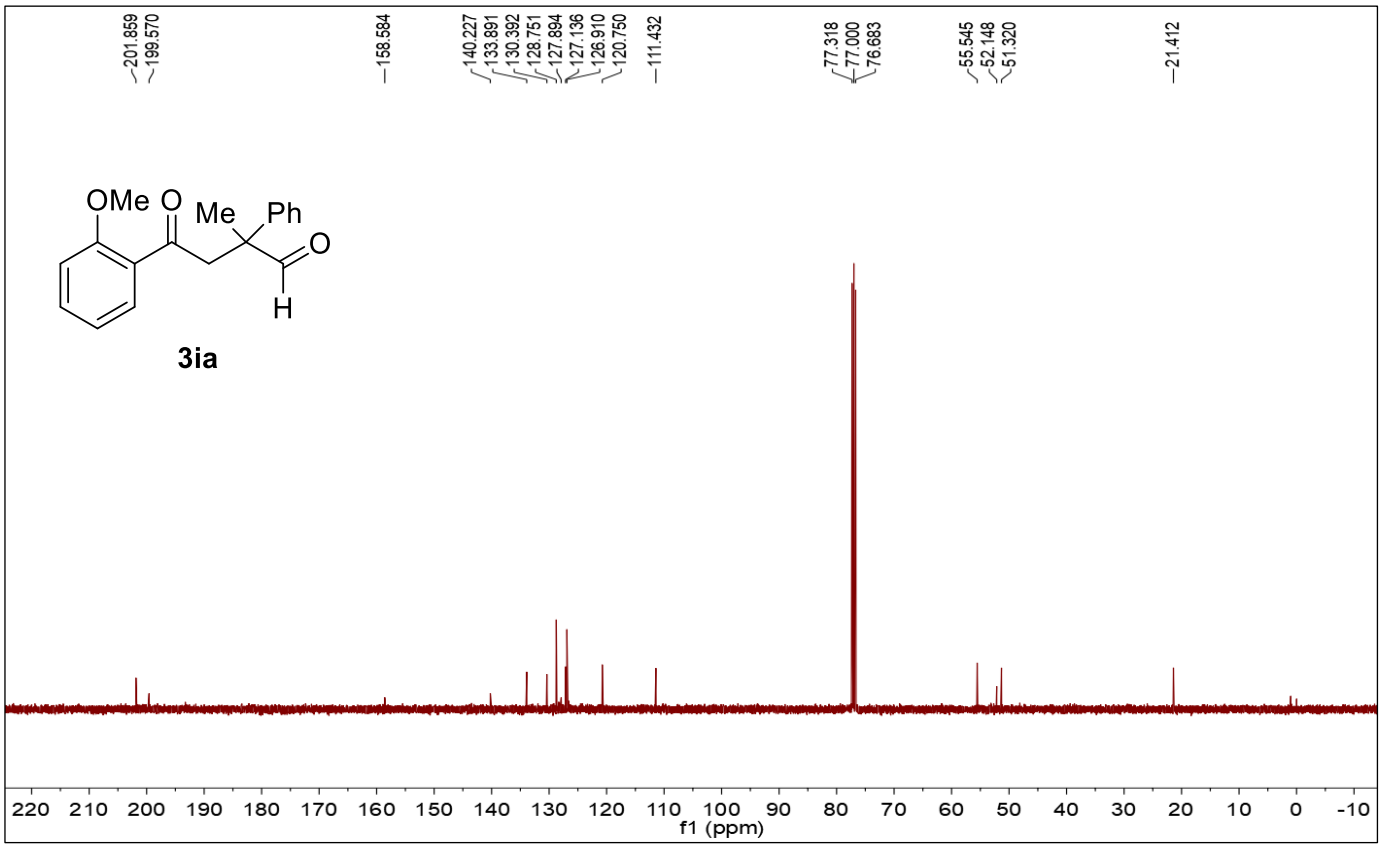

9.620
7.735
7.715
7.559
7.539
7.319
7.300
7.298
7.281
7.245
7.229
7.210
7.193
7.183
3.630
3.585
3.575
3.531
1.608
0.000
O
Me
Ph
O
H
I
3ga
0.92
2.03
2.02
2.04
3.16
2.00
3.02
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

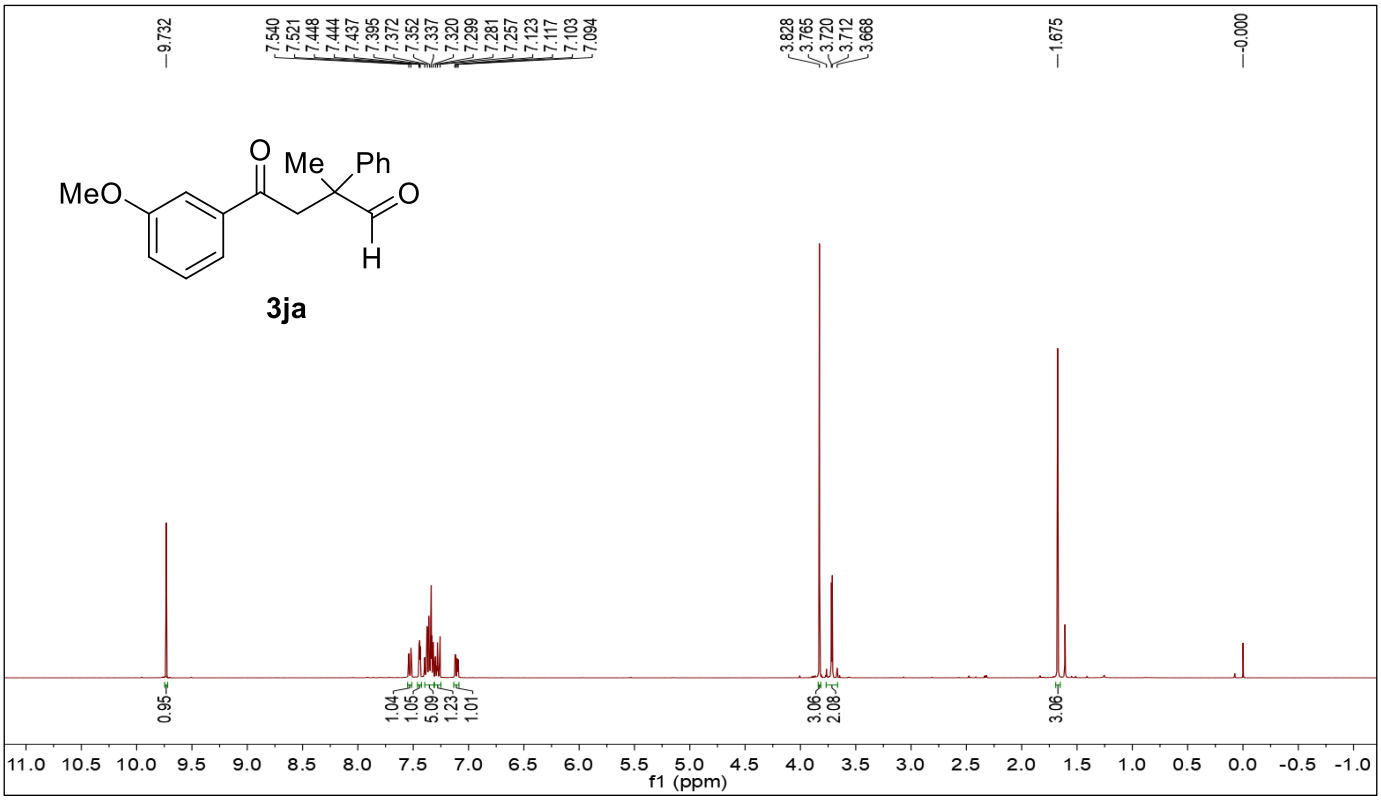

201.307
196.507
139.326
137.869
135.916
129.417
128.925
127.466
126.816
101.423
77.319
77.000
76.682
51.774
45.750
20.420
O
Me
Ph
O
H
I
3ga
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

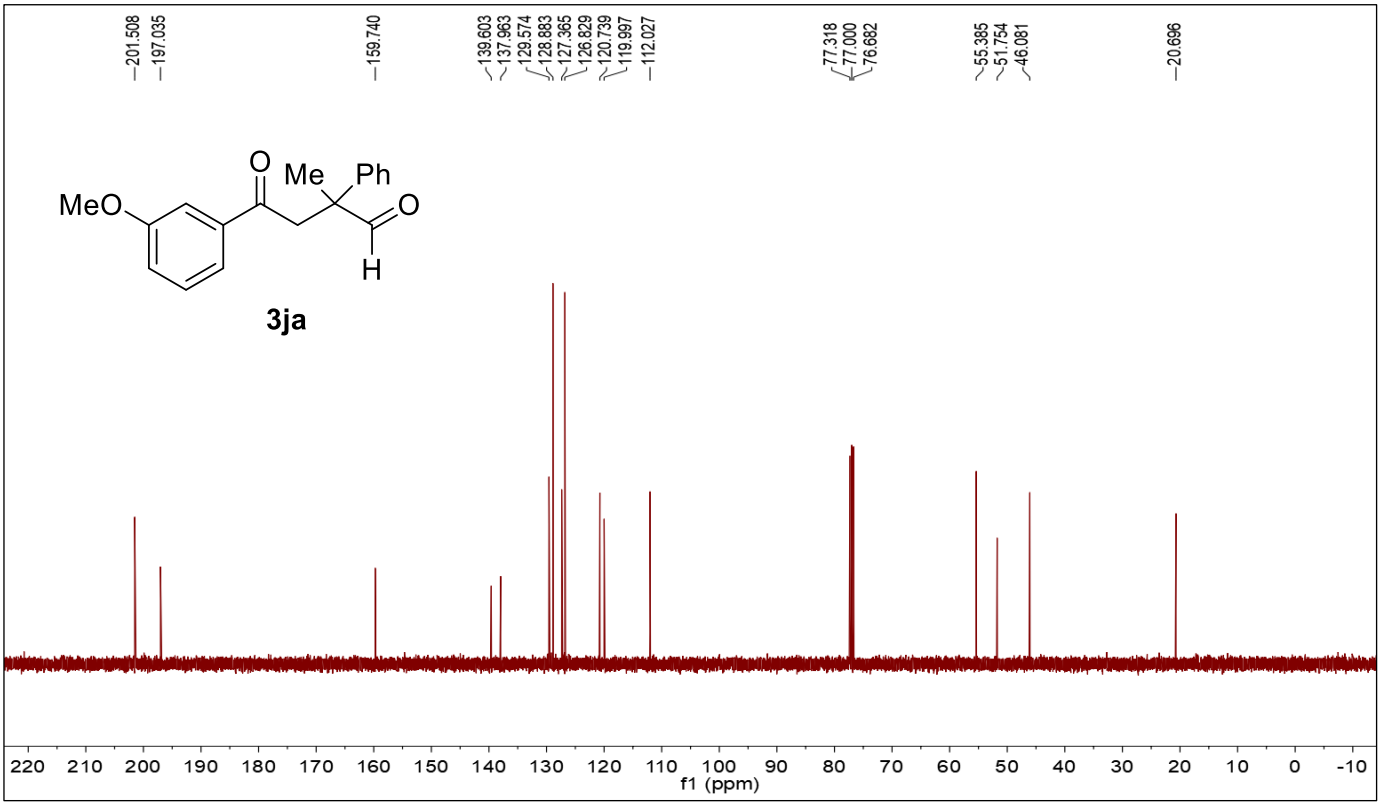

9.745
7.849
7.828
7.390
7.370
7.353
7.337
7.293
7.275
7.257
7.252
7.232
3.758
3.714
3.706
3.662
2.404
1.662
0.000
O
Me
Ph
O
H
Me
3ha
0.95
2.03
4.04
3.05
2.05
3.06
3.08
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


201.603
196.842
144.253
139.842
134.270
129.290
128.875
128.225
127.327
126.854
77.319
77.000
76.682
51.774
45.942
21.639
20.832
O
Me
Ph
O
H
Me
3ha
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


9.768
7.644
7.639
7.625
7.620
7.494
7.490
7.471
7.455
7.450
7.382
7.361
7.345
7.328
7.310
7.263
7.006
6.987
6.971
6.954
3.930
3.802
1.604
0.000
OMe
O
Me
Ph
O
H
3ia
0.96
1.12
1.28
5.33
2.49
3.02
2.01
3.19
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


201.859
199.570
158.584
140.227
133.891
130.392
128.751
127.894
127.136
126.910
120.750
111.432
77.318
77.000
76.683
55.545
52.148
51.320
21.412
OMe
O
Me
Ph
O
H
3ia
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


9.732
7.540
7.521
7.448
7.444
7.437
7.395
7.372
7.352
7.337
7.320
7.299
7.281
7.257
7.123
7.117
7.103
7.094
3.828
3.765
3.720
3.712
3.668
1.675
0.000
O
Me
Ph
MeO
O
H
3ja
0.95
1.04
1.05
5.09
1.23
1.01
3.06
2.08
3.06
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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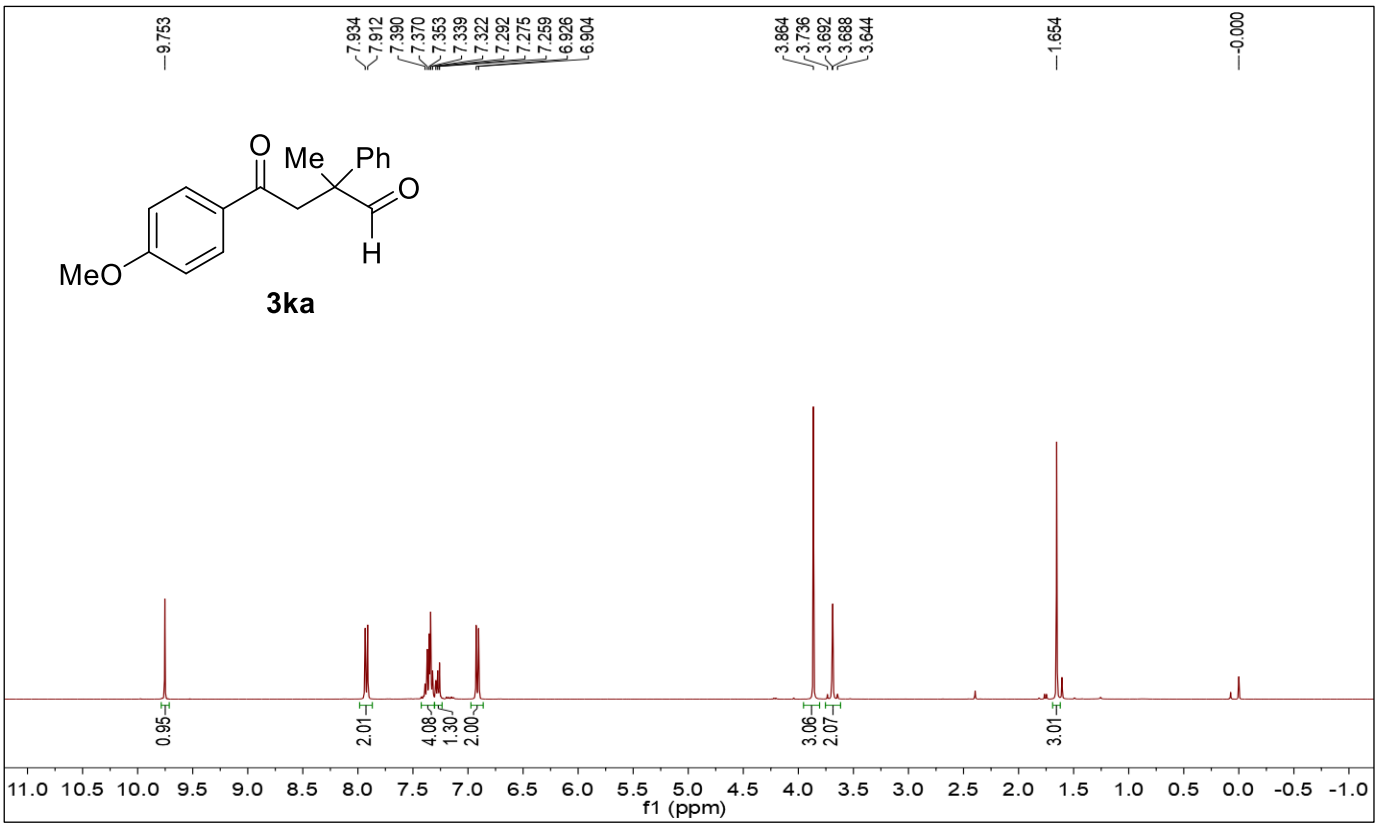
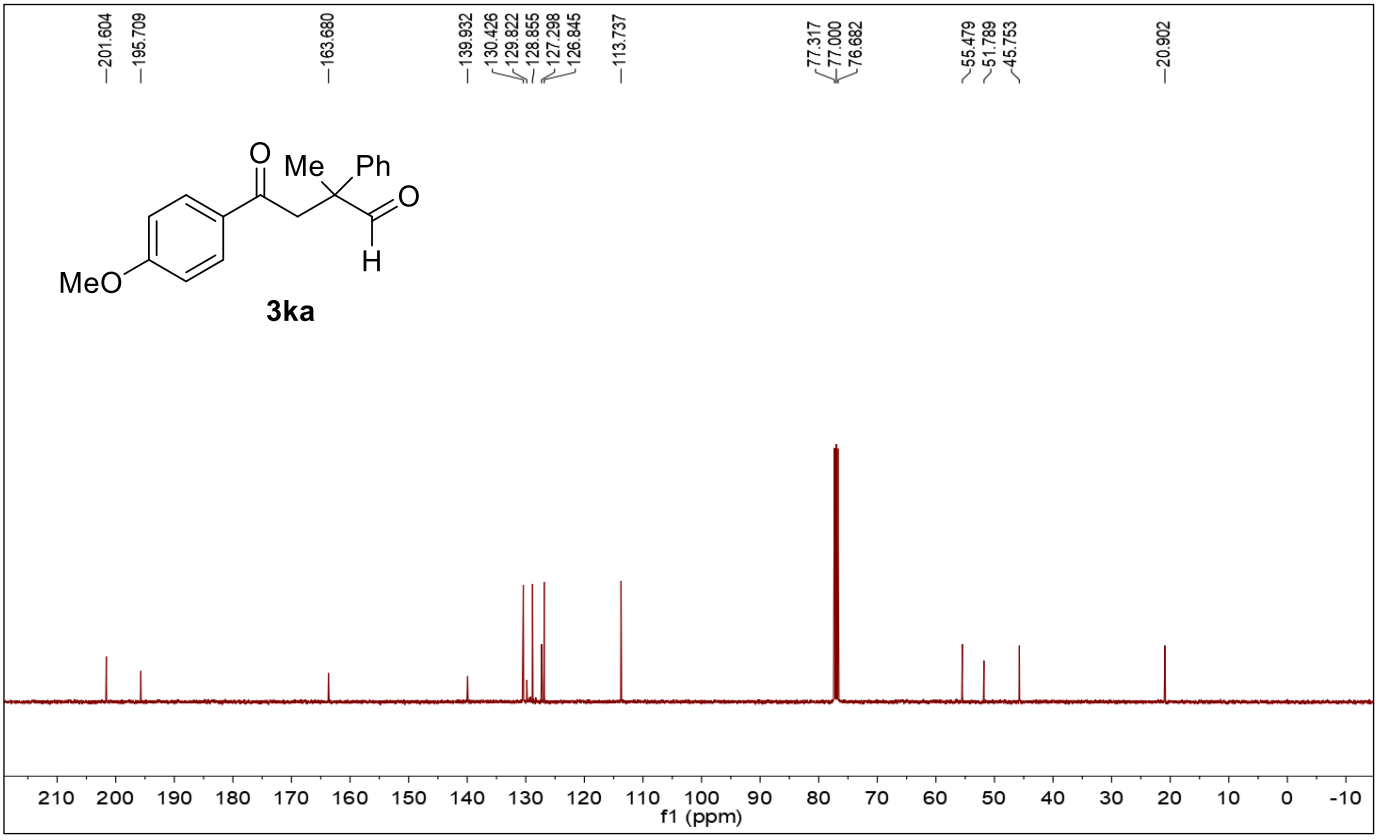
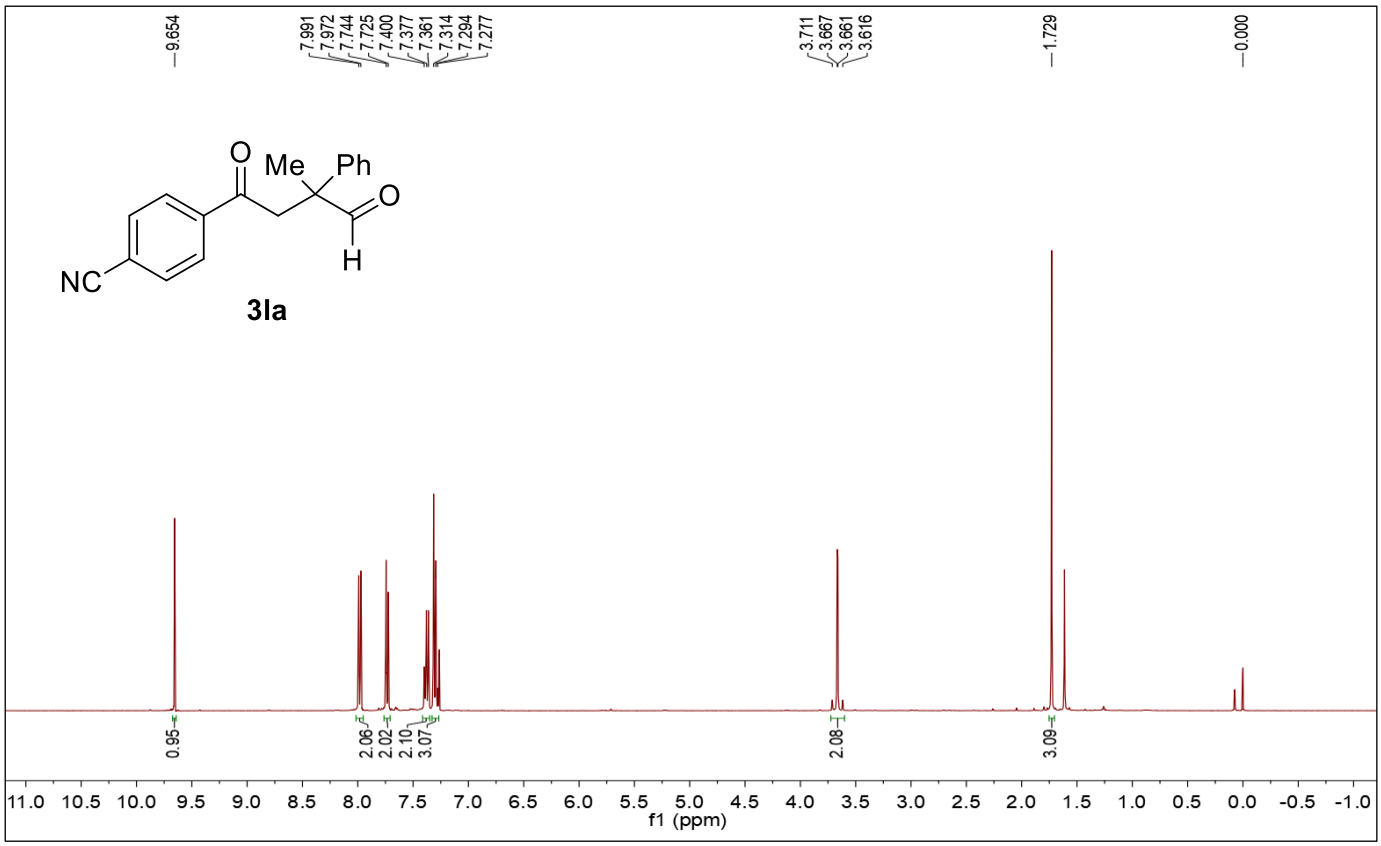
197.035
159.740
139.603
137.963
129.574
128.883
127.365
126.829
120.739
119.997
112.027
77.318
77.000
76.682
55.385
51.754
46.081
20.696
O
Me
Ph
MeO
O
H
3ja
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

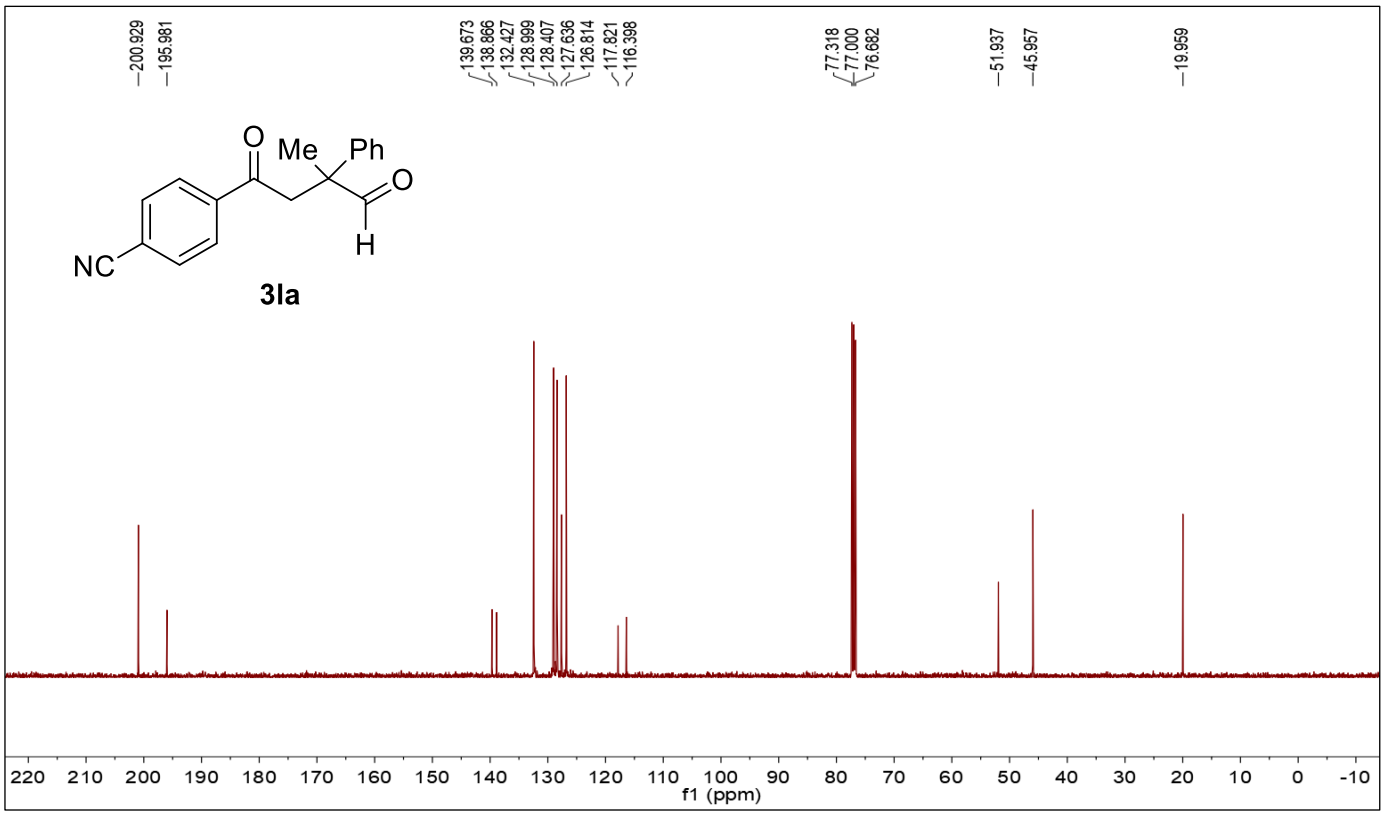

9.753
7.934
7.912
7.390
7.370
7.353
7.339
7.322
7.292
7.275
7.259
6.926
6.904
3.864
3.736
3.692
3.688
3.644
1.654
0.000
O
Me
Ph
O
H
MeO
0.95
2.01
4.08
1.30
2.00
3.06
2.07
3.01
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

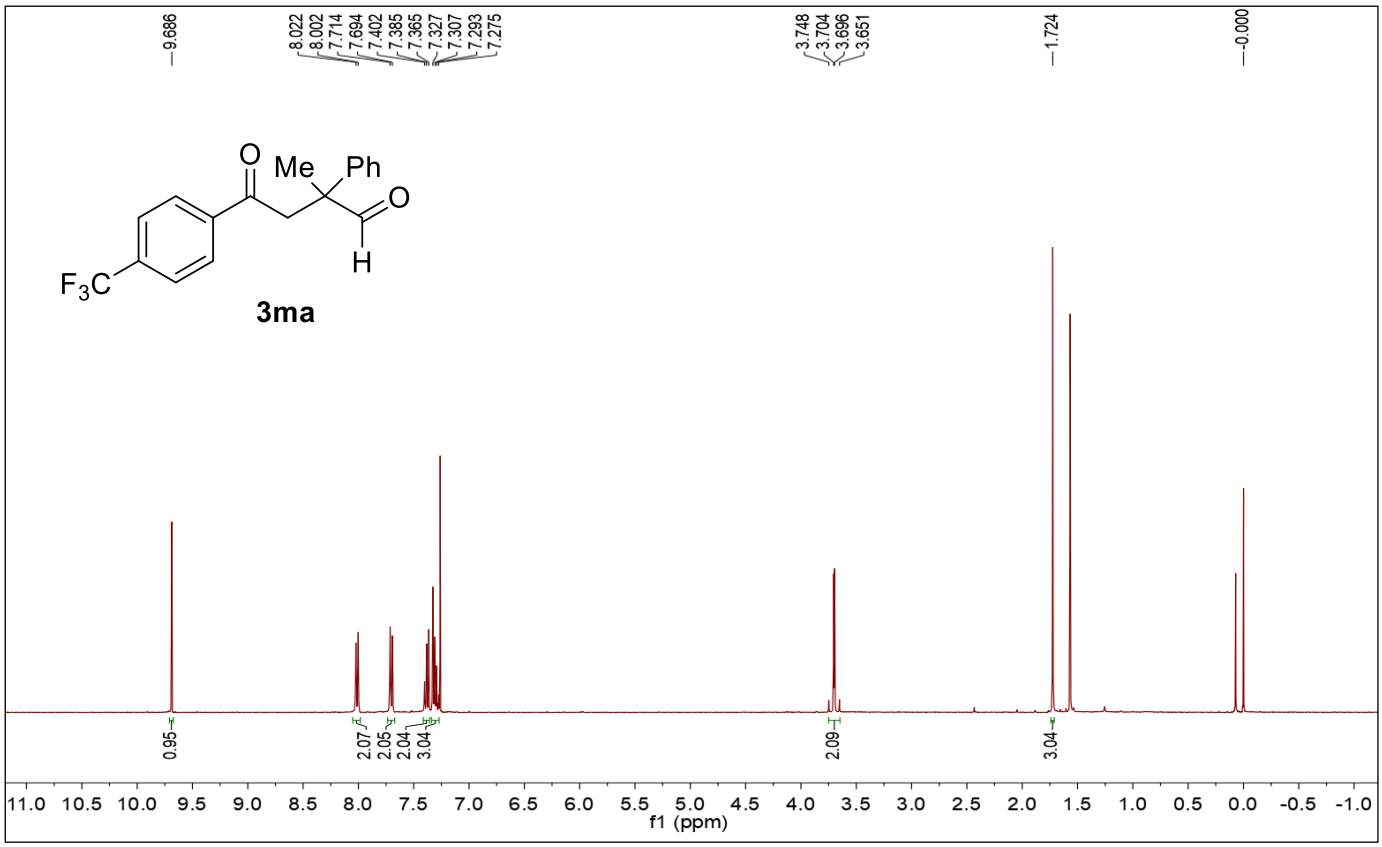
3ka

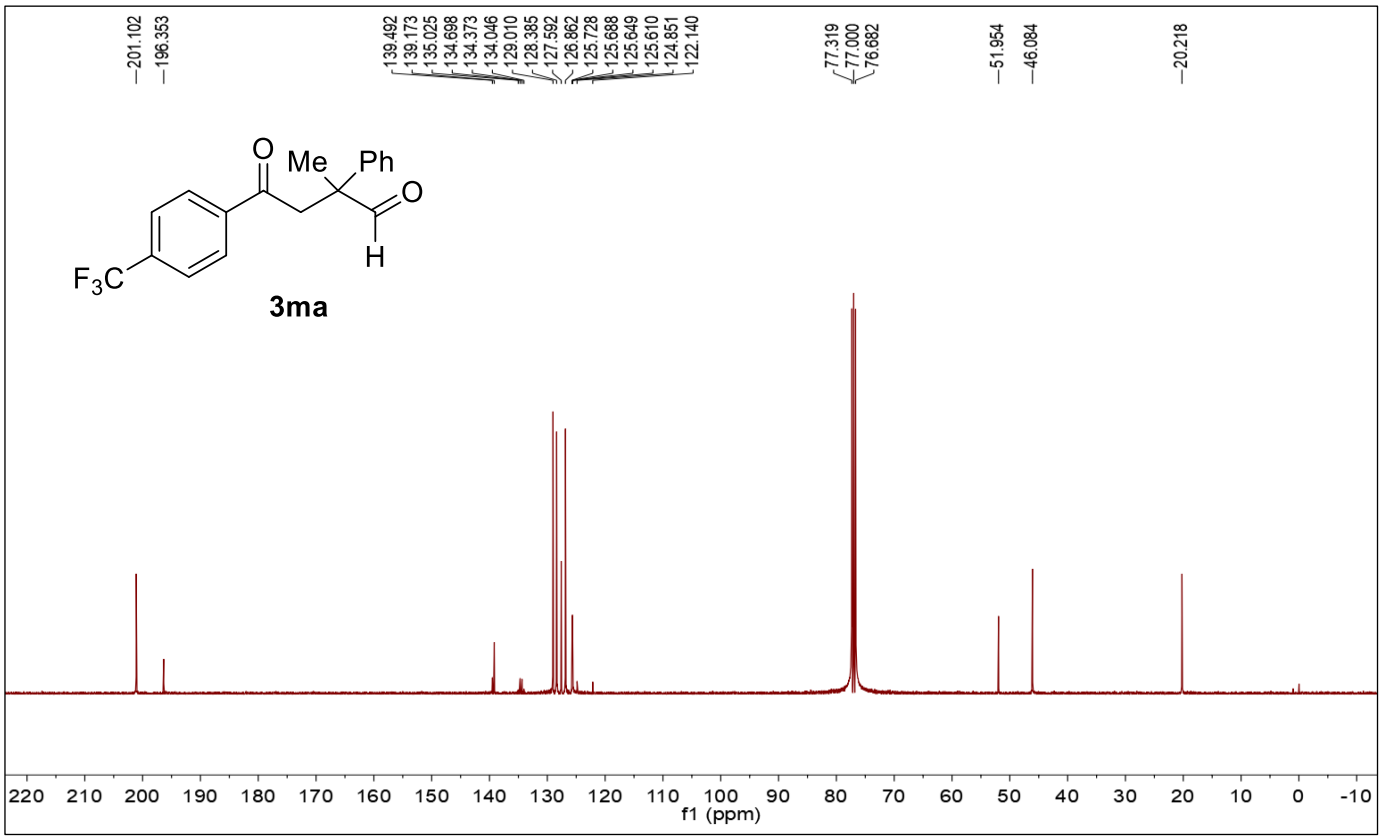

201.604
195.709
163.680
139.932
130.426
129.822
128.855
127.298
126.845
113.737
77.317
77.000
76.682
55.479
51.789
45.753
20.902
O
Me
Ph
O
H
MeO
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

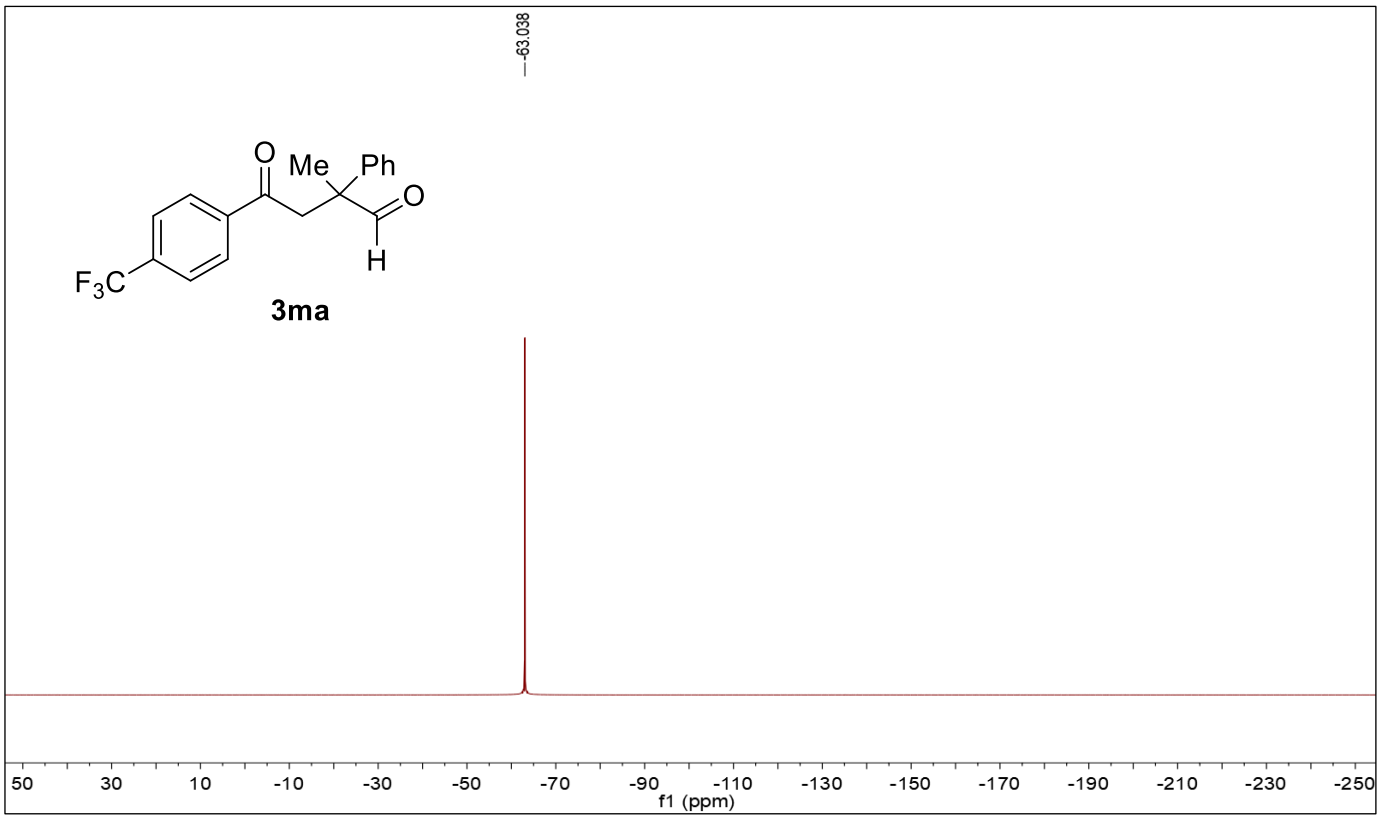
3ka

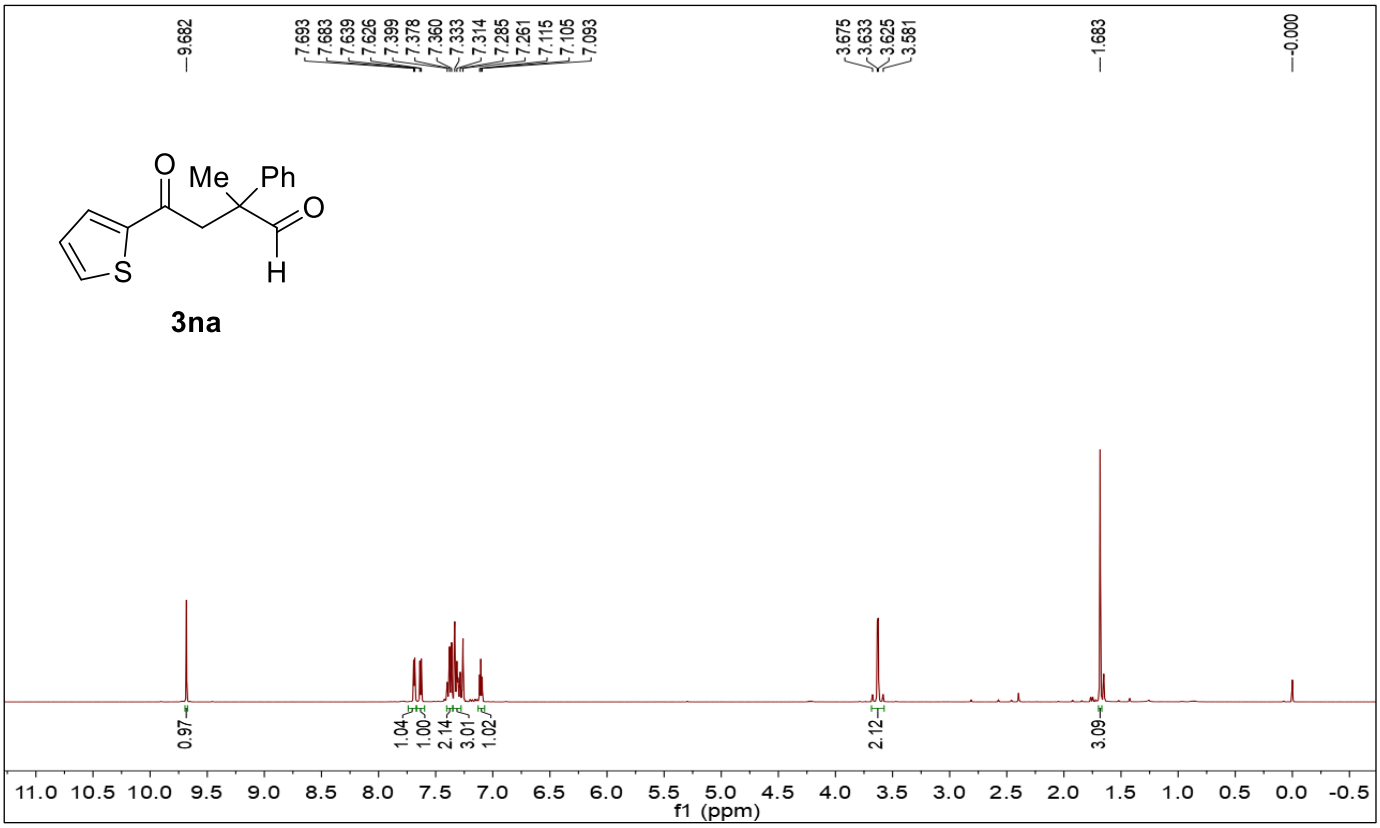

9.654
7.991
7.972
7.744
7.725
7.400
7.377
7.361
7.314
7.294
7.277
3.711
3.667
3.661
3.616
1.729
0.000
O
Me
Ph
O
H
NC
3la
0.95
2.06
2.02
2.10
3.07
2.08
3.09
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


200.929
195.981
139.673
138.866
132.427
128.999
128.407
127.636
126.814
117.821
116.398
77.318
77.000
76.682
51.937
45.957
19.959
O
Me
Ph
O
H
NC
3la
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


9.686
8.022
8.002
7.714
7.694
7.402
7.385
7.365
7.327
7.307
7.293
7.275
3.748
3.704
3.696
3.651
1.724
0.000
O
Me
Ph
O
H
F3C
3ma
0.95
2.07
2.05
2.04
3.04
2.09
3.04
f1 (ppm)


201.102
196.353
139.492
139.173
135.025
134.698
134.373
134.046
129.010
128.385
127.592
126.862
125.728
125.688
125.649
125.610
124.851
122.140
77.319
77.000
76.682
51.954
46.084
20.218
O
Me
Ph
O
H
F3C
3ma
f1 (ppm)


-63.038
O
Me
Ph
O
H
F3C
3ma
50
30
10
-10
-30
-50
-70
-90
-110
-130
-150
-170
-190
-210
-230
-250
f1 (ppm)


9.682
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7.683
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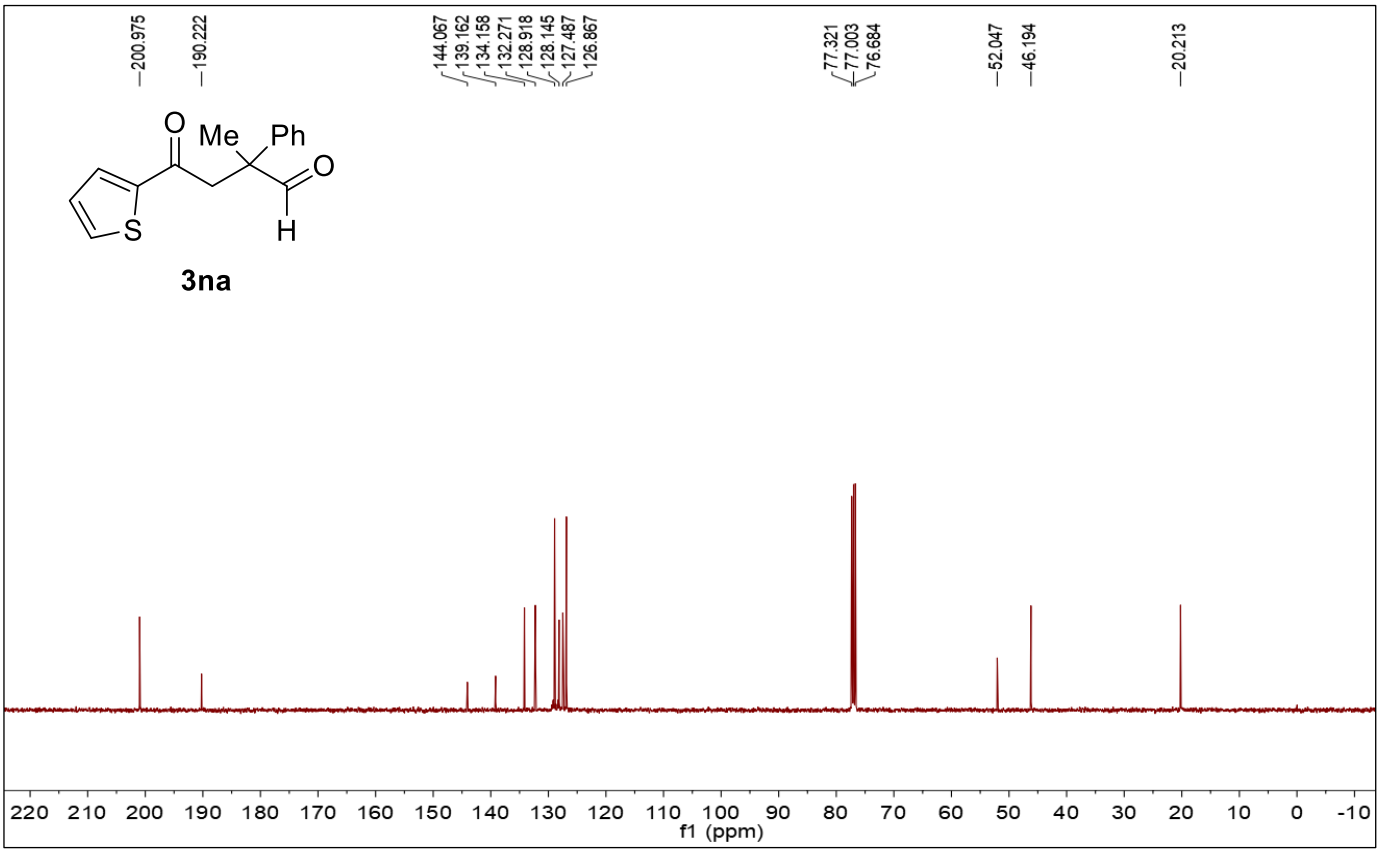
7.399
7.378
7.360
7.333
7.314
7.285
7.261
7.115
7.105
7.093
3.675
3.633
3.625
3.581
1.683
0.000
O
Me
Ph
O
S
H
3na
0.97
1.04
1.00
2.14
3.01
1.02
2.12
3.09
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5
f1 (ppm)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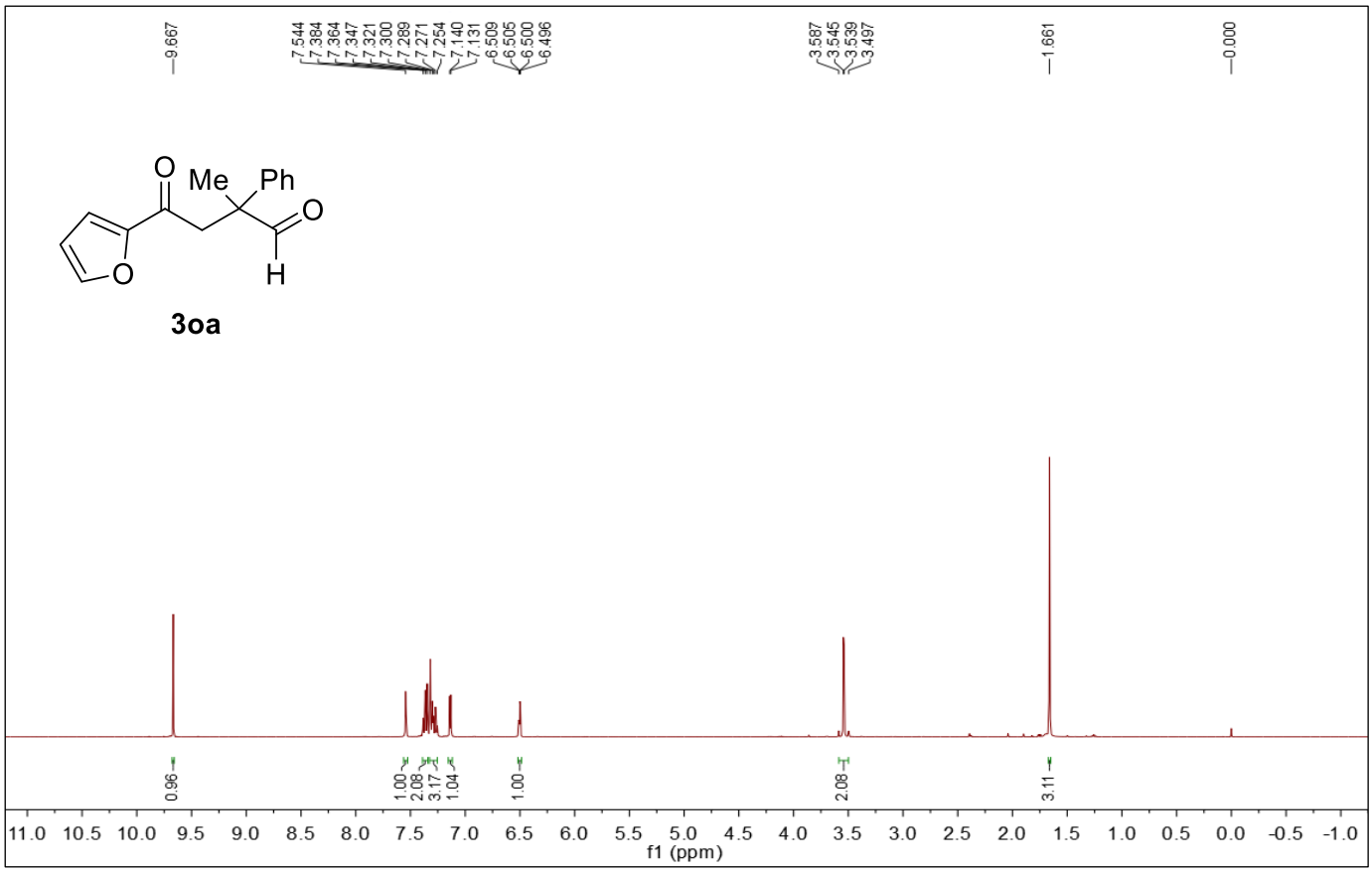
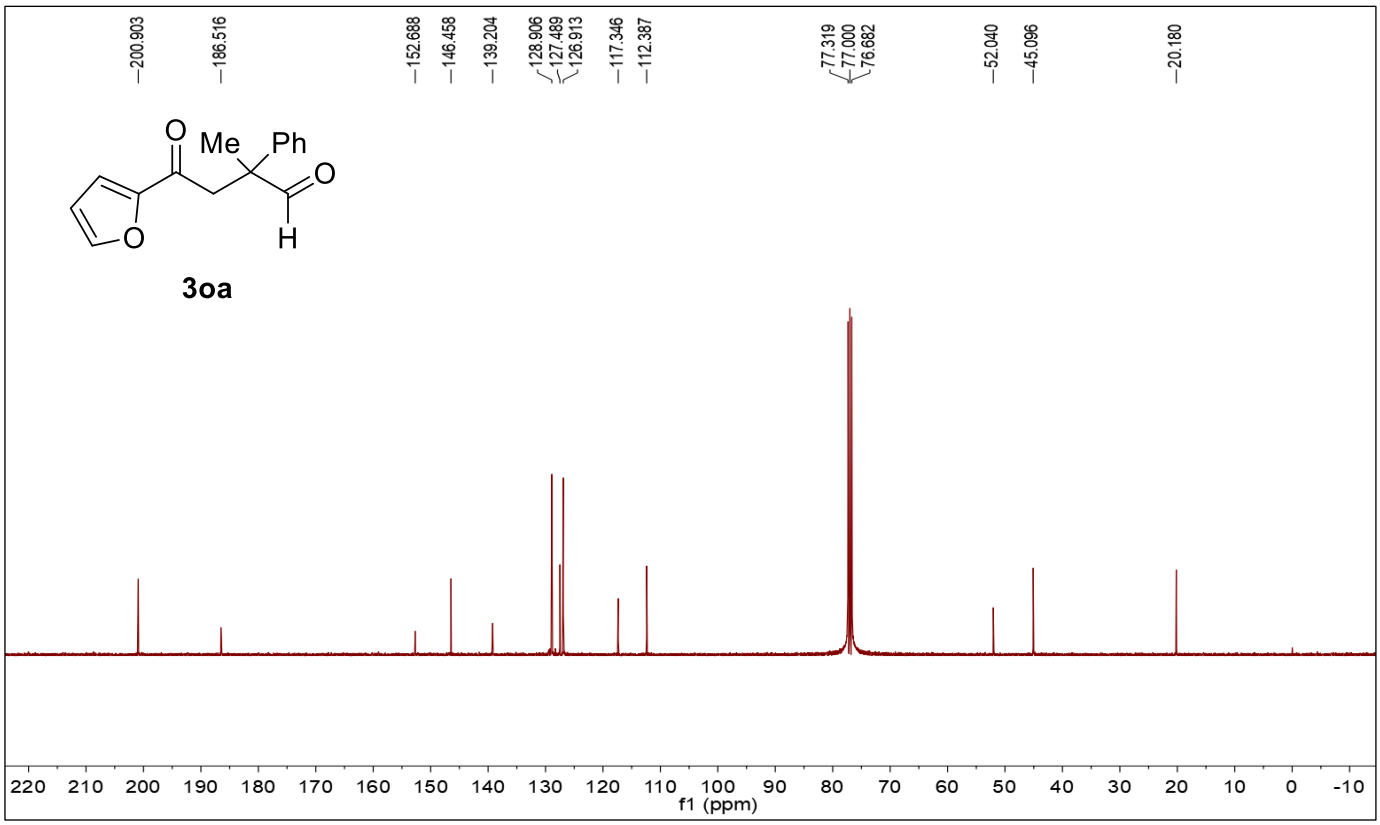
144.067
139.162
134.158
132.271
128.918
128.145
127.487
126.867
77.321
77.003
76.684
52.047
46.194
20.213
O
Me
Ph
O
S
H
3na
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


9.667
7.544
7.384
7.364
7.347
7.321
7.300
7.289
7.271
7.254
7.140
7.131
6.509
6.505
6.500
6.496
3.587
3.545
3.539
3.497
1.661
0.000
O
Me
Ph
O
O
H
3oa
0.96
1.00
2.08
3.17
1.04
1.00
2.08
3.11
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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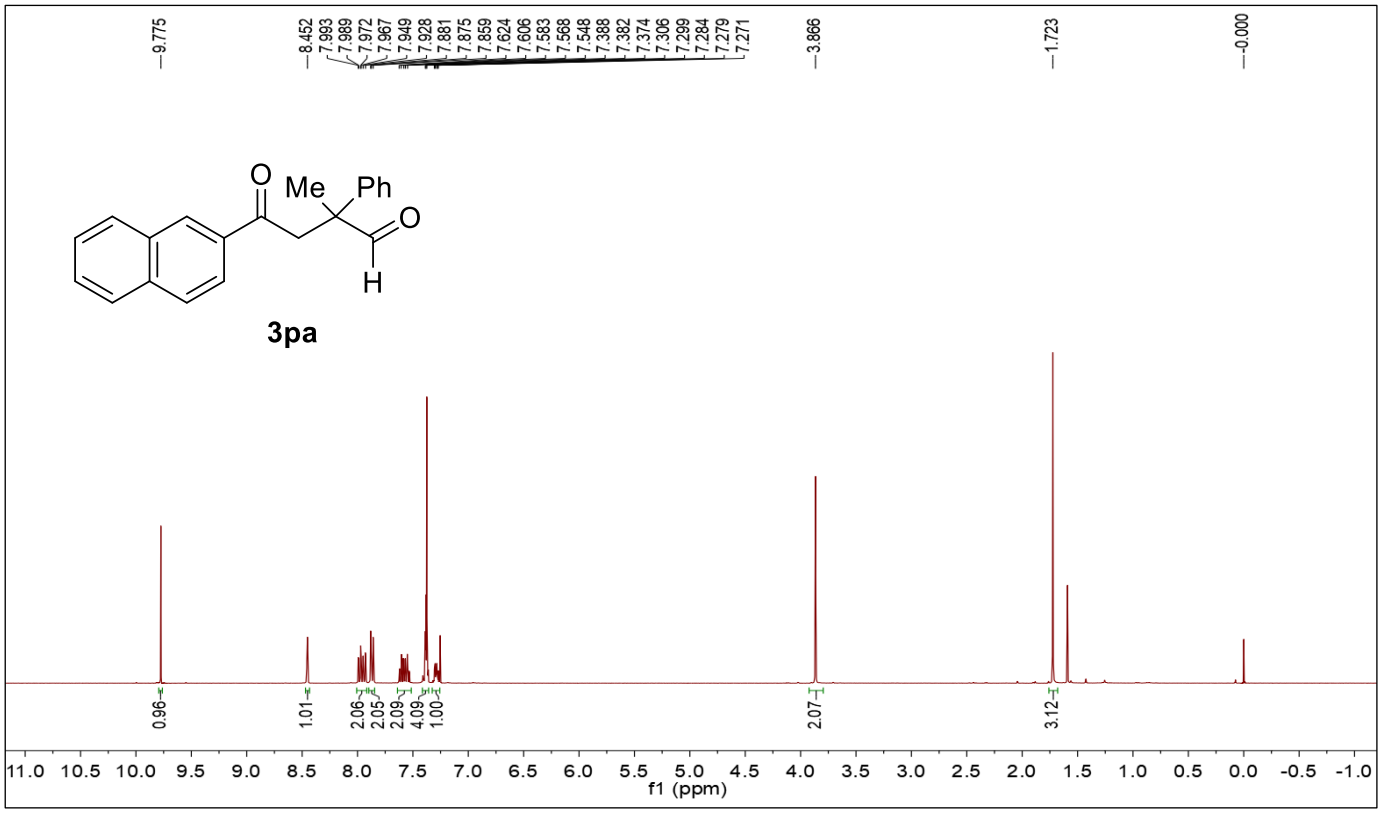
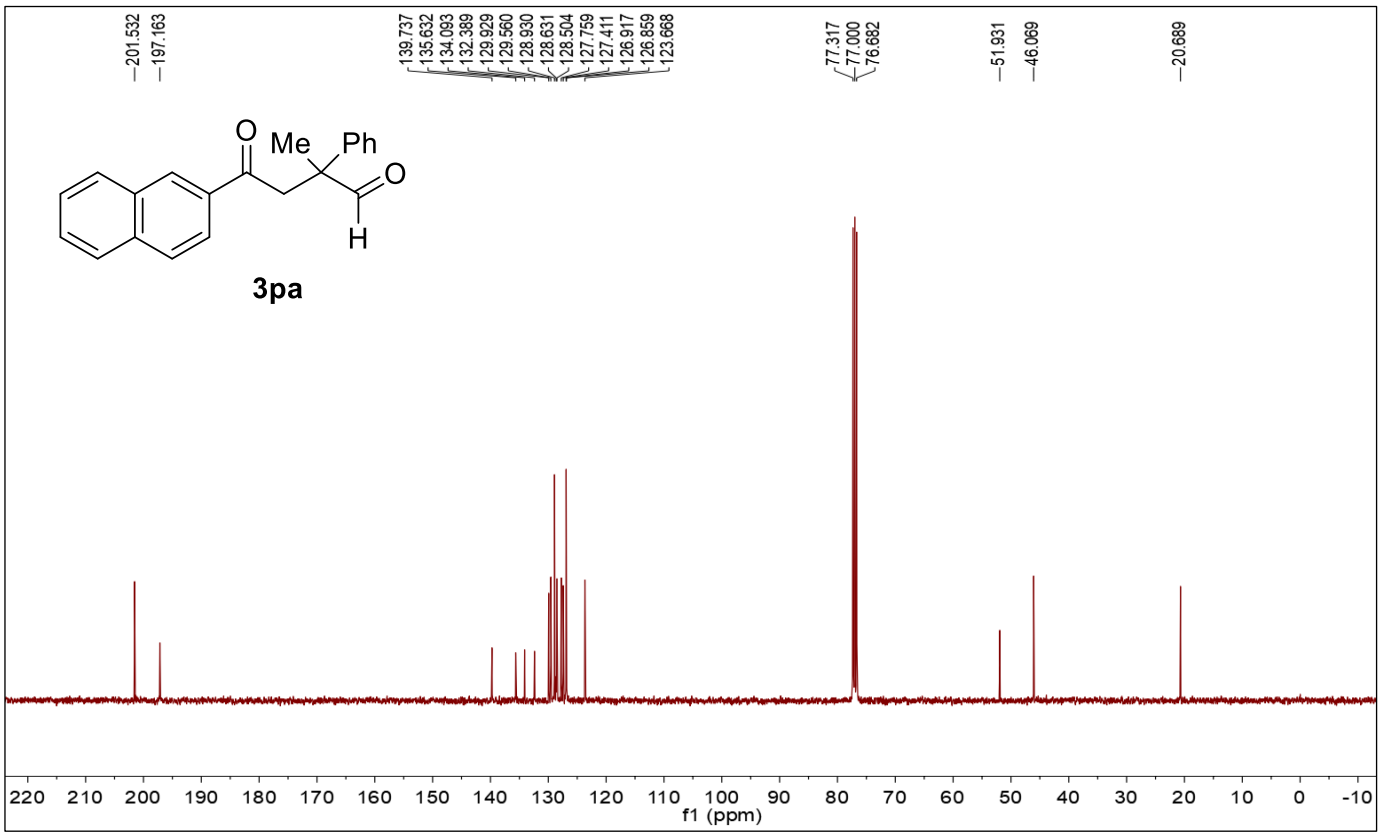
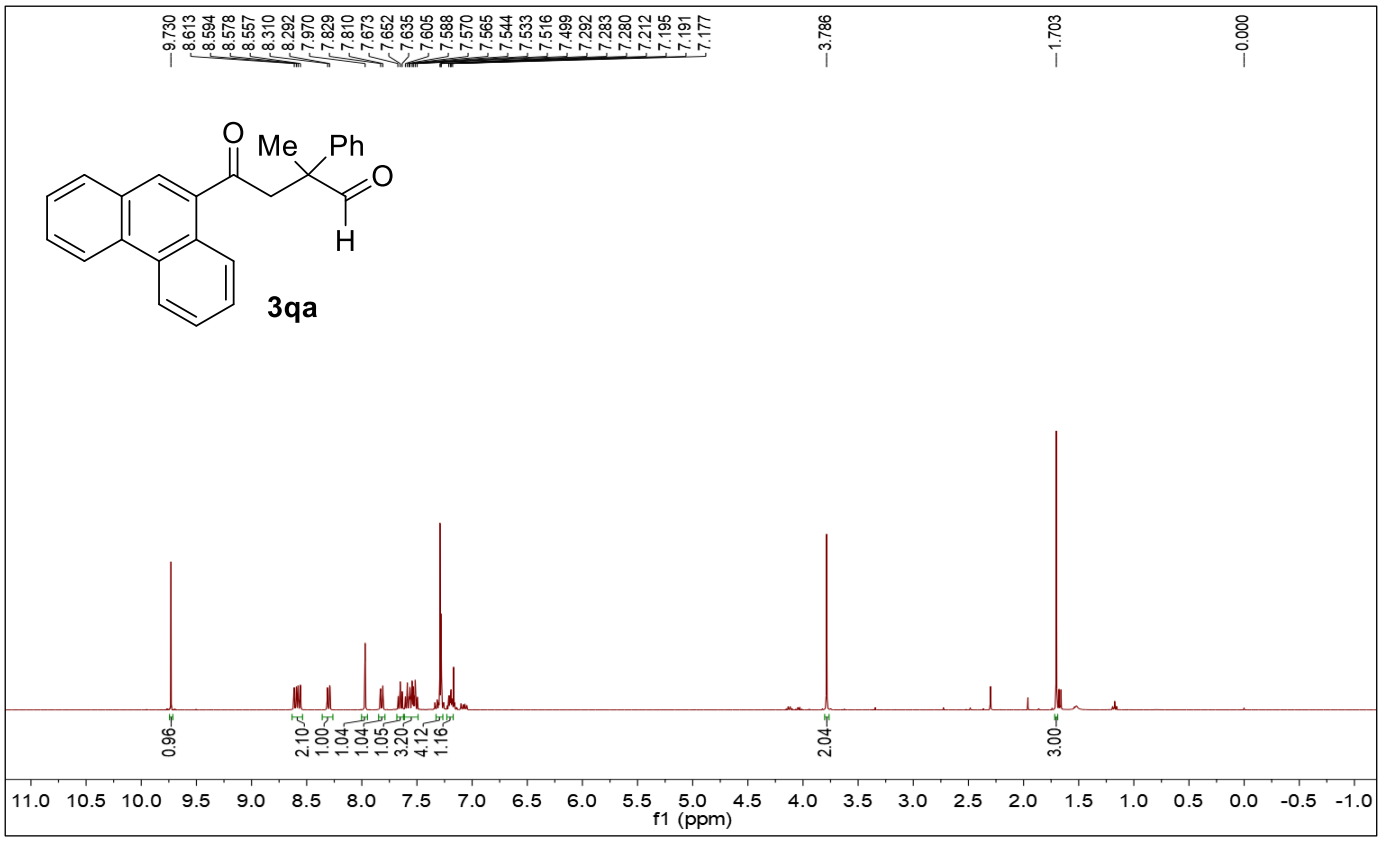
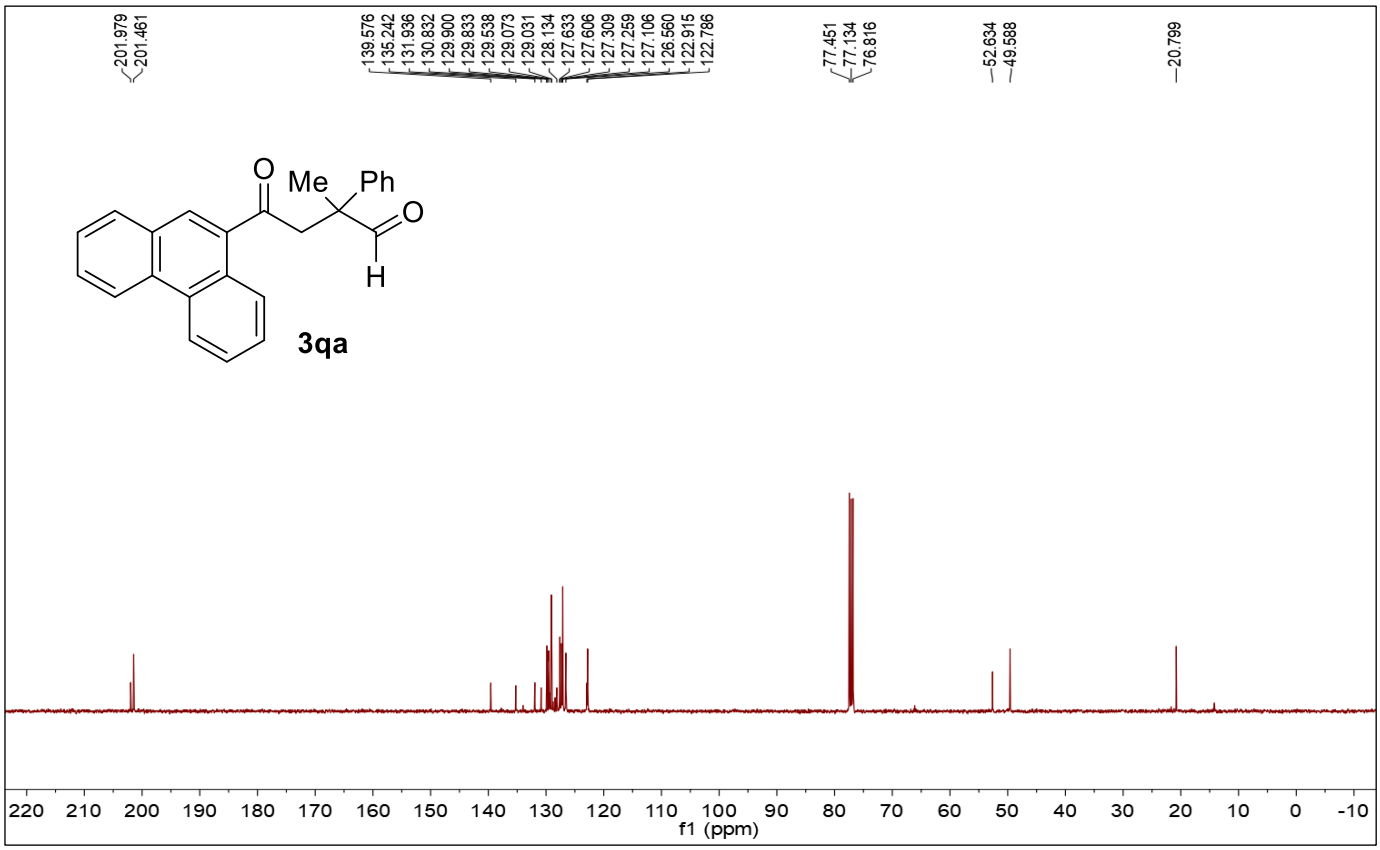
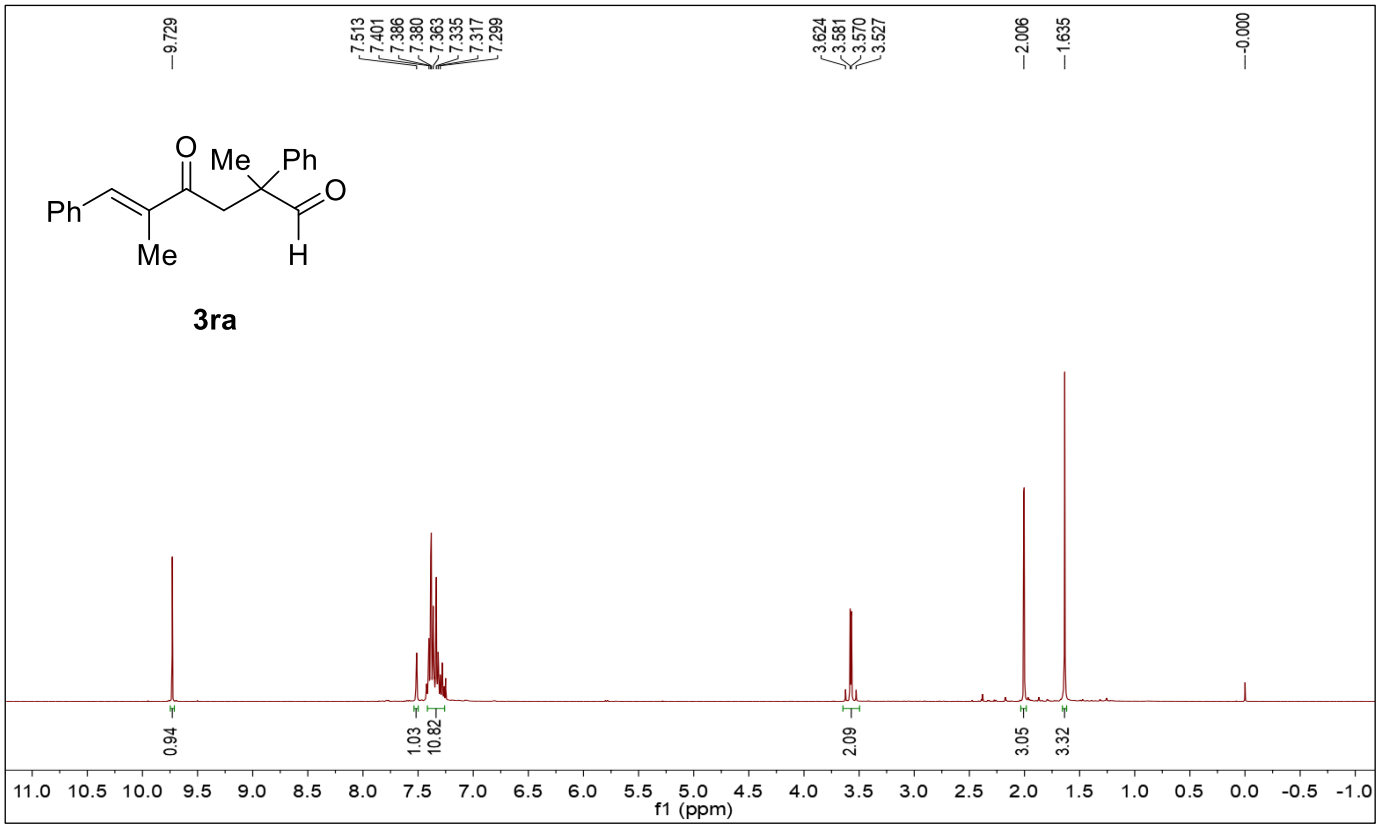
152.688
146.458
139.204
128.906
127.489
126.913
117.346
112.387
77.319
77.000
76.682
52.040
45.096
20.180
O
Me
Ph
O
O
H
3oa
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

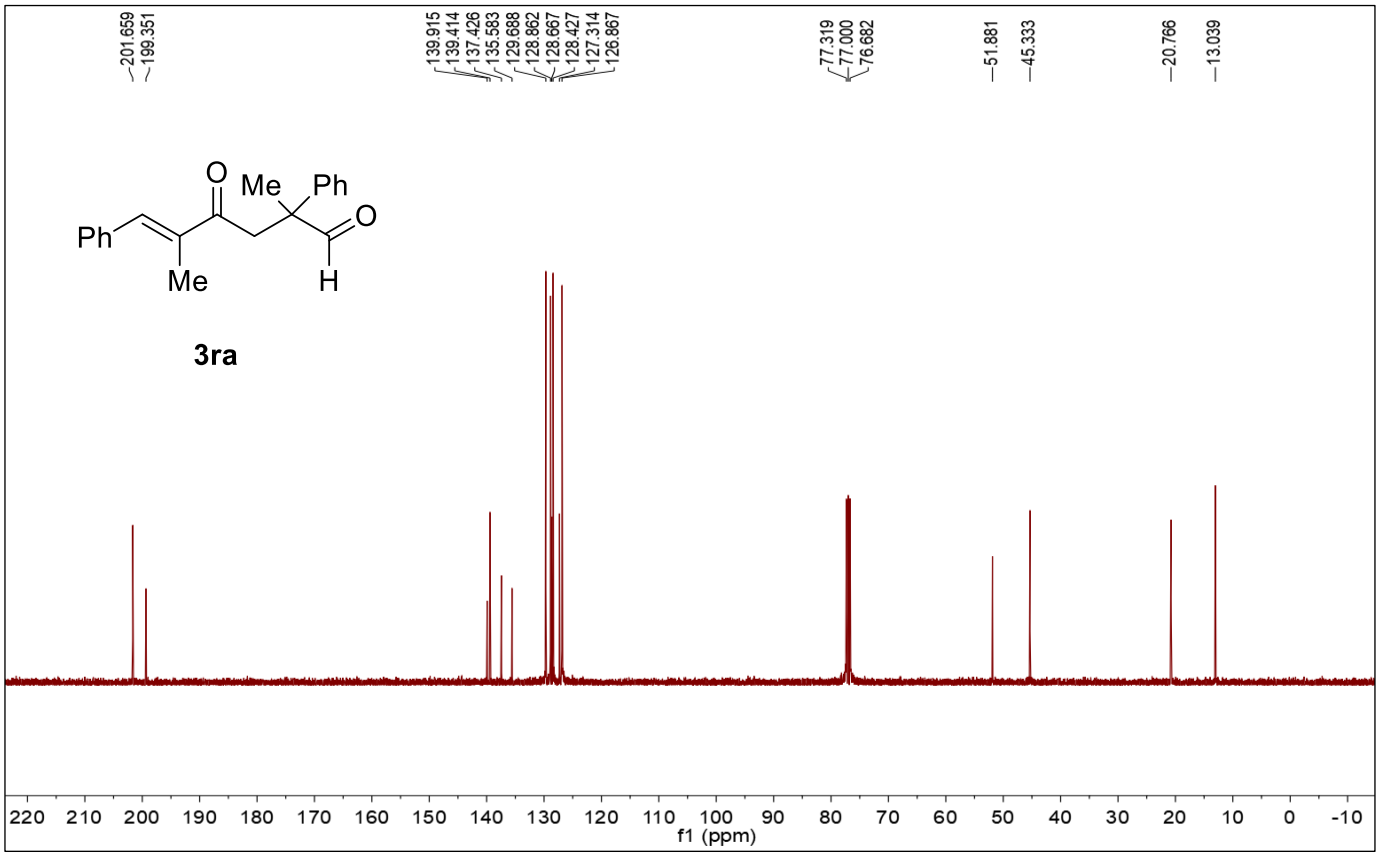

9.775
8.452
7.993
7.989
7.972
7.967
7.949
7.928
7.881
7.875
7.859
7.624
7.606
7.583
7.568
7.548
7.388
7.382
7.374
7.306
7.299
7.284
7.279
7.271
3.866
1.723
0.000
O
Me
Ph
O
H
3pa
0.96
1.01
2.06
2.05
2.09
4.09
1.00
2.07
3.12
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

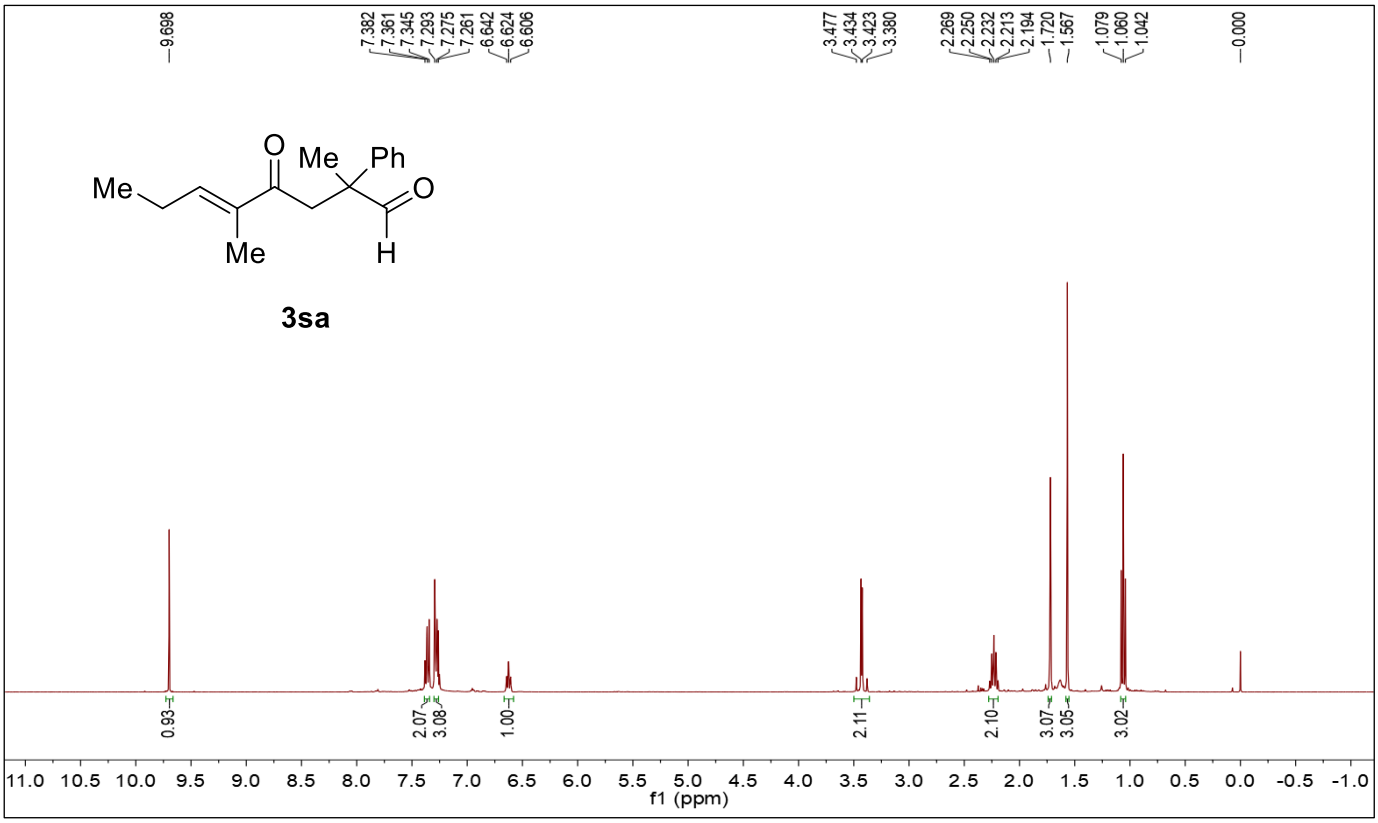

201.532
197.163
139.737
135.632
134.093
132.389
129.929
129.560
128.930
128.631
128.504
127.759
127.411
126.917
126.859
123.868
77.317
77.000
76.682
51.931
46.069
20.689
O
Me
Ph
O
H
3pa
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

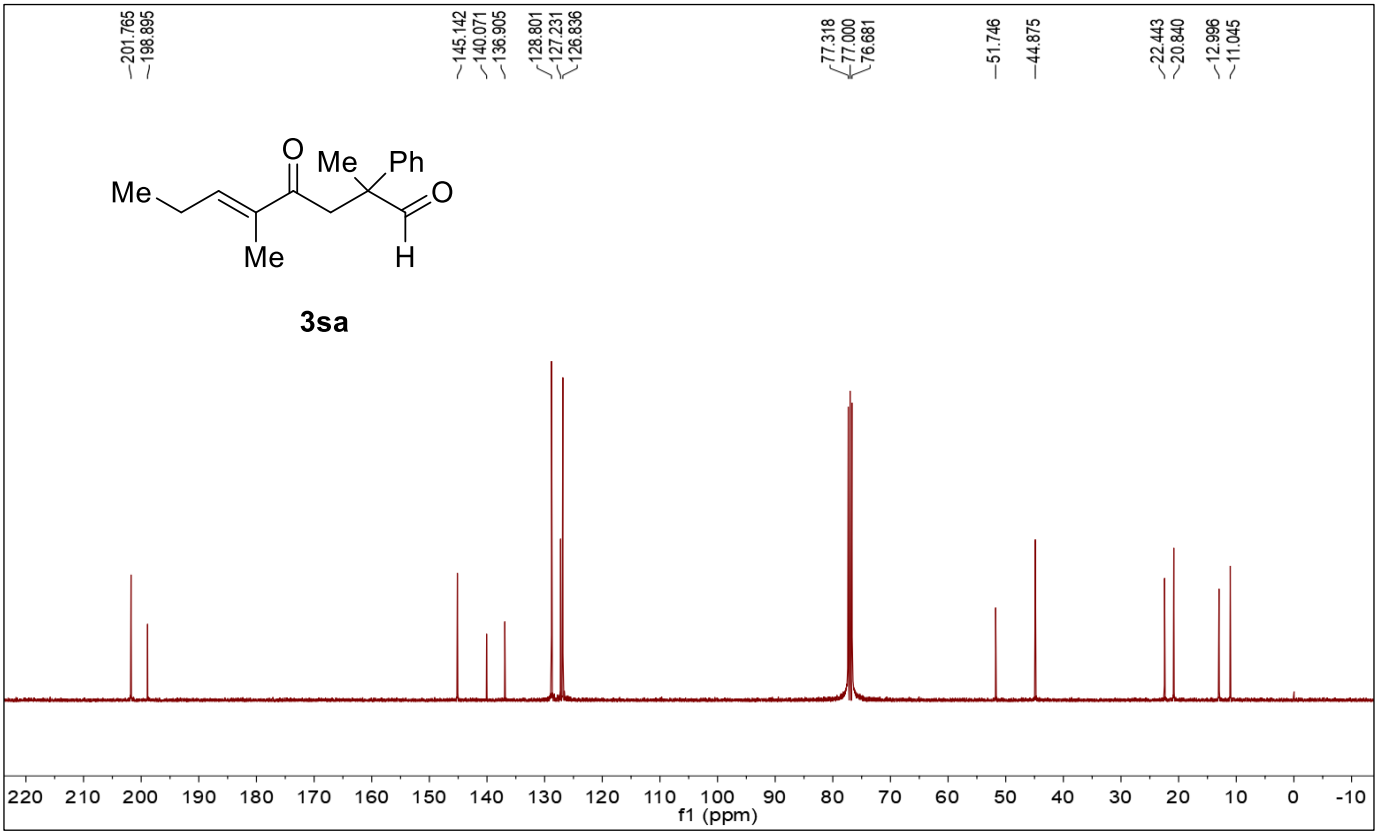

9.730
8.613
8.594
8.578
8.557
8.310
8.292
7.970
7.829
7.810
7.673
7.652
7.635
7.605
7.588
7.570
7.565
7.544
7.533
7.516
7.499
7.292
7.283
7.280
7.212
7.195
7.191
7.177
3.786
1.703
0.000
O
Me
Ph
O
H
3qa
0.96
2.10
1.00
1.04
1.04
1.05
3.20
4.12
1.16
2.04
3.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

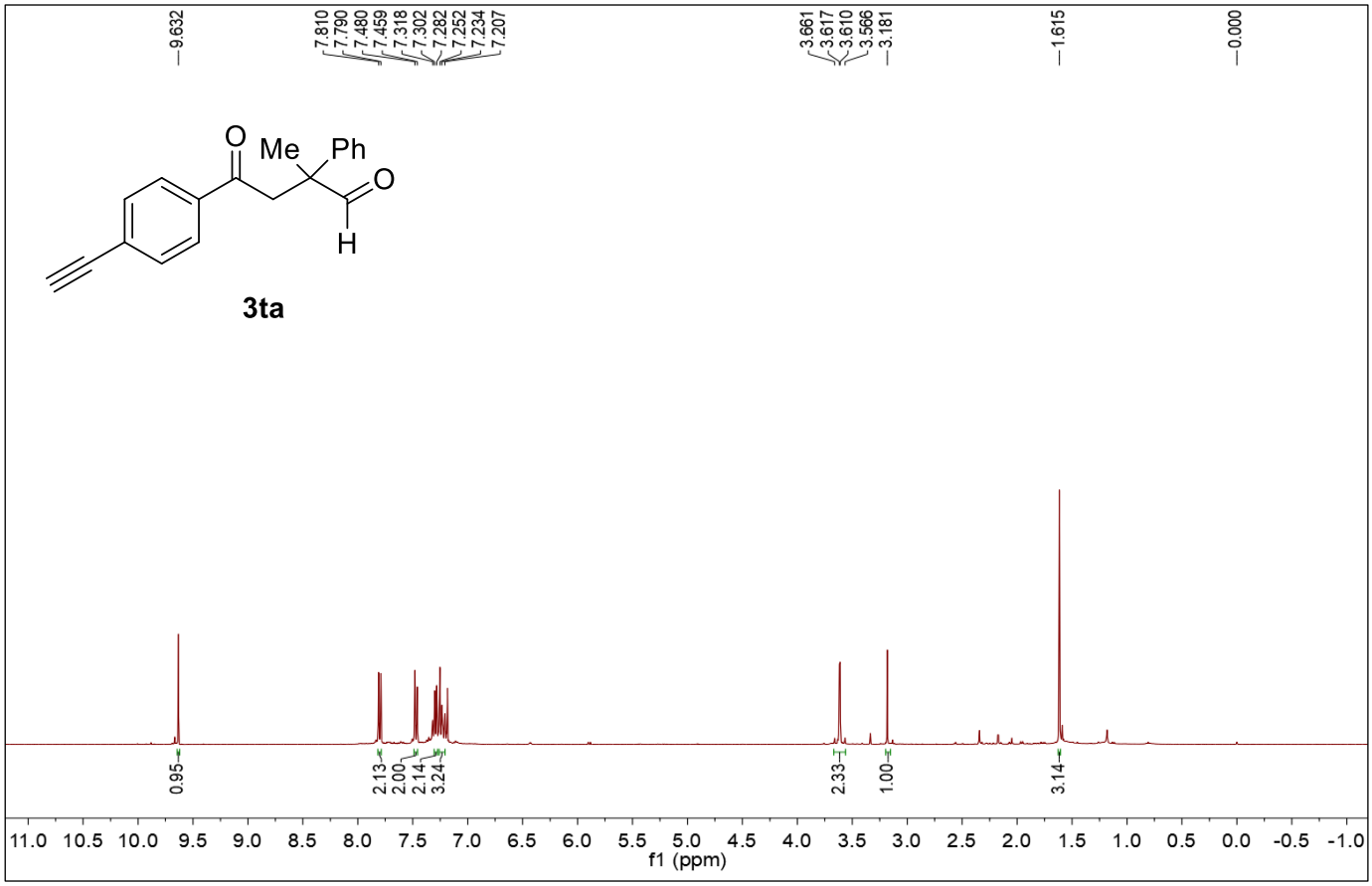

201.979
201.461
139.576
135.242
131.936
130.832
129.900
129.833
129.538
129.073
129.031
128.134
127.633
127.606
127.309
127.259
127.106
126.560
122.915
122.786
77.451
77.134
76.816
52.634
49.588
20.799
O
Me
Ph
O
H
3qa
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

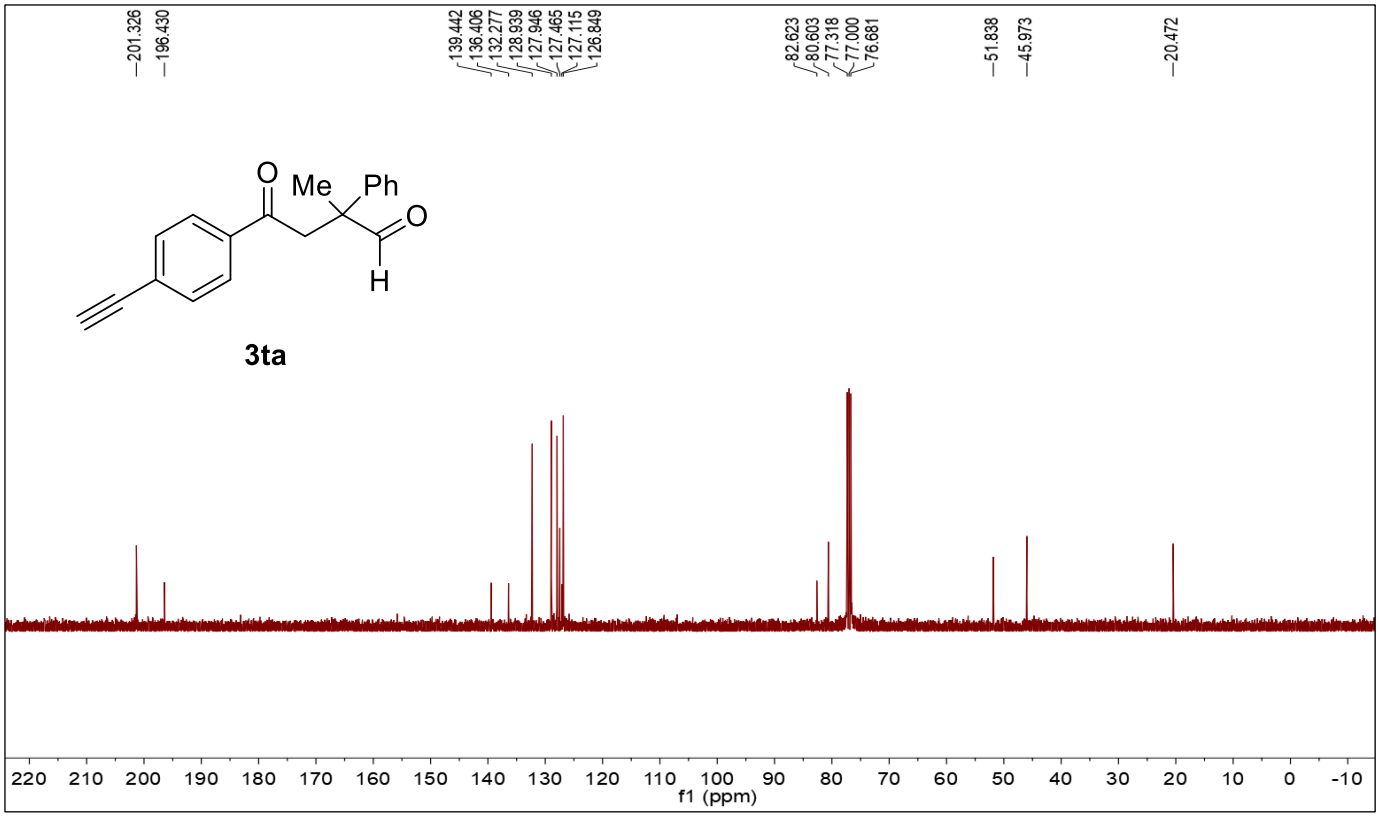

9.729
7.513
7.401
7.386
7.380
7.363
7.335
7.317
7.299
3.624
3.581
3.570
3.527
2.006
1.635
0.000
O
Me
Ph
Ph
O
Me
H
3ra
0.94
1.03
10.82
2.09
3.05
3.32
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


201.659
199.351
139.915
139.414
137.426
135.583
129.688
128.862
128.667
128.427
127.314
126.867
77.319
77.000
76.682
51.881
45.333
20.766
13.039
O
Me
Ph
Ph
O
Me
H
3ra
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

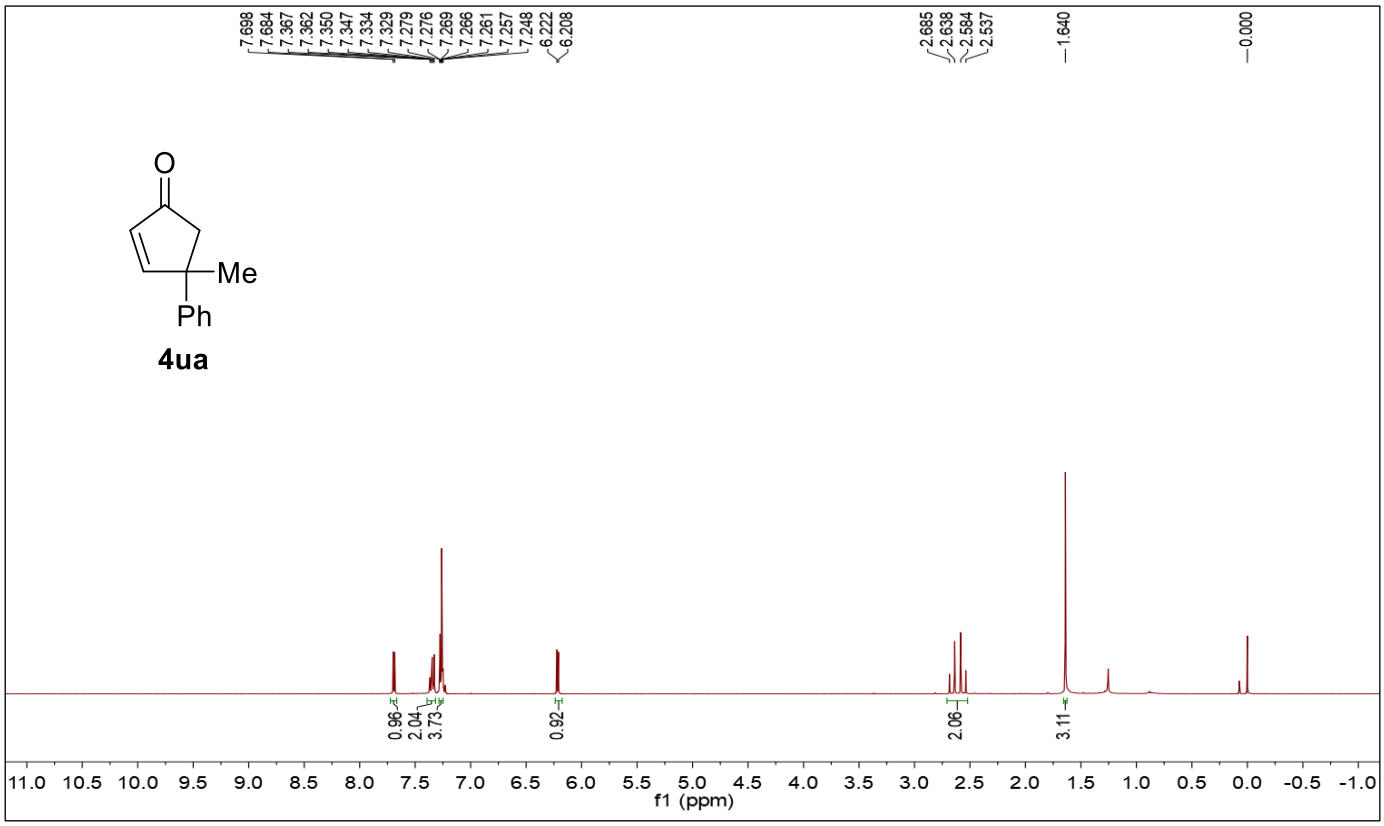

9.698
7.382
7.361
7.345
7.293
7.275
7.261
6.642
6.624
6.606
3.477
3.434
3.423
3.380
2.269
2.250
2.232
2.213
2.194
1.720
1.567
1.079
1.060
1.042
0.000
O
Me
Ph
Me
O
Me
H
3sa
0.93
2.07
3.08
1.00
2.11
2.10
3.07
3.05
3.02
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

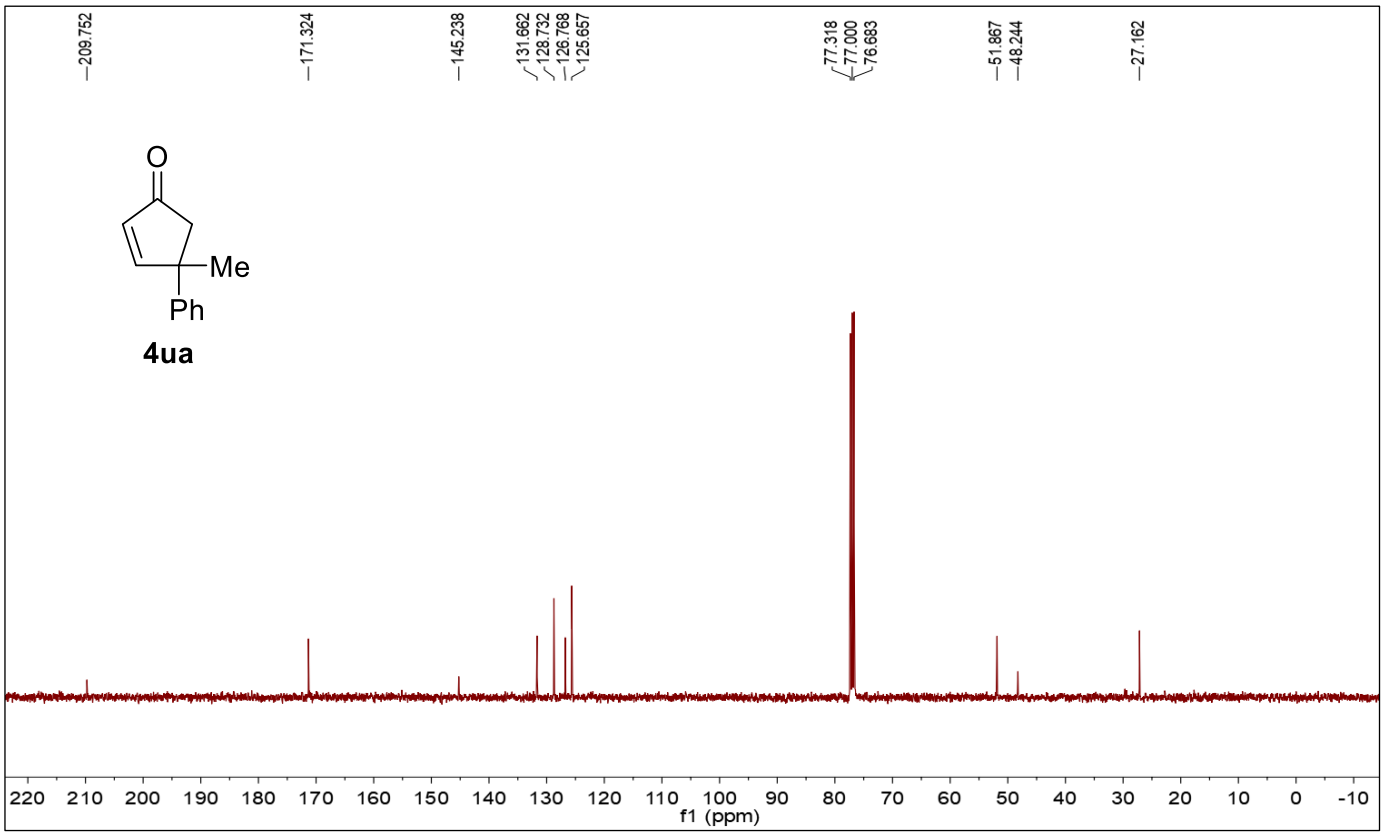

201.765
198.895
145.142
140.071
136.905
128.801
127.231
126.836
77.318
77.000
76.681
51.746
44.875
22.443
20.840
12.996
11.045
O
Me
Ph
Me
O
Me
H
3sa
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

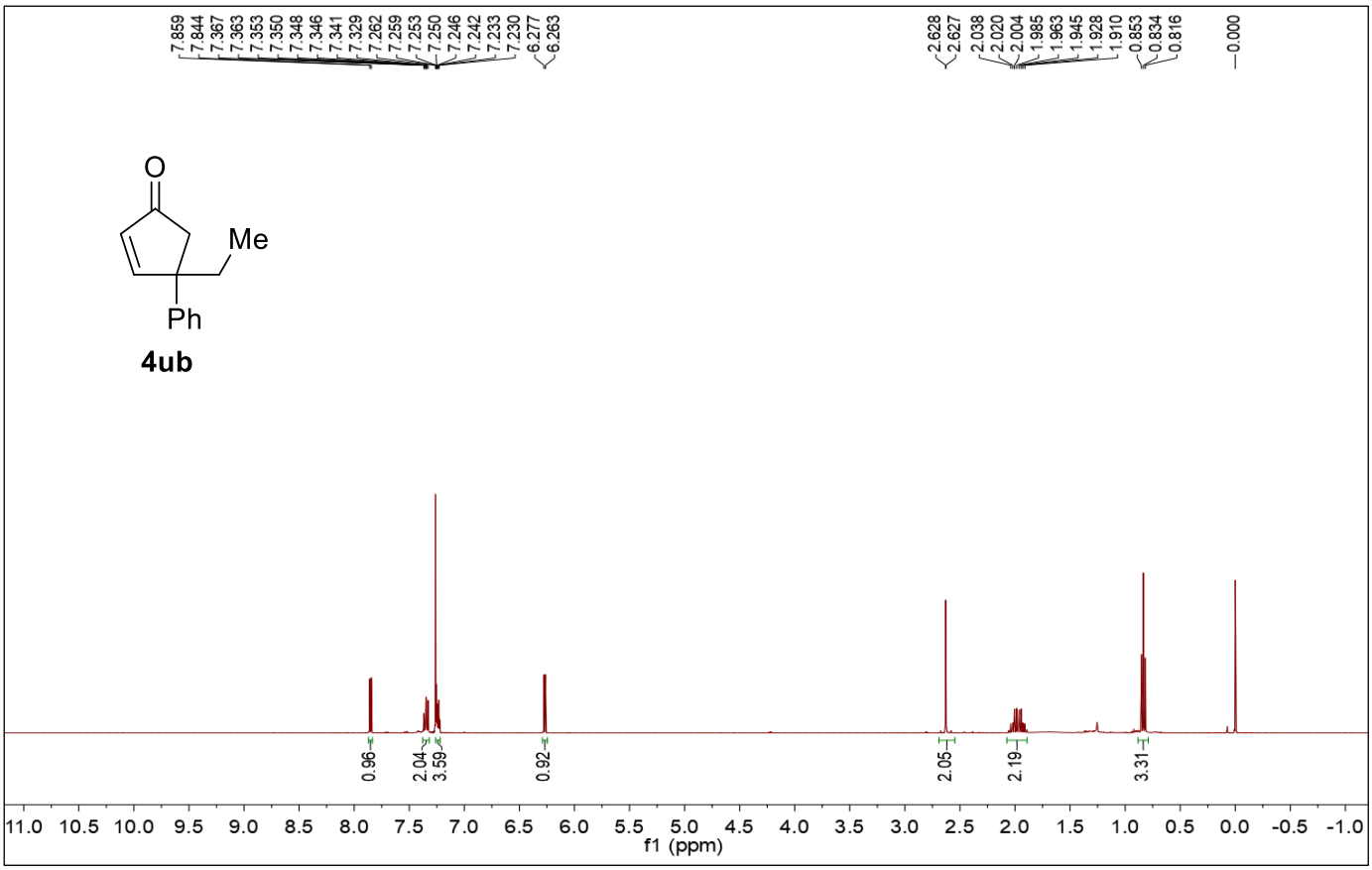

9.632
7.810
7.790
7.480
7.459
7.318
7.302
7.282
7.252
7.234
7.207
3.661
3.617
3.610
3.566
3.181
1.615
0.000
O
Me
Ph
O
H
3ta
0.95
2.13
2.00
2.14
3.24
2.33
1.00
3.14
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


201.326
196.430
139.442
136.406
132.277
128.939
127.946
127.465
127.115
126.849
82.623
80.603
77.318
77.000
76.681
51.838
45.973
20.472
O
Me
Ph
O
H
3ta
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

^{1}H and ^{13}C NMR spectra of cyclopentenones:



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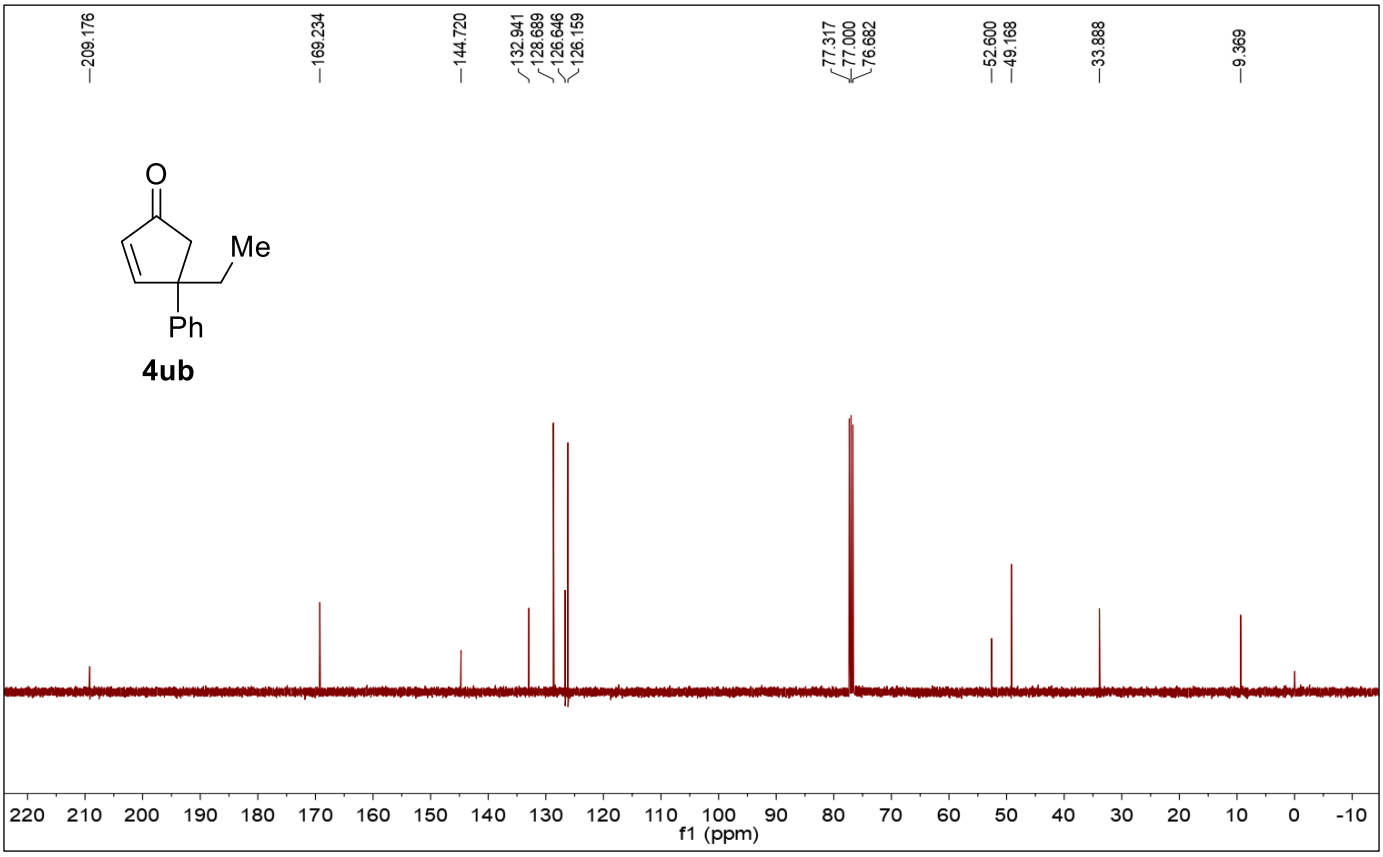
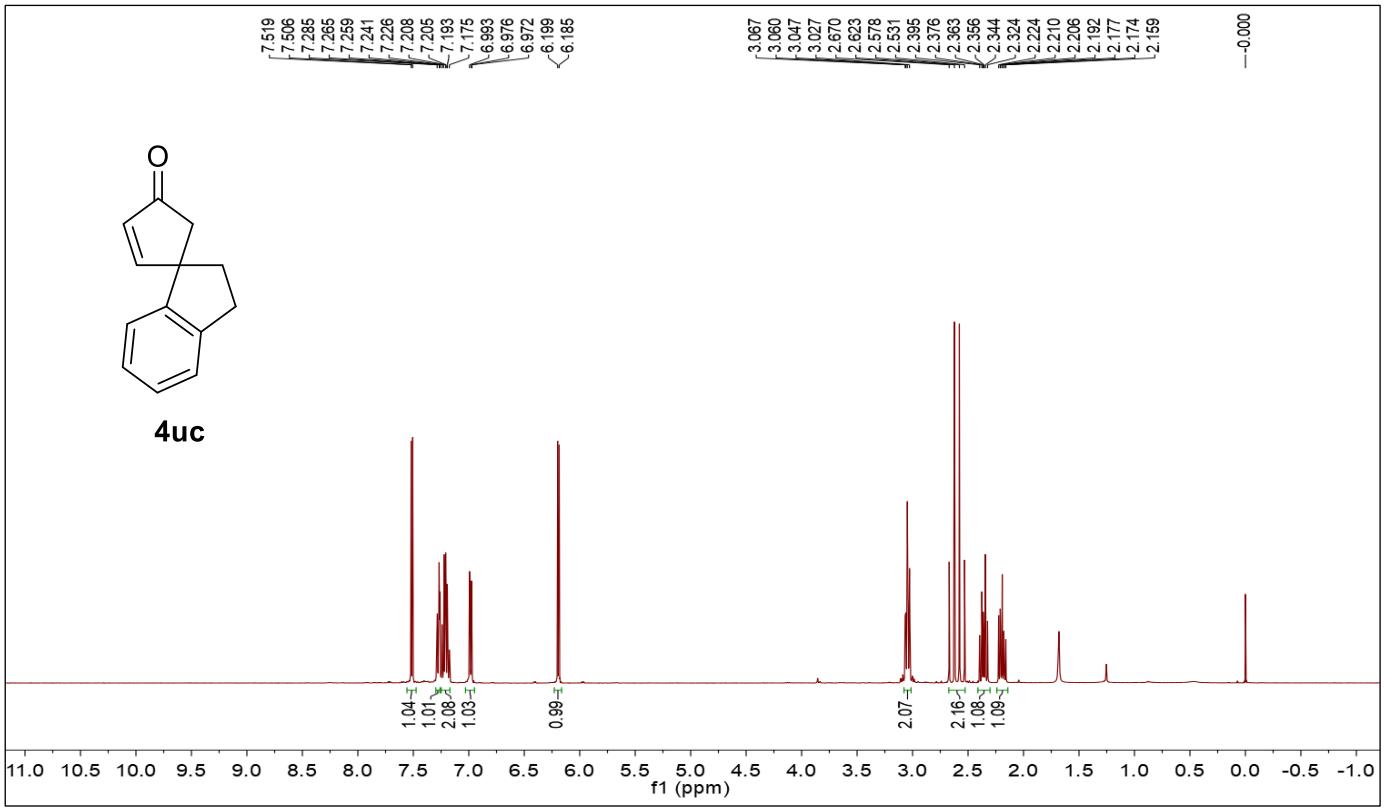
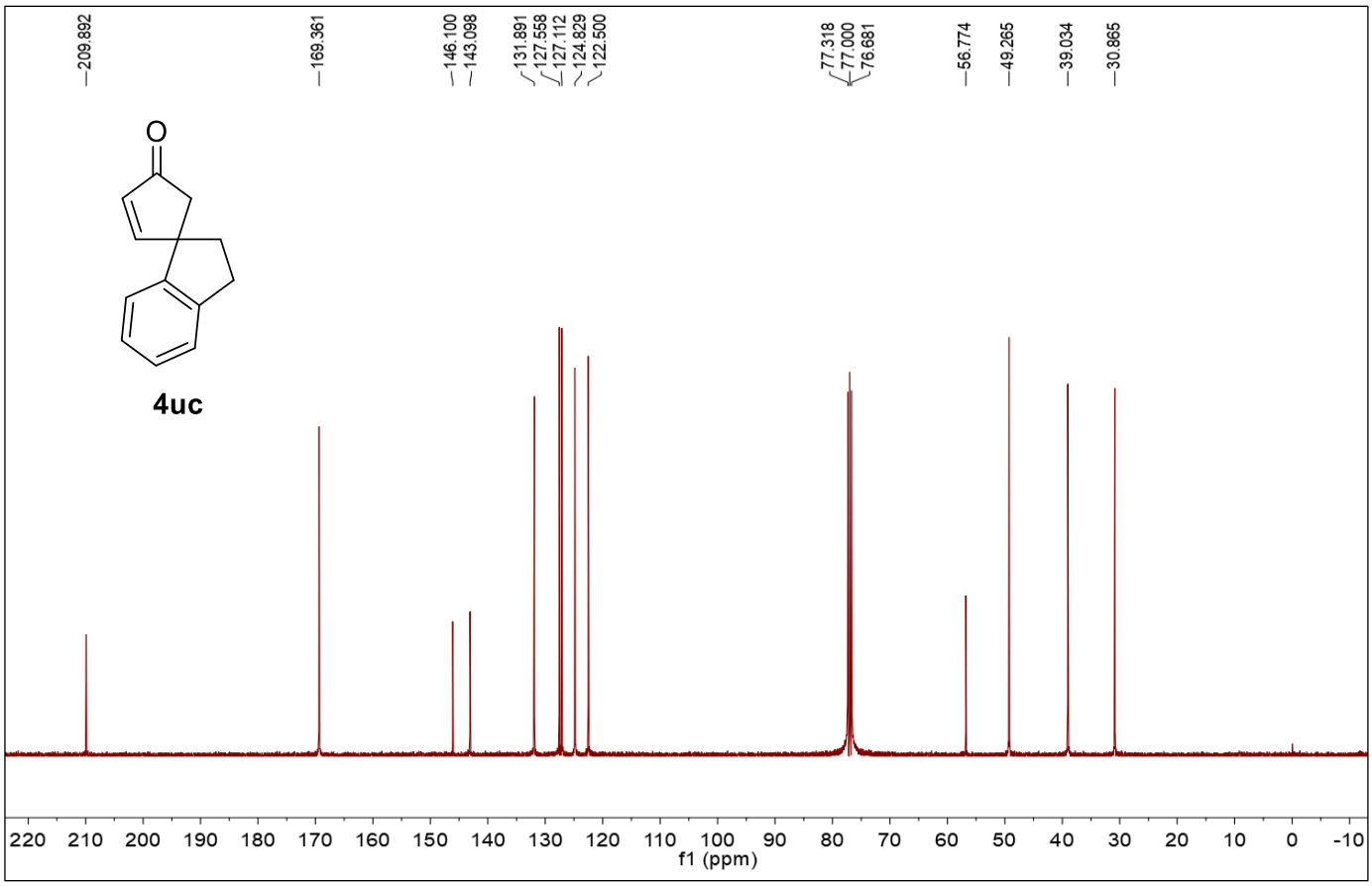
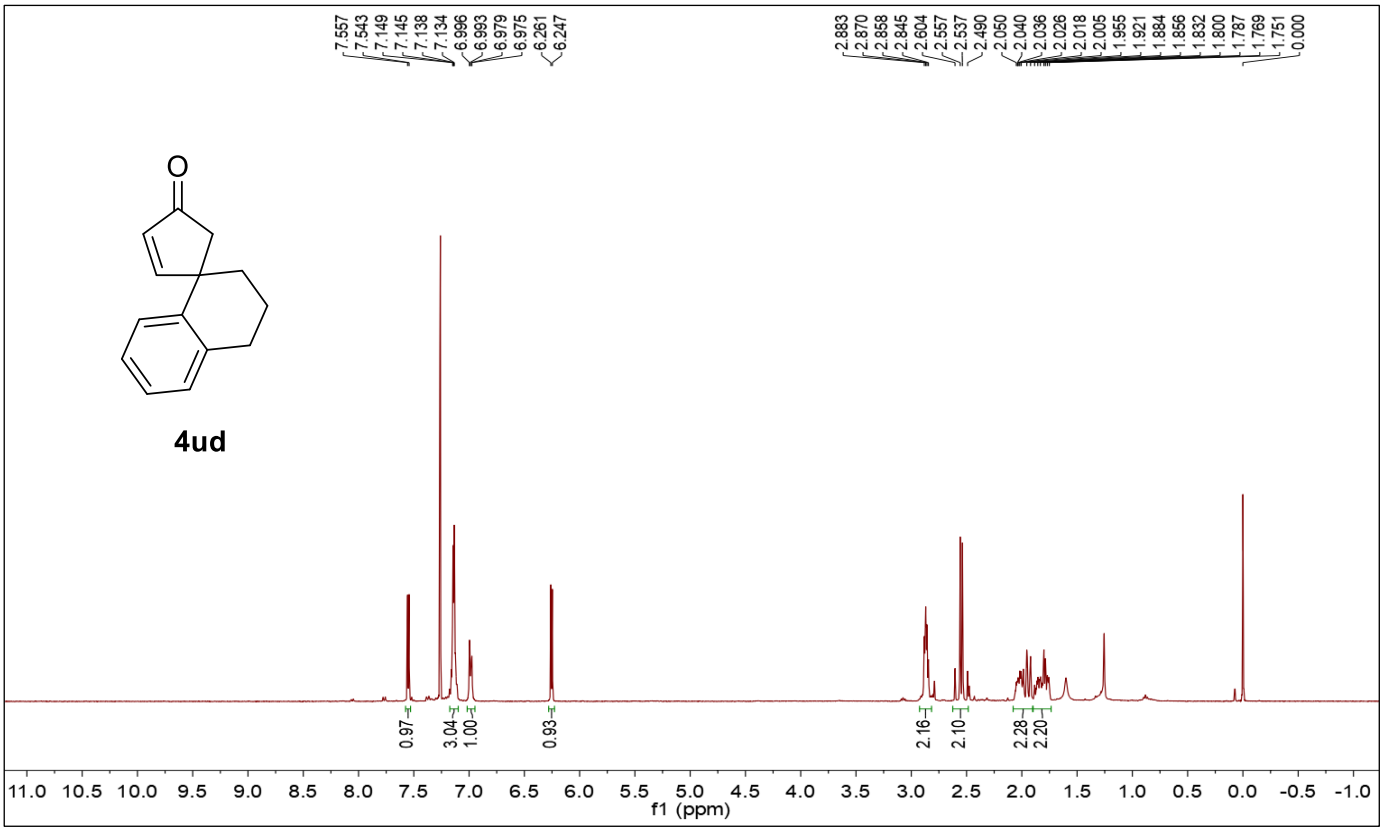
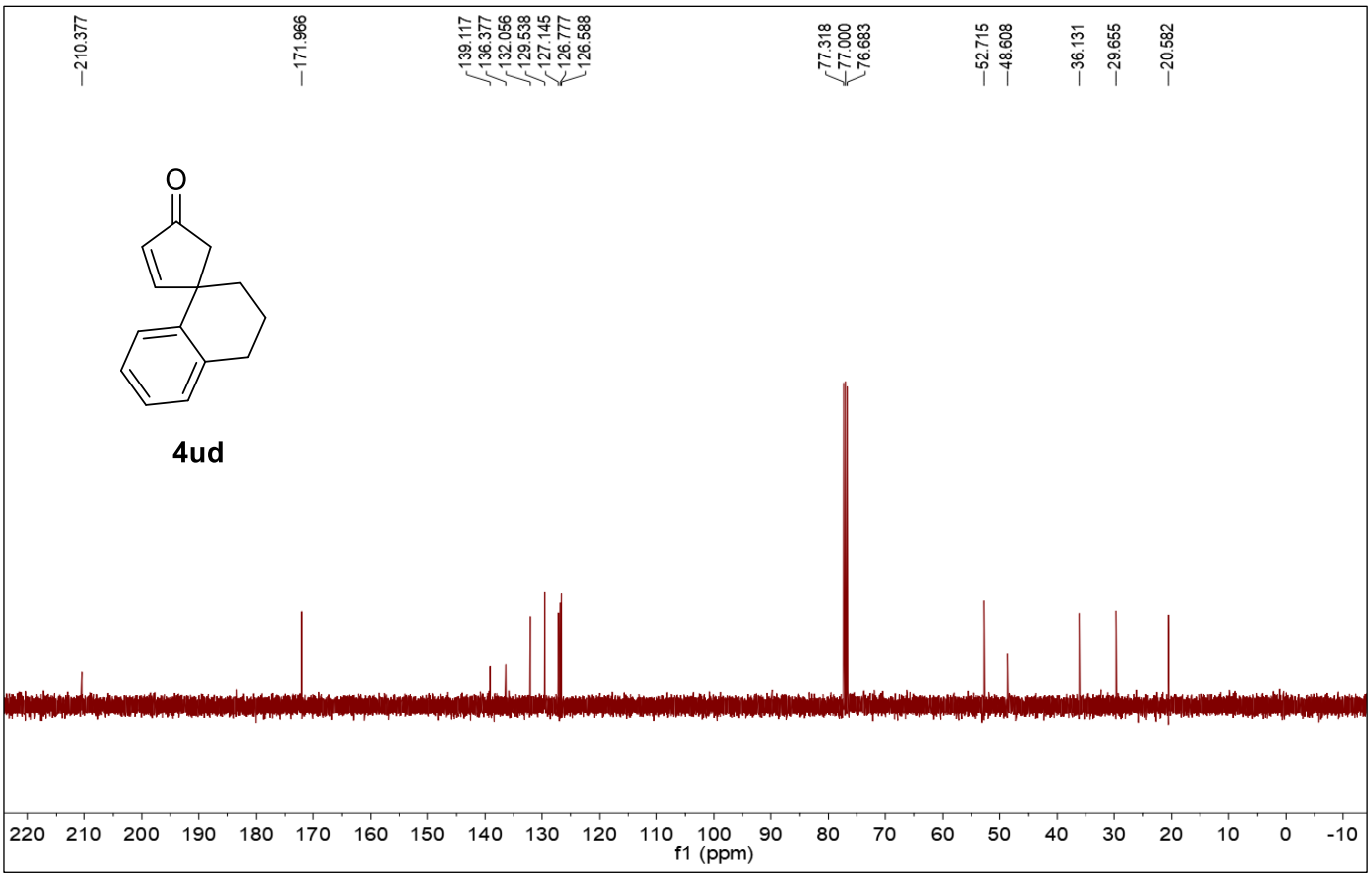
7.363
7.353
7.350
7.348
7.346
7.341
7.329
7.262
7.259
7.253
7.250
7.246
7.242
7.233
7.230
6.277
6.263
2.628
2.627
2.038
2.020
2.004
1.985
1.963
1.945
1.928
1.910
0.853
0.834
0.816
0.000
O
Me
Ph
4ub
0.96
2.04
3.59
0.92
2.05
2.19
3.31
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

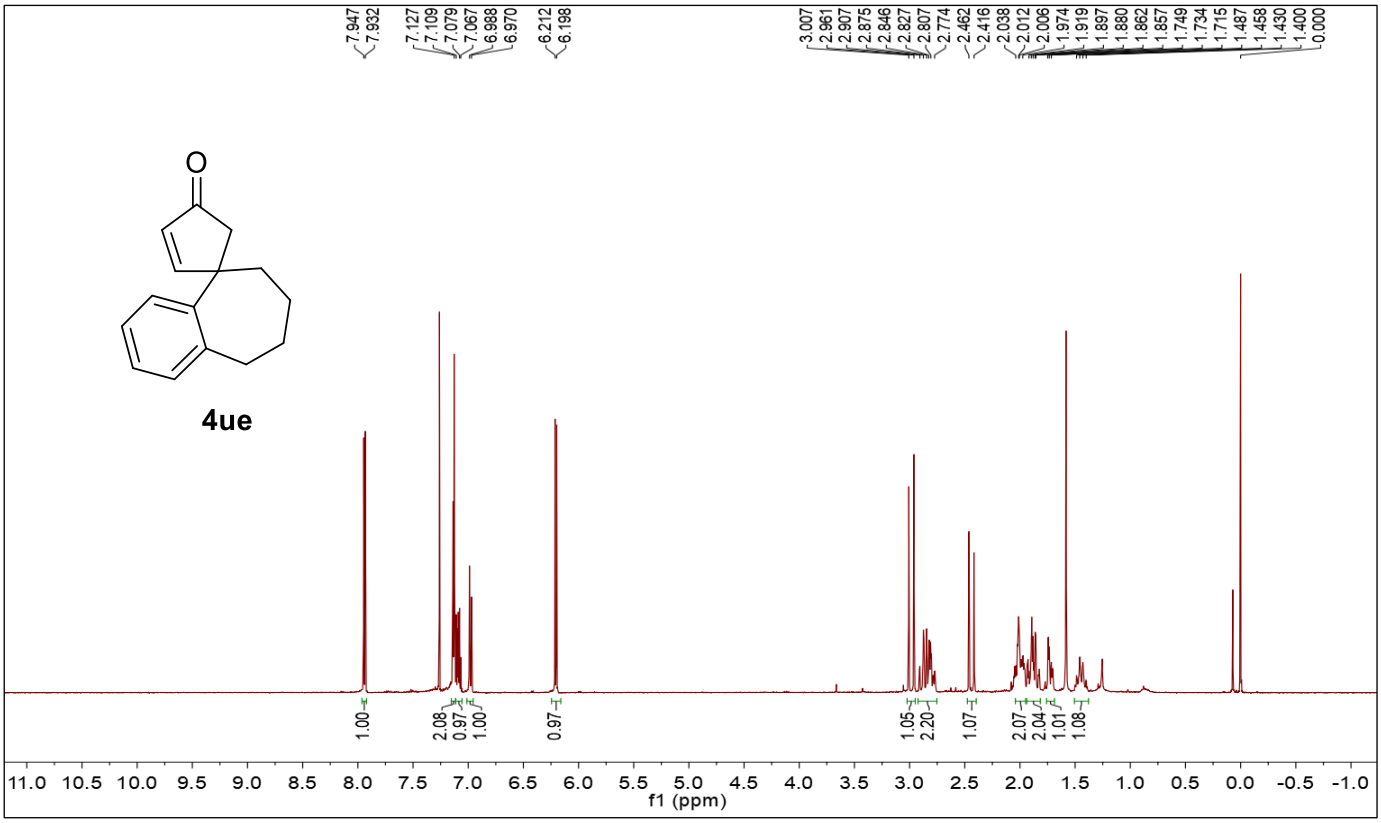

209.176
169.234
144.720
132.941
128.689
126.646
126.159
77.317
77.000
76.682
52.600
49.168
33.888
9.369
O
Me
Ph
4ub
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


7.519
7.506
7.285
7.265
7.259
7.241
7.226
7.208
7.205
7.193
7.175
6.993
6.976
6.972
6.199
6.185
3.067
3.060
3.047
3.027
2.670
2.623
2.578
2.531
2.395
2.376
2.363
2.356
2.344
2.324
2.224
2.210
2.206
2.192
2.177
2.174
2.159
-0.000
O
4uc
1.04
1.01
2.08
1.03
0.99
2.07
2.16
1.08
1.09
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


209.892
169.361
146.100
143.098
131.891
127.558
127.112
124.829
122.500
77.318
77.000
76.681
56.774
49.265
39.034
30.865
O
4uc
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


7.557
7.543
7.149
7.145
7.138
7.134
6.996
6.993
6.979
6.975
6.261
6.247
2.883
2.870
2.858
2.845
2.604
2.557
2.537
2.490
2.050
2.040
2.036
2.026
2.018
2.005
1.955
1.921
1.884
1.856
1.832
1.800
1.787
1.769
1.751
0.000
O
4ud
0.97
3.04
1.00
0.93
2.16
2.10
2.28
2.20
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


210.377
171.966
139.117
136.377
132.056
129.538
127.145
126.777
126.588
77.318
77.000
76.683
52.715
48.608
36.131
29.655
20.582
O
4ud
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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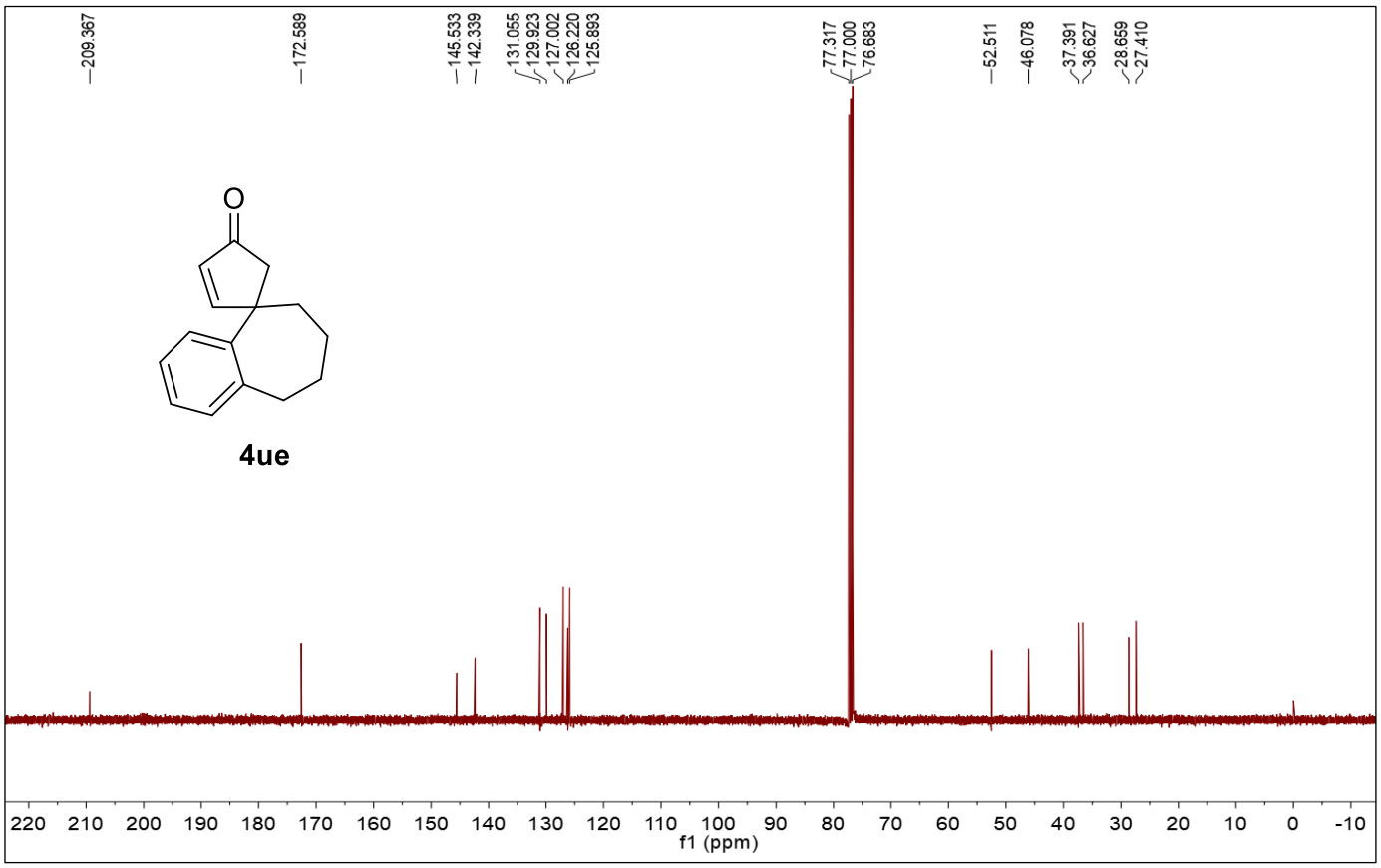
7.932
7.127
7.109
7.079
7.067
6.988
6.970
6.212
6.198
3.007
2.961
2.907
2.875
2.846
2.827
2.807
2.774
2.462
2.416
2.038
2.012
2.006
1.974
1.919
1.897
1.880
1.862
1.857
1.749
1.734
1.715
1.487
1.458
1.430
1.400
0.000
O
4ue
1.00
2.08
0.97
1.00
0.97
1.05
2.20
1.07
2.07
2.04
1.01
1.08
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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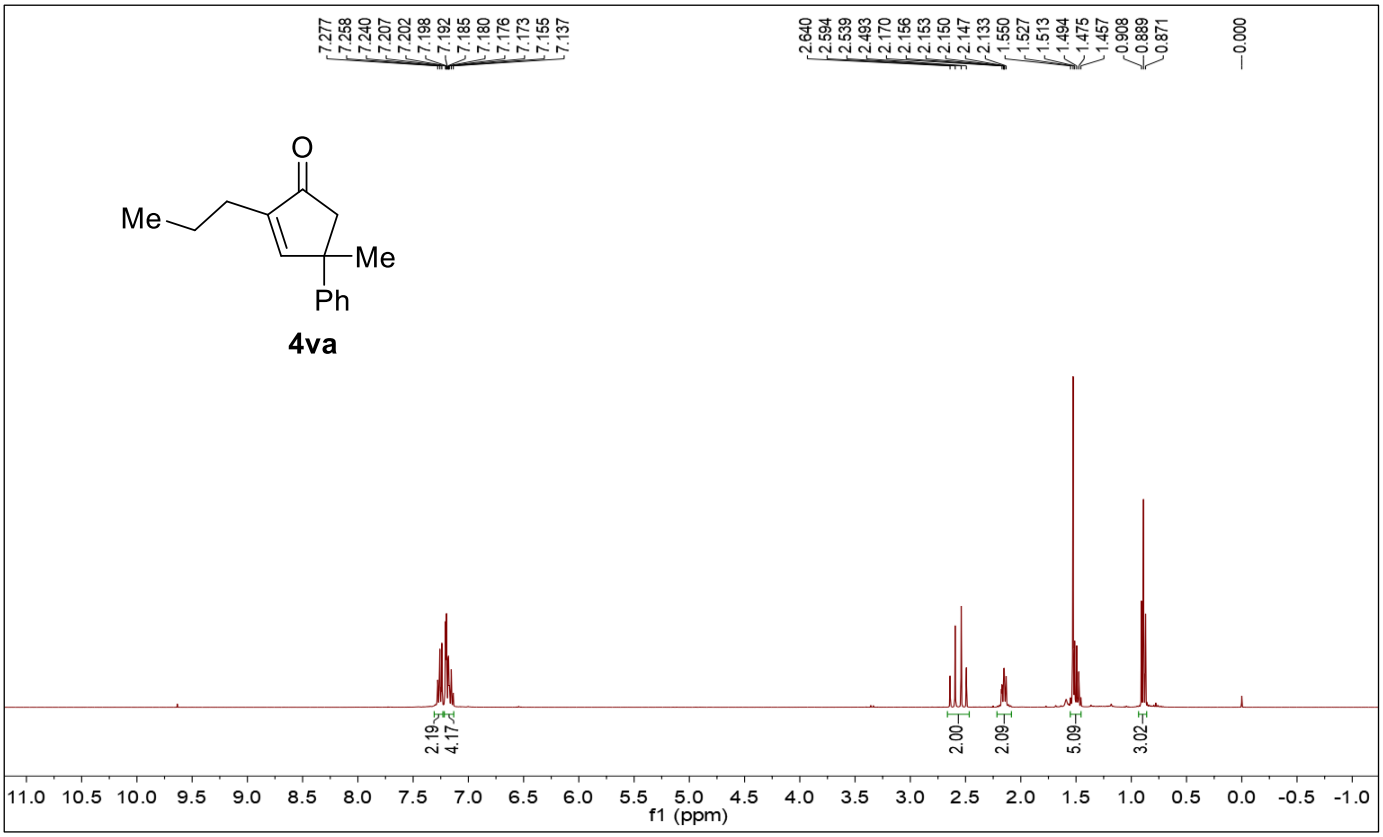
142.339
131.055
129.923
127.002
126.220
125.893
77.317
77.000
76.683
52.511
46.078
37.391
36.627
28.659
27.410
O
4ue
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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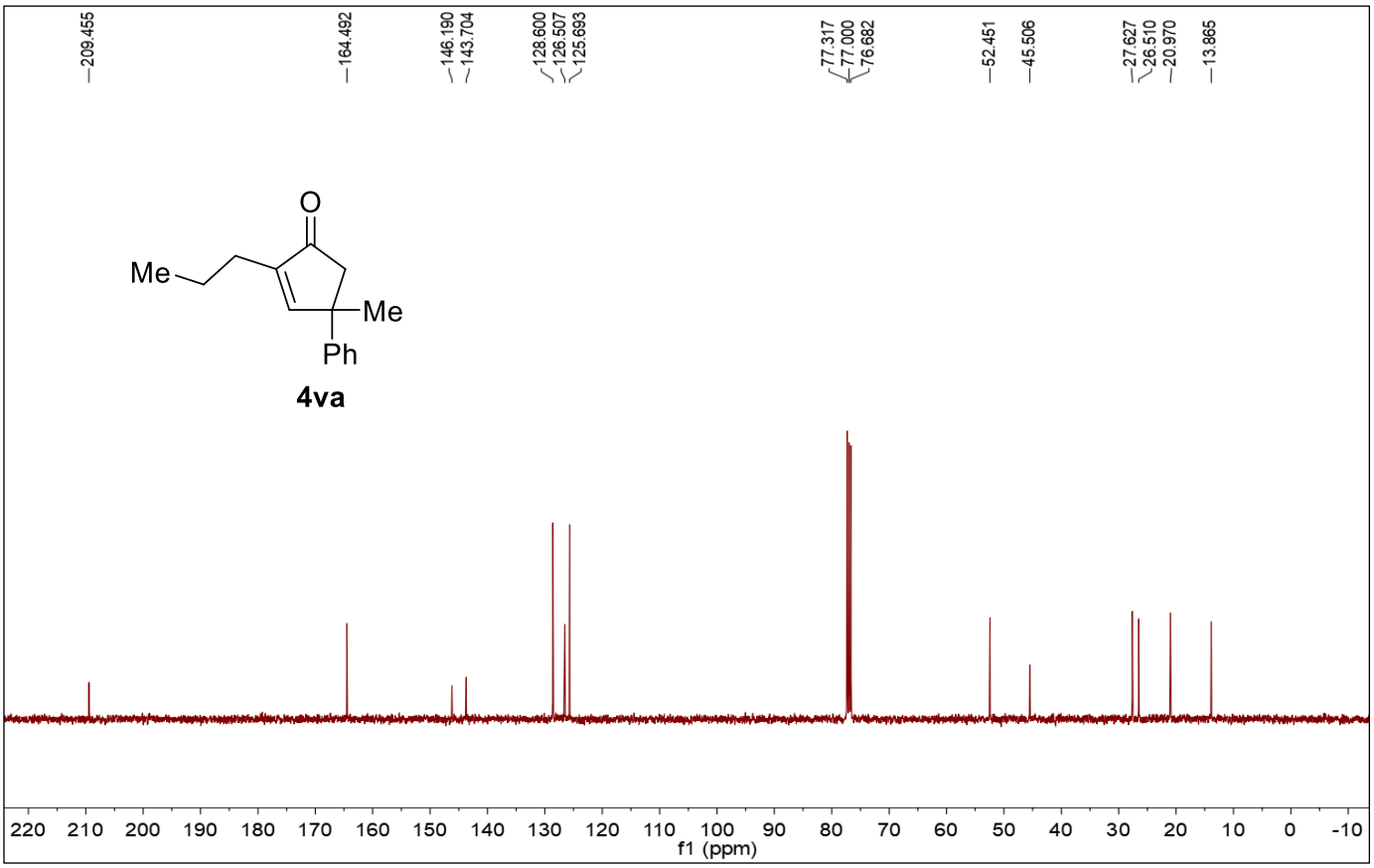
7.240
7.207
7.202
7.198
7.192
7.185
7.180
7.176
7.173
7.155
7.137
2.640
2.594
2.539
2.493
2.170
2.156
2.153
2.150
2.147
2.133
1.550
1.527
1.513
1.494
1.475
1.457
0.908
0.889
0.871
0.000
O
Me
Me
Ph
4va
2.19
4.17
2.00
2.09
5.09
3.02
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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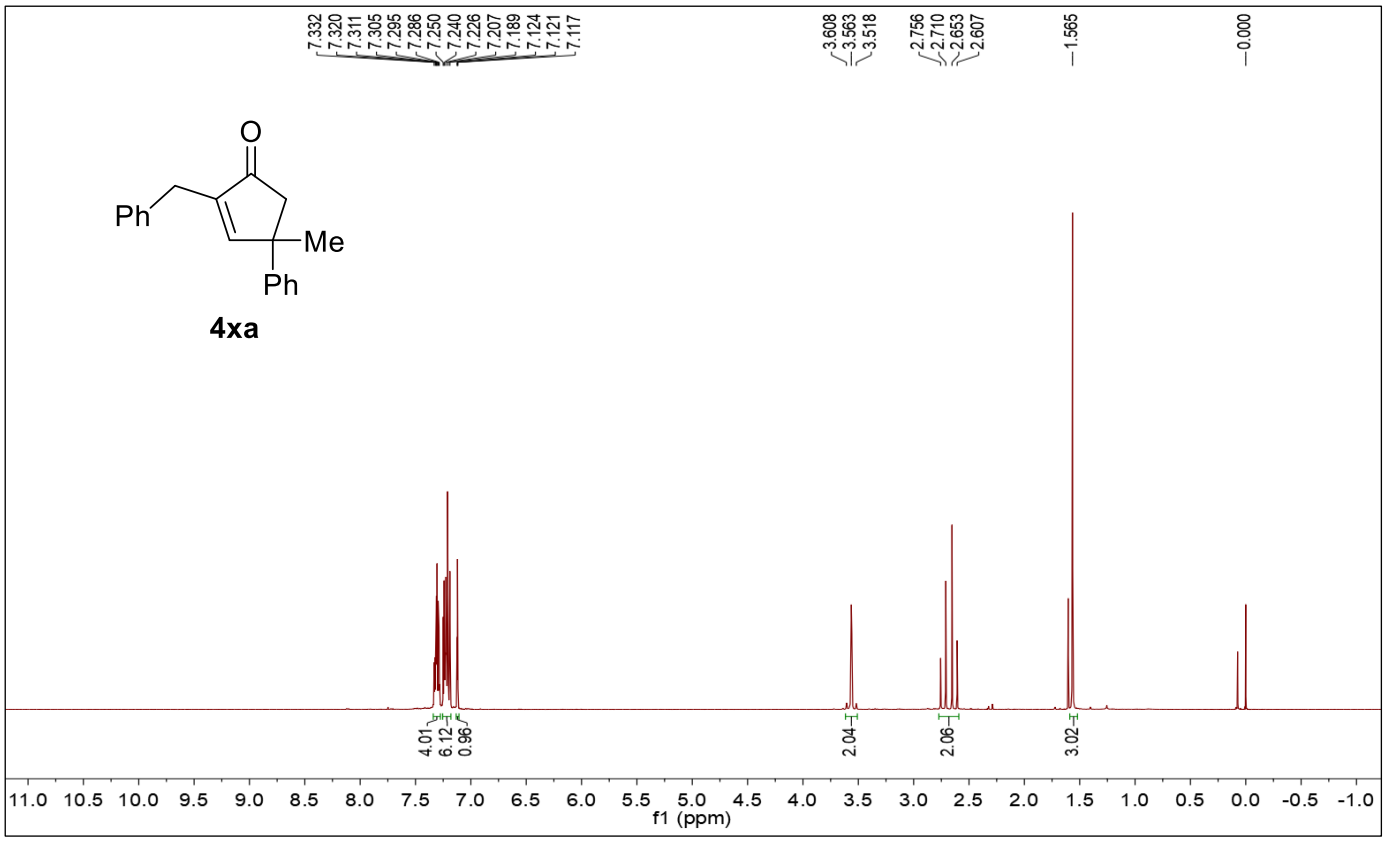
146.190
143.704
128.600
126.507
125.693
77.317
77.000
76.682
52.451
45.506
27.627
26.510
20.970
13.865
O
Me
Me
Ph
4va
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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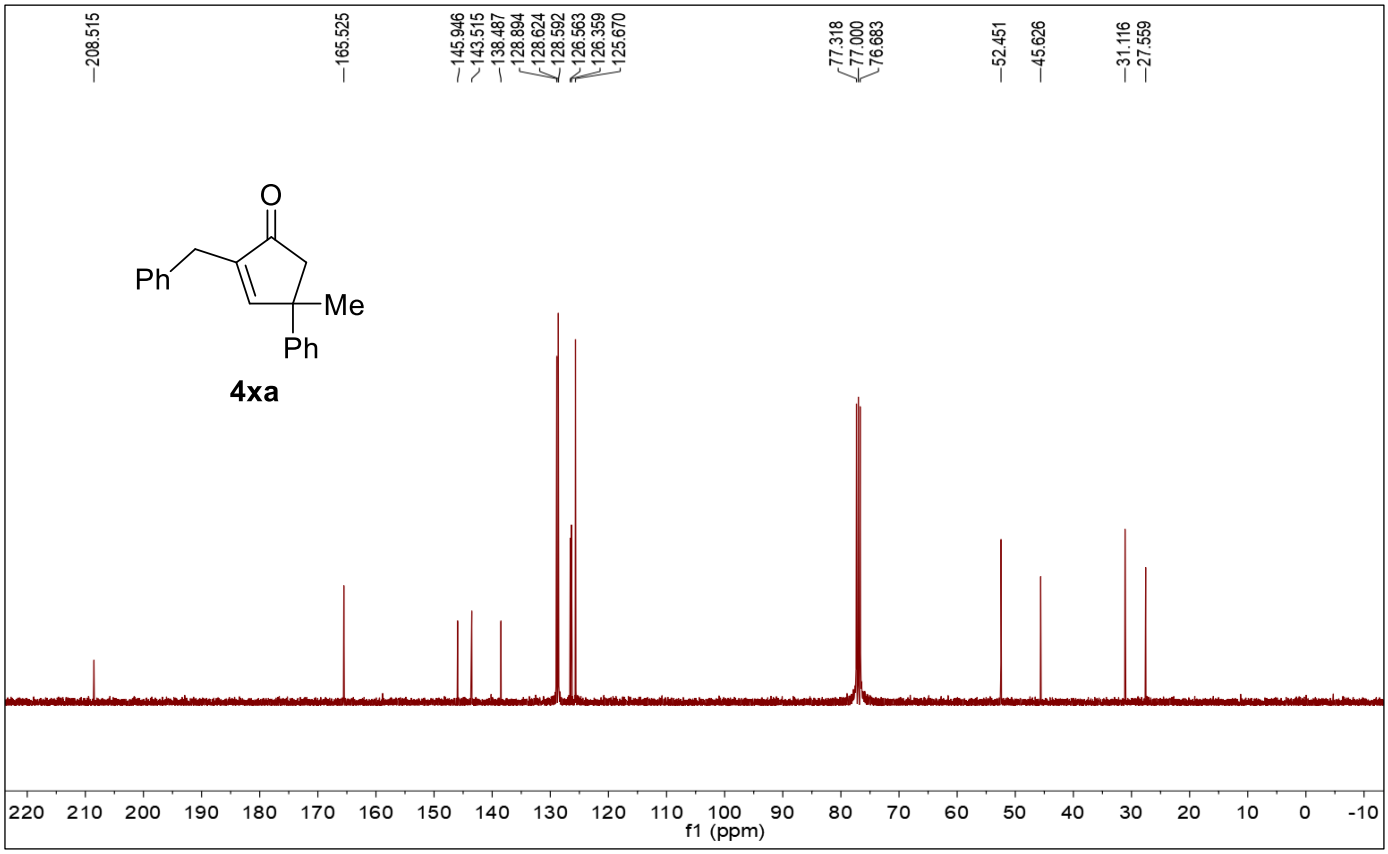
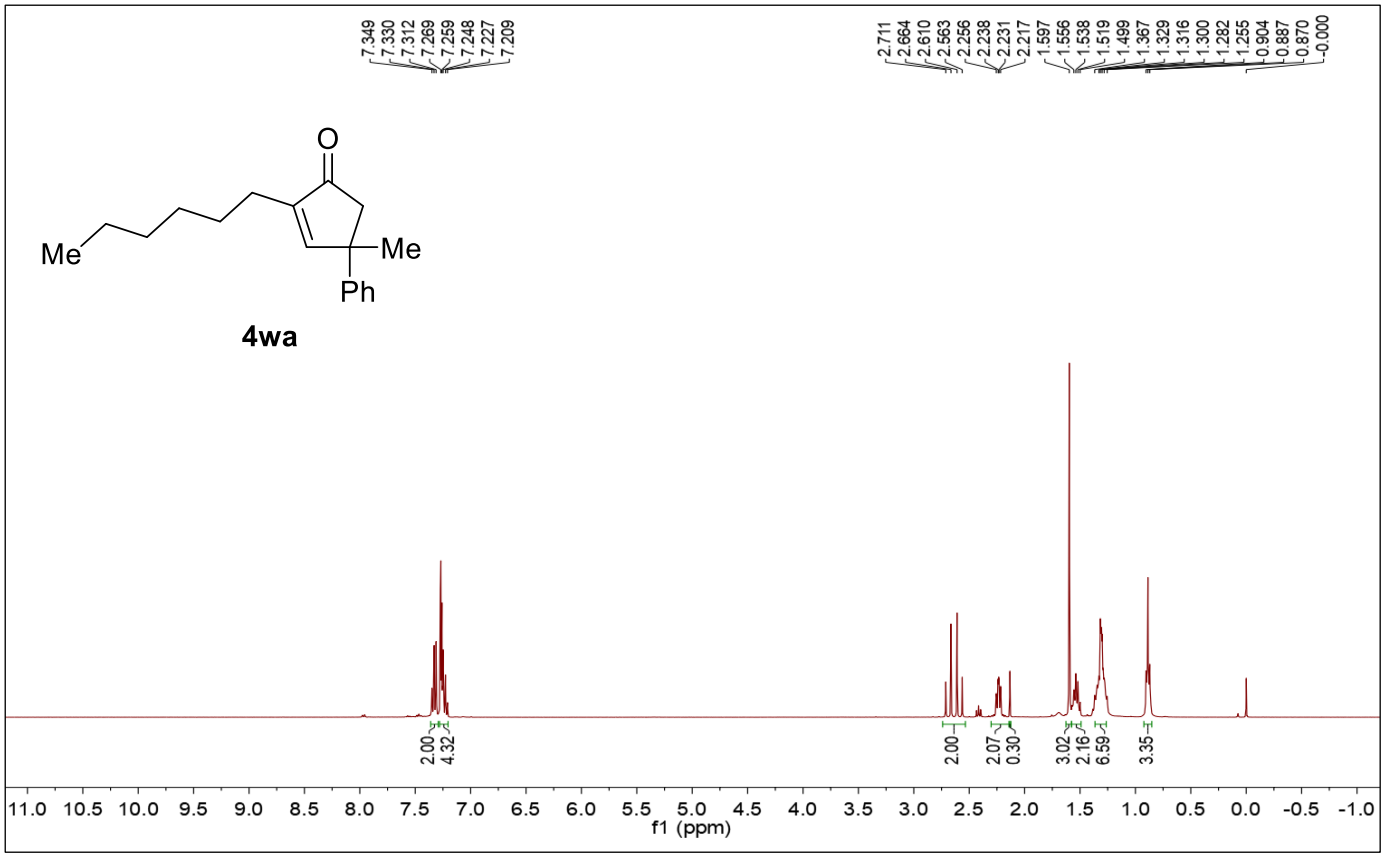
7.320
7.311
7.305
7.295
7.286
7.250
7.240
7.226
7.207
7.189
7.124
7.121
7.117
3.608
3.563
3.518
2.756
2.710
2.653
2.607
1.565
0.000
O
Ph
Me
Ph
4xa
4.01
6.12
0.96
2.04
2.06
3.02
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


208.515
165.525
145.946
143.515
138.487
128.894
128.624
128.592
126.563
126.359
125.670
77.318
77.000
76.683
52.451
45.626
31.116
27.559
O
Ph
Me
Ph
4xa
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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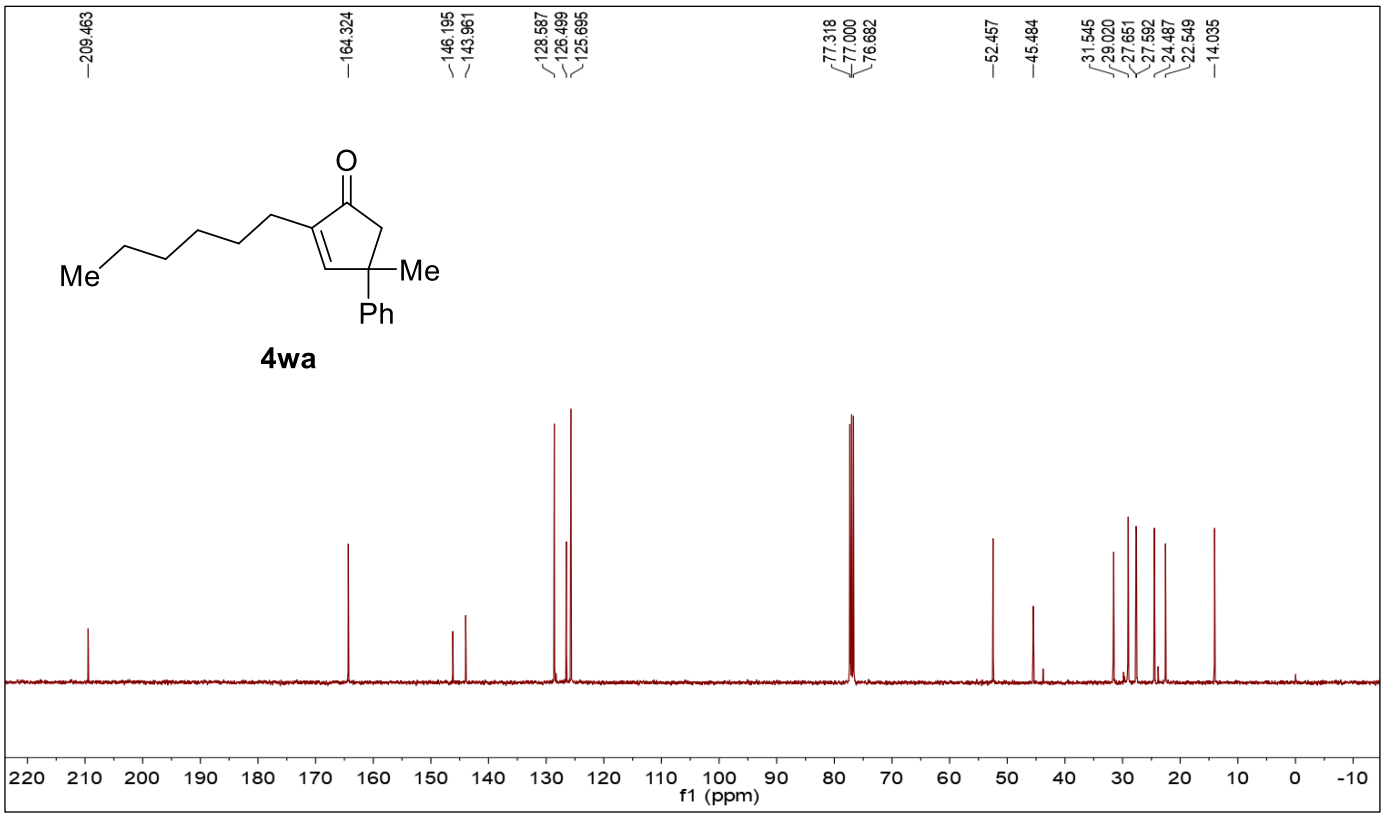
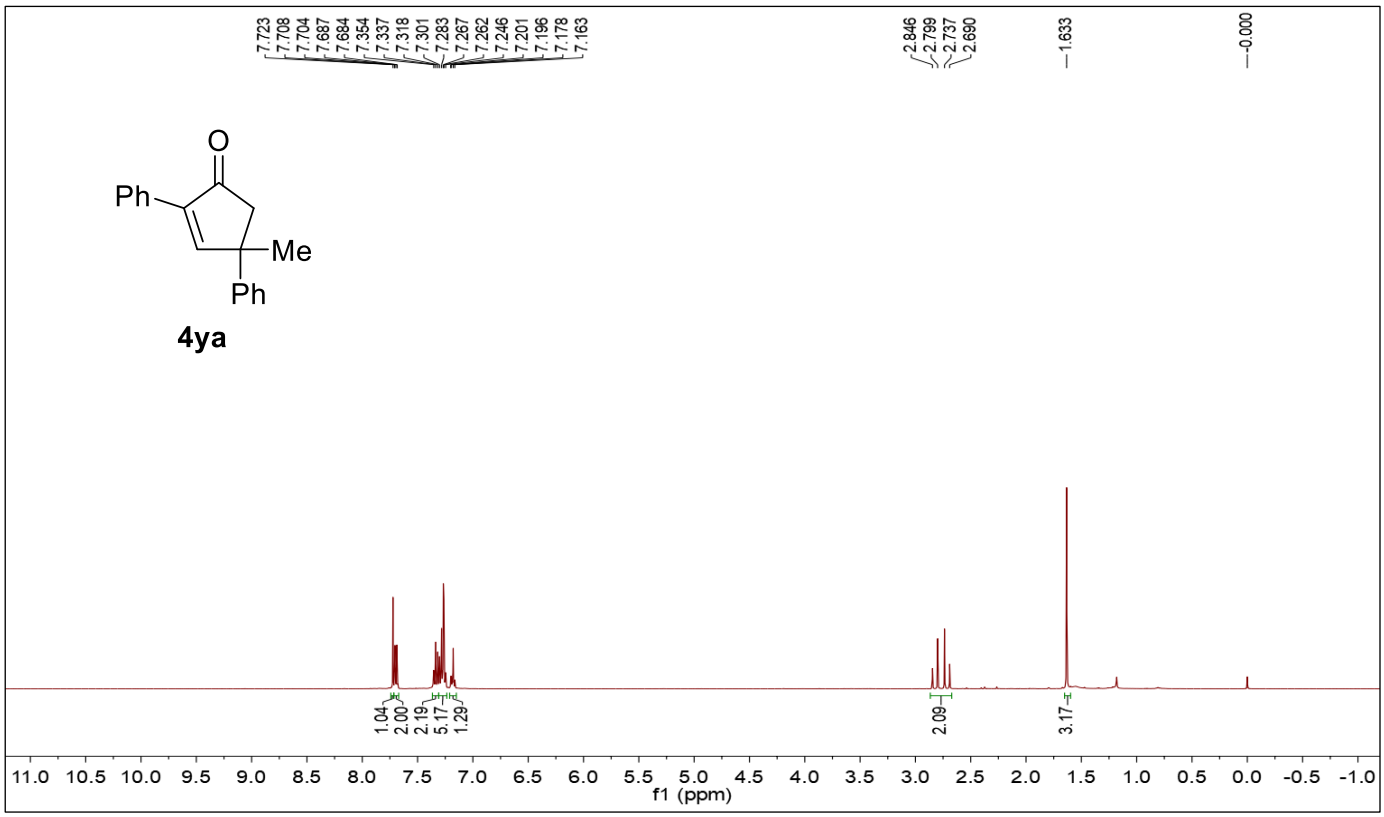
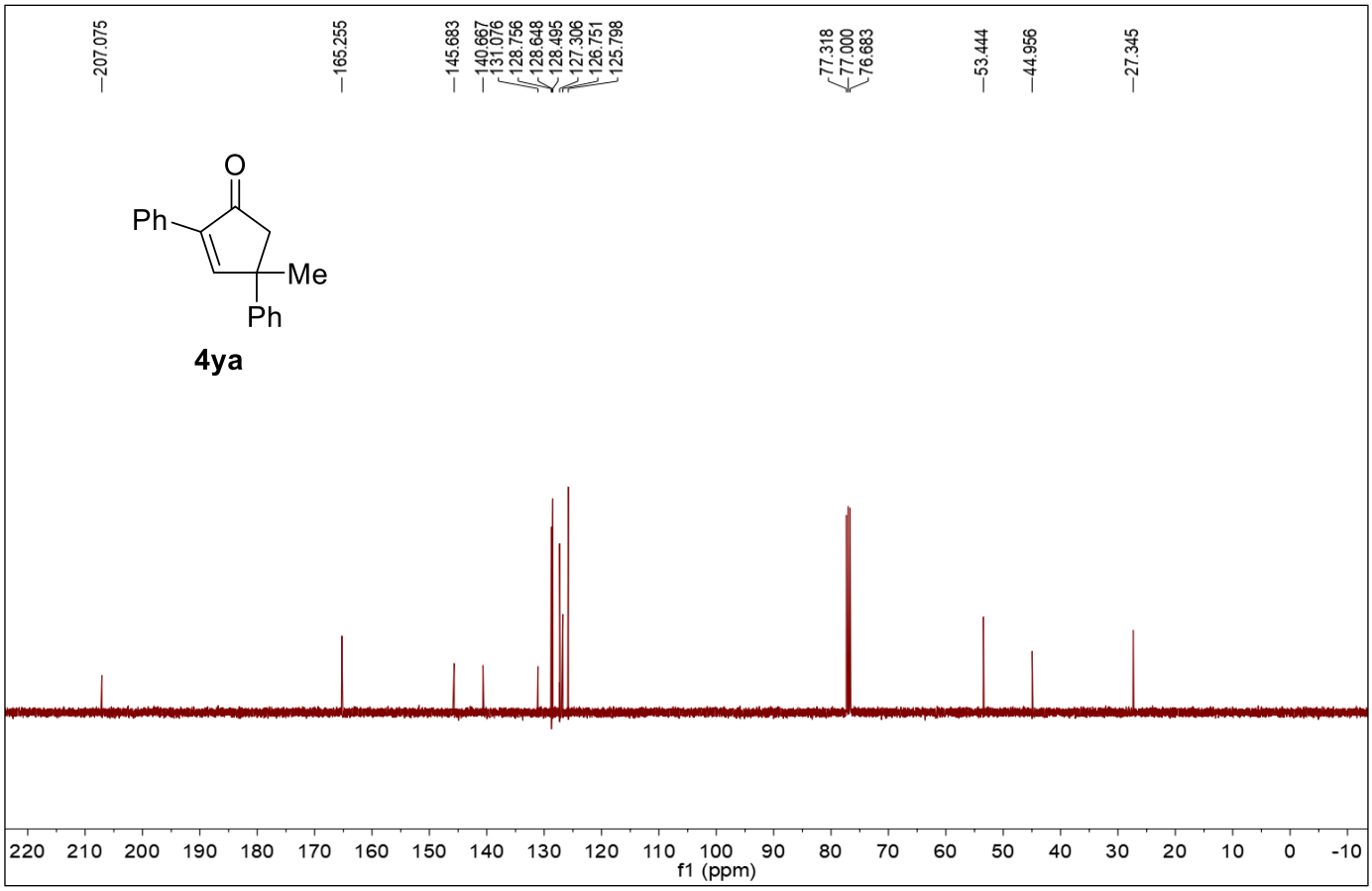
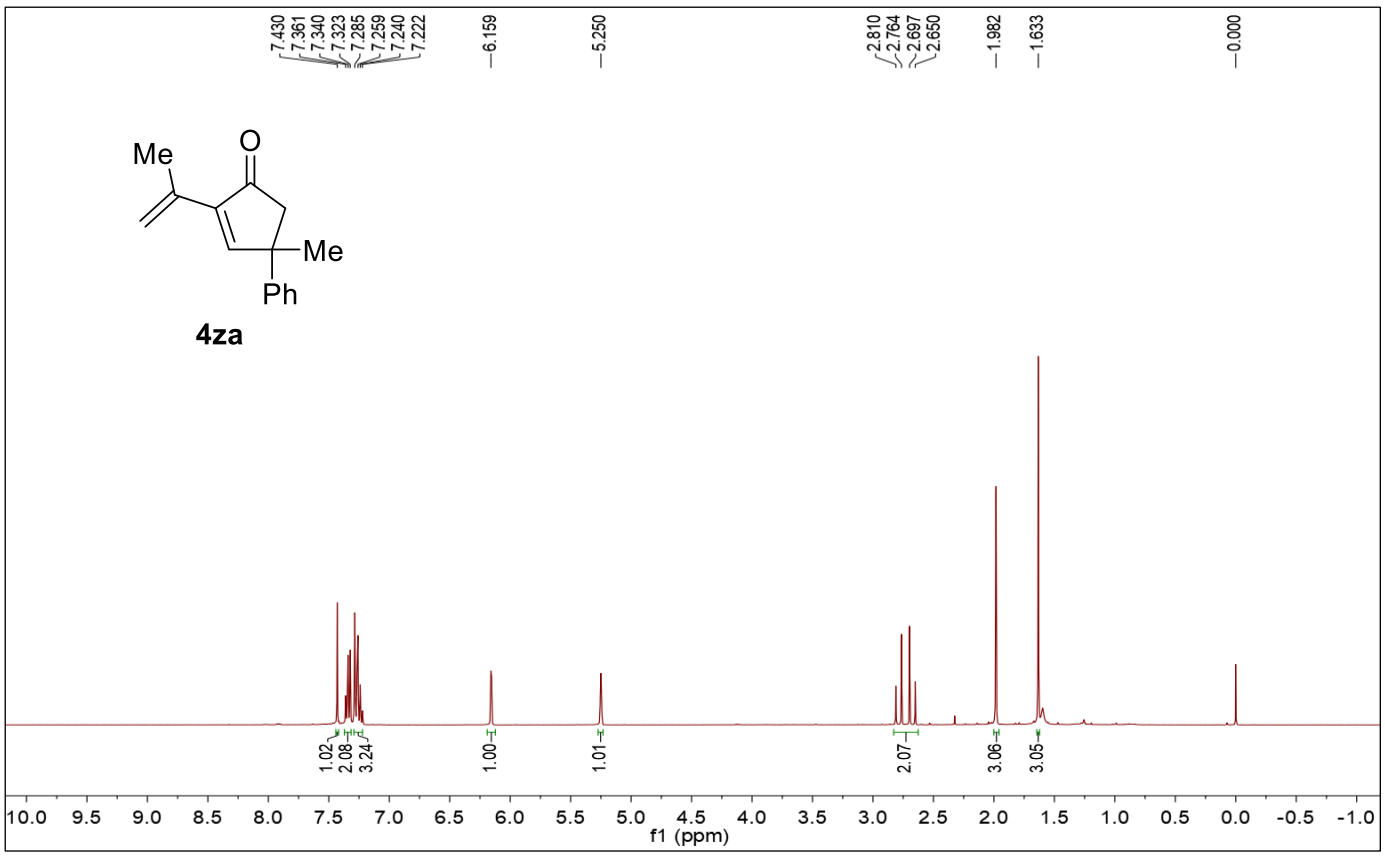
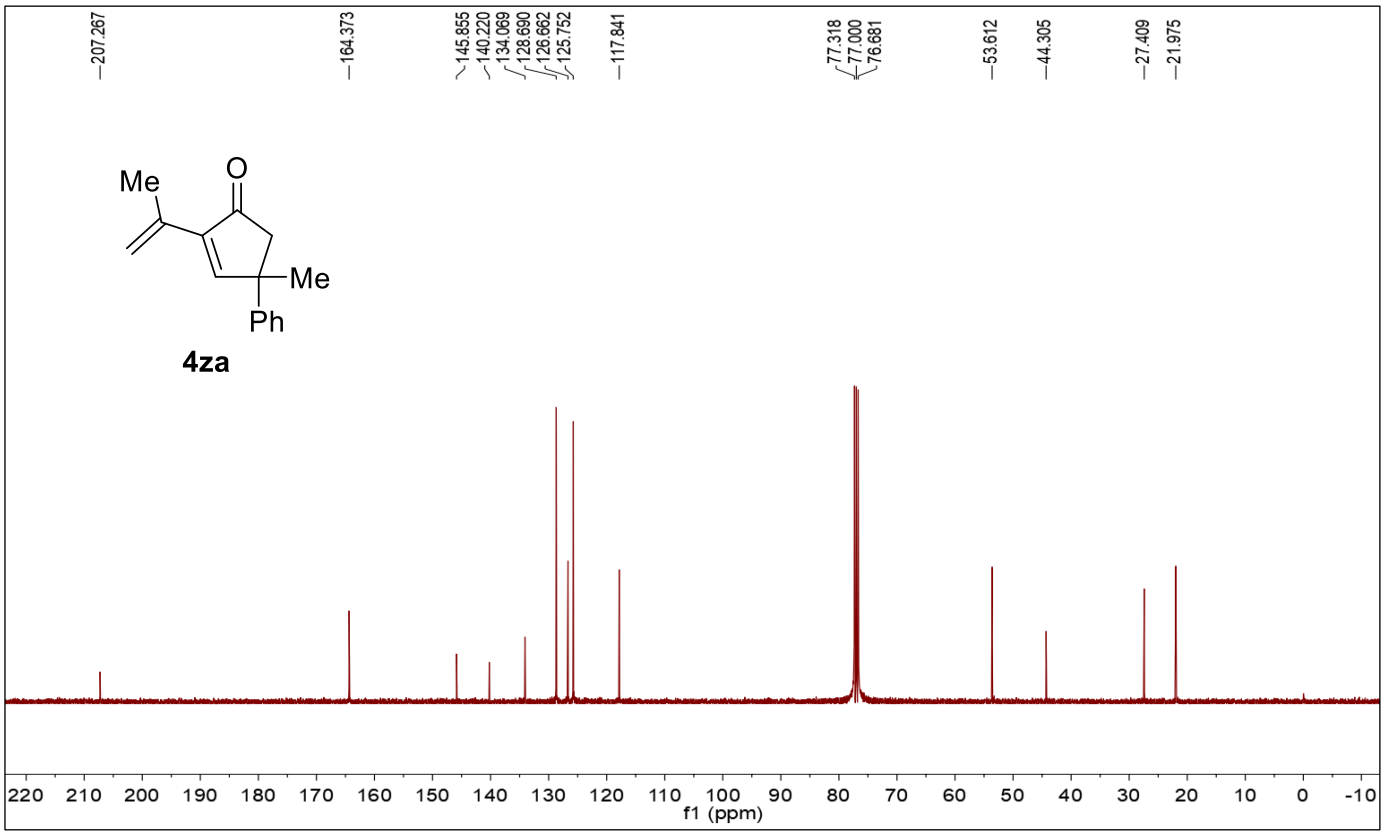
7.330
7.312
7.269
7.259
7.248
7.227
7.209
2.711
2.664
2.610
2.563
2.256
2.238
2.231
2.217
1.597
1.556
1.538
1.519
1.499
1.367
1.329
1.316
1.300
1.282
1.255
0.904
0.887
0.870
-0.000
O
Me
Me
Ph
4wa
2.00
4.32
2.00
2.07
0.30
3.02
2.16
6.59
3.35
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


209.463
164.324
146.195
143.961
128.587
126.499
125.695
77.318
77.000
76.682
52.457
45.484
31.545
29.020
27.651
27.592
24.487
22.549
14.035
O
Me
Me
Ph
4wa
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

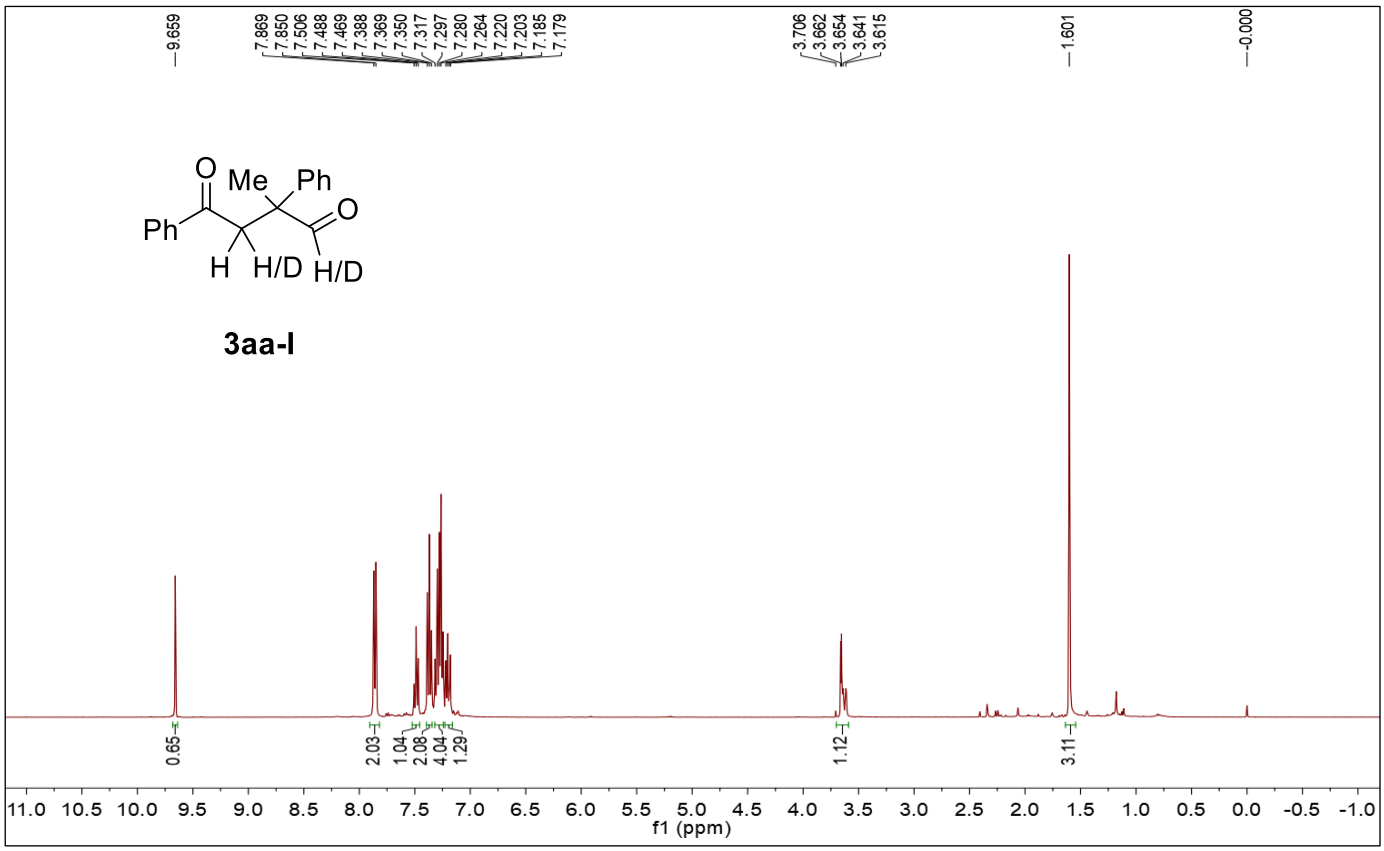

7.723
7.708
7.704
7.687
7.684
7.354
7.337
7.318
7.301
7.283
7.267
7.262
7.246
7.201
7.196
7.178
7.163
2.846
2.799
2.737
2.690
1.633
0.000
O
Ph
Me
Ph
4ya
1.04
2.00
2.19
5.17
1.29
2.09
3.17
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)

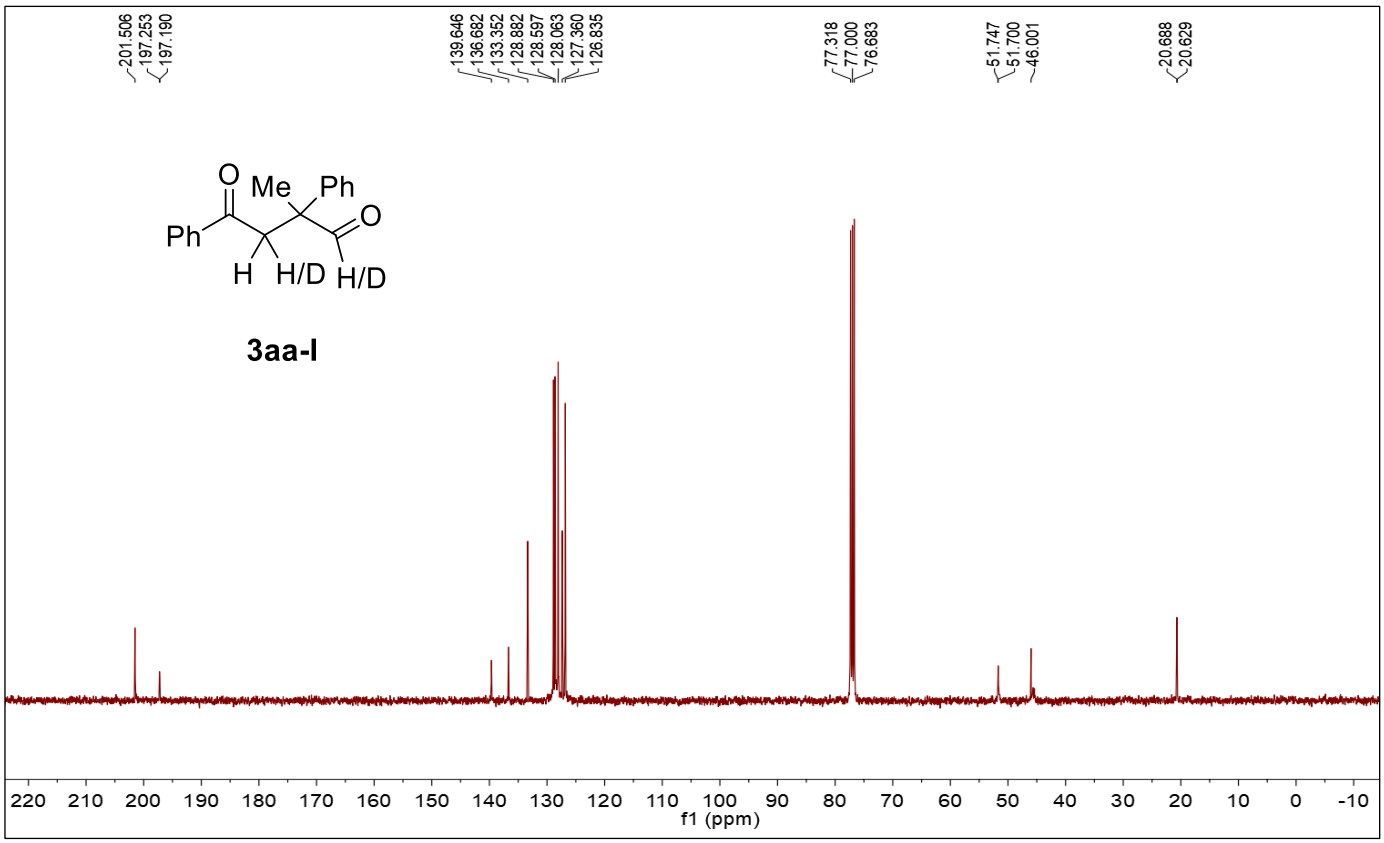

207.075
165.255
145.683
140.667
131.076
128.756
128.648
128.495
127.306
126.751
125.798
77.318
77.000
76.683
53.444
44.956
27.345
O
Ph
Me
Ph
4ya
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

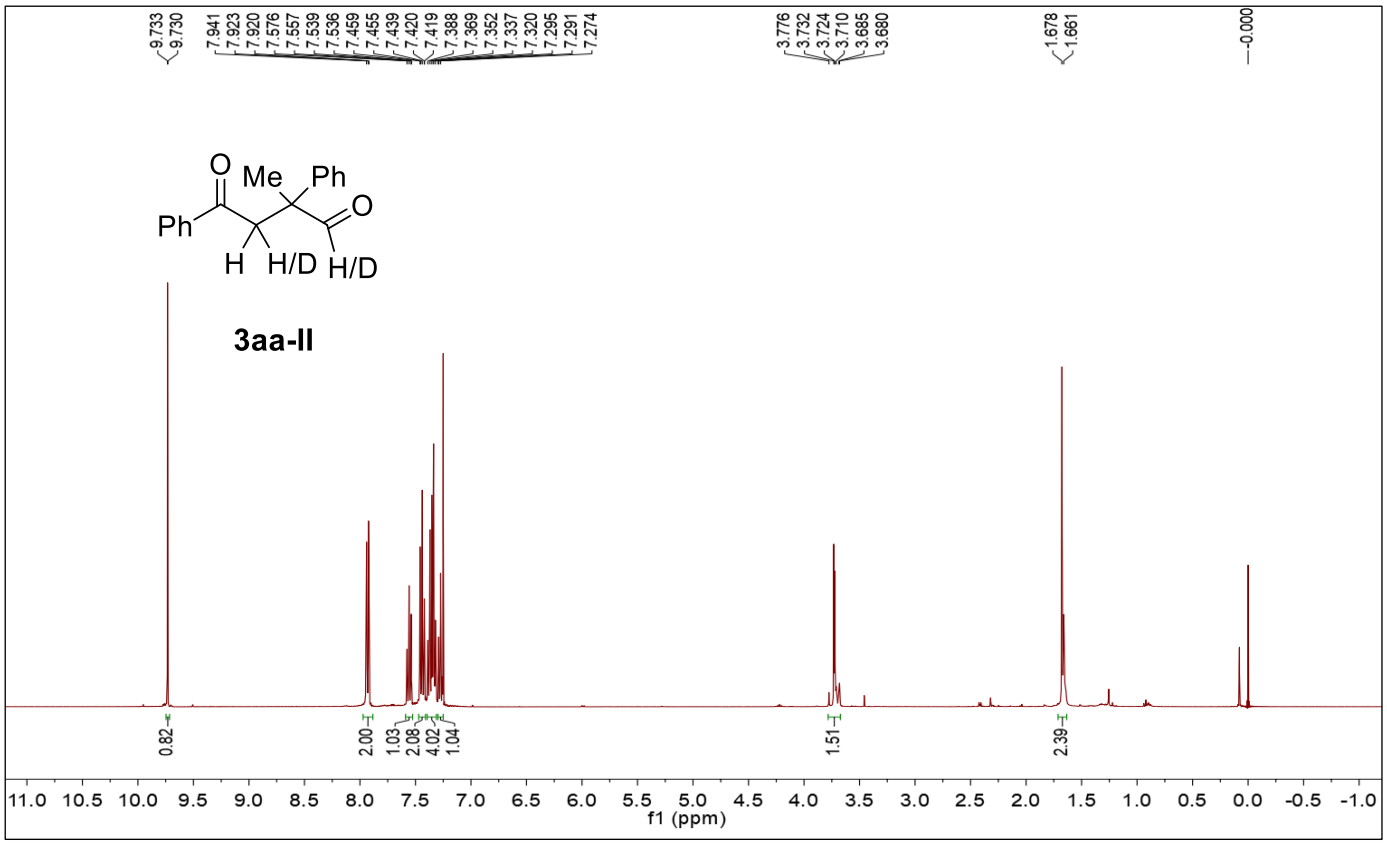

7.430
7.361
7.340
7.323
7.285
7.259
7.240
7.222
6.159
5.250
2.810
2.764
2.697
2.650
1.982
1.633
0.000
Me
O
Me
Ph
4za
1.02
2.08
3.24
1.00
1.01
2.07
3.06
3.05
f1 (ppm)

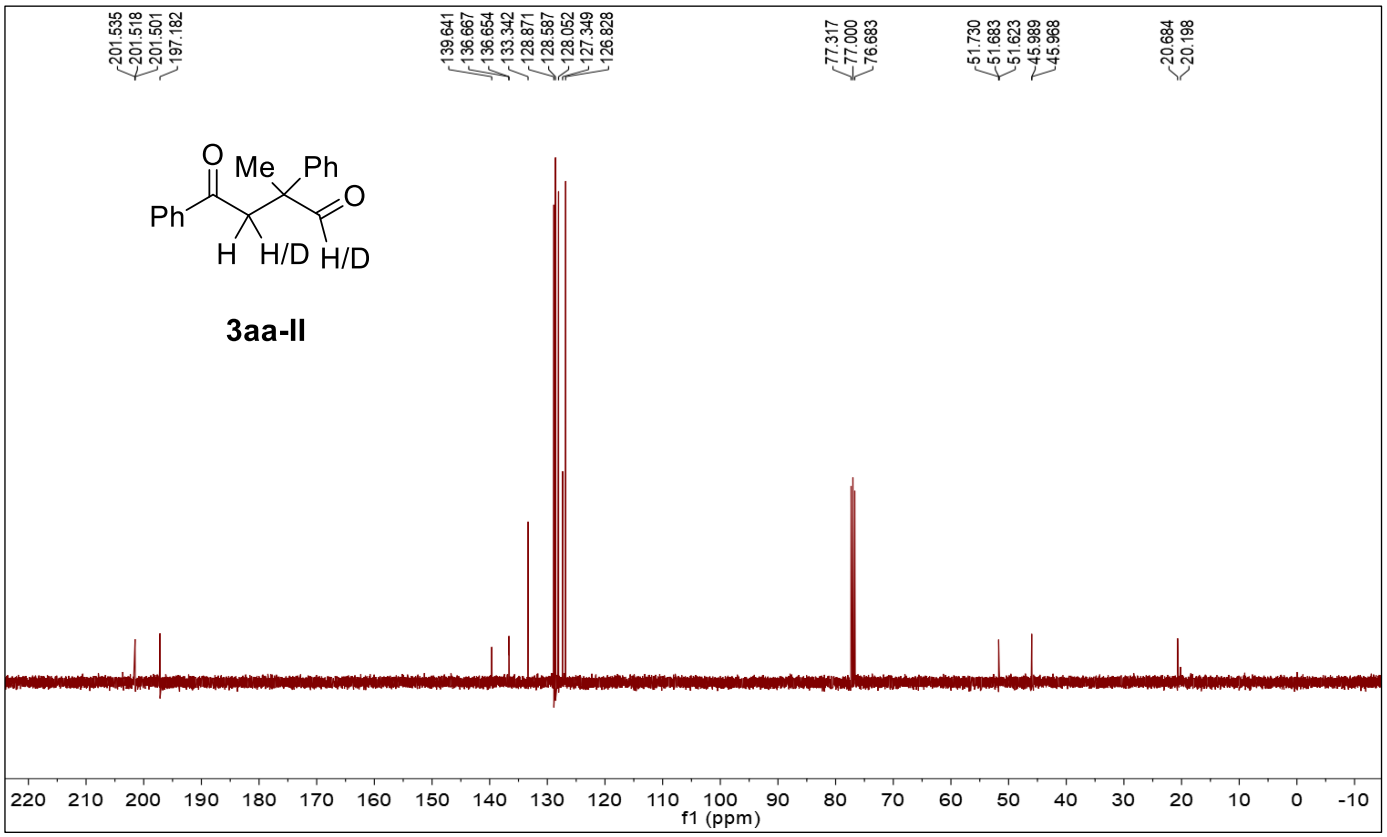

207.267
164.373
145.855
140.220
134.069
128.690
126.662
125.752
117.841
77.318
77.000
76.681
53.612
44.305
27.409
21.975
Me
O
Me
Ph
4za
f1 (ppm)

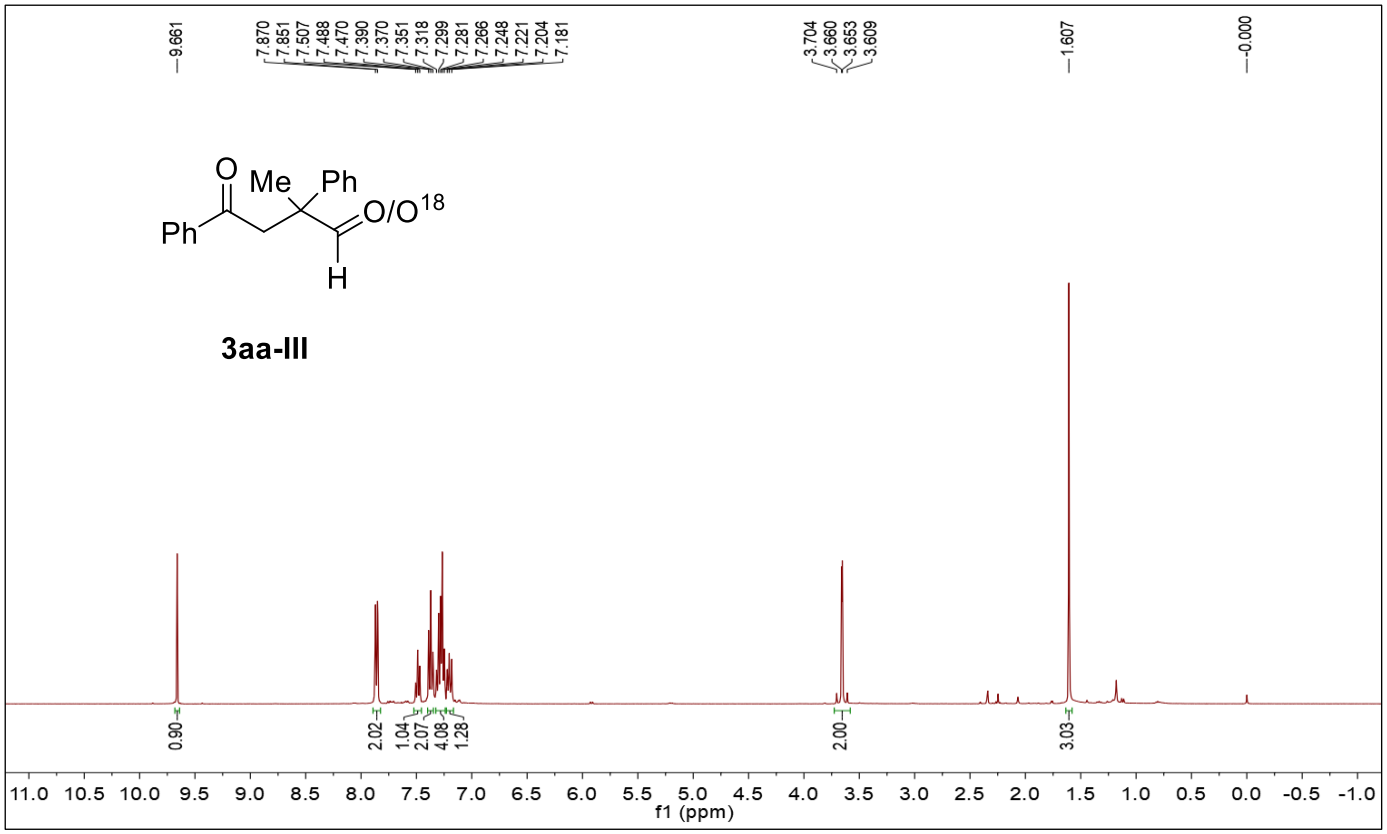
^{1}H and ^{13}C NMR spectra of mechanistic investigations

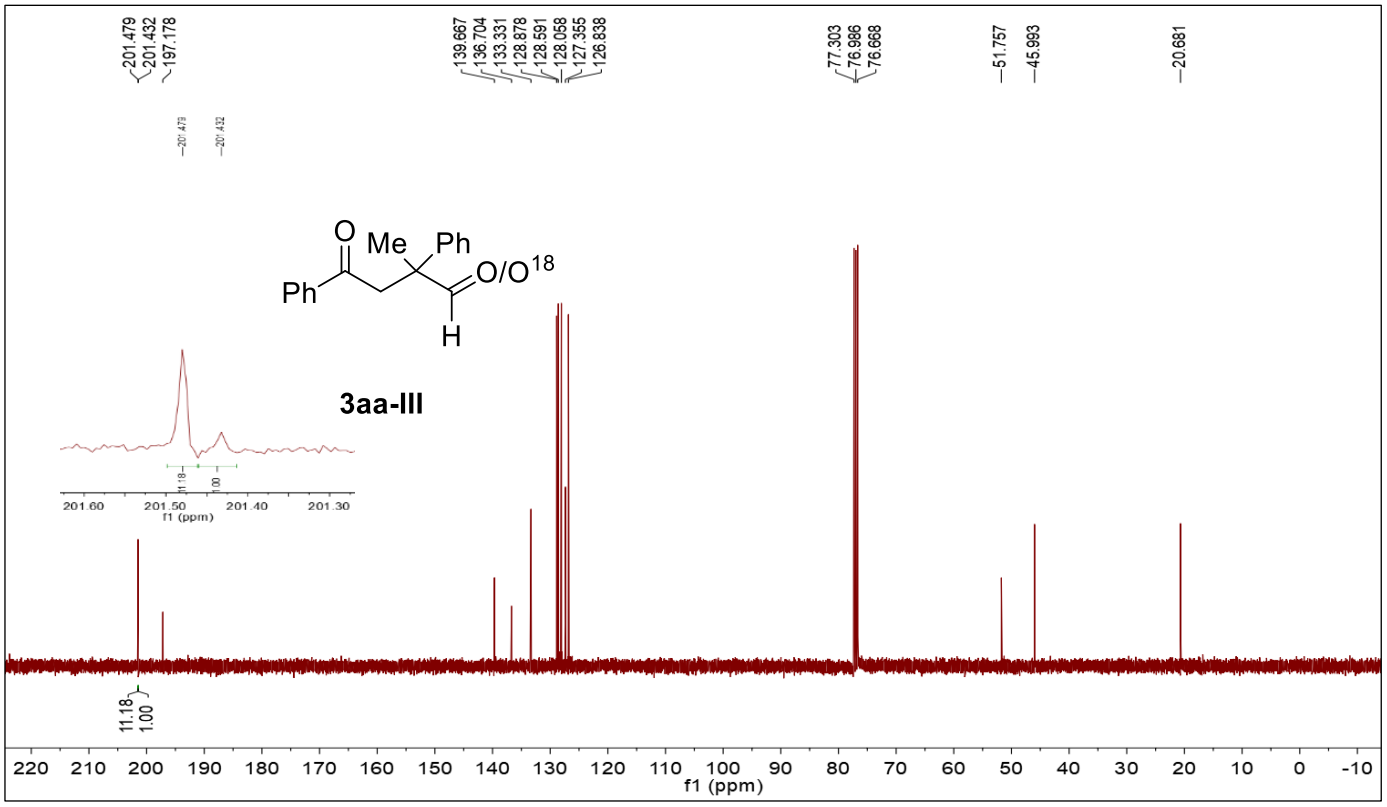





9.733
9.730
7.941
7.923
7.920
7.576
7.557
7.539
7.536
7.459
7.455
7.439
7.420
7.419
7.388
7.369
7.352
7.337
7.320
7.295
7.291
7.274
3.776
3.732
3.724
3.710
3.685
3.680
1.678
1.661
0.000
O
Me
Ph
Ph
O
H
H/D
H/D
3aa-II
0.82
2.00
1.03
2.08
4.02
1.04
1.51
2.39
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)


201.535
201.518
201.501
197.182
139.641
136.667
136.654
133.342
128.871
128.587
128.052
127.349
126.828
77.317
77.000
76.683
51.730
51.683
51.623
45.989
45.968
20.684
20.198
O
Me
Ph
Ph
O
H
H/D
H/D
3aa-II
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


9.661
7.870
7.851
7.507
7.488
7.470
7.390
7.370
7.351
7.318
7.299
7.281
7.266
7.248
7.221
7.204
7.181
3.704
3.660
3.653
3.609
1.607
0.000
O
Me
Ph
Ph
O/O18
H
3aa-III
0.90
2.02
1.04
2.07
4.08
1.28
2.00
3.03
f1 (ppm)


201.479
201.432
197.178
139.667
136.704
133.331
128.878
128.591
128.058
127.355
126.838
77.303
76.986
76.668
51.757
45.993
20.681
O
Me
Ph
Ph
O/O18
H
3aa-III
11.18
1.00
f1 (ppm)